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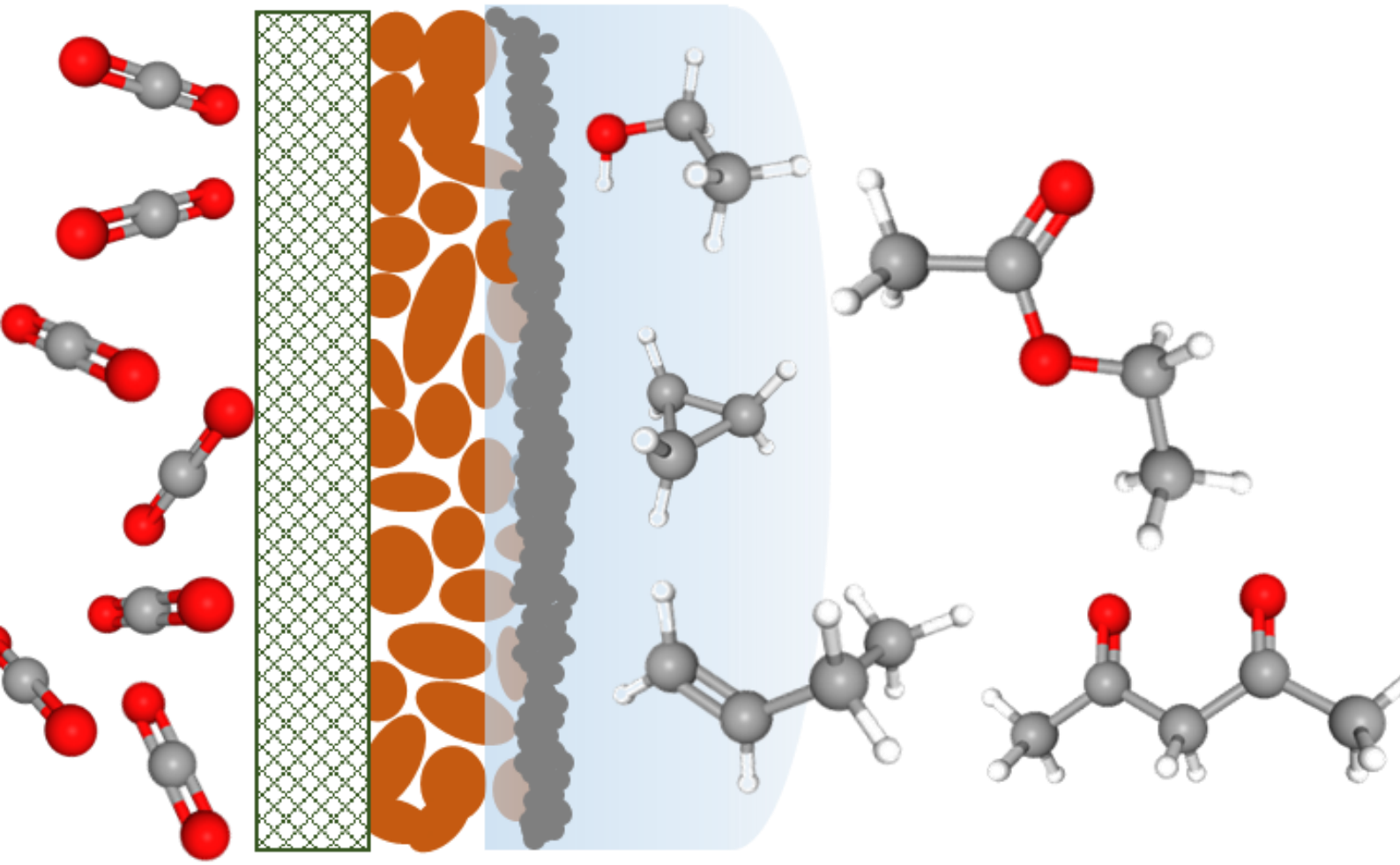
A COLLABORATION BETWEEN THE UNIVERSITY OF CAMBRIDGE,
NANYANG TECHNOLOGICAL UNIVERSITY & NATIONAL UNIVERSITY OF SINGAPORE



Biannual Research Report

April - September 2022

CAMBRIDGE
CARES

CAMBRIDGE CENTRE
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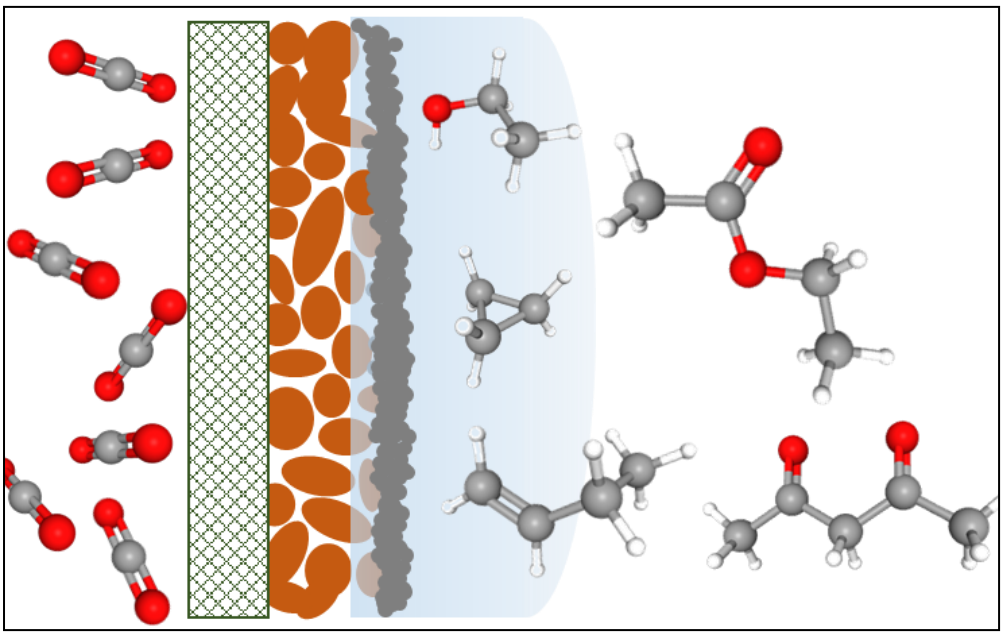

Illustration of the flow cell with gas-diffusion layer (box layer) and a copper catalyst (brown layer) converting carbon dioxide (left) to a variety of products (right).

Image by Mr Simon RIHM (PhD student, CARES, IRP 3). *See more on page 67.*

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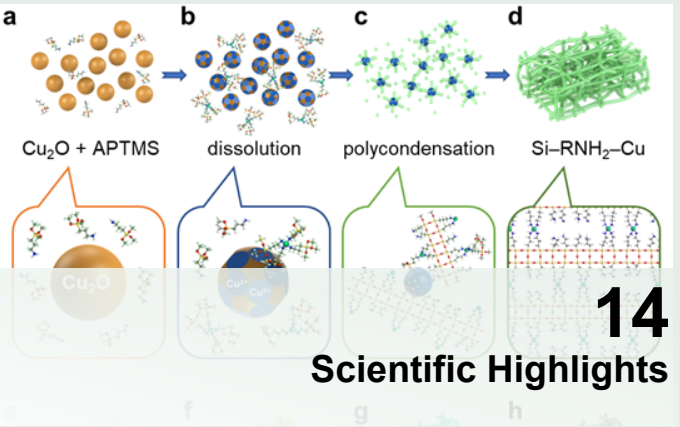
Focus on Fundamental Science



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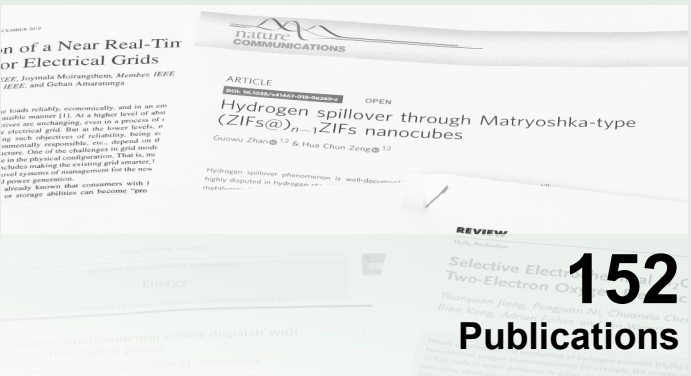

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FOREWORD

I am pleased to present the 17th Biannual Research Report for the Cambridge Centre for Advanced Research and Education in Singapore (CARES). The past six months have been much more in-person, as COVID-19 restrictions reduced in Singapore. As a result, we have enjoyed visits from many collaborators and friends, including Nobel Prize winner and Cambridge colleague Prof Didier Queloz.

New Project Opportunities

Our newest project, "Knowledge Graph Driven P2P Energy Trading" (supported by an NRF Intra-CREATE seed collaboration grant), will be led by Dr Casper Lindberg (a CARES Research Fellow who joined us as a PhD student) in collaboration with an NTU colleague, Asst Prof Hung Nguyen. I am delighted to see our students developing into independent researchers and the excellent opportunities they continue to find within our CREATE network. The project will aim to develop an energy market framework that leverages the benefits of a dynamic knowledge graph.

The World Avatar Shortlisted by WEF

The continuous efforts of the J-Park Simulator in building The World Avatar (TWA) dynamic knowledge graph have been recognised by the World Economic Forum, which has listed CARES and TWA on their list of Global Use Cases as part of its Global Digital Twin Cities initiative. Our project has not only been recognised for its current real-world uses in the CARES lab and for analysing district heating in the UK, but also for its potential to improve city governance and create a conducive urban ecosystem for industries and people.

CLIC Remote Guided Testing Publication

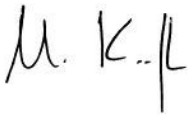
While many aspects of laboratory work had to be adapted in the last two years, the need for our CLIC Programme (Centre for Lifelong Learning and Individualised Cognition) to conduct human participants testing at the height of social distancing was a unique challenge. The team have used the opportunity to innovate a remote guided testing method in a published paper (Leong et al., 2022) that shows such testing can produce results consistent with the pre-pandemic standard of face-to-face testing. This adaptation not only allowed CLIC to continue data collection during the pandemic, but has also contributed new tools to the field.

Looking ahead

2023 will be a special year for CARES, as we will have been in operation for ten years! We are looking forward to a year of celebrations; sharing our scientific achievements and ambitions through a series of special events and communications. Please do sign up to our LinkedIn Page or our website for more details throughout the year.

I hope I have encouraged you to read more about CARES' latest work and achievements in this report. As ever, please do get in touch if you would like to know more about our work or have ideas for collaboration.

Professor Markus Kraft, CARES Director
September 2022



ABOUT US

Cambridge CARES is the University of Cambridge's presence in Singapore

The Cambridge Centre for Advanced Research and Education in Singapore (CARES) is a wholly-owned subsidiary of the University of Cambridge. Cambridge CARES is funded by the National Research Foundation as part of CREATE (Campus for Research Excellence and Technological Enterprise). We have a number of research collaborations between the University of Cambridge, Nanyang Technological University, the National University of Singapore and industrial partners.

The first programme administered by CARES is the Cambridge Centre for Carbon Reduction in Chemical Technology (C4T). The C4T programme is a world-leading partnership between Cambridge and Singapore, set up to tackle the environmentally relevant and complex problem of assessing and reducing the carbon footprint of the integrated petro-chemical plants on Singapore's Jurong Island. It brings together researchers from chemical engineering, biotechnology, chemistry, biochemistry, information engineering, electrical engineering, materials science and metallurgy.

The motivation for the C4T project is to integrate materials design and selection (i.e., for adsorbents and catalysts) with advances in process design to achieve improved selectivity and conversion. Such improvements will provide a reduced carbon footprint and energy demand for both established and new processes. Lowering the cost of CO_2 capture, and technologies and strategies for waste heat utilisation are also underlying drivers in the research. Our six collaborative Interdisciplinary Research Programmes (IRPs) combine state-of-the-art experimental analysis with advanced modelling research from Cambridge and Singapore. Whilst each IRP has clearly defined milestones and deliverables, denoted as work packages (WPs), there is significant interaction between the IRPs.

The first five-year research phase of C4T came to an end in October 2018. The programme received a further five years of funding for Phase 2, which commenced in November 2018.

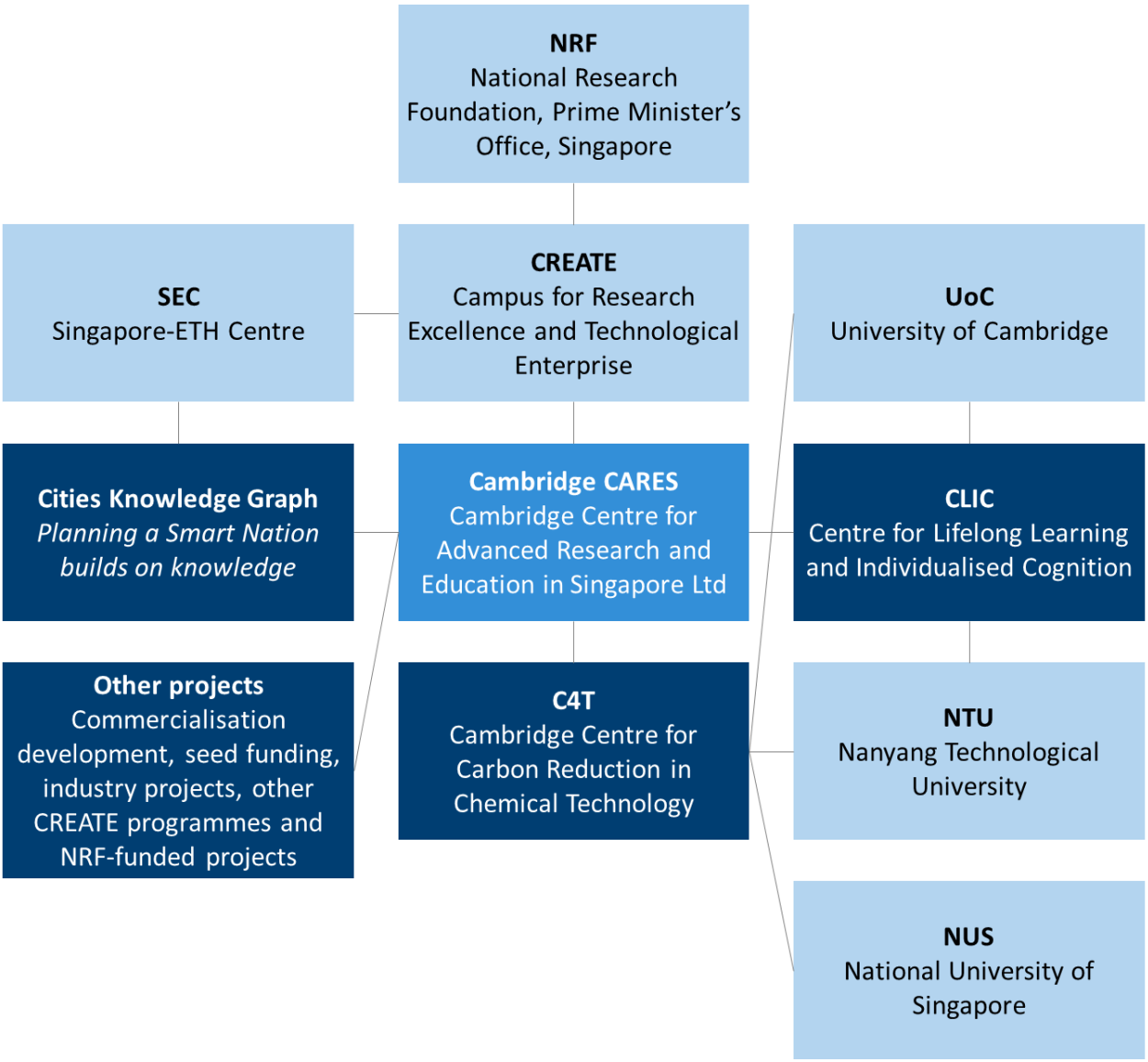
A second large CREATE-funded programme, the Centre for Lifelong Learning and Individualised Cognition (CLIC), began in October 2020. CLIC is a collaboration between University of Cambridge and NTU and focuses on the neuroscience of learning, a new research area for CARES.

In April 2020, CARES was awarded a further Intra-CREATE large grant for Cities Knowledge Graph, which brings together researchers from

University of Cambridge and ETH Zürich to harness rapidly growing and diversifying data streams to improve the planning and design of cities. Cities Knowledge Graph will do this by developing an innovative digital platform designed to combine data and share knowledge about cities, and to inject new precision and responsiveness to static instruments of planning, such as the city master-plan.

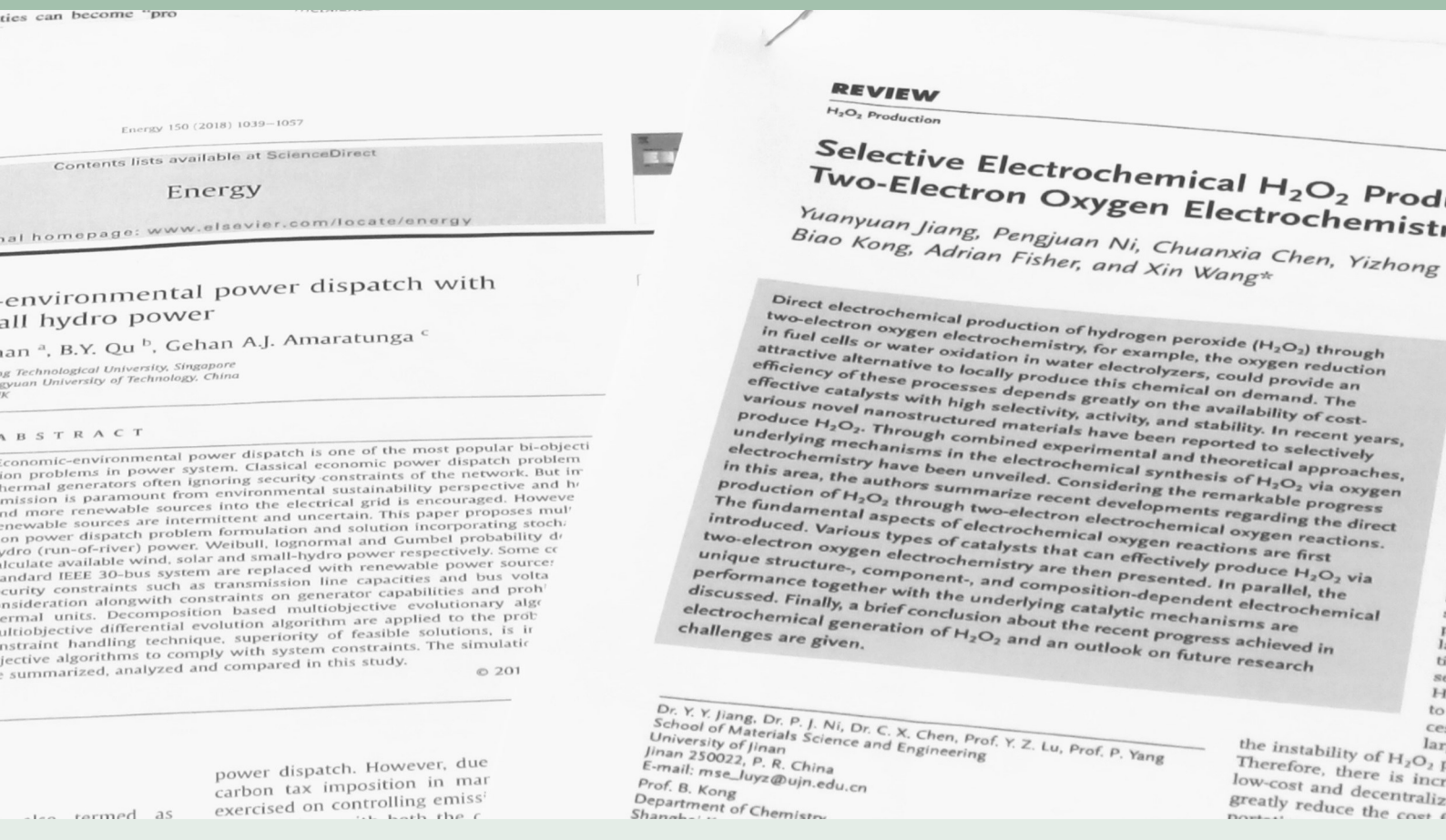
As well as this large Intra-CREATE grant, CARES is hosting AMPLE (An Accelerated Manufacturing Platform for Engineered Nanomaterials), funded by the Central Gap Fund. There are several smaller projects and spin-offs ongoing: The Intra-CREATE seed funded Knowledge Graph Driven P2P Energy Trading and Real-time Network Operation for High Renewables, and currently one ongoing stream under the Pharmaceutical Innovation Programme Singapore (PIPS) that involves industry funding. CARES also takes part in the Cooling Singapore 2.0 programme hosted by the Singapore-ETH Centre. Details and updates for these smaller projects can be found on page 135.

This report is a summary of our last half-year of research progress. It includes scientific updates from each of our researchers, along with abstracts and figures from our recent publications. There are also several articles that explain the fundamental science behind some of our work, and the impact this can have to our society.



HIGHLIGHTS

OUTSTANDING WORK FROM THE LAST SIX MONTHS OF CAMBRIDGE CARES RESEARCH



Flying into the Exhaust: Capturing In-plume Particle Characteristics

Dr Molly HAUGEN, CARES Visiting Scientist, IRP 4

Maritime emissions account for ~3% of global greenhouse gases and has led to NO_x and SO_x being directly regulated, and particle mass (PM) indirectly regulated via sulphur content within the fuel. Measuring exhaust PM has not yet been a standardised practice as collecting data on PM for in-use vessels has been a challenge within the maritime regulation community. However, here, we present an opportunity to directly measure the emitted PM in order to ensure this sector can continue towards a more sustainable future.

Capturing direct particle emissions from high particle emitting ferries at the Port of Rafina, Greece, would not have been possible without using an unmanned aerial vehicle (UAV, i.e., a drone). Our UAV acted as a tool to bring particle instruments closer to the emission source, allowing us to make measurements on particle characteristics that otherwise would not have been accessible. During the 10-day study, different particle characteristics such as particle number, lung-deposited surface area, and mass from in-use maritime emissions were measured. This was done, for the first time in literature, here in Rafina using a UAV with handheld sensors for maritime emission data collection. Comparing how these metrics change in relation to each other is key for determining particle coagulation,

A picture of Dr Haugen and the drone used in the study.

particle deposition into the water, or atmospheric suspension of particles. The UAV + handheld sensors provided novel insight into how the particles within plumes evolve as they age or as the plume moves away from the stack pipe of the ferry.

The designed study also acted as a source of validation for existing and new models, specifically for combustion sources. The data collected from the UAV were compared to an improved method of artificially emulating the plume dispersion while considering the chemical transformations within a plume. The new technique is called the Incompletely Stirred Reactor Network (ISRN) and incorporates the effects of simultaneous coagulation, mixing, and dilution downwind from the emission sources. Together, the data collected from the UAV with handheld sensors and the ISRN estimates can give researchers, policymakers, and related industrial contributors tools to monitor and explore maritime plume particle dispersion. The ISRN estimates can be used for an improved approximation of particle concentration levels for multiple metrics (particle number, lung deposited surface area, and black carbon) without requiring laborious field campaigns. This work can be used directly by the Port of Rafina to improve the air quality in the area, as well as by maritime researchers in general who are seeking new methods for measuring and building databases around in-use emissions of maritime traffic.

For more information: A paper related to this novel study is "Measurements and modelling of the three-dimensional near-field dispersion of particulate matter emitted from passenger ships in a port environment" (DOI: 10.1016/j.atmosenv.2022.119384) published in *Atmospheric Environment* by researchers from the University of Cambridge. The abstract can be found in our Research Highlights on page 20.

The drone has been developed with support from the NRF-funded CARES C4T programme and it had been originally planned to be deployed in Singapore for field work in 2020. Due to the COVID-19 pandemic, field work was initially done in Europe as described, it is however hoped that the Singapore field work plan will be able to recommence in 2023, subject to licenses.

Dr Molly Haugen is a Senior Research Associate in Emissions Measurement at the University of Cambridge. Her work with Prof Epaminondas MASTORAKOS (PI, CAM) focuses on plume dispersion in ports and urban environments. Her projects include:

1. Using drones to measure maritime emissions for use by political, academic, industrial, and regulatory industries
2. Developing an easy-to-use, low-cost particle counter (commercialisation and laboratory experiments)
3. Development of tyre and brake wear generation rigs (large-scale and bench -top versions)
4. Generating brake and tyres wear to characterise particle metrics and toxicological effects

Dr Haugen will be visiting CARES in April 2023.

Estimating Pollution from Ships using Computational Fluid Dynamics

Dr Ramesh KOLLURU, Research Fellow, IRP 4

Maritime transport is of paramount importance to international trade and the world economy. It is estimated that about 90,000 marine vessels are used to transport 80% of goods carried by sea. These vessels consume the same fossil fuels as normal road vehicles and emit up to 3% of global CO_2 emissions, among other harmful pollutants. As shipping traffic increases daily, we must ask the question: How can we assess pollutants emitted by a ship?

Common methods of assessment use experimental, analytical or numerical methods. Experimental methods require using several accurate measuring devices without interfering with the flow phenomena (water, wind, turbulence, etc.) to conduct a realistic experiment. This can sometimes be difficult or nearly impossible. In such circumstances, analytical or numerical methods are better suited to gain knowledge on the physical phenomena. Numerical simulations, which are Computational Fluid Dynamics (CFD) in this case, are used to recreate physical phenomena on a computer by solving the governing equations using numerical methods. A normal desktop computer can do the job, but it can take months to obtain basic information. In such cases, supercomputers are needed to achieve more realistic timeframes, which still range from days to a week. The decision to use a desktop or a supercomputer depends on the accuracy with which the information is sought.

Process analysis on a computer involves the following steps:

A) Creating an area - While ships sail in open oceans, we do not have the ability to model an entire area. Therefore, we select a region of interest and analyse the phenomenon in question in that region. In this case, we analyse the development of pollutants and their dispersion by a ship.

B) Creation of a grid: In the selected region, a certain number of points are chosen at which the basic equations are solved. Due to current software and hardware limitations, it is not possible to select an infinite number of points. A typical grid on a ship is shown in Figure 1.

C) Solver - One or more suitable equations are selected to be solved on a computer using an

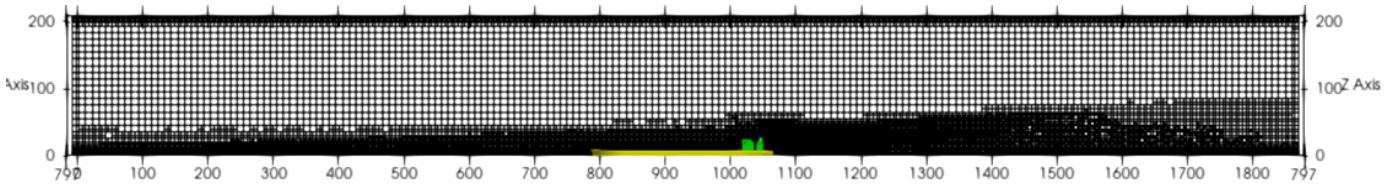


Figure 1: Adaptive grid generated on the ship (yellow and green blocks) at runtime. The spatial coordinates shown are in metres.

approximation method. CFD generally uses the finite volume method, the finite difference method, or the finite element method, depending on the complexity of the underlying physics.

D) Data analysis: After creating the grid, data is obtained when the governing equations are solved at each point. The data obtained can be represented as streamlines, line plots, and contour plots (Figure 2).

The content described is only the tip of the iceberg in the numerical simulations we call Computational Fluid Dynamics.

More information on Dr Kolluru's research can be found on page 72 of the report.

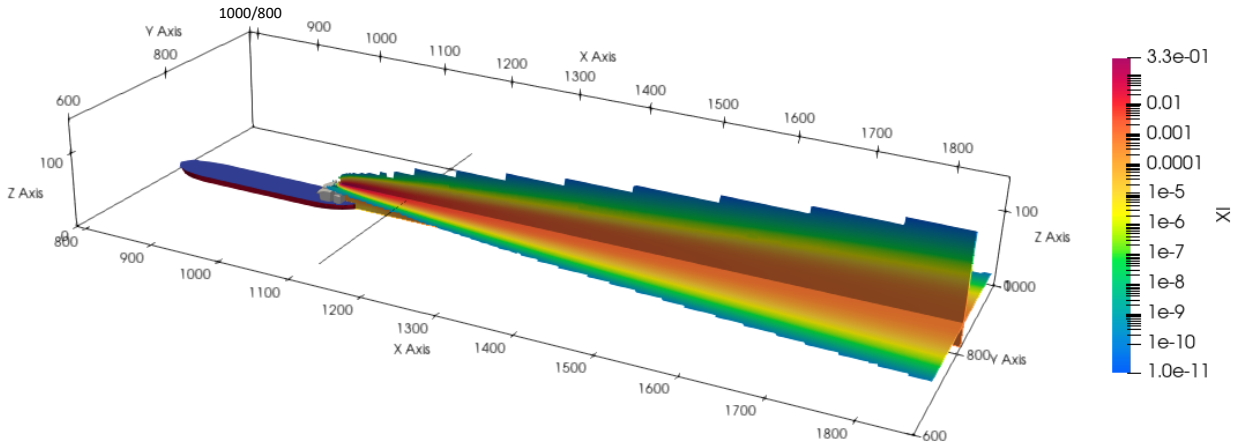


Figure 2: Cut section view of the plume evolution. The spatial coordinates are in metres and the variable Xi represents concentration of pollutants.

Dr Ramesh Kolluru obtained his PhD from the Aerospace Engineering Department at the Indian Institute of Science. He was a Postdoctoral Fellow at the department working on developing hybrid optimisers for Dassault Aviation France, using gradient-based algorithms and non-gradient-based algorithms. He also worked as an Assistant Professor at the Department of Mechanical Engineering, B.M.S. College of Engineering Bangalore from 2011 to 2019. His primary interests are in algorithm development of high-speed compressible flows and optimisation. His work at CARES focuses on numerically investigating turbulent flows related to dispersion and mixing of pollutants in the atmosphere arising from marine traffic around Singapore. He will also develop CFD models for the mixing processes in the development of reacting flows during the transformation of pollutants.

Going With the Flow in Pharmaceutical Manufacturing

Dr Dogancan KARAN, Research Fellow, PIPS

The synthesis of drug molecules involves complex multi-step processes such as chemical reactions, purifications, workups, and more. Traditionally, pharmaceutical companies employ batch processing where the product of one batch reaction is isolated and purified and used in the next batch reaction. This process is not very different than cooking food in a large pot.

Although pharmaceutical companies are comfortable with using batch processing, it comes with various interlinked problems such as heat and mass transport limitations, safety, and slow data generation for reaction optimisation. Therefore, reaction conditions are optimised by chemist intuition or one-variable-at-a-time optimisation protocols since it is extremely time consuming to test all the possible combinations of the process parameters. However, this situation generally fails to explain complex interactions between reaction parameters leading to inefficient optimisation. As a result, a combination of out of date manufacturing practices and optimisation methods increase the development time and cost of the drug molecules significantly. Therefore, there is a paradigm shift in pharmaceutical industry on continuous processing, digitalisation, process intensification, and sustainability.

Continuous processing offers various advantages over its batch counterpart – expanded operation space, enhanced safety, facile integration of process analytical technologies (PAT), and ease of

automation. The advantages offered by continuous processing open new and more sustainable process routes which are not possible to achieve in batch processing. Furthermore, with high-quality and reliable data generation offered by continuous processing, it is much easier to optimise process parameters with various methods such as first-principle modelling, mathematically-guided optimisation, or black-box optimisation.

Given the relative infancy of continuous processing in the pharmaceutical industry, there is no well-established process development strategy which is used throughout the industry. In this PIPS-Pfizer Project, we aim to fill this gap by developing continuous flow technologies and digital tools to support R&D and manufacturing activities. The main objective of the project is to demonstrate how to develop an end-to-end continuous flow process for industrially relevant drug molecules. The drug molecules chosen for the project covers a great variety of technical challenges to develop innovative solutions. First, we develop the continuous flow reactors based on the requirements of the process. The flow reactors developed allow us to obtain reliable data which we then use to optimise the process parameters with various machine learning algorithms and mathematical models. The protocols generated in this project will be used by Pfizer as common practice to support their future development of continuous flow reactors and digital tools.

More information on the PIPS-Pfizer project can be found on page 140 of the report.

Dr Dogancan Karan obtained his PhD from National University of Singapore (NUS) at the Department of Chemical and Biomolecular Engineering in 2019. He has extensive research experience in microfluidics and microreactor technology, chemical reaction engineering, multiphase systems, computational fluid dynamics (CFD) as well as organic synthesis. Prior to joining CARES, he worked on various PIPS projects to develop flow chemistry platforms for photocatalytic applications and design fluidic components of a Mid-IR sensor. Currently, he is working with Pfizer to develop novel flow chemistry platforms by using machine learning.

Highlighted research outputs from April - September 2022

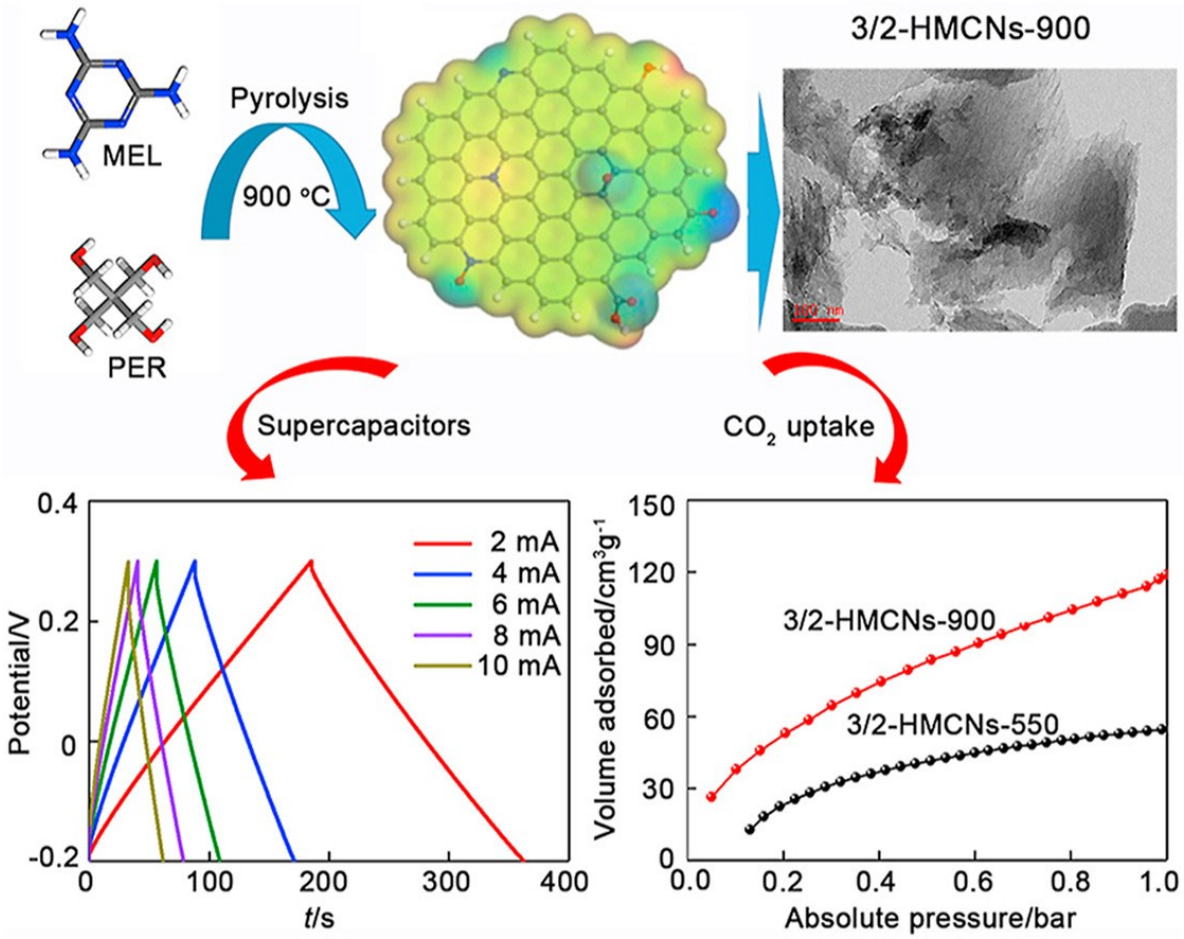
A selection of publications from across our programmes.

C4T IRP 1: Heteroatom-doped microporous carbon nanosheets derived from pentaerythritol-melamine for supercapacitors and CO_2 capture

Xiaochun Hu, Yuqing Luo, Xianyue Wu, Jiabin Niu, Mingwu Tan, Zhiqiang Sun, and Wen Liu, *Materials Today Energy*

DOI: 10.1016/j.mtener.2022.101010

Abstract: Heteroatom-doped microporous carbon nanosheets (HMCNs) are used in a wide range of applications, including gas adsorption, energy storage, and catalysis. Here, we demonstrate a solvent-free, template-free, one-pot polycondensation approach for the synthesis of HMCNs using melamine (MEL) and pentaerythritol (PER) as precursors. By varying the ratio of MEL to PER and the pyrolysis temperature, the doping amount, surface area and porosity of the HMCNs can be controlled. When used for CO_2 capture, the HMCN synthesized by the pyrolysis of a 3:2 mixture of MEL and PER at 900 °C (3/2-HMCNs-900) affords a CO_2 uptake of 5.35 mmol g^{-1} at 273 K and 1 bar CO_2 partial pressure. Density functional theory calculations suggest that the high CO_2 uptake performance of the HMCNs is associated with the chemical modification of the surface, as a result of N- and O- co-doping. When assembled in a supercapacitor, 3/2 -HMCNs-900 exhibits a high specific capacitance (475 F g^{-1} at 1.3 A) and a fast charge-discharge rate of 13.3 F s^{-1} g^{-1}. This study presents a novel, resource-efficient and environmentally friendly method for preparing HMCNs for energy and environmental applications.

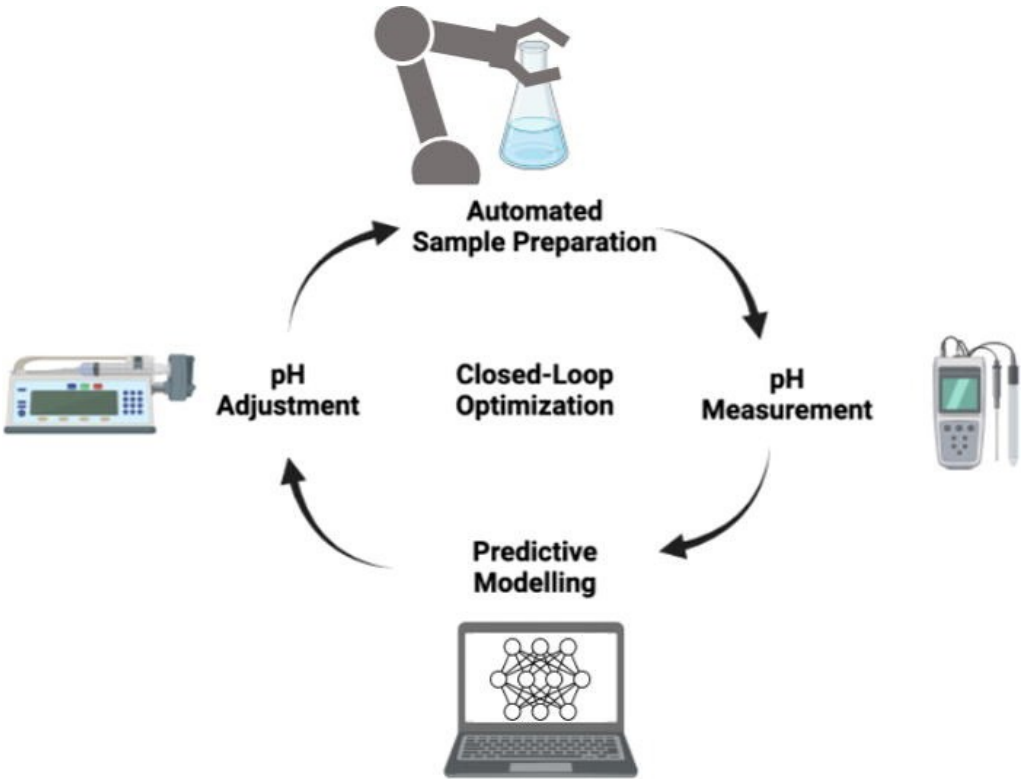


C4T IRP 1: Automated pH Adjustment Driven by Robotic Workflows and Active Machine Learning

Alexander Pomberger, Nicholas Jose, David Walz, Jens Meissner, Christian Holtze, Matthaeus Kopczynski, Philipp Müller-Bischof , and Alexei Lapkin, *Chemical Engineering Journal*

DOI: 10.1016/j.cej.2022.139099

Abstract: Buffer solutions have tremendous importance in biological systems and in formulated products. Whilst the pH response upon acid/base addition to a mixture containing a single buffer can be described by the Henderson-Hasselbalch equation, modelling the pH response for multi-buffered poly-protic systems after acid/base addition, a common task in all chemical laboratories and many industrial plants, is a challenge. Combining predictive modelling and experimental pH adjustment, we present an active machine learning (ML)-driven closed-loop optimization strategy for automating small scale batch pH adjustment relevant for complex samples (e.g., formulated products in the chemical industry). Several ML models were compared on a generated dataset of binary-buffered poly-protic systems and it was found that Gaussian processes (GP) served as the best performing models. Moreover, the implementation of transfer learning into the optimization protocol proved to be a successful strategy in making the process even more efficient. Finally, practical usability of the developed algorithm was demonstrated experimentally with a liquid handling robot where the pH of different buffered systems was adjusted, offering a versatile and efficient strategy for a pH adjustment processes.

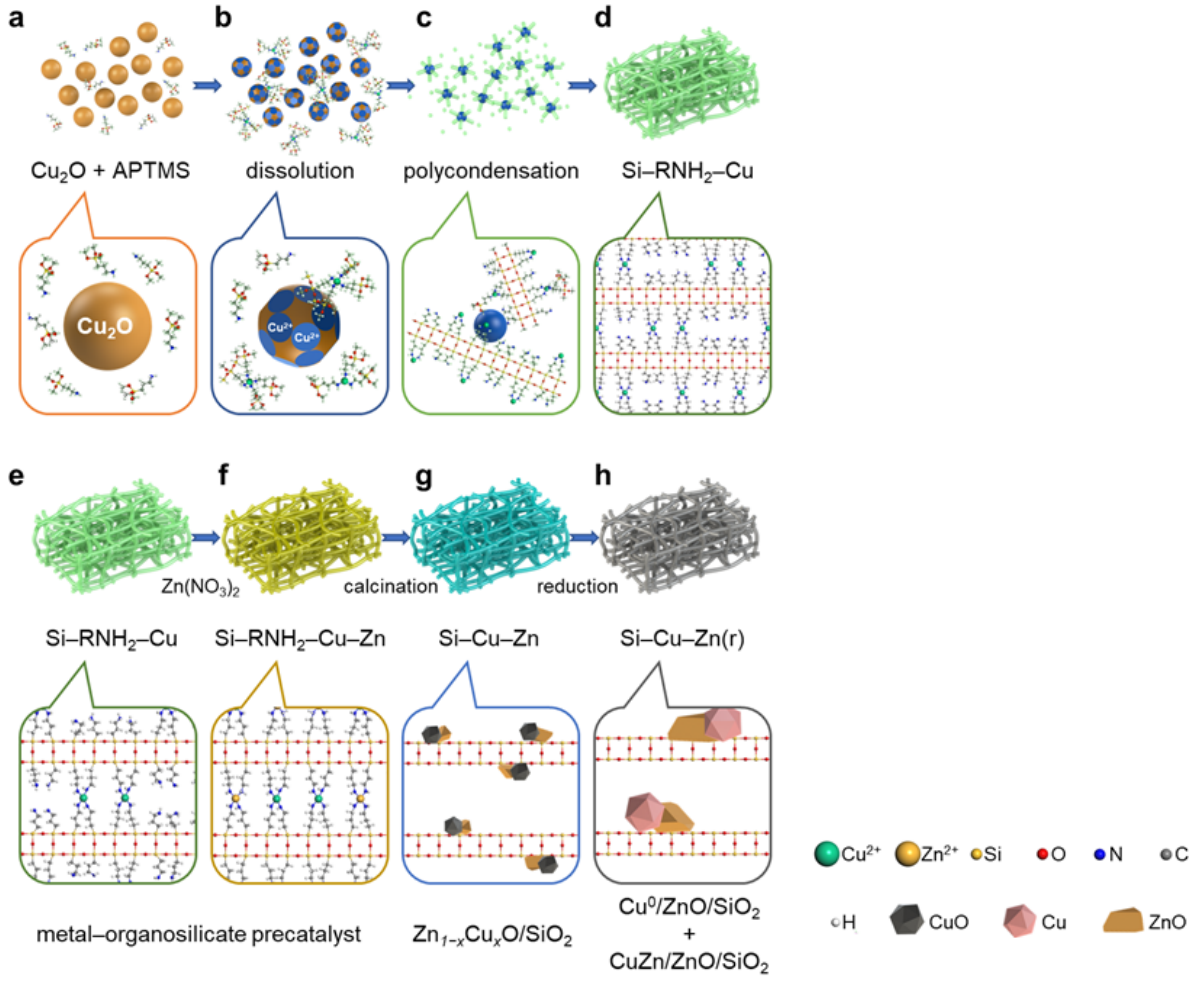


C4T IRP 1: Single Solid Precursor-Derived Three-Dimensional Nanowire Networks of CuZn-Silicate for CO_2 Hydrogenation to Methanol

Yu Shao, Mohammadreza Kosari, Shibo Xi, and Hua Chun Zeng, *ACS Catalysis*

DOI: 10.1021/acscatal.2c00726

Abstract: Hydrogenation of CO_2 to MeOH is one of the most promising technologies in mitigating the emissions of CO_2 and tackling the challenge of climate change. In this work, we present a synthetic protocol for preparing a Cu–ZnO-based heterogeneous catalyst supported by siliceous nanowire networks from a single solid precursor with a tunable composition. The resulting Si–Cu–Zn catalysts were evaluated with the MeOH synthesis from the CO_2 hydrogenation reaction operated at moderate conditions (30 barg and 200–280 °C). A specific MeOH yield of 402 $mg_{MeOH} \cdot g_{Cu}^{-1} \cdot h^{-1}$ and a MeOH selectivity of 51% were obtained at 240 °C. Such a performance was attributed to several structural and compositional merits, granted through the attentively engineered synthetic procedures. Small Cu nanoparticle (NP) size was achieved and maintained by the high dispersion of Cu to the atomic level in the precatalyst and the incorporation of ZnO as a structural promoter. Moreover, the desirable Cu–ZnO synergistic effect can be further attained from the strong metal–support interaction (SMSI) between the Cu NPs and the partially reduced ZnO phase. Lastly, the robust siliceous nanowire networks provided decent spatial confinement to contain the growth of Cu NPs while offering high accessibility with the macroscopic porous morphology. The catalyst exhibited stable performance over a week's long stability test while keeping its structural integrity intact. Overall, this study may offer an alternative design and synthesis strategy for the well-received Cu–ZnO system to approach its high performance in CO_2 hydrogenation.

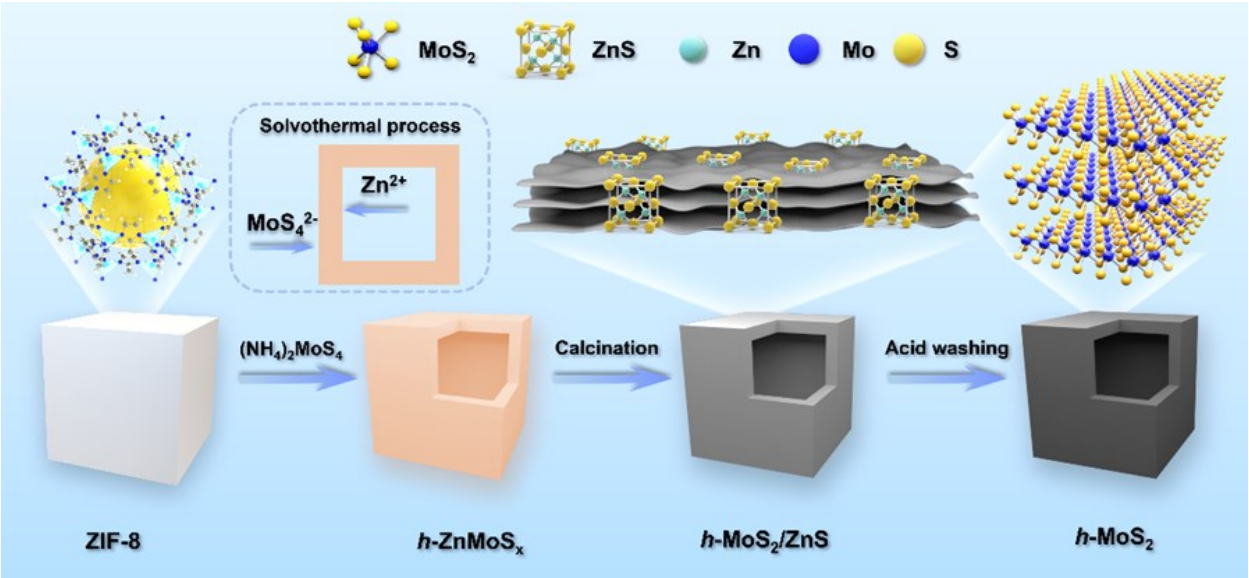


C4T IRP 1: Boxlike Assemblages of Few-Layer MoS_2 Nanosheets with Edge Blockage for High-Efficiency Hydrogenation of CO_2 to Methanol

Shenghui Zhou and Hua Chun Zeng, *ACS Catalysis*

DOI: 10.1021/acscatal.2c02838

Abstract:: Direct hydrogenation of CO_2 into methanol is a promising strategy for reducing excessive dependence on fossil fuels and alleviating environmental concerns. Recently, in-plane sulfur vacancies in two-dimensional MoS_2 nanosheets were unveiled as efficient catalytic active sites for methanol synthesis from CO_2, whereas edge vacancies facilitated hydrogenation of CO_2 to methane. Herein, we developed boxlike assemblages of quasi-single-layer MoS_2 nanosheets, which were edge-blocked by ZnS crystallites (denoted as *h*-MoS_2/ZnS) via a metal–organic framework (MOF)-engaged solvothermal route and subsequent heat treatments. The spatial confinement of the ZnS can restrain the growth and aggregation of MoS_2 and ensure the stability of few-layer or even single-layer MoS_2 in the assemblages. More importantly, the presence of ZnS can prevent reactants from approaching the edge sulfur vacancies of MoS_2. With more exposed in-plane sulfur vacancies and less edge sulfur vacancies, the *h*-MoS_2/ZnS exhibits 67.3% methanol selectively, 9.0% CO_2 conversion, and a high methanol space-time yield of up to 0.93 $g_{MeOH} \cdot g_{MoS2}^{-1} \cdot h^{-1}$ at 260 °C, 5 MPa, and 15 000 mL $\cdot g_{cat.}^{-1} \cdot h^{-1}$. The catalytic activity was stable for at least 120 h. By removing the ZnS phase from *h*-MoS_2/ZnS and thus deliberately creating more edge sulfur vacancies, it was further confirmed that edge sulfur vacancies are active catalytic sites for excessive hydrogenation of CO_2 to methane. Furthermore, the reaction mechanism of our catalyst was also investigated by a high-pressure *in situ* DRIFTS study. Thus, this MOF-templated strategy for assembling and confining quasi-single-layer MoS_2 provides insights into the development of highly efficient transition-metal dichalcogenide catalysts for CO_2 hydrogenation with excellent stability.



C4T IRP 2: Enhanced oxygen evolution over dual corner-shared cobalt tetrahedra

Yubo Chen, Joon Kyo Seo, Yuanmiao Sun, Thomas A. Wynn, Marco Olguin, Minghao Zhang, Jingxian Wang, Shibo Xi, Yonghua Du, Kaidi Yuan, Wei Chen, Adrian C. Fisher, Maoyu Wang, Zhenxing Feng, Jose Gracia, Li Huang, Shixuan Du, Hong-Jun Gao, Ying Shirley Meng and Zhichuan J. Xu, *Nature Communications*

DOI: 10.1038/s41467-022-33000-w

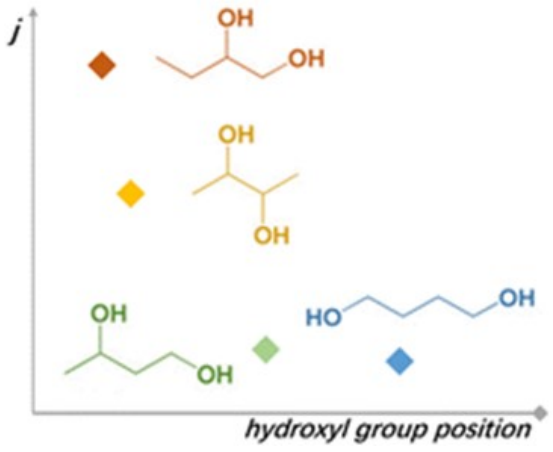
Abstract: Developing efficient catalysts is of paramount importance to oxygen evolution, a sluggish anodic reaction that provides essential electrons and protons for various electrochemical processes, such as hydrogen generation. Here, we report that the oxygen evolution reaction (OER) can be efficiently catalyzed by cobalt tetrahedra, which are stabilized over the surface of a Swedenborgite-type $YBCo_4O_7$ material. We reveal that the surface of $YBaCo_4O_7$ possesses strong resilience towards structural amorphization during OER, which originates from its distinctive structural evolution toward electrochemical oxidation. The bulk of $YBaCo_4O_7$ composes of corner-sharing only CoO_4 tetrahedra, which can flexibly alter their positions to accommodate the insertion of interstitial oxygen ions and mediate the stress during the electrochemical oxidation. The density functional theory calculations demonstrate that the OER is efficiently catalyzed by a binuclear active site of dual corner-shared cobalt tetrahedra, which have a coordination number switching between 3 and 4 during the reaction. We expect that the reported active structural motif of dual corner-shared cobalt tetrahedra in this study could enable further development of compounds for catalyzing the OER.

C4T IRP 2: The effect of the hydroxyl group position on the electrochemical reactivity and product selectivity of butanediol electro-oxidation

Shengnan Sun, Chencheng Dai, Libo Sun, Zhi Wei Seh, Yuanmiao Sun, Adrian Fisher, Xin Wang, and Zhichuan J. Xu, *Dalton Transaction*

DOI: 10.1039/D2DT02450K

Abstract: This article presents a study on the effect of the hydroxyl group position on the electro-oxidation of butanediols, including 1,2-butanediol, 2,3-butanediol, 1,3-butanediol, and 1,4-butanediol. The effect of the hydroxyl group position in butanediols on their electro-oxidation reactivities is investigated by cyclic voltammetry, linear sweep voltammetry, chronopotentiometry and chronoamperometry in 1.0 M KOH. The results show that the closer the two hydroxyl groups are, the higher the reactivity, and the lower the anodic potential butanediol has. Moreover, the oxidation products from chronoamperometry are analyzed by means of HPLC and NMR. Some value-added products, such as 3-hydroxypropionic acid/3-hydroxypropionate, are produced. The DFT calculation indicates that the oxidation of vicinal diols responds to the conversion from a hydroxyl group to a carboxylate group, followed by C–C bond cleavage, where the carbon charge decreases. These results provide an insight into reactant selection for the electrochemical synthesis of value-added chemicals.

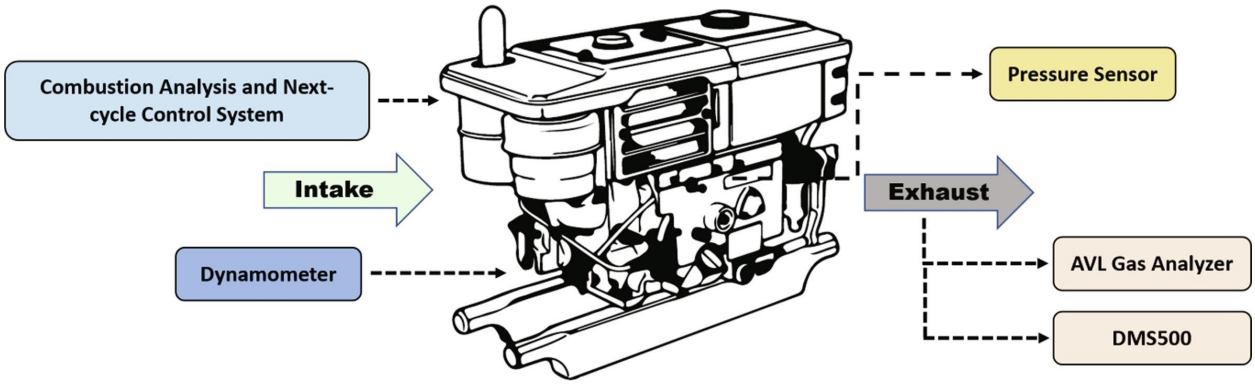


C4T IRP 3: Understanding the particulate formation process in the engine fuelled with diesel/Jet A-1 blends

Qiren Zhu, Yichen Zong, Wenbin Yu, Wenbin Yu, Wenming Yang, and Markus Kraft, *Fuel*

DOI: 10.1016/j.fuel.2021.122675

Abstract: Jet fuel has been recognized as a potential alternative for traditional diesel engines because of its ability to reduce particulate matter (PM) emissions while retaining engine power output. In this study, the particulate formation process has been studied in detail using diesel/Jet A-1 blends with evenly staggered ratios. The number concentration of the accumulation mode particle decreases exponentially when additional Jet A-1 is introduced to the blends under 30% engine load, as more fuel and particle precursors are oxidized. Additionally, the comparison of PM emissions with pilot-main and single main two injection strategies is conducted to better understand the particle formation process. The phenomenon of 'particle saturation' of nucleation mode particles is observed using the pilot-main injection strategy. With these supporting findings, we strengthen the point that the pilot-injection strategy has the potential weaken the oxidation process during the combustion process. Furthermore, this research quantifies the impact of Jet A-1 on combustion and gas emission characteristics by extracting the change rate from the data. In general, Jet A-1 tends to delay the ignition and shorten the combustion duration. The results also reveal that the rise in NO_x emissions is due to a higher proportion of premixed combustion, while the increase in HC emissions is attributed to a longer ignition delay and shorter combustion time.

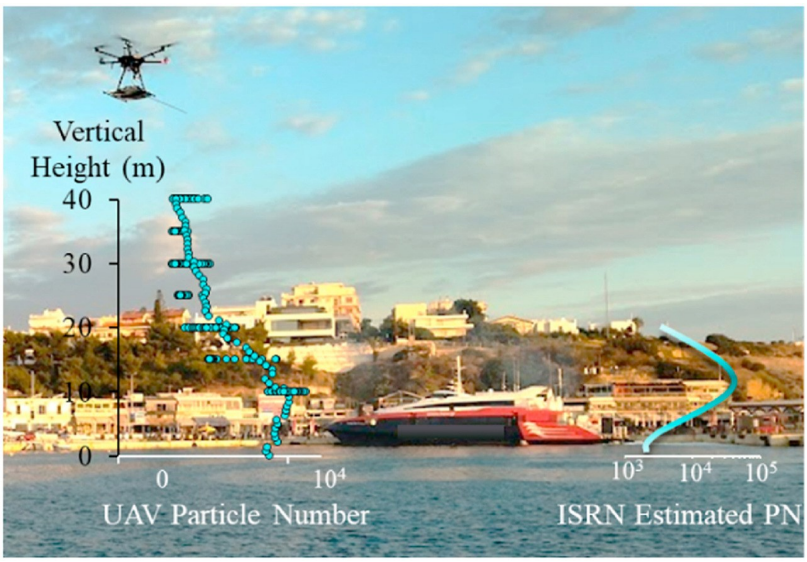


C4T IRP 4: Measurements and modelling of the three-dimensional near-field dispersion of particulate matter emitted from passenger ships in a port environment

Molly J. Haugen, Savvas Gkantonas, Ingrid El Helou, Rohit Pathania, Epaminondas Mastorakos, and Adam M.Boies, *Atmospheric Environment*

DOI: 10.1016/j.atmosenv.2022.119384

Abstract: The maritime sector poses significant challenges in controlling the emission of harmful pollutants, such as particulate matter, and reducing their impact on coastal areas and the atmospheric environment. Efforts to regulate the sector necessitate new knowledge and methods to characterise the evolution and physicochemical transformation of maritime particle emissions dispersing in port areas. An experimental campaign at the Port of Rafina, Greece, was conducted with this in mind. In this paper, we report on the first multi-characteristic particle measurements, including particle number (PN), lung deposited surface area (LDSA), and black carbon (BC), performed at both land and sea using a novel instrument set-up mounted on an unmanned aerial vehicle (UAV). Land-based measurements showed that LDSA averages, which are influenced by particle number and size, increase up to 20 times above background levels as an emission plume progresses downwind, whereas BC concentrations, which are dominated by mass, are ~12 times higher than the background concentration. Ground and UAV-based particle comparisons showed that PN and LDSA measurements exhibit greater differences than BC relative to the plume's location. Ground-based sensors had ~50% lower LDSA and PN concentrations, whereas BC was about equal. The experimental observations are further substantiated by coupling a Gaussian plume dispersion model and a new computationally attractive approach, known as the Incompletely Stirred Reactor Network (ISRN) method, to predict the three-dimensional evolution of particle characteristics considering the effects of dilution, segregation, and physicochemical transformations, such as coagulation. Based on simplifying assumptions for the particle size distribution sources within the port area, the ISRN estimates suggest that the discrepancy between various metrics can be partly explained by coagulation, responsible for a non-linear increase in particle size, depending on the local level of dilution and mixing intensity, leading up to a ~25% decrease in PN and LDSA compared to BC. Combined, measurements and modelling highlight the effect of the sampling location and the importance of monitoring more than one particle metric to characterise particle evolution.

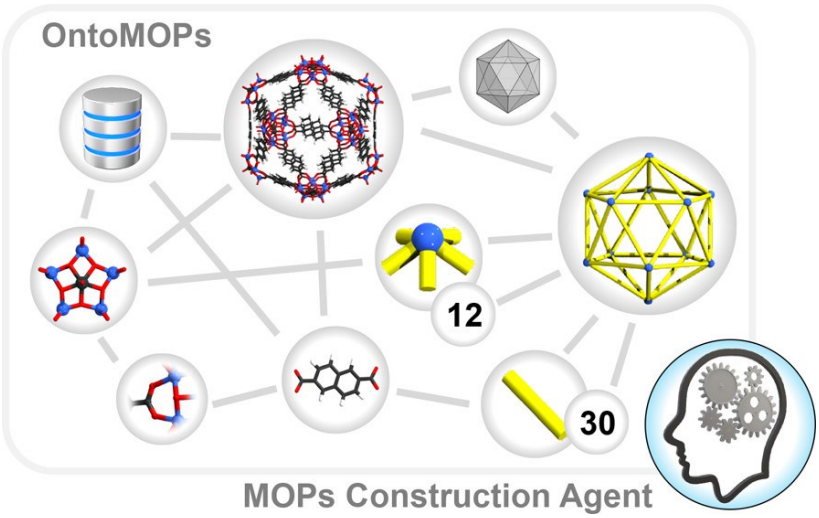


C4T IRP JPS: Automated Rational Design of Metal–Organic Polyhedra

Aleksandar Kondinski, Angiras Menon, Daniel Nurkowski, Feroz Farazi, Sebastian Mosbach, Jethro Akroyd, and Markus Kraft, *Journal of American Chemical Society*

DOI: 10.1021/jacs.2c03402

Abstract: Metal–organic polyhedra (MOPs) are hybrid organic–inorganic nanomolecules, whose rational design depends on harmonious consideration of chemical complementarity and spatial compatibility between two or more types of chemical building units (CBUs). In this work, we apply knowledge engineering technology to automate the derivation of MOP formulations based on existing knowledge. For this purpose we have (i) curated relevant MOP and CBU data; (ii) developed an assembly model concept that embeds rules in the MOP construction; (iii) developed an OntoMOPs ontology that defines MOPs and their key properties; (iv) input agents that populate The World Avatar (TWA) knowledge graph; and (v) input agents that, using information from TWA, derive a list of new constructible MOPs. Our result provides rapid and automated instantiation of MOPs in TWA and unveils the immediate chemical space of known MOPs, thus shedding light on new MOP targets for future investigations.



C4T IRP JPS: Blockchain Technology in the Chemical Industry

Xiaochi Zhou and Markus Kraft, *Annual Review of Chemical and Biomolecular Engineering*

DOI: 10.1146/annurev-chembioeng-092120-022935

Abstract: This article presents a review of the application of blockchain and blockchain-based smart contracts in the chemical and related industries. We introduce the basic concepts of blockchain and smart contracts and explain how some of their features are enabled. We review several typical or novel applications of blockchain and smart contract technologies and their enabling concepts and underlying technologies. We classify the selected literature into five categories and discuss their motivations and technical designs. We recognize that the trend of decentralization creates a need to use blockchain and smart contracts to implement trust and distributed control mechanisms. We also speculate on future applications of blockchain and smart contracts. We believe that, in the future, blockchains with different consensus mechanisms will be studied and applied to achieve more efficient and practical decentralized systems. Also, blockchain-based smart contracts will be more widely applied to enhance autonomous distributed controls in decentralized systems.

C4T IRP JPS: Universal Digital Twin: Integration of national-scale energy systems and climate data
Thomas Savage, Jethro Akroyd, Sebastian Mosbach, Nenad Krdzavac, Michael Hillman, and Markus Kraft, *Data-Centric Engineering*

DOI: 10.1017/dce.2022.22

Abstract: This article applies a knowledge graph-based approach to unify multiple heterogeneous domains inherent in climate and energy supply research. Existing approaches that rely on bespoke models with spreadsheet-type inputs are noninterpretable, static and make it difficult to combine existing domain specific models. The difficulties inherent to this approach become increasingly prevalent as energy supply models gain complexity while society pursues a net-zero future. In this work, we develop new ontologies to extend the World Avatar knowledge graph to represent gas grids, gas consumption statistics, and climate data. Using a combination of the new and existing ontologies we construct a Universal Digital Twin that integrates data describing the systems of interest and specifies respective links between domains. We represent the UK gas transmission system, and HadUK-Grid climate data set as linked data for the first time, formally associating the data with the statistical output areas used to report governmental administrative data throughout the UK. We demonstrate how computational agents contained within the World Avatar can operate on the knowledge graph, incorporating live feeds of data such as instantaneous gas flow rates, as well as parsing information into interpretable forms such as interactive visualizations. Through this approach, we enable a dynamic, interpretable, modular, and cross-domain representation of the UK that enables domain specific experts to contribute toward a national-scale digital twin.

C4T IRP 2 and IRP 3: Cobalt Quaterpyridine Complexes for Highly Efficient Heterogeneous CO_2 Reduction in Aqueous Media
Libo Sun, Vikas Reddu, Shibo Xi, Chencheng Dai, Yuan Sheng, Tan Su, Adrian Fisher, and Xin Wang, *Advanced Energy Materials*

DOI: 10.1002/aenm.202202108

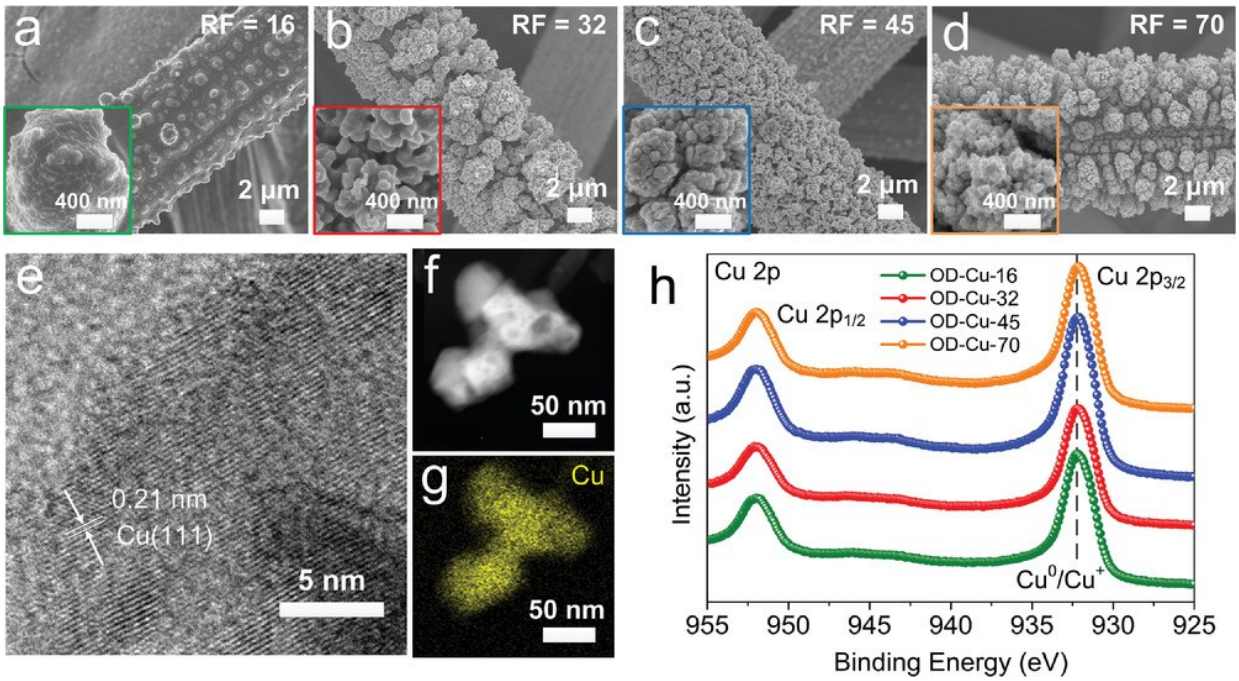
Abstract: Ligands play a critical role in the electrocatalytic CO_2 reduction reaction (CO_2RR) based on heterogeneous molecular catalysts. Previous research on heterogeneous molecular electrocatalysis has mainly dealt with N4 ligands with pyrrole as subunits (porphyrin, phthalocyanine, etc.), while ligands constructed from pyridine subunits remain uncommon. The examples for comparing active configurations are few and far between. Herein, the development of new N4 cobalt complexes based on pyridine subunits is explored. After anchoring onto carbon nanotubes, they can exhibit CO_2RR activity at a low overpotential of 140 mV, and high activity from -0.30 to -0.60 V versus reference hydrogen electrode with a selectivity of above 98%. Excellent performance at large current densities can also be observed in a flow cell. In situ attenuated total reflectance-Fourier transform infrared spectroscopy proves that such electrocatalysts exhibit CO production at lower overpotential and moderate CO adsorption ability over a wide potential range. From density functional theory calculations, it is shown that a pyridine-based cobalt complex on a carbon substrate can reduce the Gibbs free energy for reactions further than its counterpart pyrrole-based ones. Further analysis proves that the semimetal behavior of optimized d-orbitals may facilitate charge transfer and increase the activity. This provides a new insight for understanding catalytically active moieties in heterogeneous molecular catalysts with ligands constructed from pyridine subunits.

C4T IRP 1 and IRP 3: Elucidating Reaction Pathways of the CO_2 Electroreduction via Tailorable Tortuosities and Oxidation States of Cu Nanostructures

Guanyu Liu, Peace Adesina, Noushin Nasiri, Haojing Wang, Yuan Sheng, Shuyang Wu, Markus Kraft, Alexei Lapkin, Joel Ager, and Rong Xu, *Advanced Functional Materials*

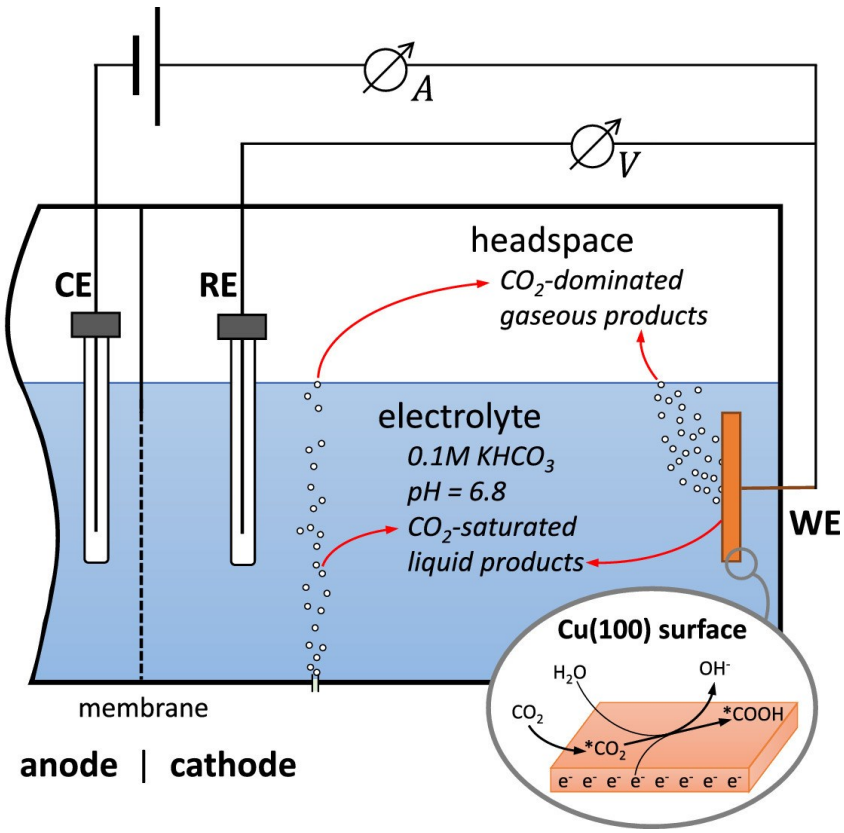
DOI: 10.1002/adfm.202204993

Abstract: Copper-based 3D fractal nanostructures are integrated on the electrodes using a scalable and ink-free flame aerosol synthesis technique for electrochemical CO_2 reduction. The effects of tortuosity and oxidation state of copper are respectively investigated by isolating each effect from the others. By balancing the intermediate confinement and local availability of CO_2, CuO-derived Cu with optimal tortuosity exhibits a Faradaic efficiency of 65% toward C_{2+} products at an applied potential of −1.04 V versus reversible hydrogen electrode. A subsequent study of the effects of the oxidation state, which is free from the influence of tortuosity, reveals that Cu^{2+}-derived Cu demonstrates suppressed hydrogen evolution reaction and a higher C_{2+}/CH_4 ratio than metallic Cu. The preference for the formation of both ethanol and n-propanol versus ethylene, is found to follow the trend from metallic Cu > Cu^{2+}-derived Cu > Cu^{+}-derived Cu toward alcohols' formation. These findings elucidate the underlying causes for the effects of tortuosity of porous Cu electrodes on selectivity and provide insights into the specific effects of the initial oxidation state on various reaction pathways during electrochemical CO_2 reduction.



C4T IRP 3 and IRP JPS: Modelling a detailed kinetic mechanism for electrocatalytic reduction of CO_2
Simon Rihm, Jethro Akroyd, and Markus Kraft, *Proceedings of the Combustion Institute*
DOI: 10.1016/j.proci.2022.07.096

Abstract: For the first time a fully-elementary reversible kinetic model for electrocatalytic CO_2 reduction towards a multitude of different products has been established and verified with experimental data. The detailed reaction mechanism was generated by compiling hypothesized reaction paths and intermediates from many different sources. Thereby a focus was put on distinguishing different embodiments of similar elementary steps: For proton-coupled electron transfer three hydrogenation mechanisms were considered and for intermediates with unclear molecular structure separate paths were modelled. The micro-kinetic model was fed with tabulated energy parameters and results of DFT calculations to simulate CO_2 reduction on a Cu(100) surface for constant applied potentials. The operating conditions were chosen according to published experimental results in order to compare Faradaic Efficiencies. With these, the model parameters were successfully calibrated across a wide potential range while keeping all values within a tight interval of theoretical bounds derived from ab initio calculations and other theoretical considerations. The calibrated model was found to be in good qualitative agreement with the measurement data and also captures trends of surface coverages reported for in-situ measurements. Most interestingly, it finds the widely accepted hypothesis of dimerization via *CO intermediates to be inaccurate. Instead, coupling reactions of *CHO and *CH2 intermediates are observed. The shifting of dimerization routes with varying applied potential especially towards ethylene is supported by other experimental studies. Furthermore, this work establishes a methodology of creating and calibrating complex electrochemical micro-kinetic models.

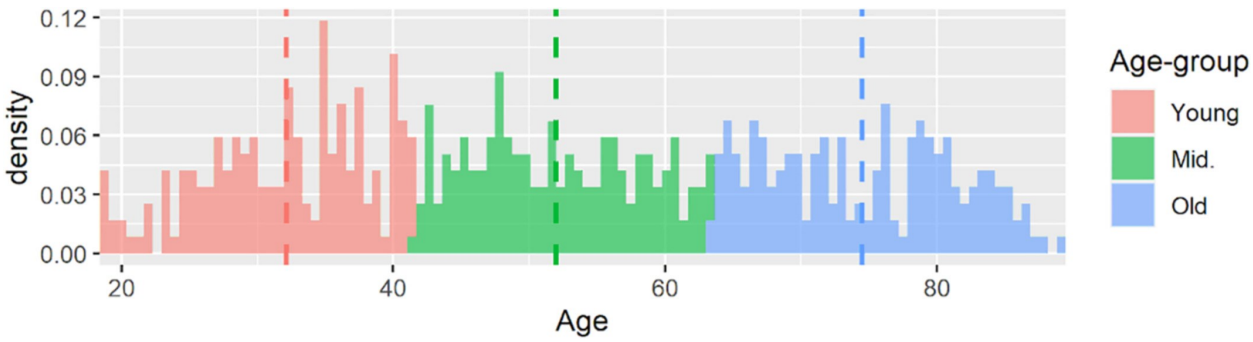


CLIC: Asymmetric generalizability of multimodal brain-behavior associations across age-groups

Junhong Yu and Nastassja Lopes Fischer, *Human Brain Mapping*

DOI: 10.1002/hbm.26035

Abstract: Machine learning methods have increasingly been used to map out brain-behavior associations (BBA), and to predict out-of-scanner behavior of unseen subjects. Given the brain changes that occur in the context of aging, the accuracy of these predictions is likely to depend on how similar the training and testing data sets are in terms of age. To this end, we examined how well BBAs derived from an age-group generalize to other age-groups. We partitioned the CAM-CAN data set (N = 550) into the young, middle, and old age-groups, then used the young and old age-groups to construct prediction models for 11 behavioral outcomes using multimodal neuroimaging features (i.e., structural and resting -state functional connectivity, and gray matter volume/cortical thickness). These models were then applied to all three age-groups to predict their behavioral scores. When the young-derived models were used, a graded pattern of age-generalization was generally observed across most behavioral outcomes—predictions are the most accurate in the young subjects in the testing data set, followed by the middle and then old-aged subjects. Conversely, when the old-derived models were used, the disparity in the predictive accuracy across age-groups was mostly negligible. These findings hold across different imaging modalities. These results suggest the asymmetric age-generalization of BBAs—old-derived BBAs generalized well to all age-groups, however young-derived BBAs generalized poorly beyond their own age-group.



IRP 1

SUSTAINABLE REACTION ENGINEERING FOR CARBON NEUTRAL INDUSTRY

IRP 1 is focused on chemical technologies that allow rapid decarbonisation of chemical industry and the chemical supply chain. Our target is to deliver innovative solutions to direct utilisation of carbon dioxide as a feedstock, as well as to significantly increase the efficiency in conversion of methane to bulk intermediates. We are also exploring the options for the emerging circular economy, by developing new transformations of molecules available in different bio-waste resources into high-value functional molecules. Potential impact on carbon emissions reduction is evaluated by life cycle assessment tools.

IRP 1 Principal Investigators:

Professor Alexei LAPKIN
University of Cambridge

Asst Professor Paul LIU Wen
Nanyang Technological University

Assoc Professor YAN Ning
National University of Singapore

OVERVIEW

In this reporting period the focus of work within IRP1 shifted towards design of catalytic materials and processes for carbon reduction chemistry. Our joint team between CAM, NTU and CARES is collaborating with colleagues in Canada and India on a project to unlock the potential of inverse computational design of catalytic materials and the teams in NTU, NUS and CARES are working on new chemical reactions and new catalytic material. This will be the foundation for the final phase of the project.

Professor Alexei Lapkin, PI
University of Cambridge

Update on work package 1.1

Design of nano-structured catalysts

Dr Quan ZHANG (Research Fellow, NUS) has been focusing on developing highly efficient catalysts for conversion of renewable resources. Recently, he has synthesised many kinds of alloy catalysts including bimetallic NiCo alloy nanoparticles (NPs) and Ru-based high entropy alloy NPs. The structures of the alloy nanoparticles were characterised by TEM, XRD, EDX and XPS. The results confirmed the success of synthesising these alloy nanoparticles. Furthermore, he has loaded the synthesised alloy catalysts on various different supports such as, carbon black, CeO_2, and MgO-CeO_2. The catalytic performance of the alloy NPs will be investigated in several important reactions.

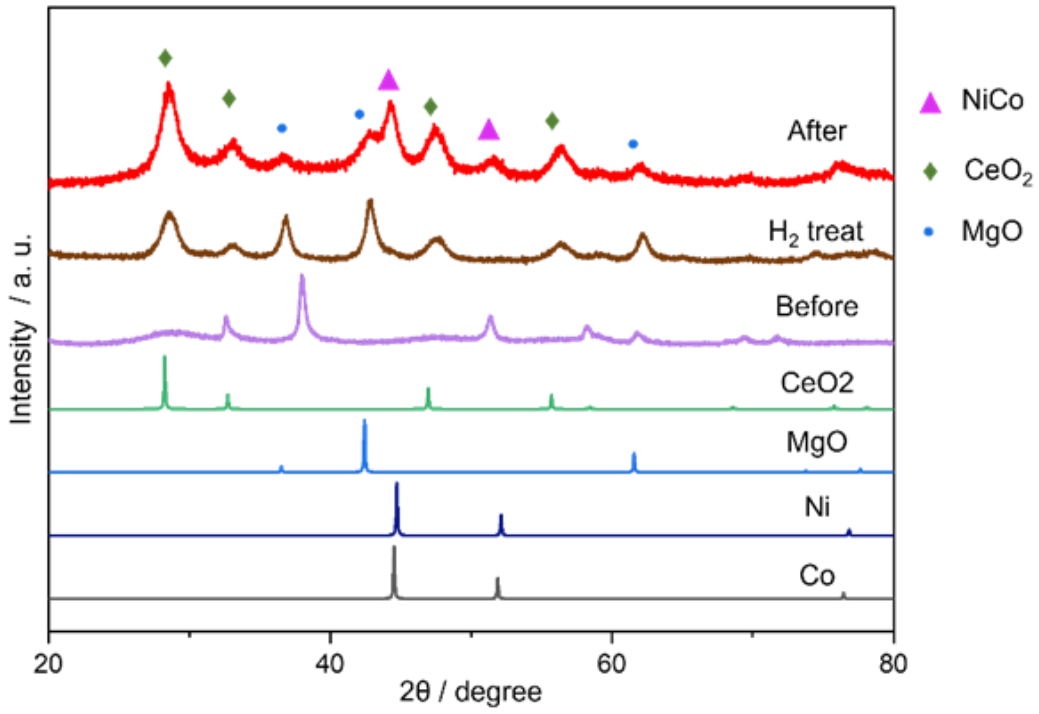


Figure 1.1: XRD pattern of the NiCo alloy NPs loaded on MgO-CeO_2.

Dr Quan ZHANG

Update on work package 1.2

Novel reactions and functional molecules

Guided by **Prof Alexei LAPKIN (PI, CAM)**, **Dr Zhen GUO (Senior Research Fellow, CARES)** focused on developing a tool for chemical route searching. Tasks that have been conducted include:

1. Development of Application Programming Interfaces (APIs).

2. Testing, building and deployment of the tool as an application (currently for a single user).

3. Database Restructure. Reaction data and molecular data were refined and stored in a more structured database. The revamp improves the speed and usability of the algorithm.

4. Started the development of a reaction condition recommendation system (RCRS) for gathering relevant reaction conditions given a target reaction as a request.

5. Started extraction of reaction templates. The quality of reaction templates is determined by several factors such as data quality, extraction algorithm, standardisations and denotations. At this stage, ongoing work includes improvement of data quality and development of template extraction algorithms.

In addition, a paper based on the work in collaboration with **Assoc Prof Ning YAN's (PI, NUS)** group has been submitted.

Dr Guo will continue to improve the tool for searching of chemical routes, mainly targeting safety, scalability and reliability. Development of methods for extraction of reaction templates will be another main task as this technique is identified as an important component based on feedback from IRP members and industrial partners.

Mr Aniket CHITRE'S (PhD student, CAM) main research interests lie in applying machine learning (ML) methods to accelerating the development of liquid formulations, and in the future, towards more sustainable products. The key requirement of such a methodology is a good formulations dataset. Mr Chitre and **Prof Alexei LAPKIN (PI, CAM)** are working in collaboration with BASF using industrial formulations ingredients to develop just such a dataset. The work is being conducted via an exchange in Prof Kedar HIPPALGAONKAR'S (Non-C4T PI, NTU) AI for Accelerated Materials lab at the Institute of Materials Research and Engineering (IMRE), A*STAR. Here, Mr Chitre has been developing a modular and semi-automated workflow to prepare and characterise the liquid formulations. Notable challenges have arisen from working with viscous liquids and ingredients with a complex behaviour. Highlights from the past six months include several ongoing works with the Opentrons automated pipetting robot, retrofitted with a mass balance, and the pictured platform for pH adjustment. In collaboration with Dr Jayce CHENG (A*STAR) at IMRE, Mr Chitre has developed a state-of-the-art pH robot for automated, ML-driven pH adjustment of viscous formulations. This is a key step in the overall workflow, which will soon be ready for 'production-quality' data to be generated.

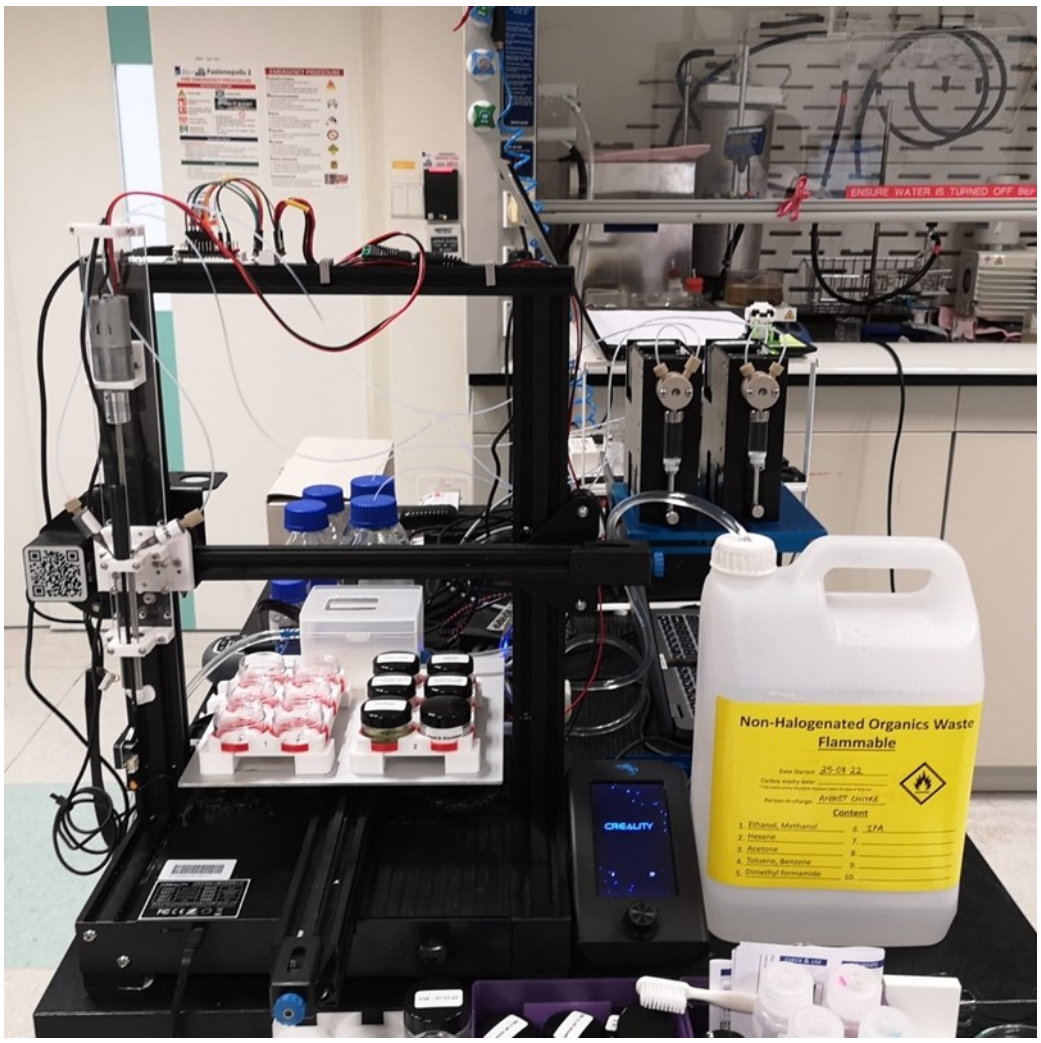


Figure 1.2: Custom-built pH platform for automated, ML-driven adjustment of viscous formulations

Mr Aniket CHITRE

Mr Adarsh ARUN (PhD student, CAM) commenced his PhD in January 2021 and focuses on identifying sustainable routes from biowaste to chemicals using networks and knowledge graphs (KGs). Over the past several months, he has been investigating and developing an ontology/ schema to represent the required data, integrating a variety of existing ontologies from a variety of domains (location, agronomy, reactions, processes). He has also begun populating a preliminary KG based on the results of a case study on biowaste sources in Singapore, Malaysia and Indonesia which he undertook in his first year. Concurrently, he has also continued his work on enriching reaction networks by data mining large chemical databases such as Reaxys to predict impurities and byproducts in chemical reactions. He has a preprint of the work pending publication as well as peer review.

Update on work package 1.3

Novel reactors and process technology

Mr Qianwenhao FAN (PhD student, NTU) is writing up his thesis "Development of Advanced Fe-Based Oxygen carriers for Chemical Looping Applications" and is expected to submit in late 2022.

Mr Syed SAQLINE (PhD student, NTU) is writing up his thesis "Chemical Looping Combustion of Syngas using Low Cost Oxygen Carriers" and is expected to submit in early 2023.

Dr Hui Ling TAN's (Research Fellow, NTU) main research interest lies in the development of (photo)catalysts and technologies for renewable energy generation and pollutant abatement. In the project of identifying the adsorption structures of formic acid on TiB_2 and TiB_2-supported Pd catalysts, she has shown that formate is the only species that formed on the catalysts' surfaces from formic acid dissociation via in-situ Diffuse Reflectance Infrared Fourier Transform Spectroscopy (DRIFTS) technique. This was verified by adsorption of deuterated DCOOH on the catalysts followed by D-H scrambling experiment.

Ms Xianyue WU (CARES Visitor, NTU) has been actively working on the development of a two-step CO_2 capture and methanation process using Ni-supported-on-alkaline earth metal carbonate dual-function materials, demonstrated in Figure 1.3. She has conducted a series of characterisation work to investigate the composition change of Ni/$CaCO_3$ DFMs after long-term CO_2 capture and methanation cycles. The Ni dispersion was increased after hydrogenation and a clear drop in $CaCO_3$ component and emergence of CaO was detected by XRD, TEM as well as in situ DRIFT (Figure 1.4). The Ni metal nanoparticles were mostly surrounded by CaO after hydrogenation, whereas the $CaCO_3$ further away from the Ni particles did not decompose. Ms Wu proposes a model for Ni/$CaCO_3$ during this scheme of CO_2 hydrogenation and carbonation process with a formate reaction pathway (Figure 1.5). She will continue to study the detailed mechanism of CO_2 hydrogenation on Ni-supported-on-alkaline earth metal carbonate DFMs and investigate the effects of basicity of the support carbonates.

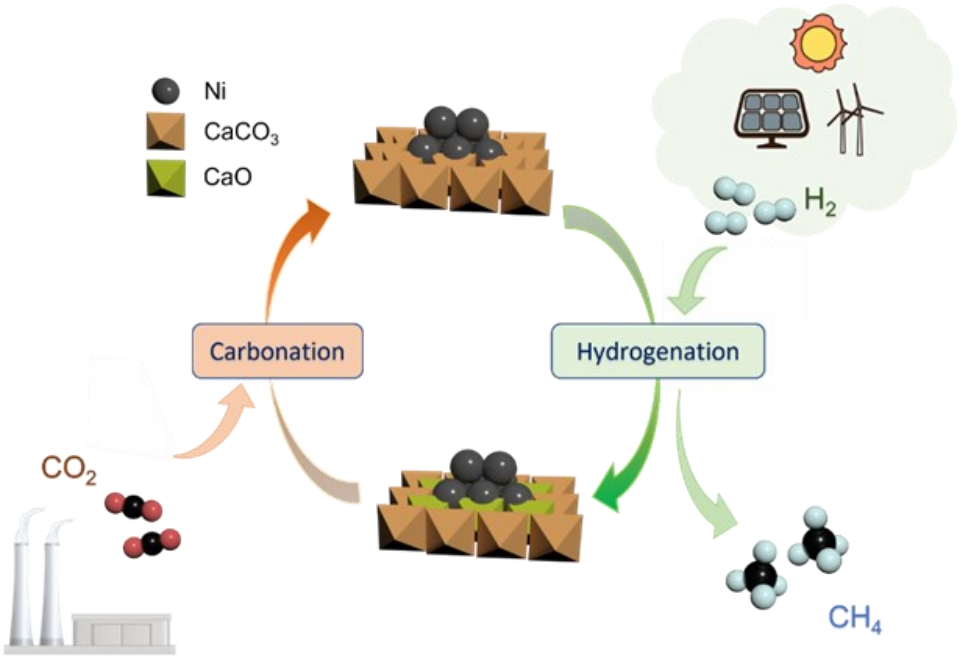


Figure 1.3: Scheme of In-situ CO_2 Capture and Catalytic Methanation Using Ni/alkaline Earth Carbonate Dual Function Materials.

Ms Xianyue WU

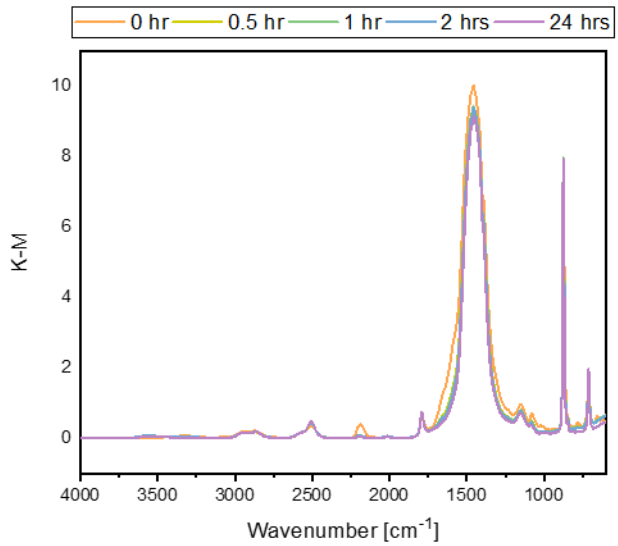


Figure 1.4: in situ DRIFTS measurements on $Ni/CaCO_3$ and (b) Ni/Mg at 400 °C in 10 vol% H_2/N_2 for 24 h.

Ms Xianyue WU

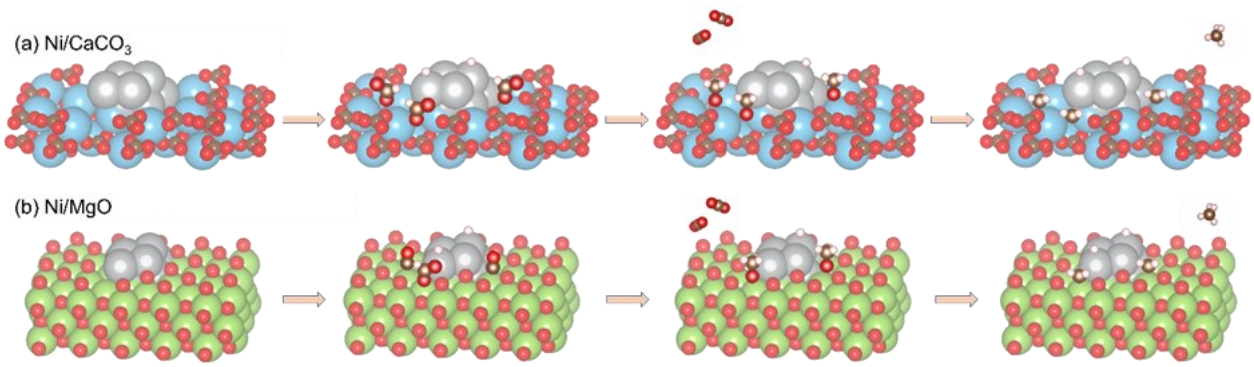


Figure 1.5: Reaction mechanisms of two-step CO_2 carbonation-hydrogenation at 400 °C on (a) $Ni/CaCO_3$ and (b) $Ni/MgCO_3$.

Ms Xianyue WU

Asst Prof Tej CHOKSI's (Co-PI, NTU) group is funded by an Emerging Opportunities Fund grant. The objective of this grant is to design supported metal catalysts that enable CO_2 conversion using first principles methods. In the last six months, his group has made progress with two of the project aims: (1) predicting catalytic stability, and (2) understanding the selectivity trends among CO_2 reduction catalysts. The group has developed a generalised model that can estimate the stability and determine the shape of gold nanoparticles dispersed on 2D and 3D carbide/nitride supports. This model uses fundamental physico-chemical properties of the gold-carbide/nitride system as inputs, and is thus applicable across a wide-range of materials. Using thermodynamic constraints, Asst Prof Choksi's group has also created a selectivity map of CO_2 reduction to CO, H_2, (syn-gas) and HCOOH. This selectivity map uses molecular descriptors like the adsorption energies of CO* and OH* as inputs. The models for stability and selectivity, when taken together, will enable the high-throughput screening of gold/support heterostructures that are not only thermodynamically stable, but selectively transform CO_2 into either syn-gas or HCOOH, a potential hydrogen carrier.

The group is also collaborating with an experimentalist, Prof Lydia WONG (Non-C4T PI, NTU) on designing sulphur-doped Cu_xSb_y catalysts that convert CO_2 into either syn-gas or HCOOH.

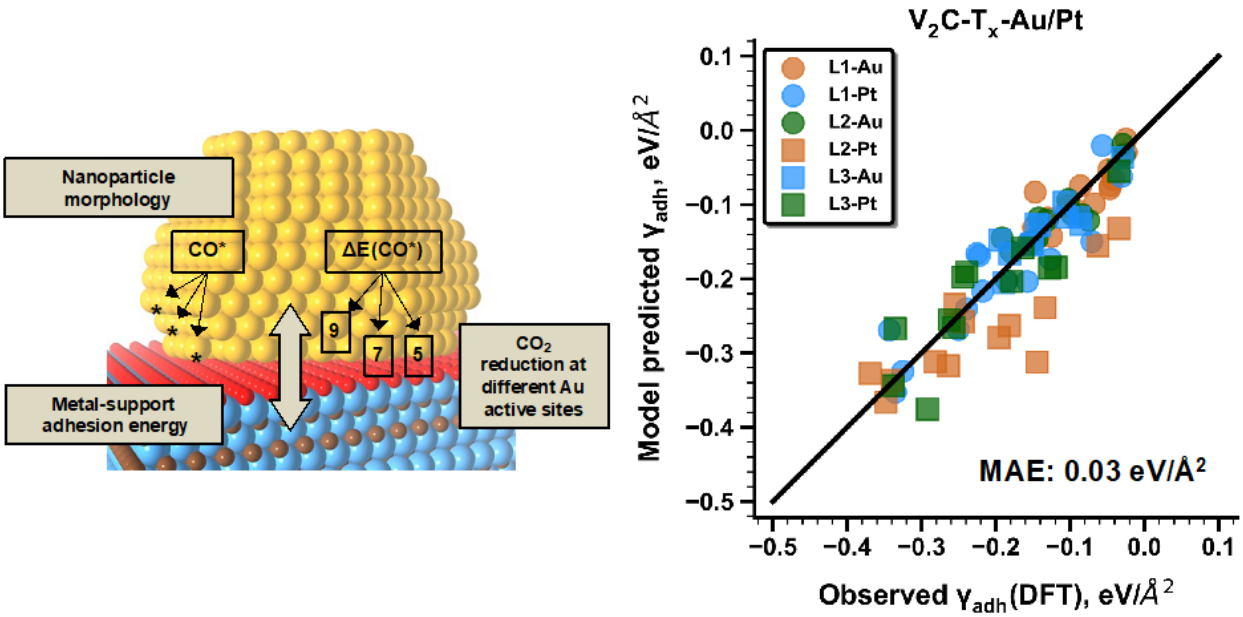


Figure 1.6: (left). Gold nanoparticle supported on vanadium carbide terminated by oxygen atoms. (right). A parity plot comparing model-predicted adhesion energies with those estimated using density functional theory. The adhesion energies reflect the stability of gold on the support. Furthermore, these adhesion energies also yield the morphology of the supported gold nanoparticle.

Asst Prof Tej CHOKSI

Dr Mikhail KOVALEV (Technical Development Manager, AMPLE) was previously working under IRP 1 before moving to the AMPLE project (see page 136). He was working on electrochemical CO_2 reduction (ECO_2R) and focusing on a newly observed effect of the enrichment of the ^{13}C isotope. Carbon-13 isotope has a few unique properties that allows its use in many modern applications, mainly as a labelling agent in organic chemistry as it is observable in NMR due to its nuclei's half-spin. Current methods of ^{13}C enrichment are very expensive and lengthy, for example, a cryogenic distillation of CO (carbon monoxide) can take a few weeks to reach a concentration of 20% of ^{13}CO and a few months to reach 99% of ^{13}CO. Seeking a new approach for the enrichment of ^{13}C may open doors to more affordable isotope sources that will boost its use.

Discovering that ECO_2R discriminates $^{13}CO_2$ opens the opportunity to cheaper sources of the heavier carbon isotope. On Figure 1.7 the proposed scheme allows the use of continuous flow of CO_2 to enrich carbon-13 in a faster and cheaper method than currently known techniques. By placing electroreduction cells and separation units in a consecutive chain, it is possible to achieve high degrees of $^{13}CO_2$ enrichment with the enrichment ratio of 0.4% in each step.

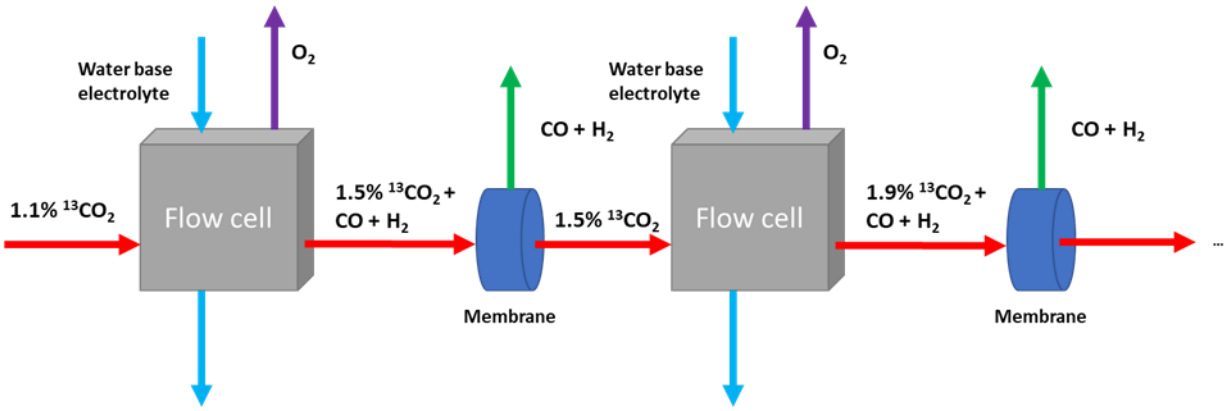


Figure 1.7: Schematic continuous flow $^{13}CO_2$ enrichment process where flow-cells can be placed consequently to reach high content of ^{13}C after the chain.

Dr Mikhail KOVALEV

Mr Alvin Ming Hao LIM (in-kind PhD student, NUS) and **Prof Hua Chun ZENG (Co-PI, NUS)** have recently functionalised the as-synthesised zinc-silicate (ZnSiO) nanoflowers (ca. 600 to 800 nm in diameter) with Cu and Zn for CO_2 hydrogenation for methanol production. Through simple incipient wetness impregnation, Cu and Zn-based nanoparticles were well-dispersed on the petals of these zinc-silicate nanoflowers with the diameter of these metallic nanoparticles kept at around 5 nm (Figure 1.8). The Zn to Cu ratio could be varied between 0.1 to 0.3 with the Cu loading maintained at around 9 wt% to investigate optimal metal ratio through experimental testing for this catalytic reaction. The preliminary results indicate a high specific activity when the

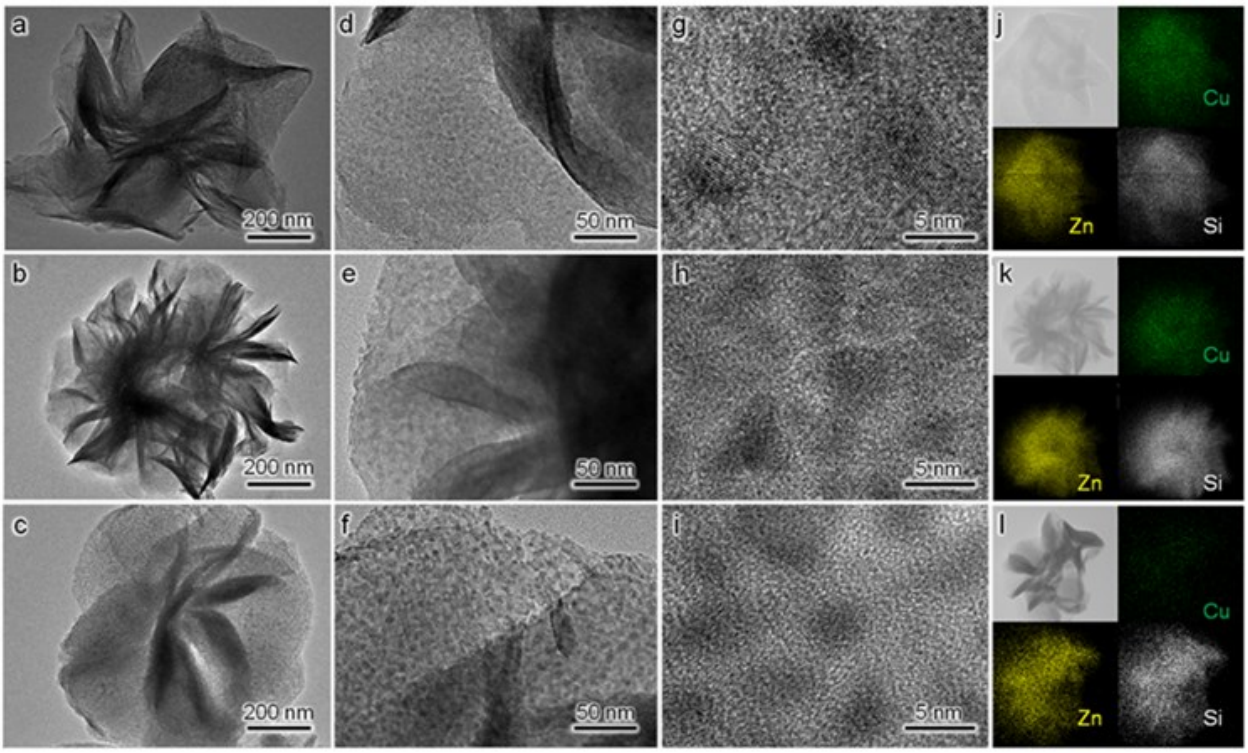


Figure 1.8 (a-c) TEM image, (d-i) HRTEM images and elemental mappings of ZnSiO/0.1CuZn (a, d, g, j), ZnSiO/0.2CuZn (b, e, h, k), and ZnSiO/0.3CuZn (c, f, i, l).

Mr Alvin Ming Hao LIM

ratio of Zn to Cu is tuned at 0.2 and performs better when compared against an industrial catalyst (Alfa Aesar 45776) as depicted in Figure 1.9. Duplicate experimental runs were performed to confirm performance repeatability at different temperature points. This project demonstrated the structure-activity relationship for nanocatalysts, the application potential for zinc-silicate material to be embedded with other transition metals to achieve small and well-dispersed metallic nanoparticles onto the petals of zinc-silicate nanoflower, and its utility and functionality for other catalytic applications.

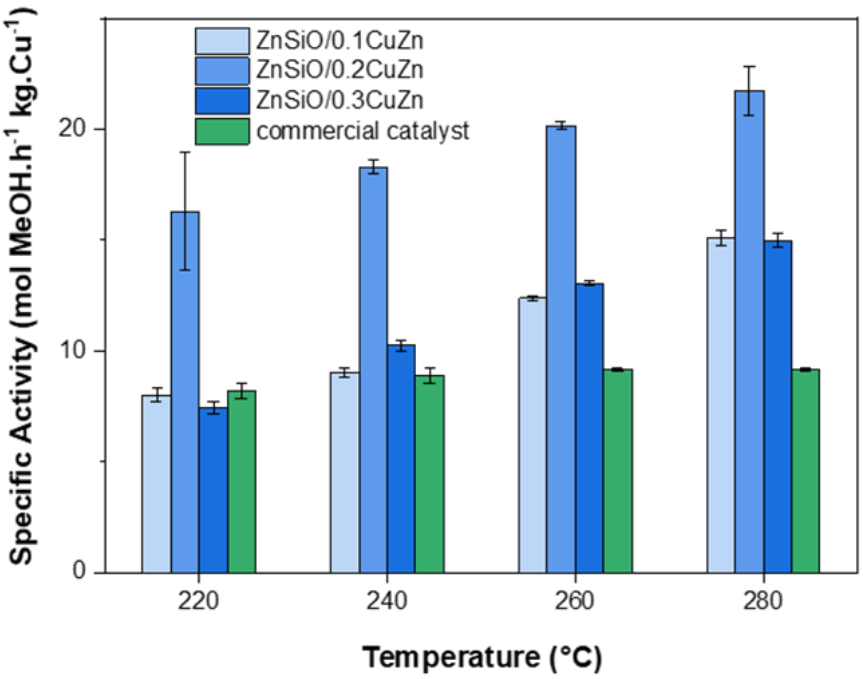


Figure 1.9. Specific activity of different ZnSiO catalysts with varying Zn/Cu atomic ratios and a commercial catalyst (Alfa-Aesar 45776).

Mr Alvin Ming Hao LIM

Dr Shenghui ZHOU (Research Fellow, NUS) and **Prof Hua Chun ZENG (Co-PI, NUS)** have proposed a novel MOF-templated strategy for the synthetic architecture of hollow ZnS confined quasi-single-layer MoS_2 nanoboxes. Firstly, they developed a fast solvothermal treatment process to obtain hollow $ZnMoS_x$ nanoboxes using the ZIF-8 nanocube as a template and ammonium tetrathiomolybdate (ATM) both as a Mo source and as a sulfidation agent. This hollow precursor could then be further transformed into higher crystalline h-MoS_2/ZnS after the annealing treatment in an inert atmosphere. FESEM image showed that h-MoS_2/ZnS had a hollow morphology with rougher surfaces (Figure 1.10a). The well-defined structure with a central cavity and 20 nm thick shell was further confirmed by the TEM technique (Figure 1.10b, c). Closer inspection (Figure 1.10d) of some randomly titled MoS_2 nanosheets revealed that only 1–3 layers of MoS_2 were stacked to form the walls of the nanoboxes, noting that these nanosheets laid largely parallel on the surfaces. Furthermore, the high-resolution TEM (HRTEM) image (Figure 1.10e) exhibited a typical lamellar structure with a well-resolved d-spacing of 0.63 nm, which could be ascribed to the lattice fringes of d_{002} of MoS_2 (JCPDS No. 37-1492). In the center surrounded by MoS_2, one could also see another clear lattice stripe of 0.31 nm, which was indexed to the d_{111} of the ZnS (JCPDS No. 05-0566) phase, suggesting that the Zn^{2+} in ZIF-8 nanocubes had been successfully sulfurised to zinc sulfide by ATM. An analysis of HRTEM-EDX line-scanning profiles and corresponding elemental mappings (Figure 1.10f–h) confirmed the homogeneous distribution of Zn,

Mo, S, N, and C elements in h-MoS_2/ZnS nanoboxes. When evaluated as catalysts in selective hydrogenation of CO_2 to methanol, the obtained h-MoS_2/ZnS can achieve STY_{MeOH} up to 0.93 $g_{MeOH} \cdot g_{MoS2}^{-1} \cdot h^{-1}$ with a CO_2 conversion of 9.0% and a methanol selectivity of 67.3% at 260°C, 5 MPa, and 15 000 mL $\cdot g_{cat.}^{-1} \cdot h^{-1}$. The good performance of h-MoS_2/ZnS makes it a prospective candidate for potential industrial application in CO_2 hydrogenation.

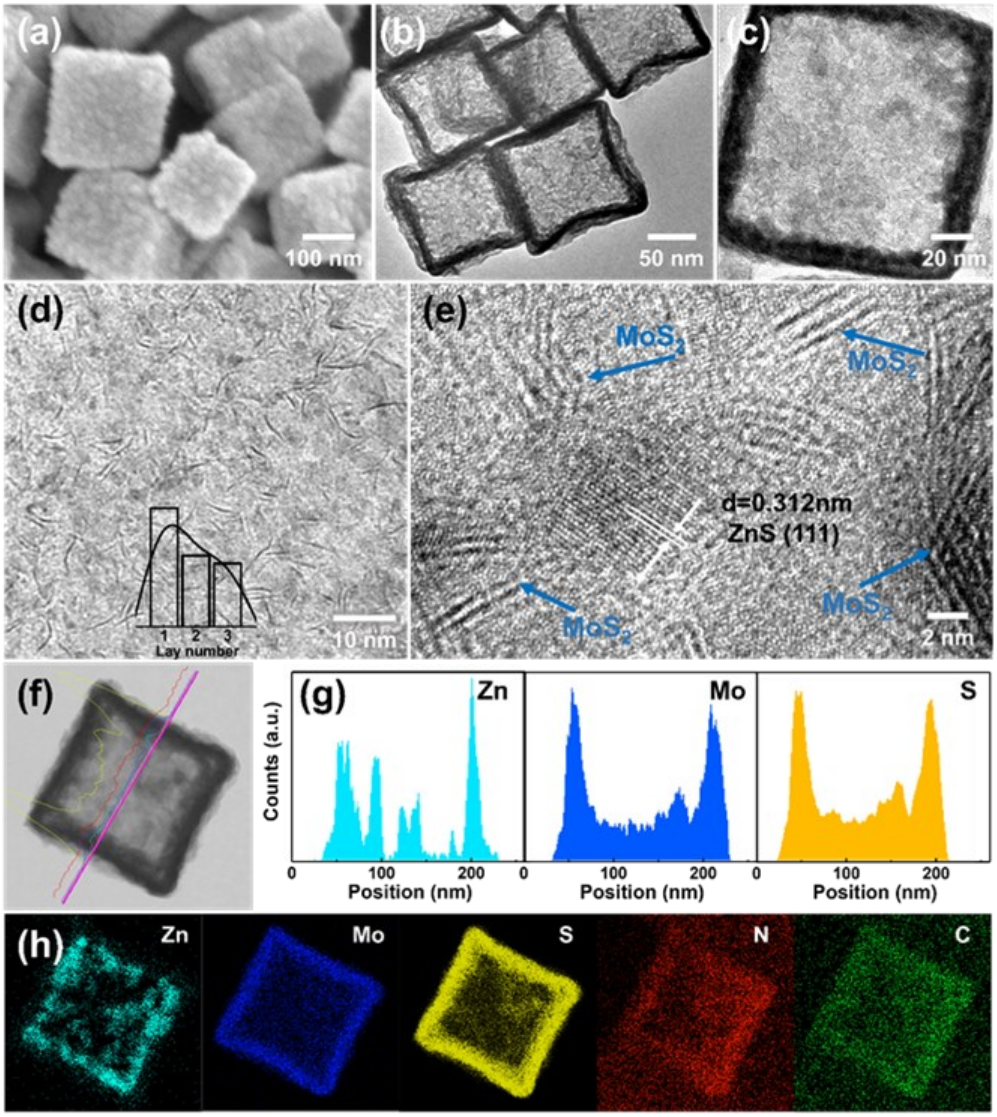


Figure 1.10: (a) FESEM, (b, c) TEM and (d, e) HRTEM images of h-MoS_2/ZnS nanoboxes. (f) Scanning TEM image, (g) corresponding EDX line scans and (h) chemical mappings of h-MoS_2/ZnS nanoboxes.

Dr Shenghui ZHOU

Mr Chao WANG (CARES Visitor, NUS) and **Prof Hua Chun ZENG (Co-PI, NUS)** have been investigating the unique properties of ZrO_2 and its synergistic effect with Cu. Substantial progress has been made in designing high performance ZrO_2-containing catalysts for CO_2-to-methanol conversion. From the aspect of structural designing, in this work, a silica-boosted ZrO_2 host matrix with a highly thermal stable nanostructure was synthesised from monodisperse Zr-MOFs (metal-organic frameworks) (Figure 1.11). This ZrO_2-based host material possesses a well-developed porosity and significant thermal stability. It was then loaded with Cu to be used as a catalyst candidate for CO_2 hydrogenation to methanol under relatively low temperature and

pressure. The as-prepared Cu catalyst presented a more efficient catalytic performance in CO_2 hydrogenation to methanol comparing with other ZrO_2-containing Cu catalysts with varied architectural forms and the commercial Cu/ZnO/Al_2O_3 catalyst under the same reaction conditions. On the other hand, the copper component was highly dispersed within the host frameworks, and owing to its robust structure, the aggregation of either copper phase or the host material was effectively prevented during the stability test. In addition to catalytic performance, in-depth investigations on the copper site and reaction mechanism were also carried out using in situ X-ray absorption spectroscopy and DRIFTS.

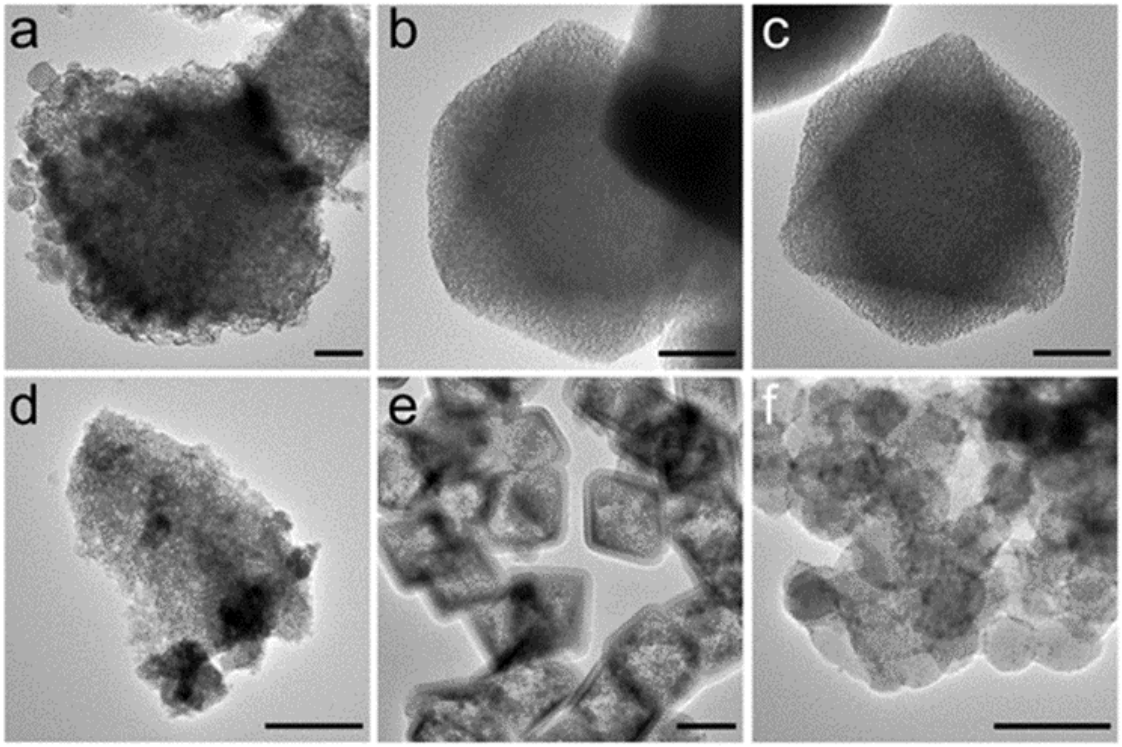


Figure 1.11: Representative HRTEM images of ZrO_2-containing catalysts with different structures. Catalysts of Cu being loaded on (a) UiO-66, (b) ligand-removed UiO-66, (c) silica-boosted UiO-66, (d) amorphous ZrO_2, (e) core-shell ZrO_2@$mSiO_2$ (m denotes "mesoporous"), and (f) ZrO_2-coloaded $mSiO_2$. Scale bars in (a-c) are 50 nm and in (d-f) are 200 nm.

Mr Chao WANG

Ms Ziyu GAO (Non-C4T PhD student, NUS) and **Prof Hua Chun Zeng (Co-PI, NUS)** focused on carbon nanotubes (CNTs) which are generally recognised as one of the best and most easily available one-dimensional (1D) nanostructures having a long, tubular shape. In this project, CNTs are used as a structural template and mesoporous SiO_2 ($mSiO_2$) was coated on surfaces of CNTs to obtain a hybrid CNTs@$mSiO_2$ with 1D mesoporous structure. It is clear that the uniform $mSiO_2$ shell of ca. 20 nm in thickness was formed on the outer surface of CNTs, affirming a complete coverage. ZIF-8 layer (ca. 5-7 nm) with microporous structure was then introduced on the outer surface of $mSiO_2$ at room temperature to obtain the hybrid CNTs@$mSiO_2$@ZIF-8 with micro-mesoporous structure. A second ligand was also introduced in ZIF-8 through a post-synthesis modification method, and metal Cu was also introduced in the CNTs@$mSiO_2$@ZIF-8-A (after modification) with the assistance of amino groups in the framework structure. The obtained CNTs@$mSiO_2$@ZIF-8-A (Zn, Cu) will be subsequently processed and used as an efficient catalyst in the CO_2 hydrogenation reaction to methanol.

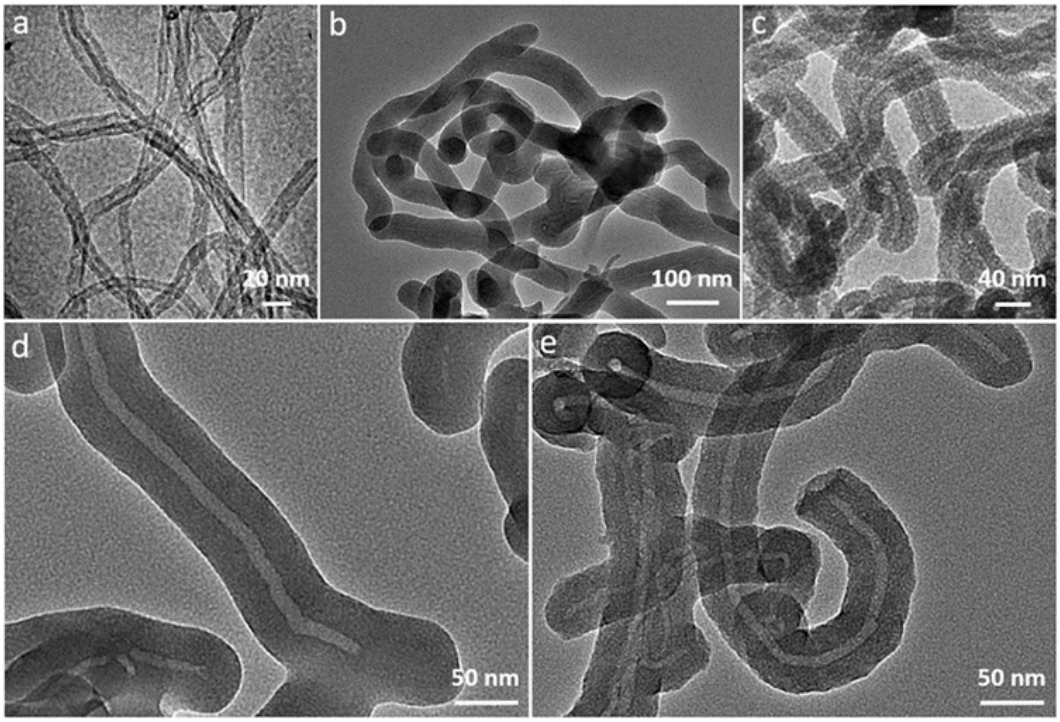


Figure 1.12: Representative TEM images of (a) pristine CNTs, (b)CNTs@mSiO$_2$, (c) CNTs@mSiO$_2$ (after calcining), (d) CNTs@mSiO$_2$@ZIF-8, and (e) CNTs@mSiO$_2$@ZIF-8-A.

Ms Ziyu GAO

Dr Nicholas JOSE (Research Fellow, CARES) has been working on a number of different projects over the previous six months:

Automation: Dr Jose has continued working on *flab* (a coding framework for rapid automation of laboratory equipment), increasing the number of case studies and working on the following functionalities:

- A user-friendly interface for real-time operation of devices (*flab Console*)
- Network communication for remote operation of instruments with *flab Console*

Carbon quantum dots (CQDs): A rapid synthetic method for CQD synthesis has been under development, now focusing on their applications in biological settings, as potential sensors or antimicrobials. Recent work has focused on characterising the effect of pH shifts on the dots, which have shown complex effects on their fluorescent behaviour.

Wohl Alliance: The team consisting of Dr Jose, **Dr Mikhail KOVALEV (Technical Development Manager, AMPLE)**, and **Prof Alexei LAPKIN (PI, CAM)** have been awarded a Wohl Clean Growth Alliance grant for a collaborative project with Prof Ovadia LEV (Hebrew University of Jerusalem). The US$25k funded project, "Improving clean water availability for future growth with new materials - Wohl Clean Growth Alliance Grants" seeks to develop cost-effective processes for producing materials developed by HUJ, and could kickstart further collaborative grant funding on the topic. The collaboration between HUJ and CARES members began as a direct result of interaction with HUJ/SHARE within CREATE.

Simulations: Work continues on simulations of annular flow microreactor hydrodynamics in collaboration with PhD researcher Mr Zhai SONG (RWTCH Aachen University). This is a continuation of a previous Masters project with University of Cambridge, in which Mr Song led the development of a model for computational prediction of shear rate and mixing time within the reactor. Dr Jose expects to finish long-term simulations this fall, with a manuscript draft by the end of the year.

Scientific output

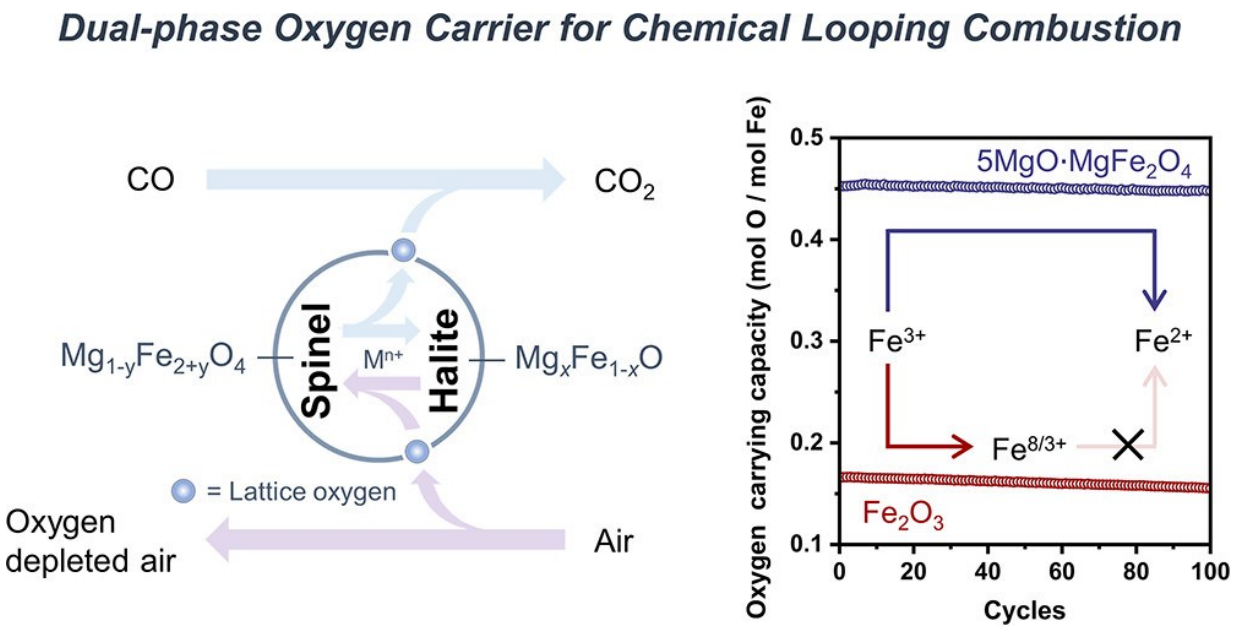
The following are the CREATE-acknowledged publications generated by IRP 1 during the reporting period, excluding those already featured in the Scientific Highlights section on page 14.

Breaking the Stoichiometric Limit in Oxygen-Carrying Capacity of Fe-Based Oxygen Carriers for Chemical Looping Combustion using the Mg-Fe-O Solid Solution System

Qianwenhao Fan, Chuande Huang, Shibo Xi, Yong Yan, Jijiang Huang, Syed Saqline, Longgang Tao, Yihu Dai, Armando Borgna, Xiaodong Wang, and Wen Liu, *ACS Sustainable Chemistry & Engineering*

DOI: 10.1021/acssuschemeng.2c00271

Abstract: The performance of oxygen carriers contributes significantly to the efficiency of chemical looping combustion (CLC), an emerging carbon capture technology. Despite their low cost, Fe_2O_3-based oxygen carriers suffer from sintering -induced deactivation and low oxygen-carrying capacity (OCC) during CLC operations. Here, we report the development of a sintering-resistant MgO-doped Fe_2O_3 oxygen carrier with an optimal composition of $5MgO{\cdot}MgFe_2O_4$, which exhibits superior cyclic stability and an OCC of 0.45 mol O/mol Fe (2.25 mmol O/g_{solid}), exceeding the widely accepted OCC limit of 0.167 mol O/mol Fe (2.08 mmol O/g_{solid}) of unmodified commercial Fe_2O_3. This result distinguishes this report from all past studies, in which efforts to enhance the cyclic stability of Fe-based oxygen carriers would always result in dilution of the OCC. The capacity enhancement by MgO is attributed to the unique mixtures of $Mg_xFe_{1-x}O$ (halite) and $Mg_{1-y}Fe_{2+y}O_4$ (spinel) solid solutions, which effectively reduce the exergonicity for the reduction from Fe^{3+} to Fe^{2+}, while preventing any irreversible structural transformations during the redox process. This hypothesis-driven oxygen carrier design approach provides a new avenue for tailoring the lattice oxygen activities of oxygen carriers for chemical looping applications.

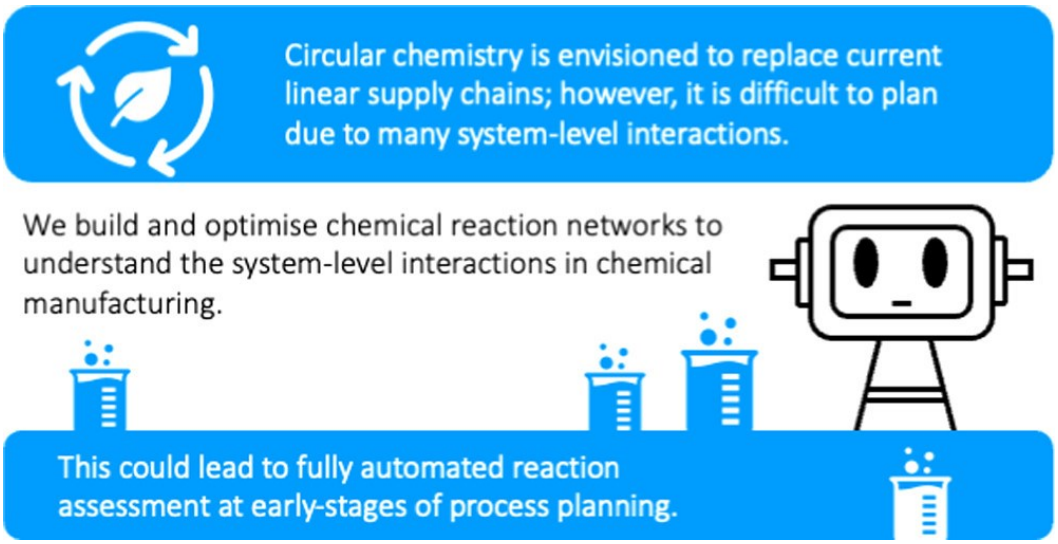


Discovering Circular Process Solutions through Automated Reaction Network Optimization

Jana Weber, Zhen Guo, and Alexei Lapkin, *ACS Engineering Au*

DOI: 10.1021/acsengineeringau.2c00002

Abstract:: The transition toward a circular and biobased chemical industry is needed to cut global CO_2 emissions and limit the chemical industry's overall impact on the environment. However, the development of circular chemical reaction systems is challenging as it requires symbiotic sets of novel chemical reaction pathways and involves unconventional processing steps. We present a methodological pipeline for automated reaction network optimization. The tools can guide the development of circular processes on the reaction pathway level. Chemical big data combined with energetic assessment metrics and state-of-the-art decision-making has the potential to efficiently identify the most promising reaction systems. We mine large-scale chemical reaction data from Reaxys database and automate the screening of pathways based on chemical rules. We then approximate thermodynamic properties for exergy calculations of the prescreened pathways and formulate the optimization problem as linear programming and mixed-integer linear programming problem. The methodological workflow is illustrated in a case study on the conversion of β-pinene to citral. Our results show that the tools are well suited to model circular process interactions within different environment scenarios.

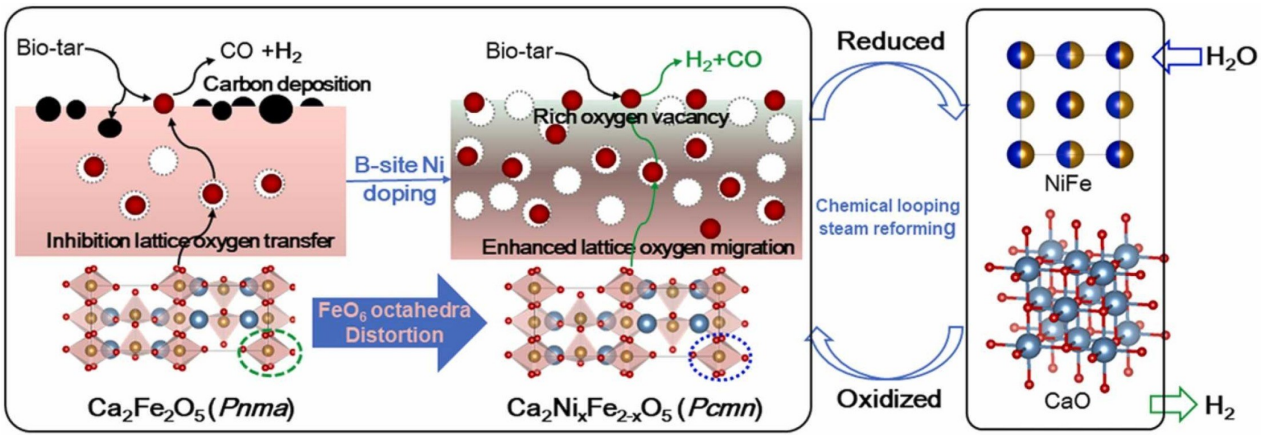


Modulating lattice oxygen activity of $Ca_2Fe_2O_5$ brownmillerite for the co-production of syngas and high purity hydrogen via chemical looping steam reforming of toluene

Tingting Xu, Xun Wang, Haibo Zhao, Bo Xiao, Dong Liu, and Wen Liu, *Applied Catalysis B: Environmental*

DOI: 10.1016/j.apcatb.2022.122010

Abstract: The chemical looping steam reforming (CLSR) of biomass tar enables the process intensification for the co-preparation of syngas and high purity hydrogen. The practical application of brownmillerite-structured $Ca_2Fe_2O_5$ is hindered by activity-related issues such as low fuel conversion and oxygen transfer capacity. Here, the doping of heteroatoms, e.g. Ni induces structural changes to the brownmillerite lattice, transforming it from a *Pnma* phase to a *Pcmn* one, with increased distortion of the FeO_6 octahedra. The structural changes lead to the upwards shifts of the O 2*p* band of oxygen carrier, and subsequently improved lattice oxygen activity as well as oxygen transfer capacity. The formation of oxygen vacancy is a rate determining step during CLSR, while the Ni-doped $Ca_2Fe_2O_5$ reduces the energy of oxygen vacancy formation and energy barrier for lattice oxygen migration through the bulk. During CLSR, $Ca_2Ni_{0.25}Fe_{1.75}O_5$ lead to significant improvement in syngas productivity, hydrogen purity and fuel conversion.

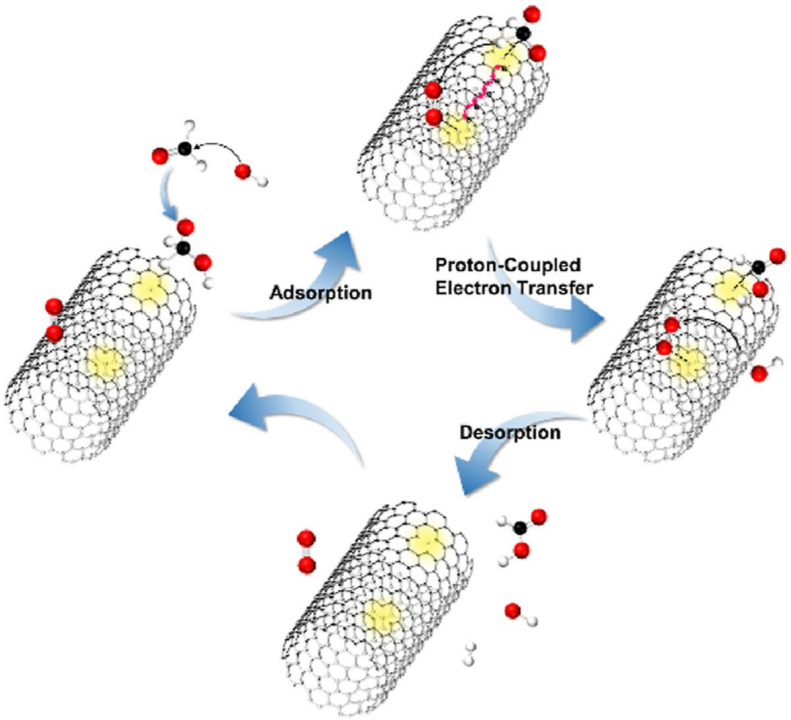


Carbon-catalyzed oxygen-mediated dehydrogenation of formaldehyde in alkaline solution for efficient hydrogen production

Nan Lu, Xiaoqing Yan, Hui Ling Tan, Hisayoshi Kobayashi, Xuehan Yu, Yuezhou Li, Jiemei Zhang, Zhengxin Peng, Jing Sui, Ziying Zhang, Wen Liu, Renhong Li, and Benxia Li, *International Journal of Hydrogen Energy*

DOI: 10.1016/j.ijhydene.2022.06.134

Abstract:: We report an efficient process to dehydrogenate formaldehyde in alkaline solution, catalyzed by carbon nanotubes (CNTs) via a unique reaction mechanism involving molecular O_2. The superior catalytic performance of carbon nanotubes (CNTs) compared to the other carbon-based catalysts is attributed to their sp^2-carbon-rich surface, hydrophilicity and abundant surface defects, which are the most plausible active sites. Peroxide species originating from the activation of adsorbed molecular oxygen on the CNTs is found to be a key to C–H activation, leading to efficient hydrogen production. The cost-effective carbon-based dehydrogenation catalysts offer new opportunities to the development of novel liquid organic hydrogen carrier technologies.

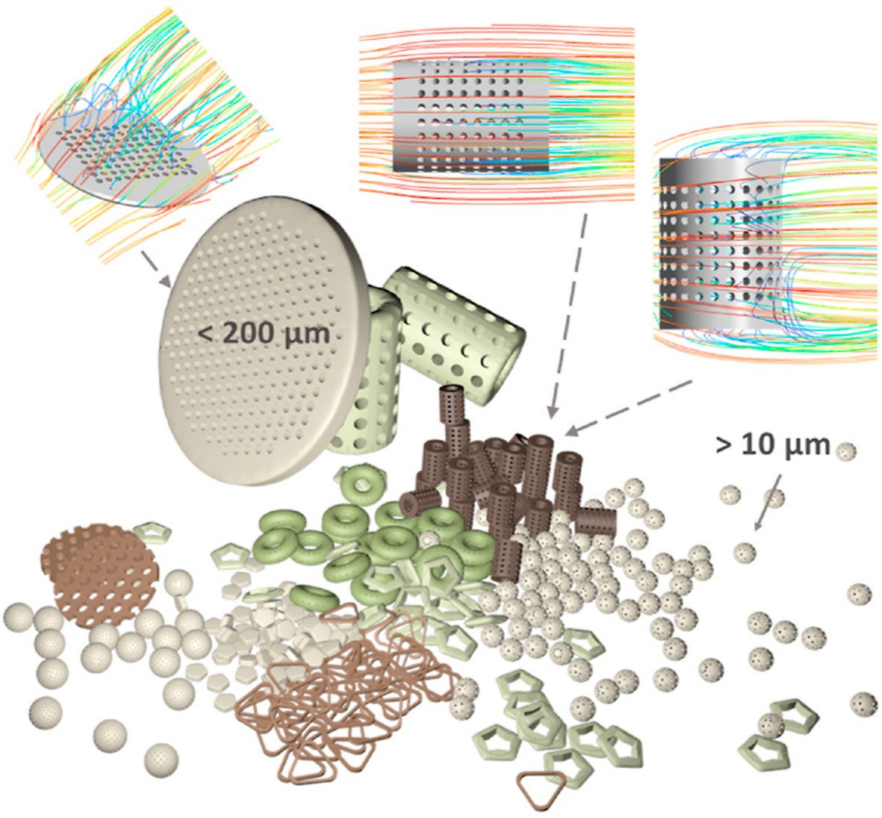


Surface reconstruction, modification and functionalization of natural diatomites for miniaturization of shaped heterogeneous catalysts

Bowen Li, Tian Wang, Qiujian Le, Runze Qin, Yuxin Zhang, and Hua Chun Zeng, *Nano Materials Science*

DOI: 10.1016/j.nanoms.2022.05.001

Abstract: Since the discovery of mesoporous silica in 1990s, there have been numerous mesoporous silica-based nanomaterials developed for catalytic applications, aiming at enhanced catalytic activity and stability. Recently, there have also been considerable interests in endowing them with hierarchical porosities to overcome the diffusional limitation for those with long unimodal channels. Present processes of making mesoporous silica largely rely on chemical sources which are relatively expensive and impose environmental concerns on their processes. In this regard, it is desirable to develop hierarchical silica supports from natural minerals. Herein, we present a series of work on surface reconstruction, modification, and functionalization to produce diatomite-based catalysts with original morphology and macro-meso-micro porosities and to test their suitability as catalyst supports for both liquid- and gas-phase reactions. Two wet-chemical routes were developed to introduce mesoporosity to both amorphous and crystalline diatomites. Importantly, we have used computational modeling to affirm that the diatomite morphology can improve catalytic performance based on fluid dynamics simulations. Thus, one could obtain this type of catalysts from numerous natural diatoms that have inherently intricate morphologies and shapes in micrometer scale. In principle, such catalytic nanocomposites acting as miniaturized industrial catalysts could be employed in microfluidic reactors for process intensification.

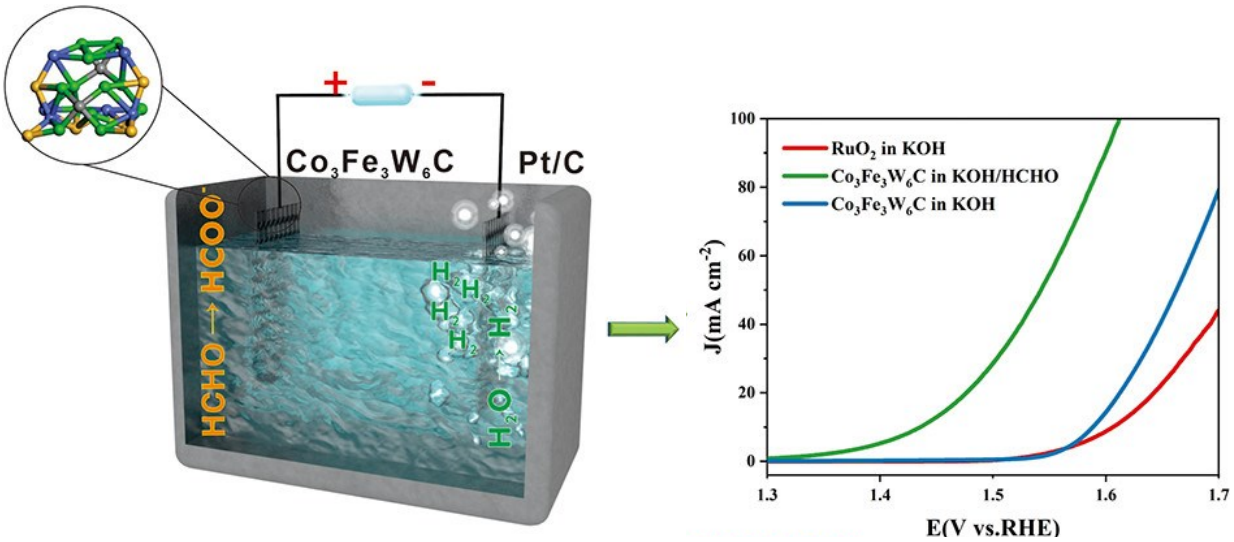


Boosting Electrocatalytic Hydrogen Evolution with Anodic Oxidative Upgrading of Formaldehyde over Trimetallic Carbides

Xiangbowen Du, Tong Wei, Mingwu Tan, Hisayoshi Kobayashi, Zhengxin Peng, Hongliang Zhu, Zhikang Jin, Junjie Song, Wen Liu, and Renhong Li, *ACS Sustainable Chemistry & Engineering*

DOI: 10.1021/acssuschemeng.2c01229

Abstract: Coupling the electrochemical oxidative upgrading of organic molecules with hydrogen evolution reaction could enable the energy-efficient production of H_2 from renewable electricity with simultaneous chemical production. This work shows that a trimetallic carbide ($Co_3Fe_3W_6C$), derived from one-pot synthesis, could act as a robust electrocatalyst for formaldehyde upgrading reaction (FUR) to produce formate at a high faradaic efficiency (>98%), without any production of CO_2 or O_2. Compared to OER, the input voltages of $Co_3Fe_3W_6C$-catalyzed FUR are 150 and 120 mV lower to achieve current densities of 10 and 50 mA cm^{-2}, respectively, thereby facilitating a significant boost in the energy efficiency of electrochemical H_2 production from water. Density functional theory calculations reveal that the trimetallic carbide system modulates the d band of the transition-metal active sites to achieve optimal adsorption toward the selective oxidation of formaldehyde, while suppressing the further formation of CO_2. $Co_3Fe_3W_6C$ was also found to be highly stable under considerably high-throughput electrochemical conditions in an alkaline electrolyte. This work offers a new strategy of synergizing water electrolysis with the oxidative upgrading of organic molecules to simultaneously boost the cost competitiveness of green hydrogen production and the electrochemical upgrading of organic feedstocks.



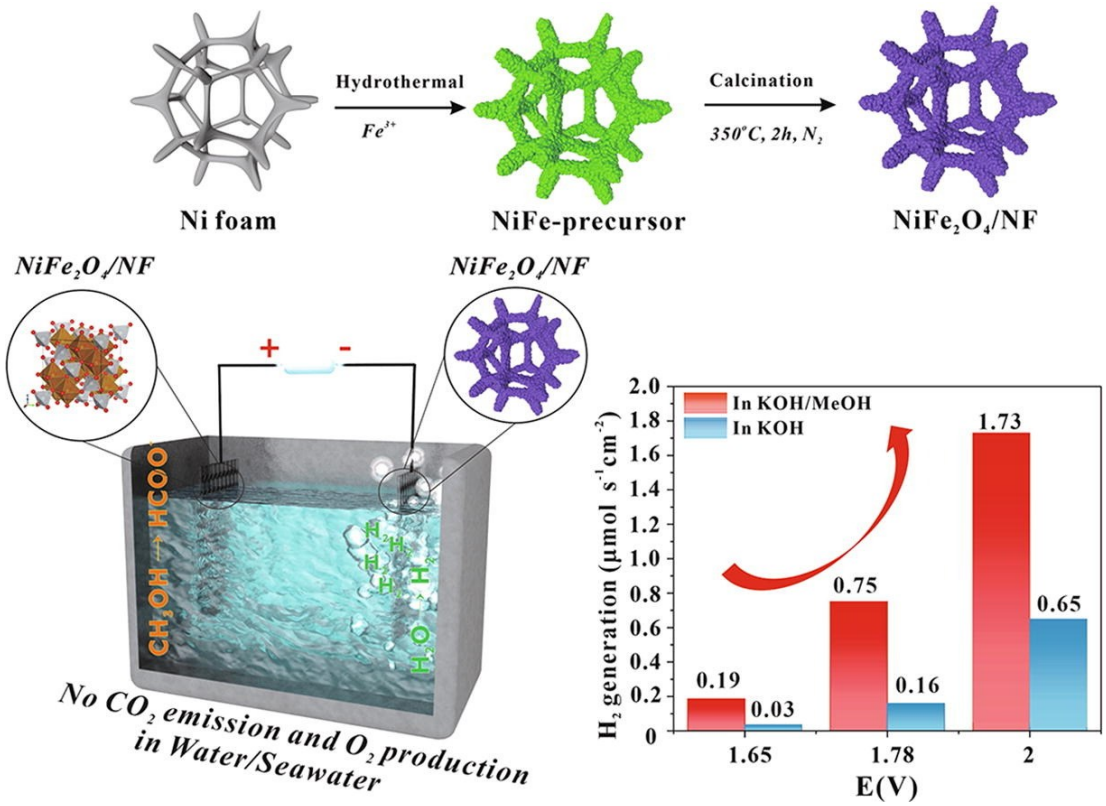
No CO_2 emission and O_2 production

Highly Efficient and Robust Nickel-Iron Bifunctional Catalyst Coupling Selective Methanol Oxidation and Freshwater/Seawater Hydrogen Evolution *via* CO-Free pathway

Xiangbowen Du, Mingwu Tan, Tong Wei, Hisayoshi Kobayashi, Junjie Song, Zhengxin Peng, Hongliang Zhu, Zhikang Jin, Renhong Li, and Wen Liu, *Chemical Engineering Journal*

DOI: 10.1016/j.cej.2022.139404

Abstract: The production of green hydrogen by water electrolysis is often kinetically limited by the sluggish oxygen evolution reaction (OER) at the anode. Here, we prepared a bifunctional nickel foam supported $NiFe_2O_4$ spinel catalyst (*i.e.* $NiFe_2O_4$/NF) that is capable of facilitating the coupling of hydrogen evolution reaction (HER) with selective methanol oxidation reaction (SMOR) in seawater to produce formate via a CO -free pathway. At a cell potential of 2.0 V, the $NiFe_2O_4$/NF||$NiFe_2O_4$/NF catalyzed HER-SMOR system produces remarkably high current density (>800 mA cm^{-2}) with high Faradaic efficiencies (FE) at both electrodes (> 96% for HER and >95% for SMOR to formate). The $NiFe_2O_4$/NF||$NiFe_2O_4$/NF HER-SMOR system also exhibits excellent stability over 48 h of continuous operation. With H_2 being the only gaseous product, the HER-SMOR electrolysis system could operate in the absence of a membrane. Furthermore, the $NiFe_2O_4$/NF||$NiFe_2O_4$/NF electrodes are sufficiently robust for continuously catalyzing HER-SMOR in seawater electrolysis, showing no sign of deactivation or chlorine oxidation reactions over 6 h of continuous operation at a high current density of 700 mA cm^{-2}. Mechanistic investigation and DFT calculations reveal that SMOR proceeds via a CO-free pathway, with Ni and Fe as the active sites for methanol and OH activation, respectively.



Other activities and achievements

Asst Prof Paul LIU (PI, NTU) presented a talk (virtual) titled "CO_2 Hydrogenation to Methanol on Tungsten-Doped Cu/CeO_2" at the CO_2 Activation Workshop hosted by UK Catalysis Hub.

Prof Manish CHHOWALLA (Co-PI, CAM) was elected as a Fellow of the Royal Academy of Engineering.

Asst Prof Tej CHOKSI's (Co-PI, NTU) group gave in-person presentations at the conferences listed below:

- 27th North American Catalysis Society Meeting in New York from 22 - 27 May 2022, presenting "Designing Stable, Active, and Selective Low-Dimensional Gold-Heterostructures for CO_2 Reduction to C_1 Products" and "Uncertainty Quantification of Stability Metrics of Nanoparticle Catalysts from First Principles".
- 9th Singapore Catalysis Society Forum on 26 May 2022, presenting "Generalized Principles for Designing Bifunctional Supported Metal Nanoparticles".
- 264th American Chemical Society National Meeting in Chicago from 21 - 25 August 2022, presenting "Generalized Design Principles for Determining the Stability and Reactivity of Supported Gold Catalysts"
- 2nd International Conference on Materials for Humanity in Singapore from 19 - 21 September 2022, presenting "Designing Stable, Active, and Selective Low-Dimensional Gold-Heterostructures for CO_2 Reduction to C_1 Products" and "Uncertainty Quantification of Stability Metrics of Nanoparticle Catalysts from First Principles".

Dr Mingwu TAN (Research Fellow, NTU) won 2nd prize for the poster presentation titled "Hydrogen spillover assisted by oxygenate molecules over nonreducible oxides" at the 9th Singapore Catalysis Society Forum on 26 May 2022.

Mr Adarsh ARUN (PhD student, CAM) won the Early Career Research Talks prize sponsored by Digital Discovery (Royal Society of Chemistry) for presenting his work "Reaction impurity prediction using a data mining approach" at the 5th Machine Learning and AI in Bio(Chemical) Engineering Conference hosted by the University of Cambridge.

Mr Aniket CHITRE (PhD student, CAM) won Best Oral Presentation Award for the talk "Surfactants Featurisation and Machine Learning Driven Exploration of Personal Care Formulations" at the 2nd International Conference on Materials for Humanity in Singapore from 19 - 21 September 2022. He also attended the ML/AI in Formulations and Formulating Colloids conference days hosted by the Society of Chemical Industry (SCI) in London, 25 - 26 April 2022.

Mr Chitre's PhD is also funded by BASF Shanghai and his work is in close collaboration with BASF's Formulations, Molecular Modelling & Digitalization of R&D groups. He is working with formulations ingredients which BASF has agreed to publish an open-source dataset with, including the molecular structures. While the pH project could lead to a technical invention, he may only disclose limited details for now. He is also on exchange at IMRE, A*STAR under Prof Kedar HIPPALGAONKAR'S (Non-C4T PI, NTU) group.

IRP 2

ELECTROSYNTHETIC PATHWAYS FOR ADVANCED LOW-CARBON CHEMICAL MANUFACTURING

In IRP 2, low carbon electrosynthetic processes and technologies are developed which target local, on-scale and on-demand conversion of electricity to commodity or specialty chemicals. As the contribution of renewables to the total electricity generation capacity continues to grow, novel technological opportunities arise for direct chemical conversion of the newly available low carbon electrons. This project addresses core challenges to the implementation of low carbon, on-demand driven advanced manufacturing of chemical targets via electrosynthesis.

IRP 2 Principal Investigators:

Professor Adrian FISHER
University of Cambridge

Professor WANG Xin
Nanyang Technological University

Asst Prof ZHANG Sui
National University of Singapore

OVERVIEW

Research in the IRP2 team targets new low carbon manufacturing routes using electrosynthesis for applications in the production of future fuels, clean water treatment methods and next generation agrochemicals. In this reporting period work has delivered a new integrated model approach to synthesiser development, next generation tools for inline frequency-time analysis for industrial chemical manufacture and novel membrane structures, electrode architectures and catalytic approaches for electrosynthetic pathways.

Dr Chencheng Dai and Dr Libo Sun's research in IRP2 focused on electrode architectures and synthesis reactors. In this reporting period they have been exploring potential electrochemical energy vectors such as alcohols for future fuel applications. In one study the effect of the hydroxyl group position on the electro-oxidation of butanediols, has been explored for selectivity and yield optimisation. The effect of the hydroxyl group position in butanediols on their electro-oxidation reactivities was investigated by advanced voltammetry techniques. Further work targets a rational method to manipulate the spin state and optimise the catalytic ability of materials. Work has focused on the molecular complex nickel phthalocyanine and activity for CO_2 reduction. In depth analysis on the investigation into the role of spin state of active centres in the electrocatalytic process is reported.

Asst Prof Sui Zhang's research in IRP2 focused on engineering membranes for electrochemical device applications. In particular research has focused on the development of self cleaning and disinfecting structures by the incorporation of electrocatalytic materials within the electrode membranes. These catalytic structures can be electrically activated driving redox chemistry within the solution to induce chemical cleaning. In this reporting period, work has targeted precision molecular sieving in harsh organic solvents. These pioneering studies offer unprecedented opportunities for widening the application of organic solvent nanofiltration (OSN). Her work has introduced tris(4-carbazoyl-9-ylphenyl)amine (TCTA) as the monomer to form robust conjugated microporous polymer (CMP) membranes via electrochemical polymerisation.

Ms Freyja Björk Dagbjartsdóttir has extended her research in the field of reactor engineering, in collaboration with one of our industrial partners (Syngenta), to develop new methods for analysis and assessment of the sensitivity of electrochemical parameters for electrolysis reactions. The complexity of parameters, ranging from geometric design to operating conditions, raises considerable challenges for optimising the performance of an electrochemical system. During this reporting period, a mathematical model has been built using a finite element method. Monte Carlo Simulations have been performed with 14 stochastic parameters as input. Different types of sensitivity analysis were then conducted by varying an individual parameter (One-Factor-at-a-time Method) to investigate the influence of each key input parameters in electrolysis systems. Rankings in terms of cell performance are given, based on different sensitivity analysis methods. Rankings are produced by One-Factor-at-a-time Method. Operating temperature, the charge transfer coeffi-

cient for the Oxygen Reduction Reaction and porosity of the catalyst layer are the top 3 parameters in both methods. The Scatter Plots method indicates that operating temperature and the charge transfer coefficient for Oxygen Reduction Reaction are two of the key influential parameters.

Alongside sensitivity analysis new analytical tools have been developed to allow frequency-time domain characteristics of electrolysers to be monitored in realtime. Three approaches have been developed using different transformation methods (Hilbert-Huang (HHT), (continuous) wavelet (CWT) and synchrosqueezing transforms (SST)). Each approach has been computationally and comparatively investigated.

IRP 2 Singapore-based start-up company Datum ElectroniX, which was launched by Dr Kamal Elouarzaki and Prof Adrian Fisher, has also progressed with a range of new industrial collaborations underway.

Prof Adrian Fisher, PI
University of Cambridge

Update on work package 2.1

Advanced electrode architectures

Dr Chencheng DAI (Research Fellow, NTU) studied the effect of the hydroxyl group position on the electro-oxidation of butanediols, including 1,2-butanediol, 2,3-butanediol, 1,3-butanediol, and 1,4-butanediol. The effect of the hydroxyl group position in butanediols on their electro-oxidation reactivities is investigated by cyclic voltammetry, linear sweep voltammetry, chronopotentiometry and chronoamperometry in 1.0 M KOH. The results show that the closer the two hydroxyl groups are, the higher the reactivity and the lower the anodic potential butanediol has. Moreover, the oxidation products from chronoamperometry are analysed by means of HPLC and NMR. Some value-added products, such as 3-hydroxypropionic acid/3-hydroxypropionate, are produced. The DFT calculations indicate that the oxidation of vicinal diols responds to the conversion from a hydroxyl group to a carboxylate group, followed by C–C bond cleavage, where the carbon charge decreases. These results provide an insight into reactant selection for the electrochemical synthesis of value-added chemicals.

Dr Dai has also been working on the electrochemical production of ammonia from nitrate reduction reaction (NO_3RR) as a green and decentralised supplement to the traditional Haber-Bosch process. He has obtained a maximal ammonia faradaic efficiency (FE) of ~ 93% at -0.35 V compared to a reversible hydrogen electrode with an ammonia yield rate (YR) of ~ 5.9 mg h^{-1} mg_{cat}^{-1}. The ^{15}N isotope labelling experiment suggests that the ammonia is generated from nitrate ions but not other resources. The catalyst also shows excellent stability as the ammonia FE and YR in 20 consecutive electrolysis cycles under the optimal ammonia selectivity reaction condition remain stable.

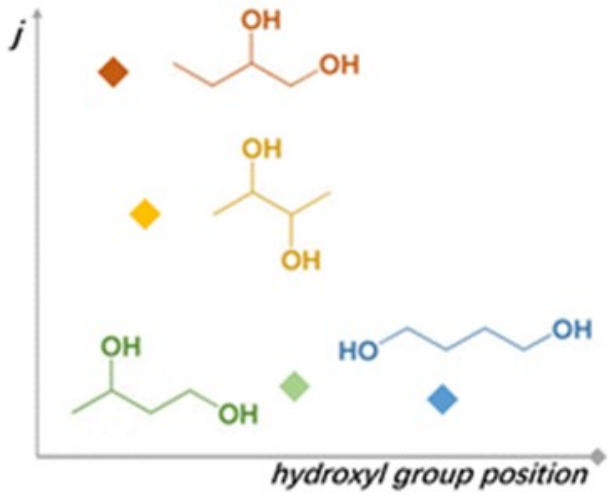


Figure 2.1: In the electro-oxidation of butanediols, closer hydroxyl groups lead to higher electrochemical reactivities. Vicinal hydroxyl groups facilitate the formation of a carboxylate group, and the following C–C bond cleavage.

Dr Chencheng DAI

Ms Yanqiu LU (Non-C4T PhD student, NUS) focuses on how engineering membranes for precise molecular sieving in harsh organic solvents offer unprecedented opportunities for widening the application of organic solvent nanofiltration (OSN). In her work with **Asst Prof Sui ZHANG (PI, NUS),** she introduced tris(4-carbazoyl-9-ylphenyl)amine (TCTA) as the monomer to form robust conjugated microporous polymer (CMP) membranes via electrochemical polymerisation for OSN in a challenging environment. The method enables the use of rigid CMP membranes in harsh dimethylformamide (DMF) with temperatures up to 100°C. The resulting membranes show size-dependent selectivity towards charged dyes and pharmaceutical molecules. Moreover, the membranes present excellent stability in DMF solution containing a high base content of triethylamine at different temperatures due to their rigid crosslinked chemical structure. The optimal membrane can reach 94.4 ± 2.2% rejection of Allura Red AC (496.42 g mol-1) at a dimethylformamide (DMF) permeance of 33.1 ± 1.2 L m-2 h-1 bar-1 at 100 °C. To the best of our knowledge, this is the first report to experimentally recognise the unique advantages of CMPs in high-temperature OSN applications.

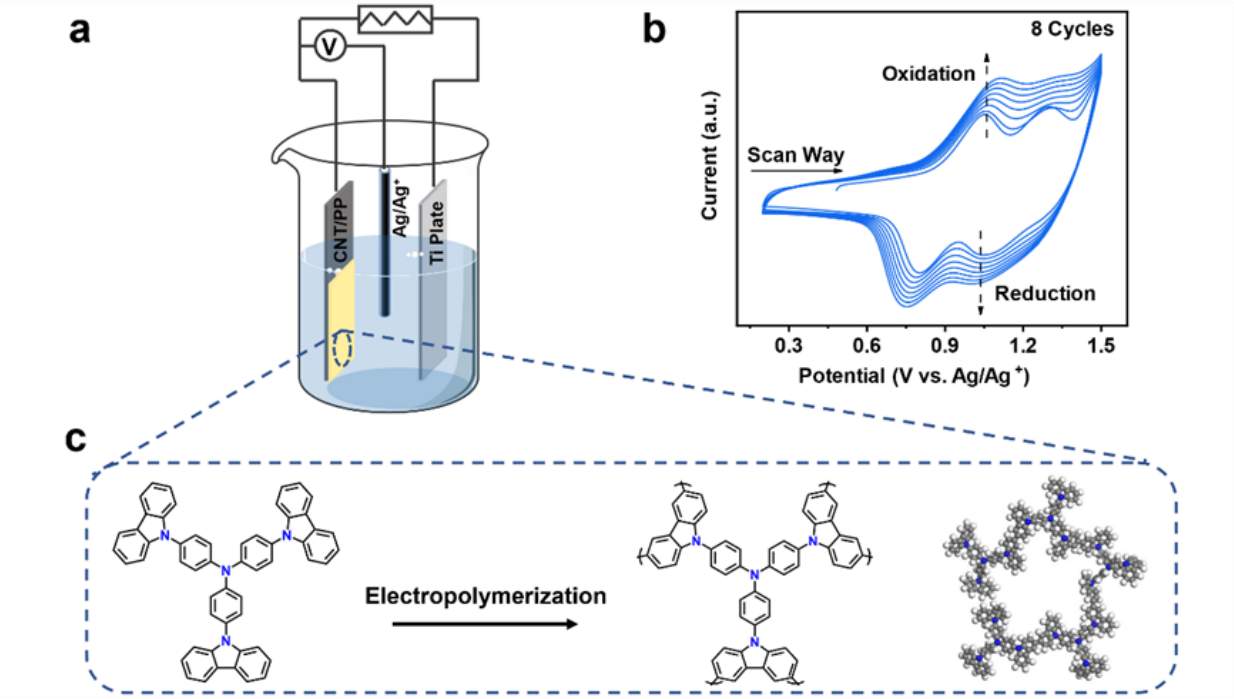


Figure 2.2: (a) An illustration of the three-electrode electrochemical cell used for TCTA polymerisation on the CNT/PP film. (b) Multicycle cyclic voltammetry profiles of TCTA with the scanning potential from 0.2 to 1.5 V vs. Ag/Ag+, at a scan rate of 50 mV s-1. (c) A schematic representation of the electrochemical polymerisation of TCTA-EP films and their elementary pore structures.

Ms Yanqiu LU

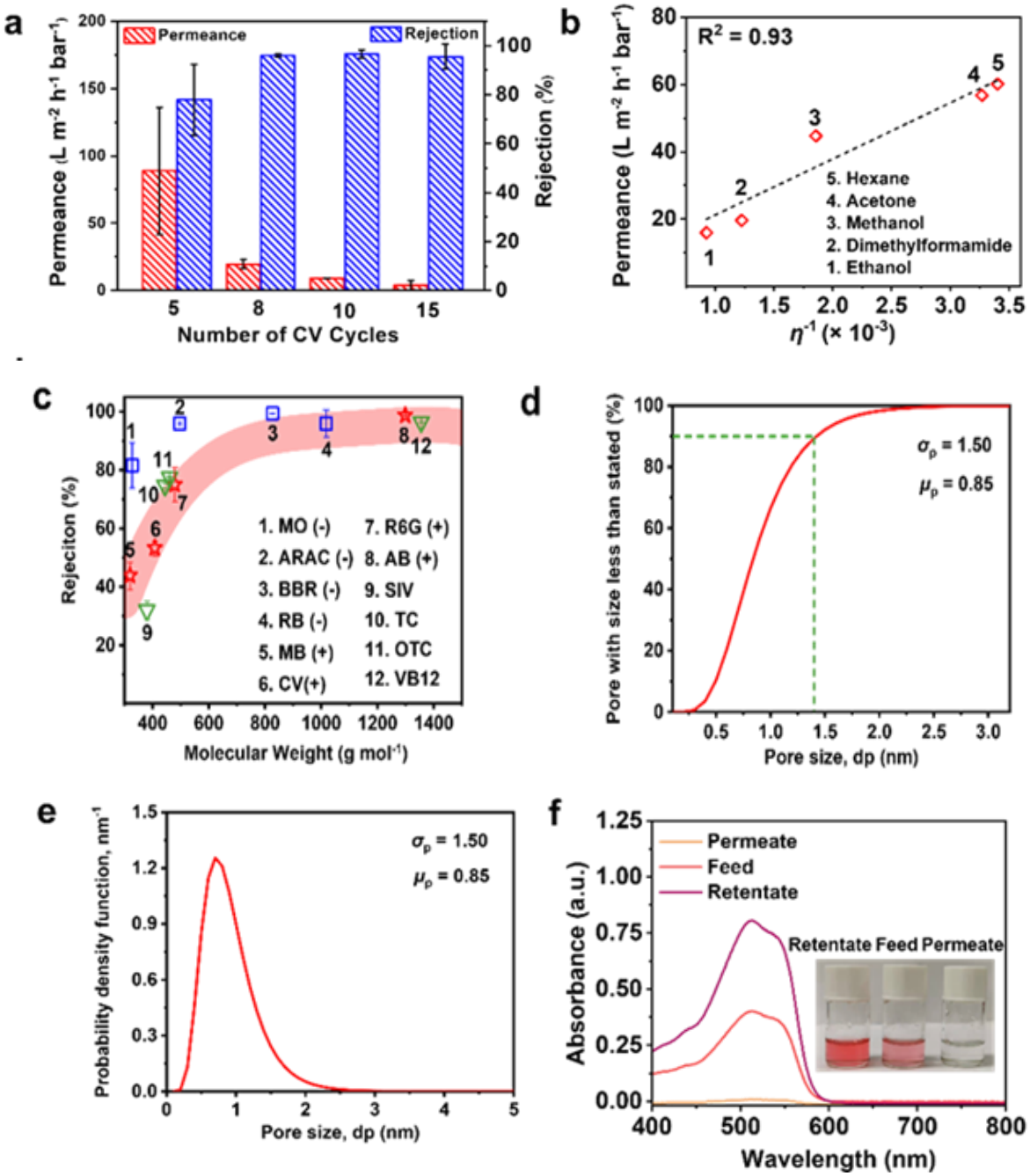


Figure 2.3: (a) Rejection and permeance of TCTA-EP films with different CV cycle number. The feed solution was 50 ppm Allura Red AC (MW = 496.42 g mol⁻¹) in DMF. (b) The organic solvent permeance of TCTA-EP-8 membrane against the inverse of viscosity. (c) Rejection of the TCTA-EP-8 film towards different dyes and pharmaceutical molecules in DMF. (d) Cumulative pore size distribution curves and (e) probability density function curves of the TCTA-EP-8 membrane. (f) UV absorption spectra of the feed, permeate and retentate after 50 ml permeate was collected from the 100 ml feed with TCTA-EP-8 membrane. The feed solution was 50 ppm Allura Red AC (MW = 496.42 g mol⁻¹) in DMF.

Ms Yanqiu LU

Update on work package 2.2

Co-generation and electrolytic synthesis reactor engineering

Dr Libo SUN (Research Fellow, NTU) reports that spin state of metal species plays a key role in affecting the activity of electrocatalytic reactions. However, developing a rational method to manipulate the spin state and optimise the catalytic ability of materials still remains a great challenge. While the molecular complex nickel phthalocyanine shows poor advantages, single-atom catalysts with nickel element could exhibit excellent activity towards CO_2 reduction. This prompts the investigation into the role of spin state of active centres in the electrocatalytic process. Molecular complexes may allow the spin state of the chelated metal species to be regulated by surrounding functional groups. Herein, several nickel phthalocyanine derivatives with regulated spin state of nickel centres were fabricated. Among them, the high-spin-state nickel phthalocyanine (NiDMAPc) exhibits the best activity than the others. Neat 100% selectivity towards CO production over a wide potential range could be obtained for NiDMAPc with higher current density of nearly 5 times than NiPc at -0.85 V compared to RHE in traditional H-type cell tests. Furthermore, the flow cell setup measurements of NiDMAPC exhibit remarkable stability and selectivity (>99%) for more than 40 hours. This research may disclose the key role of spin state in electrocatalytic reactions and help deepen our understanding of its mechanisms.

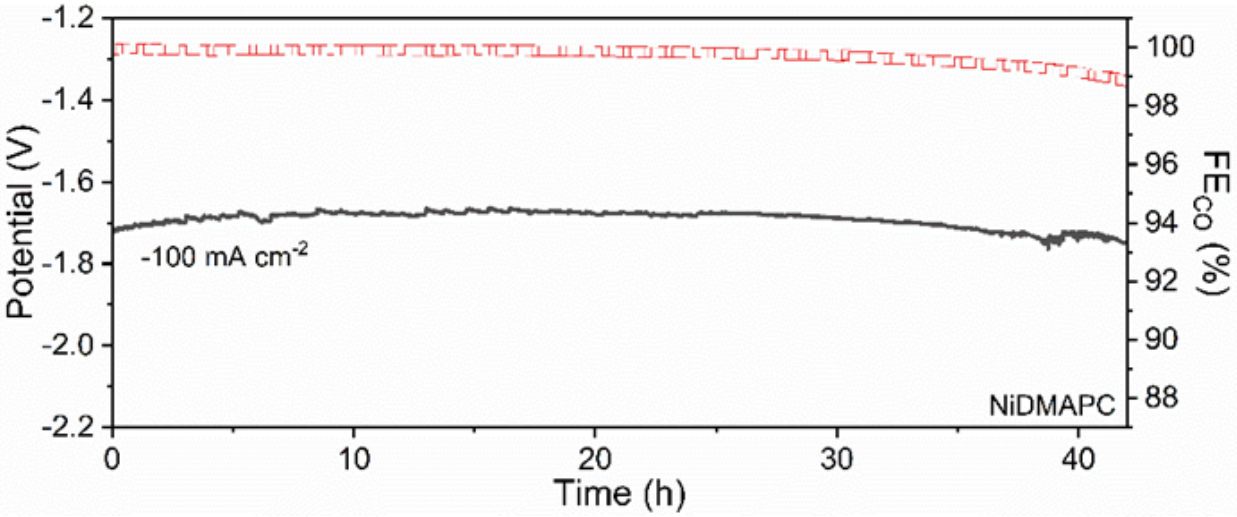


Figure 2.4: The long-term stability test of NiDMAPC under -100 mA cm^{-2} in a flow cell setup.

Dr Libo SUN

Update on work package 2.3

Micro-variable pressure and temperature electrosynthesis plant

Ms Freyja Björk DAGBJARTSDÓTTIR's (PhD student, CAM) research interests lie in investigating novel electrochemical systems where a complex relationship exists between chemistry and mass transport. The aim is to make mathematical descriptions of electrochemical systems that can be used to investigate, design, and monitor these systems.

One system of interest is the direct electrochemical synthesis of hydrogen peroxide, H_2O_2. It has been demonstrated in experiments that it is possible to create up to 20% w/w hydrogen peroxide in water solution using direct electrolysis and the aim is to build a device that can produce enough hydrogen peroxide at low cost on site for users by just using electricity, water and oxygen.

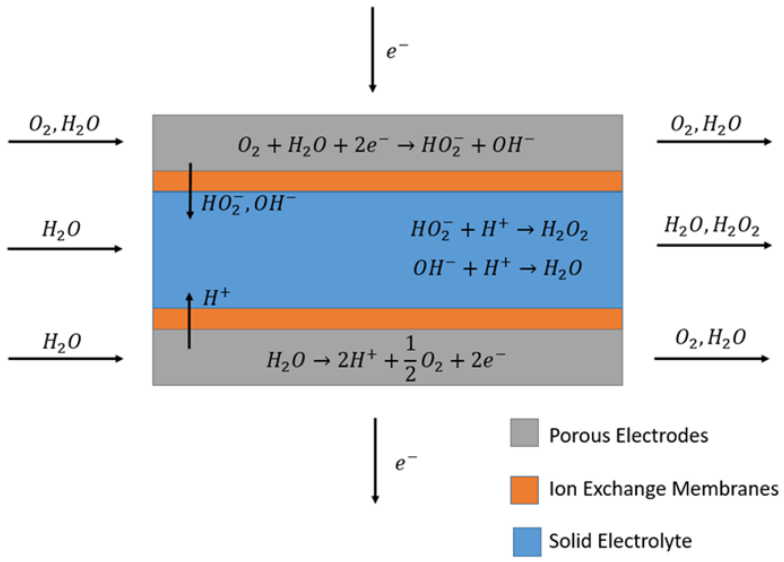


Figure 2.5: A schematic of chemical and current flow in the H_2O_2 electrolyser

Ms Freyja Björk DAGBJARTSDÓTTIR

Ms Dagbjartsdóttir has been building a model describing this system by first building a base case and conducting a sensitivity analysis on this base case, abiding by the following principles: (1) moderate manufacturing and running costs (2) convenience and robustness, (3) accessibility to control and monitor the performance, (4) simplicity and versatility, (5) feasibility for future updates. The following steps were used in the design approach:

- Build a mathematical model of direct electrosynthesis of a hydrogen peroxide device
- Discover influential parameters by performing Monte Carlo Simulations
- Quantify the effects of input parameters on output
- Rank the parameters using Sensitivity Analysis and therefore optimise the device in terms of cell performance, conversion efficiency, product concentration, etc.

The indicators or the outputs of interest for the system are cell performance, conversion efficiency and concentration of H_2O_2.

Monte Carlo Simulation (MCS) were introduced for capturing the deviations that would occur in real systems and providing optimal design on this basis. A MCS of the parameter set was developed to generate random samplings as inputs to predict or quantify numerical outcomes. Table 1 shows the system characterisation for the H_2O_2 reactor system.

Numerical characterisation of the polarisation curve and H_2O_2 concentration are shown in Figure 2.6 as the applied cell voltage is changed from 0.4 V to -0.6 V, the total current density becomes larger which means the rate of this electrochemical reaction is increasing.

Parameter	Symbol (X_i)	Mean Value (μ_i)	Unit	RSD (σ_i/μ_i)
Geometry				
Channel length	L	2×10^{-2}	m	5%
Channel width	W	5×10^{-3}	m	5%
Operating conditions				
Temperature	T	298	K	5%
Inlet H_2 flow rate	Q_{H_2}	50	sccm	5%
Inlet O_2 flow rate	Q_{O_2}	50	sccm	5%
Material Properties				
Porosity of the gas diffusion layer [23]	ϵ_{gdl}	0.8	-	5%
Porosity of the catalyst layer [51]	ϵ_{cl}	0.6	-	5%
Electrical conductivity of the gas diffusion layer [34]	σ_{gdl}	250	S m^{-1}	5%
Electrical conductivity of the catalyst layer [15]	σ_{cl}	90	S m^{-1}	5%
Electrical conductivity of the solid electrolyte [33]	σ_{se}	0.27	S m^{-1}	5%
Electrochemistry				
Charge transfer coefficient for HOR [29]	α_{H_2}	0.47	-	5%
Charge transfer coefficient for ORR [54]	α_{O_2}	0.61	-	5%
Exchange current density for H_2 [25] &	$i^{ref}_{H_2}$	1.03	A m^{-2}	5%
Exchange current density for O_2 [38]	$i^{ref}_{O_2}$	3×10^{-5}	A m^{-2}	5%

Table 1: Stochastic parameters used in simulation with means and deviations.

Ms Freyja Björk DAGBJARTSDÓTTIR

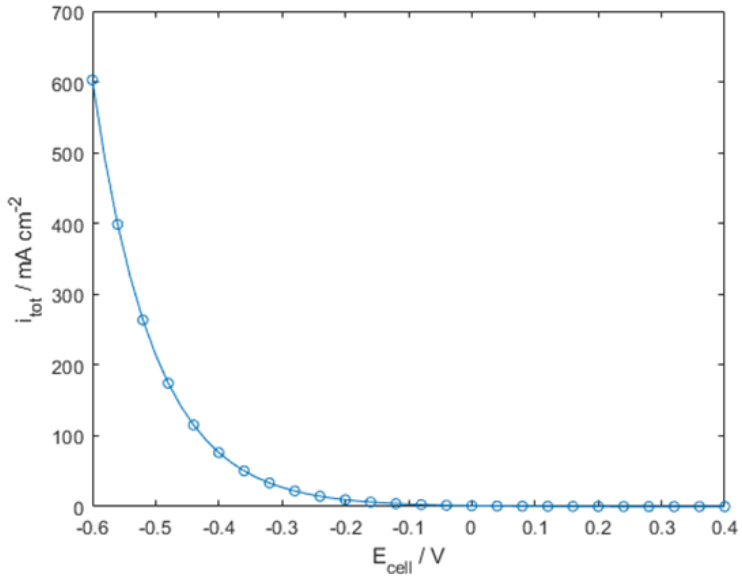


Figure 2.6: Polarisation curve of direct H_2O_2 electrosynthesis cell with same parameters as the mean values in Table 1.

Ms Freyja Björk DAGBJARTSDÓTTIR

The dependence of H_2O_2 concentration on the outlet boundary layer on the deionised water flow rate is shown in Figure 2.7.

With the data generated by MCS, the one factor at a time method was conducted by examining the effect of all the input parameters individually in terms of cell performance. Each stochastic parameter has a sample size of 200, subject to the One-sample Kolmogorov-Smirnov test. And after that, every parameter is ranked based on its coeffi-

cients of variation. To be concise, a ranking of the first 8 parameters is given in decreasing order as shown in Figure 2.8.

Despite the significant role the operating temperature plays in cell performance, it is advised not to optimise the device by adjusting the operating temperature for the sake of stability. Therefore, optimisation by changing the charge transfer coefficient for ORR is recommended as far as this setting is concerned.

Ms Dagbjartsdóttir has also been collaborating with a MPhil student Mr Leopold KLOYER (Non-C4T student, CAM) in the IRP2 research thrust on advanced analytical tools for inline electrochemical control and real time monitoring.

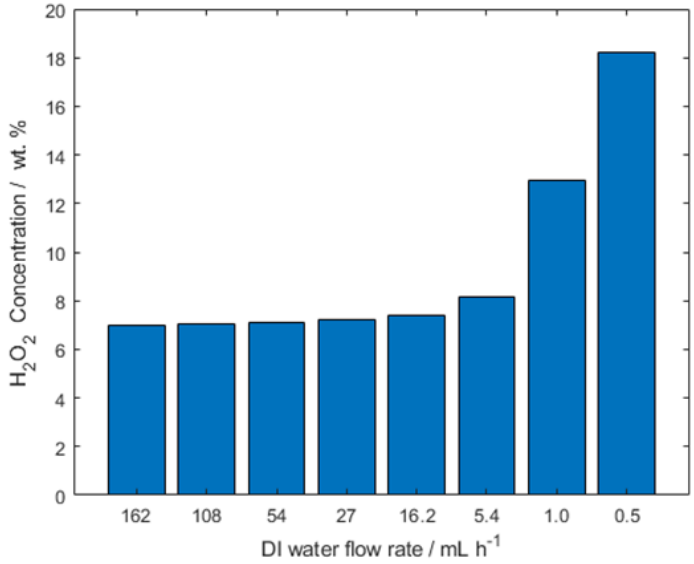


Figure 2.7: Dependence outlet H_2O_2 concentration on the deionised water flow rate at E_{cell} = −0.5V.

Ms Freyja Björk DAGBJARTSDÓTTIR

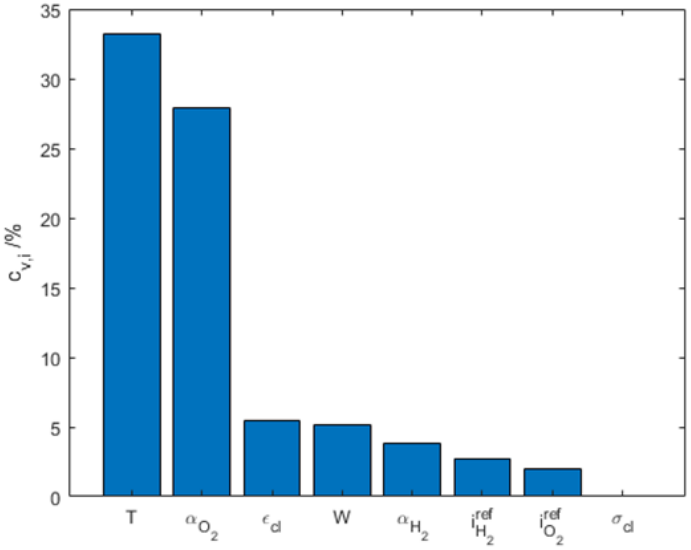


Figure 2.8: Coefficients of variation of the cell performance under different input parameters at E_{cell} = −0.5 V, ranked in decreasing order. Top 8 parameters are T (temperature), a_{O2} (charge transfer coefficient for ORR), ε_{cl} (porosity of the catalyst), W (channel width), a_{H2} (charge transfer coefficient for HOR), i^{ref} (exchange current density for H_2 and O_2), and σ_{cl} (conductivity of the catalyst layer).

Ms Freyja Björk DAGBJARTSDÓTTIR

They have extended this topic to explore advanced tools which allow a time-frequency response to be analysed, in comparison to the frequency only domain output achieved through the Fourier Transform AC Voltammetry approach. Three approaches were addressed using different transformation methods which remain unexploited in the field of electrolysis: (Hilbert-Huang (HHT), (continuous) wavelet (CWT), and synchrosqueezing transforms (SST)). The approaches were investigated for the analysis of electrochemical data to identify oscillatory subcomponents, which most accurately represent possible underlying physical processes occurring in electrochemical synthesis cells. We anticipate these new tools will allow development of an advanced computer models and analysis of physical/ chemical processes occurring during electrolysis.

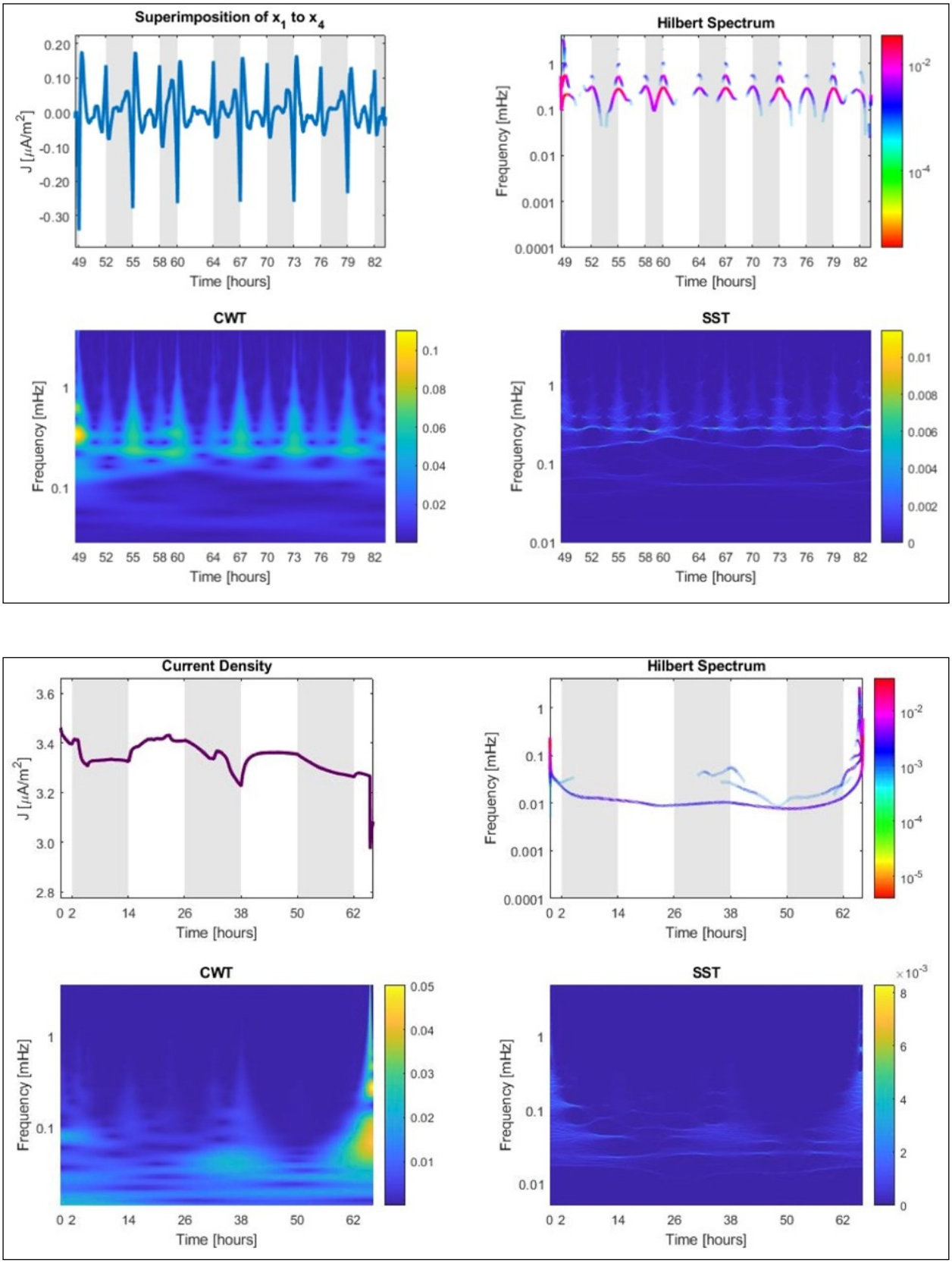


Figure 2.9: Current density profile, Hilbert spectrum, CWT and SST for the large amplitude perturbation in an electrochemical cell (top) and decaying electrochemical signal in a synthesis cell (bottom).

Ms Freyja Björk DAGBJARTSDÓTTIR

Scientific output

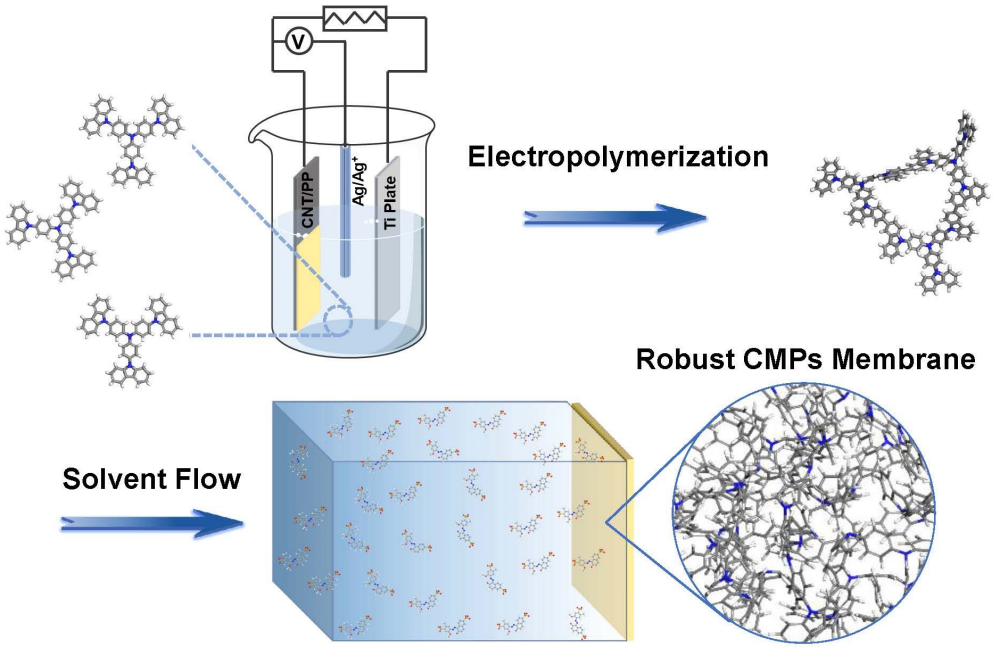
The following are the CREATE-acknowledged publications generated by IRP 2 during the reporting period, excluding those already featured in the Scientific Highlights section on page 14.

Electropolymerized thin films with a microporous architecture enabling molecular sieving in harsh organic solvents under high temperature

Yanqiu Lu, Wei Liu, Kaiyu Wang, and Sui Zhang, *Journal of Materials Chemistry A*

DOI: 10.1039/D2TA02178A

Abstract: Engineering membranes for precise molecular sieving in harsh organic solvents offer unprecedented opportunities for widening the application of organic solvent nanofiltration (OSN). Here, we introduce tris(4-carbazoyl-9-ylphenyl)amine (TCTA) as the monomer to form robust conjugated microporous polymer (CMP) membranes *via* electrochemical polymerization for OSN in a challenging environment. The method enables the use of rigid CMP membranes in harsh dimethylformamide (DMF) with temperature up to 100 °C. The resulting membranes show size-dependent selectivity towards charged dyes and pharmaceutical molecules. Moreover, the membranes present excellent stability in DMF solution containing a high base content of triethylamine at different temperatures due to their rigid crosslinked chemical structure. The optimal membrane can reach 94.4 ± 2.2% rejection of Allura Red AC (496.42 g mol^{-1}) at a dimethylformamide (DMF) permeance of 33.1 ± 1.2 L m^{-2} h^{-1} bar^{-1} at 100 °C. To the best of our knowledge, this is the first report to experimentally recognize the unique advantages of CMPs in high-temperature OSN applications.



The 2022 solar fuels road map

Gideon Segev, Jakob Kibsgaard, Christopher Hahn, Zhichuan J Xu, Wen-Hui (Sophia) Cheng, Todd G Deutsch, Chengxiang Xiang, et al, *Journal of Physics D: Applied Physics*

DOI: 10.1088/1361-6463/ac6f97

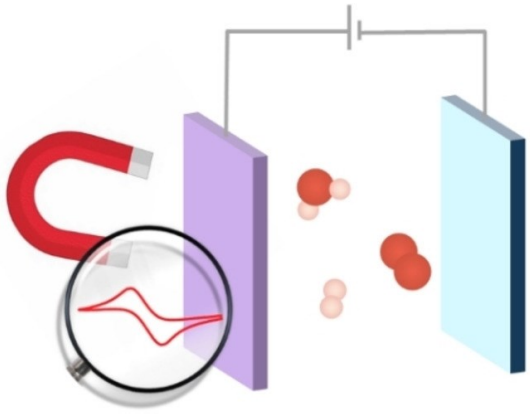
Abstract: Renewable fuel generation is essential for a low carbon footprint economy. Thus, over the last five decades, a significant effort has been dedicated towards increasing the performance of solar fuels generating devices. Specifically, the solar to hydrogen efficiency of photoelectrochemical cells has progressed steadily towards its fundamental limit, and the faradaic efficiency towards valuable products in CO_2 reduction systems has increased dramatically. However, there are still numerous scientific and engineering challenges that must be overcame in order to turn solar fuels into a viable technology. At the electrode and device level, the conversion efficiency, stability and products selectivity must be increased significantly. Meanwhile, these performance metrics must be maintained when scaling up devices and systems while maintaining an acceptable cost and carbon footprint. This roadmap surveys different aspects of this endeavor: system benchmarking, device scaling, various approaches for photoelectrodes design, materials discovery, and catalysis. Each of the sections in the roadmap focuses on a single topic, discussing the state of the art, the key challenges and advancements required to meet them. The roadmap can be used as a guide for researchers and funding agencies highlighting the most pressing needs of the field.

Electrochemistry in Magnetic Fields

Songzhu Luo, Kamal Elouarzaki, and Zhichuan J. Xu , *Angewandte Chemie International Edition*

DOI: 10.1002/anie.202203564

Abstract: Developing new strategies to advance the fundamental understanding of electrochemistry is crucial to mitigating multiple contemporary technological challenges. In this regard, magnetoelectrochemistry offers many strategic advantages in controlling and understanding electrochemical reactions that might be tricky to regulate in conventional electrochemical fields. However, the topic is highly interdisciplinary, combining concepts from electrochemistry, hydrodynamics, and magnetism with experimental outcomes that are sometimes unexpected. In this Review, we survey recent advances in using a magnetic field in different electrochemical applications organized by the effect of the generated forces on fundamental electrochemical principles and focus on how the magnetic field leads to the observed results. Finally, we discuss the challenges that remain to be addressed to establish robust applications capable of meeting present needs.

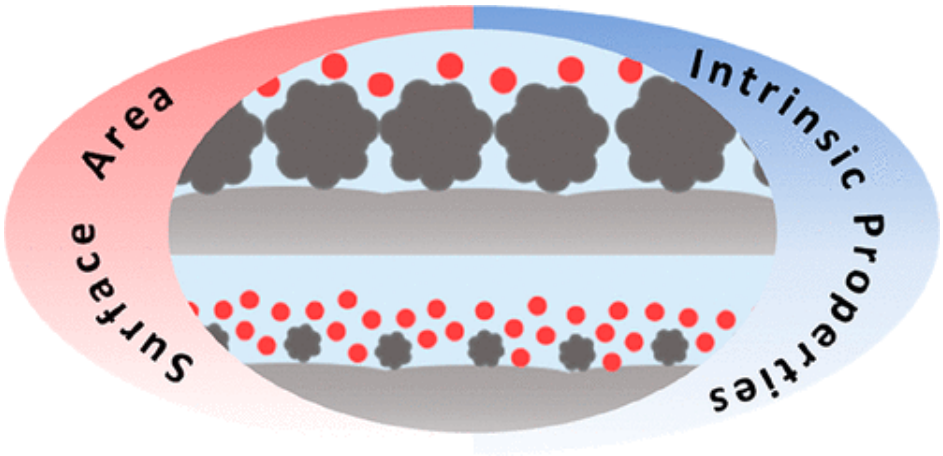


Size Effects of Electrocatalysts: More Than a Variation of Surface Area

Tianze Wu, Ming-Yong Han, and Zhichuan J. Xu, *ACS Nano*

DOI: 10.1021/acsnano.2c04603

Abstract: The efficiency of electrocatalytic reactions has been continuously improved in recent years due to the great effort in the development of electrocatalysts. A popular strategy is engineering the size of electrocatalysts for better electrochemical performance and lower cost. Nanosized electrocatalysts with high specific surface area have been widely used in state-of-the-art electrochemical devices such as fuel cells. From an engineering aspect, nanosizing electrocatalysts increases the surface area of the electrode and improves the electrode/device performance. Beyond an engineering scope, this perspective highlights the size effects of certain scientific fundamentals in electrocatalytic reactions. The paper summarizes the representative examples in studying the size effects of electrocatalysts and sheds light on the change of intrinsic properties of electrocatalysts caused by the size variation. The size effects of electrocatalysts should be investigated in terms of both engineering and fundamental aspects; that is, the observed activity change is more than a result of surface area variation, and it is interesting to investigate the link between the intrinsic activity and the properties of the catalysts.



Other activities and achievements

Asst Prof Sui ZHANG (PI, NUS) was awarded the AIChE SLS Outstanding Young Faculty Award and a Faculty Research Award by NUS College of Design and Engineering

Prof Zhichuan Jason XU (Co-PI, NTU) joins the journal *EES Catalysis* as an Associate Editor.

Ms Freyja Björk DAGBJARTSDÓTTIR's (PhD student, CAM) PhD is jointly funded by CARES and Syngenta.

The work in IRP 2 aims to develop novel electrocatalytic routes to produce cleaner synthesis of specialty chemicals for the chemical industry.

IRP 3

COMBUSTION FOR CLEANER FUELS AND BETTER CATALYSTS

To formulate the fuel of the future, IRP 3 looks at new molecules that can be produced within the techno-economic constraints of a refinery and that have the potential to reduce pollutant emissions when added to fossil-derived fuels. This research will help to identify the best fuels (or fuel mixtures) for low-emission energy conversion, and to design and manufacture optimised cost-effective nanostructured materials for catalysis.

IRP 3 Principal Investigators:

Professor Markus KRAFT
University of Cambridge

Professor XU Rong
Nanyang Technological University

Assoc Professor YANG Wenming
National University of Singapore

OVERVIEW

In the present reporting period, we have continued and broadened our investigations into oxygenated fuels as additives to diesel and jet fuels and their role in mitigating the formation of soot in internal combustion engines. We have considered various additives including poly (oxymethylene) dimethyl ether (PODE), ethanol and others. For comparative studies, we ensured the same amount of oxygen in each experiment in order to separate the effect of mere presence of oxygen from the effect of chemical structure of the additive. While previously we had conducted such studies only in laboratory flame setups, we can now perform particle size distribution measurements in the exhaust stream of a compression ignition engine. In agreement with previous work, we have found that ethanol is most effective in reducing particulate emissions, although this depends on engine load.

Furthermore, we have continued our research on the electrocatalytic reduction of CO_2 into useful products such as fuels and other chemicals both experimentally and computationally. In the lab, we have studied the properties of a commercial gas diffusion layer that we have previously coated with flame-synthesised copper oxide nanoparticles using our own rig. We found that increasing the hydrophobicity of the catalytic film extends its lifespan. At the same time, we are also continuing to investigate the microkinetic mechanism underlying electrochemical CO_2 reduction. Having previously developed a detailed kinetic model, we have now managed to experimentally identify numerous additional minor products of the process, which can help us to refine our detailed model even further and thus deepen our understanding.

Professor Markus Kraft, PI
University of Cambridge

Update on work package 3.1

Refinery, fuel and engine of the future — experimental

Properties of surrogate fuels, marine engine after-treatment

Mr Yong Ren TAN (PhD student, CAM) is currently investigating the impact of blending oxygenated fuels with Jet A1 in a compression ignition engine. Four oxygenated fuels were studied, namely dimethyl carbonate (DMC), ethanol (EtOH), polyoxymethylene dimethyl ether 1 (PODE1) and polyoxymethylene dimethyl ether 4 (PODE4). The oxygenated fuels were blended to make up fuel blends with 5% oxygen content to study the molecular effect of different oxygenated fuels in affecting soot formation. Figure 3.1 shows the particle size distributions (PSDs) for the fuel blends at three engine loads (30%, 45% and 60%). There is a strong reduction of the larger particles at above 10 nm (accumulation and coarse modes) for EtOH, especially at the low engine load of 30%. The reduction diminishes with the increase in the engine load up to 60%, i.e., the PSDs for all the oxygenated fuel blends are similar to the case for Jet A1. The rest of the oxygenated fuels have an almost negligible effect on the PSDs at all fuel engine loads. Another notable observation is an increase in smaller particles (below 10 nm, nucleation mode) for the EtOH and PODE1 fuel blend at an engine load of 30%. This suggests that the molecular structure of EtOH may have the strongest effect on the PSDs when compared to the rest of the oxygenated fuels at a low engine load.

Dr Yichen ZONG (Research Fellow, NUS) has been leading experimental research on future fuels for low emission energy utilisation. The research activities are conducted under the collaboration of NUS and Cambridge researchers. The major experiments have been successfully finished in the last few months, covering a wide range of additives (DMM, DMC, PODE3, PODE4, Ethanol) and their impact on diesel and jet fuel in engine combustion. A research paper from previous experiments has been published in *Fuel* which systematically investigates the particulate emissions from diesel/butanol blend combustion. Another summary paper on the comparison of all additives is in preparation. Moreover, Dr Zong is also working on engine simulations with CMCL engineers (an R&D company) and air quality monitoring with the collaboration of the C4T JPS team.

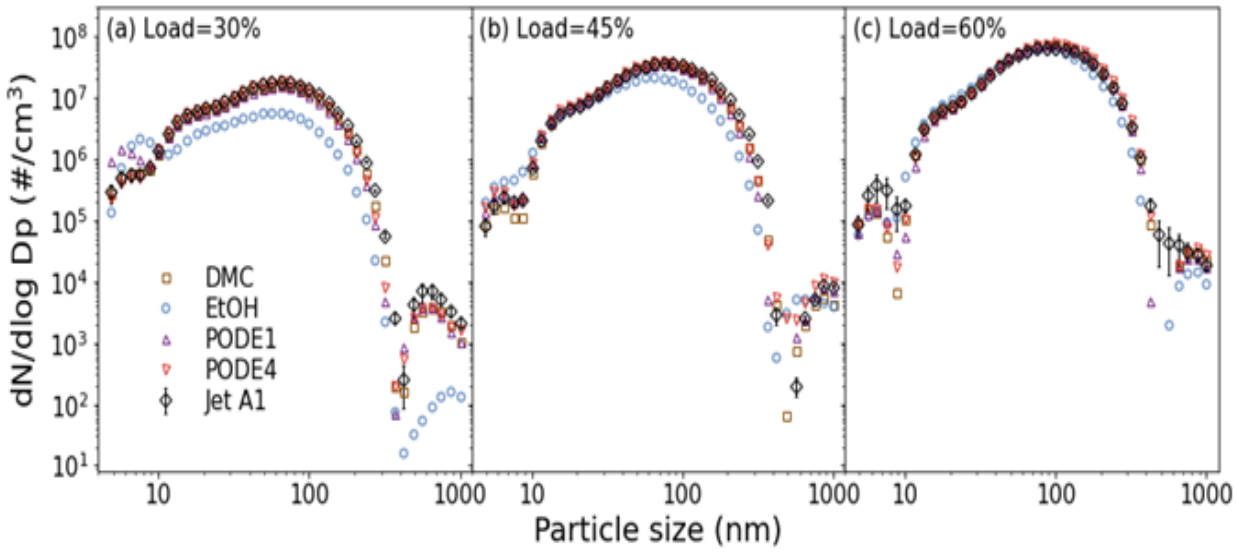


Figure 3.1: Particle size distributions measured using a DMS500 at the exhaust of the engine with different fuel blends. The data in each panel are grouped by the engine load at (a) 30%, (b) 45%, and (c) 60%.

Mr Yong Ren TAN

Update on work package 3.2

Refinery, fuel and engine of the future — modelling

Chemical mechanisms, PAH chemistry, after-treatment

Dr Laura PASCAZIO's (Research Fellow, CARES) main research interest lies in the study of combustion-generated carbonaceous nanoparticle (also known as soot) formation using computational methods. Understanding the soot inception mechanism remains one of the most debated topics in the combustion scientific community.

Recently, she submitted a paper on the development of a knowledge-graph based framework for the automated parameterisation of reactive force fields derived from relaxed potential energy surface (PES) scans. Jointly with **Dr Angiras MENON (PDRA, CAM)** and CMCL, an ontological representation for Potential Energy Surface scans, OntoPESScan, has been developed that allows for the semantic enrichment of quantum chemical calculations within The World Avatar project (theworldavatar.com). Following this, she developed a software agent able to perform PES scans results retrieval and reactive force field calibration tasks. She presented a poster on this project at the 39th International Symposium on Combustion (see "Other activities and achievements" in this chapter section for more details).

Currently, Dr Pascazio is working with Dr Ali NASERI (Non-C4T Research Fellow, CAM) and CMCL on extending The World Avatar capabilities in the chemistry domain. The goal is to retrieve data from other chemistry databases such as PubChem or Open Reaction Database and store the data in The World Avatar so that this data can be queried and used by software agents for reaction prediction, chemical synthesis planning and experimental design.

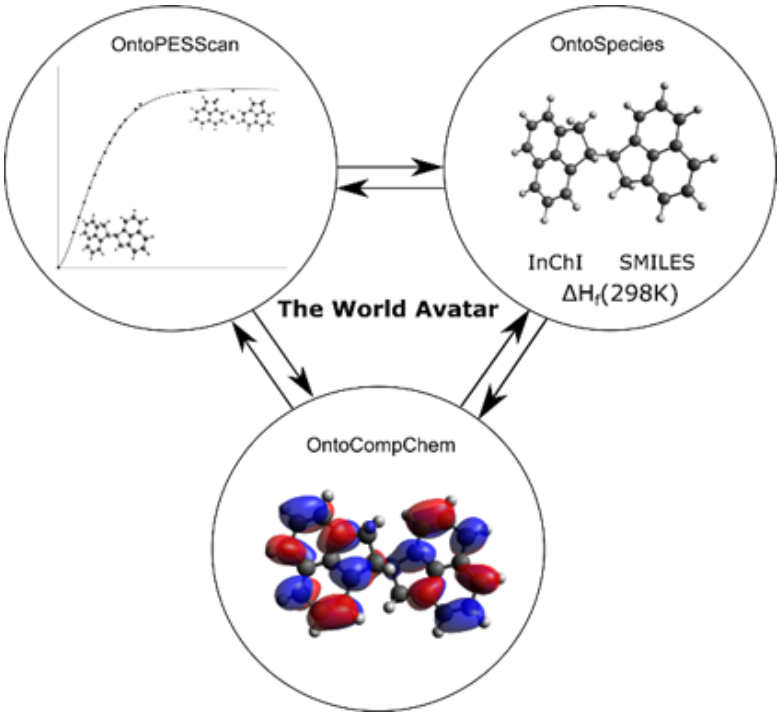


Figure 3.2: OntoPESScan ontology, linked to OntoSpecies and OntoCompChem, is developed in The World Avatar framework to extend the chemistry domain.

Dr Laura PASCAZIO

Update on work package 3.3

Better, cheaper, cleaner nanostructures — experimental

Flame synthesis of thin films of mixed metal oxide nanoparticles

Dr Yuan SHENG's (Senior Research Fellow, NTU) recent work has been mainly on the durability of flame synthesised CuO_x nanoparticulate films during the electrochemical CO_2 reduction reaction (eCO_2RR). Fixation of the film on the gas diffusion layer (GDL) with a suitable amount of binder and the hydrophobic properties of the GDL were found critical to the durability. Decorating commercial GDL with a highly hydrophobic conductive coating extended the lifespan of the films by increasing their resistance to flooding. Trace amount of O_2 in the feed gas, on the other hand, accelerated flooding of the electrode. To study the role of hydrophilicity further, Dr Sheng has developed PTFE-modified formulations of catalyst inks based on commercial CuO nanoparticles so that the catalytic film itself can be rendered hydrophobic. The resulting films showed high selectivity toward ethylene at 0.5A/cm² for at least 2 h, better than the unmodified films (Figure 3.3). Dr Sheng co-authored a paper with **Dr Guanyu LIU (past-C4T Research Fellow, NTU)** about the effect of pore tortuosity and oxidation states of flame-synthesised Cu/CuO nanoparticulate films on the selectivity of eCO_2RR products. In addition, Dr Sheng studied the degradation behaviour of NiFe hydroxide films as the oxygen evolution catalyst during sustained alkaline electrolysis in industrial conditions. Migration of Fe species from anode to cathode was found to play an important role.

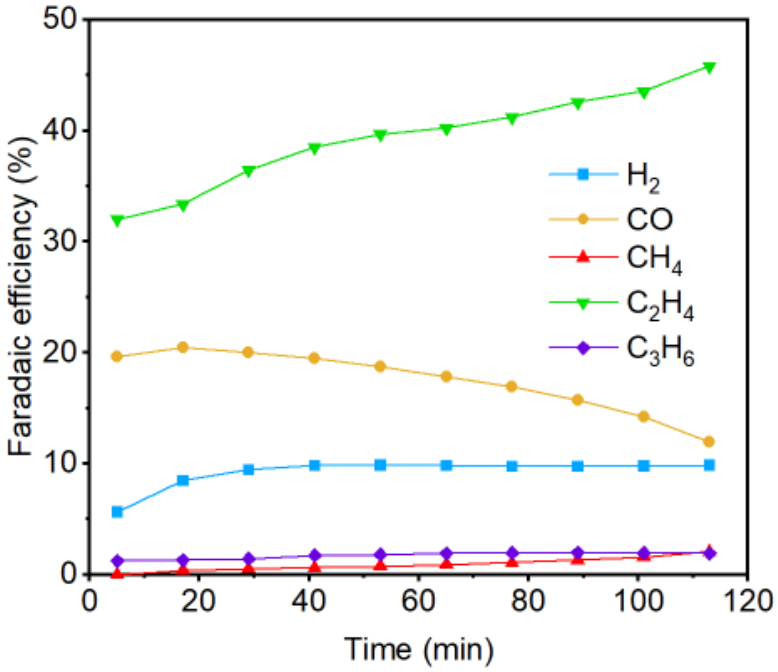


Figure 3.3: The eCO_2RR performance of a PTFE-modified CuO nanoparticulate film on commercial GDL. Total current density: 0.5 A/cm². Electrolyte: 3.5 M KOH.

Dr Yuan SHENG

Update on work package 3.4

Better, cheaper, cleaner nanostructures — modelling

Gas- and surface-phase kinetics, molecular modelling and reactor optimisation

Mr Simon RIHM (PhD student, CARES) successfully built a reversible and fully-elementary micro-kinetic model in the previous report. He continues his research in advancing the understanding of electrocatalytic CO_2 reduction (eCO_2R) – specifically the reaction mechanism. Together with **Dr Mikhail KOVALEV (Technical Development Manager, AMPLE)**, Mr Rihm has been working on the characterisation of minor products of an eCO_2R flow reactor via gas-chromatography mass-spectrometry (GC-MS). The mass-spectrometry method used is based on proton-transfer reactions (PTRs) and can reveal previously unknown products of very small concentrations which are separated via the GC column. The analysis of the data generated is challenging because it is spread out over multiple dimensions (time, mass, applied potential, temperature) and greatly varying concentrations between species lead to complex effects through the PTR mechanism. For this reason, an automated process of species identification and analysis was developed and deployed, by which many additional species were identified that seem to constitute minor eCO_2R products which have not been reported before. This is important for the mechanism investigation as well as designing around potential catalyst poisoning.

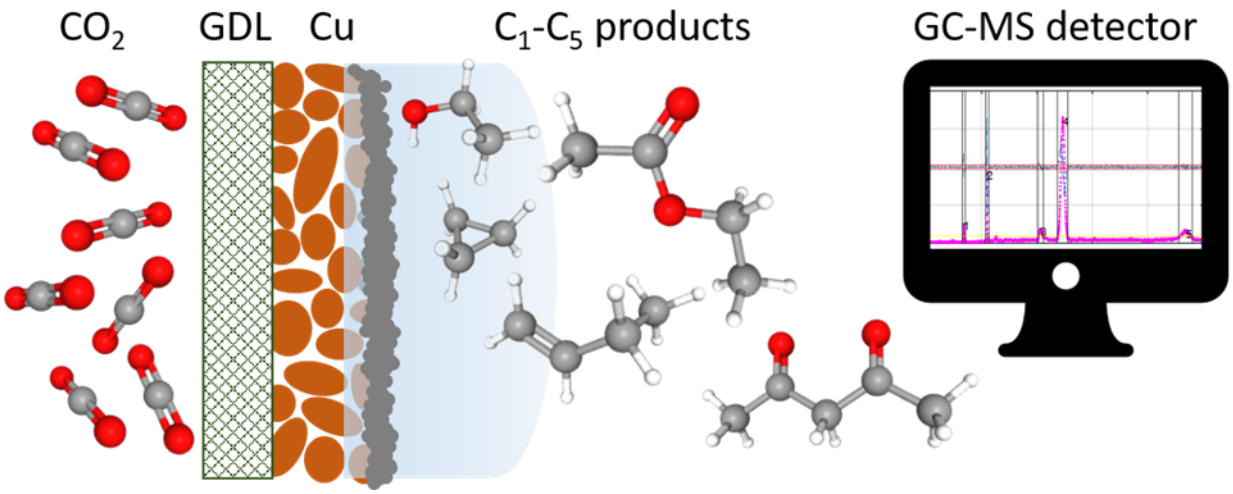


Figure 3.4: Illustration of the flow cell with gas-diffusion layer (GDL) and a copper catalyst (Cu) converting carbon dioxide to a variety of products that can be detected and analysed within a GC-MS setup.

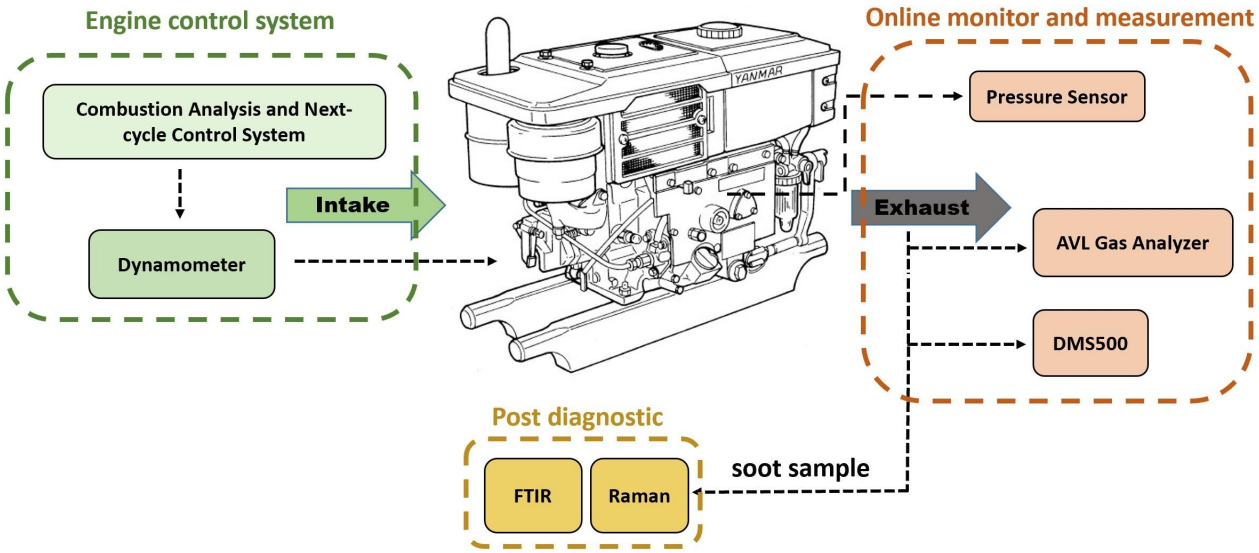
Mr Simon RIHM

Scientific output

The following are the CREATE-acknowledged publications generated by IRP 3 during the reporting period, excluding those already featured in the Scientific Highlights section on page 14.

Evaluating the effect of *n*-butanol additive on particulate matter emission in diesel engine
Qiren Zhu, Yichen Zong, Yong Ren Tan, Jieyao Lyu, Wenbin Yu, Wenming Yang, and Markus Kraft, *Fuel*
DOI: 10.1016/j.fuel.2022.126003

Abstract: In this study, we perform experiments on a common-rail diesel engine to investigate the effect of diesel/*n*-butanol blends on particulate matter (PM) emission under pilot-main injection strategy. Three different blends (diesel, D80B20, D50B50) are compared under two distinct loads (~30 %, ~60 %) and two injection pressures (40 MPa, 60 MPa). Under identical load and injection pressure, the engine fueled with D80B20 has the longest ignition delay and the shortest combustion duration, resulting in the highest HC emissions. The results indicate that the addition of *n*-butanol can improve the mixing process of fuel and oxidizer and result in a lower mean chamber temperature. The addition of *n*-butanol can also reduce NO_x emission (up to 25 %) because the lower mean chamber temperature caused by the lower heating value of *n*-butanol tends to reduce NO_x emission although there is extra oxygen content in *n*-butanol. Moreover, the addition of *n*-butanol can reduce PM emission (up to 69 %) and the engine with D50B50 under low load has the lowest PM emission. We further sample the soot particles from the engine emission. The result of the Raman spectroscopy investigation of soot samples reveals that *n*-butanol can increase the oxidative reactivity of the particles and the shorter combustion duration of engine can increase the level of disorder of the particles. The result of FTIR spectra shows that the aromatic C=C functional group has the highest signal intensity when the engine is fueled with D80B20 and is diffusion combustion dominated.



Other activities and achievements

Several IRP 3 researchers attended the 39th International Symposium on Combustion from 24 - 29 July 2022 in Vancouver.

- **Prof Markus Kraft (PI, CAM)** co-chaired a session on "Soot, Nanomaterials & Large Molecules"
- **Dr Laura PASCAZIO (Research Fellow, CARES)** presented a poster titled "Reactive force field for localized π-radicals".
- **Mr Yong Ren TAN (PhD student, CAM)** presented posters titled "Injection of charge from non-thermal plasma into a soot forming laminar coflow diffusion flame" and "Influence of the types of oxygenated fuels on the characteristics of soot emitted from a CI engine".
- **Mr Simon RIHM (PhD student, CARES)** delivered an oral presentation titled **"**Modelling a detailed kinetic mechanism for electrocatalytic reduction of CO_2".

Prof Markus KRAFT (PI, CAM) was invited to give a series of lectures on the formation of soot as part of the annual Tsinghua-Princeton-CI Summer School on Combustion. The programme attracts over 400 attendees each year and is one of the most reputable summer schools in the field of combustion.

Dr Yichen ZONG (Research Fellow, NUS) is starting a new collaboration with Prof Tat Loon CHNG (NUS Mechanical Engineering). This collaboration aims to study the NO_x reduction and soot formation in flame under non-equilibrium plasma.

Dr Laura PASCAZIO (Research Fellow, CARES) delivered an oral presentation titled "Review of soot and carbon black formation: role of aromatic π-diradicals" at Carbon 2022 in London held from 3 - 8 July 2022.

IRP 4

BETTER, CLEANER HEAT USAGE

Better, Cleaner Heat Usage is a new IRP 4 for Phase 2, replacing the former energy/electricity focus in Phase 1. This work is focused on high-performance thermal management and waste heat recovery research for improved, i.e., cleaner and more efficient heat usage in energy conversion technologies. IRP 4 addresses two key challenges in power generation systems: a) the efficient management of heat and b) the emission of harmful pollutants, which is particularly problematic in fuel-based technologies such as diesel engine power plants or marine engines. Regulations are increasingly stringent for these systems and a full understanding of the underlying phenomena is necessary to tackle this problem.

IRP 4 Principal Investigators:

Professor Epaminondas MASTORAKOS
University of Cambridge

Professor Alessandro ROMAGNOLI
Nanyang Technological University

Professor LEE Poh Seng
National University of Singapore

OVERVIEW

The activities for better energy efficiency, lower pollution, and decarbonisation in the marine sector is becoming of huge importance worldwide and for Singapore. The last few years have seen a huge increase in research and dissemination activity in this area. This IRP addresses these significant problems by a series of connected work packages, including fundamental studies on particulate emissions from marine engines burning fossil or alternative fuels, waste heat utilisation methods such as the use of Organic Rankine Cycles and the associated turbomachinery, high-efficiency heat exchangers, and estimates and measurements of pollutant dispersion from ships and its reception in port and urban areas.

During the reporting period, some of the Tasks were not active due to shortage of personnel. However, new post-docs have now been appointed and the work will pick up pace again. The heat exchanger is designed in direct context of an ORC. The CARES emissions measurement drone has been developed further and the results supplemented with new modelling. This activity feeds into a process model, which is important for modelling pollutant dispersion in ports and coastal areas.

Professor Epaminondas Mastorakos, PI
University of Cambridge

Update on work package 4.1

Engine combustion — best fuel, best operating condition

Dr Ramesh KOLLURU (Research Fellow, CARES reports that the contribution to air pollution by marine traffic is significant and an often overlooked aspect, as these heavy vessels produce the same pollutants as any other IC engine. In this project, an attempt is made to study the dispersion of pollutants evolving from a typical heavy marine vessel. Computational fluid dynamic studies on total dispersion and distribution of pollutants were carried out on a typical crude oil tanker Maran Helen which was used as a model for ship geometry.

Numerical Simulation of flow over a ship with headwind case was simulated with a single non-reactive passive scalar emerging from the chimney of the main engine. This passive scalar can be assumed as one of the components of the pollutant that is being discharged from the main engine. Figure 4.1 shows a typical passive scalar emerging from the engine chimney.

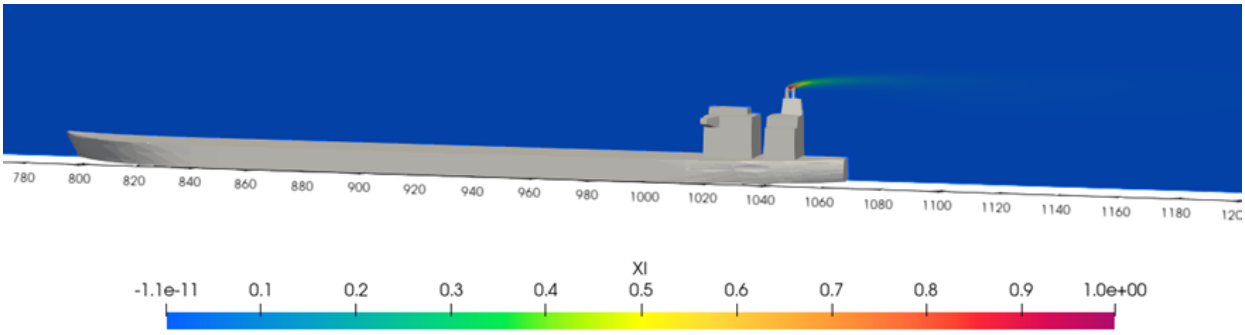


Figure 4.1: Passive scalar from chimney of the main engine. The spatial coordinates are in metres and the variable Xi represents concentration of pollutants.

Dr Ramesh KOLLURU

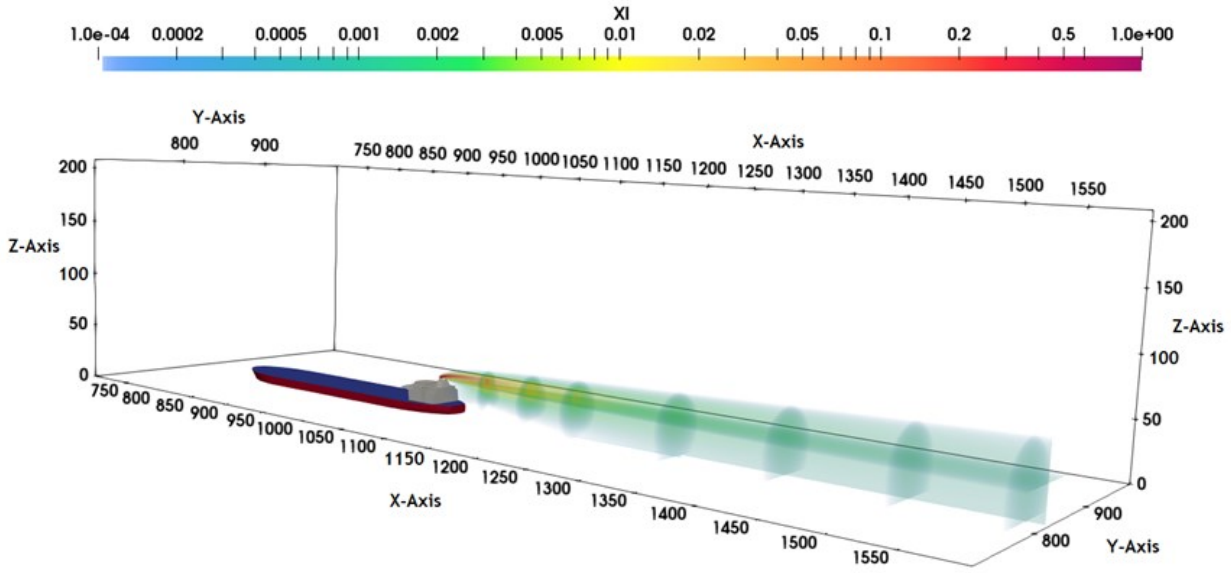


Figure 4.2: Pollutant distribution on log scale. The spatial coordinates are in metres and the variable Xi represents concentration of pollutants.

Dr Ramesh KOLLURU

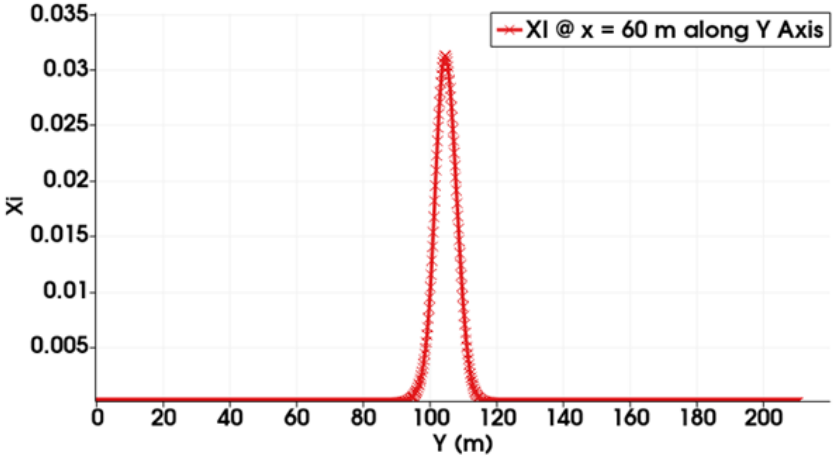


Figure 4.3: Passive scalar distribution behind the ship at 60m away from the chimney.

Dr Ramesh KOLLURU

Detailed analyses of the passive scalar are being carried out to understand pollutant dispersion. A typical distribution of the pollutant in the wake of the ship is shown in Figure 4.3. CFD analysis with different wind conditions like crosswind and realistic wind will be carried out and Dr Kolluru will continue to study the dispersion of pollutants along with the chemical reactions that would happen as the pollutant travels.

Update on work package 4.2

Closed power cycles—selection and analysis

There are no updates for work package 4.2 in this report due to recruitment difficulties in the last few months.

Update on work package 4.3

High-efficiency heat exchanger

Dr Yu HAN (Research Fellow, NUS) has been working on the performance of high efficiency heat exchangers using waste heat from engines in 80kW organic Rankine cycle (ORC). The heat exchanger can be used to reduce heat rate at the ORC evaporator and achieve better cycle efficiency. Additionally, the gliding temperature of the heat exchanger could also reduce the irreversibility between the heat exchanges at the ORC evaporator and condenser. Thus, the cycle working temperatures come closer to the temperatures of the heat source and the heat sink. During heat exchange in the evaporator and condenser of the ORC cycle, temperature differences between the heat exchanging fluids generate irreversibility at the cycle components, destroying some parts of the cycle available work. Use of the heat exchanger in the ORC is a feasible and effective method to reduce the temperature differences during heat exchanges. Figure 4.4 (a) shows the other two types of ORC processes with the positive slope of the saturated vapour curve in the T–S diagram. State points 1 and 2 correspond to the ORC system. Starting from state 2, the working fluid is heated in the evaporator at constant sub-critical pressure until it becomes saturated (state point 3), or it is superheated (state point 3). It is then expanded to state point 4, which is in the superheated vapour region. In this cycle, if the temperature T4 is markedly higher than temperature T1, it may be rewarding to implement a heat exchanger into the cycles as shown in Figure 4.4 (b). This heat exchanger is also represented by the additional state points 4a and 2a. The turbine exhaust flows into the heat exchanger and cools in the process (4–4a) by transferring heat to the compressed liquid that is heated in the process (2 –2a).

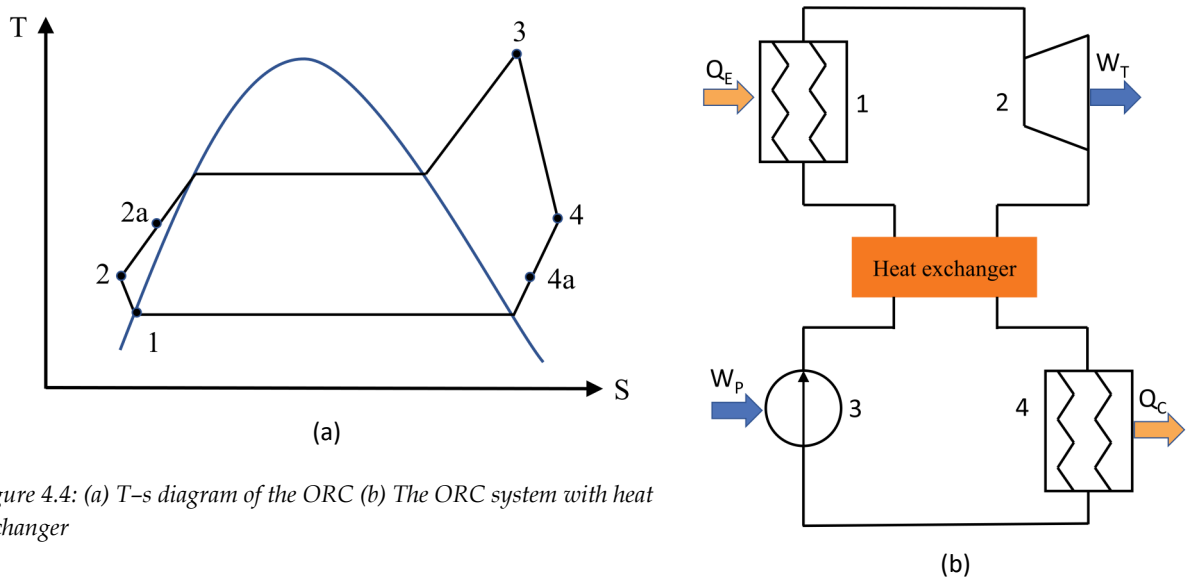


Figure 4.4: (a) T–s diagram of the ORC (b) The ORC system with heat exchanger

Dr Yu HAN

Update on work package 4.4

Process system model for the J-Park Simulator

Dr Molly HAUGEN (CARES Visiting Scientist, CAM), Dr Savvas GKANTONAS (Research Associate, CAM), Prof Epaminondas MASTORAKOS (PI, CAM), and Dr Adam BOIES (Co-PI, CAM) focused on both data collection for particles from an unmanned aerial vehicle (UAV) and using the incompletely stirred reactor network (ISRN) estimates to understand maritime plume dispersion in a port setting. The primary progress of this work has been the publication of the work from September 2021. This was a joint effort ensuring the publication was of the highest quality. During this time, the work was disseminated at an international conference. The conclusion from Figure 4.5 is that the dispersion of the plume in the vertical direction can be modelled using the ISRN, and that the stack height of the emission source dictates where the maximum concentration of particles is, as well as the relative concentrations below the stack height. Together, this information can be used to understand how plumes disperse into ports and urban environments.

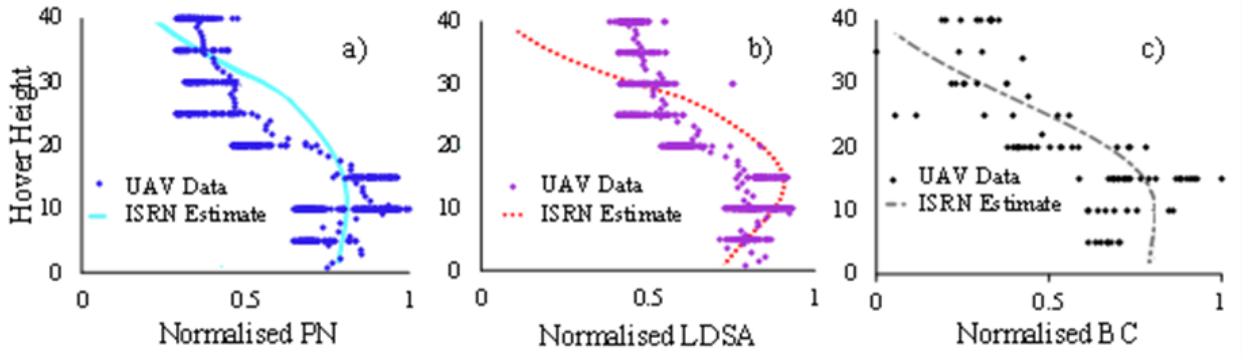


Figure 4.5: Vertical stratification of the plume for (a) particle number, (b) lung deposited surface area (LDSA), and (c) black carbon. The scatter plots are the collected data and the solid and dashed lines are the ISRN estimates.

Dr Molly HAUGEN, Dr Savvas GKANTONAS, Prof Epaminondas MASTORAKOS and Dr Adam BOIES

Dr Ghulam MAJAL (Software Developer, CARES) developed a 'NUSDavisWeatherStation' agent to periodically retrieve weather measurement data from a weather station near the National University of Singapore (NUS) (shown in Figure 4.6). The weather station transmits measurement data to a website called 'WeatherLink', and the data is retrieved periodically by the NUSDavisWeatherStation agent using an API. Dr Majal is also conducting a literature review on open-source Gaussian plume packages to estimate the dispersion of pollutants in the environment with a relatively inexpensive method. The primary objective of this study is to identify open-source models that can be incorporated into The World Avatar (see IRP JPS section on page 82) with relative ease.

Figure 4.6: A weather station located in the vicinity of the National University of Singapore (NUS)

Dr Ghulam MAJAL

Other activities and achievements

Prof Epaminondas MASTORAKOS (PI, CAM) was invited to be a panel member and speaker at the EU-Shipping-BCE conference held in Athens, Greece from 19 – 21 September 2022. He presented **Dr Li Chin LAW's (Research Engineer, CARES)** work titled "The role of Low Carbon Fuels in Decarbonising Maritime Transport" (see EOF 05- Future marine economy on page 147 for more details). The conference was attended by approximately 50 in-person attendees and 200 remote participants.

Prof Mastorakos was also elected as a Fellow of the Royal Academy of Engineering.

Dr Molly HAUGEN (CARES Visiting Scientist, CAM) presented a poster presentation of their work on particulate dispersion in Port Rafina, Greece at the International Aerosol Conference in Greece held from 4 - 9 September 2022. The conference was not funded by CARES, but there was significant interest in the work during the poster session.

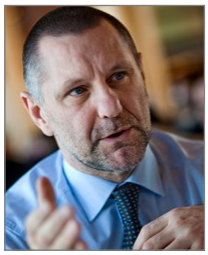
The work carried out in IRP 4 could help us better understand the effects of the shipping industry on air quality in Singapore.

IRP BB

BETTER BUSINESS: PATHWAYS TO INDUSTRIAL DECARBONISATION

The Better Business IRP acts as an incubator for ideas from all other IRPs and will support the acceleration and scaling of the technology outputs from the programme. It will examine different possible business models and compare the situation in Singapore with other important chemical clusters worldwide, engaging with stakeholders to identify the potential benefits and co-benefits of each technology arising from the programme.

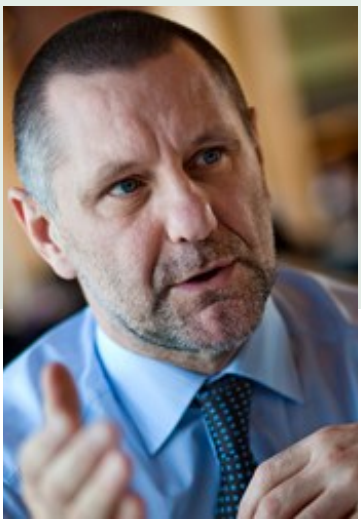
IRP BB Principal Investigators:



Professor Steve EVANS
University of Cambridge

Professor S. VISWANATHAN
Nanyang Technological University

Professor Kenneth HUANG Guang-Lih
National University of Singapore



OVERVIEW

Over the last few months, we have continued to proceed with the existing projects and start new ones.

For the research on business model innovations related to solar energy adoption, we have revised the manuscript on the previous work. Meanwhile, we have also started a new research project on business models for remanufactured products. We have revised and improved the paper for re-submission based on the reviews for the survey project regarding policy formulation, customer, and industry perceptions. Our new project has entered a conclusion for submission on how strengthening institutions for VC investment influence the pollution behaviour of firms in China's chemical and energy-intensive sectors. We made substantial revisions based on the comments from several friend reviewers and seminars. The manuscript is almost ready and will be submitted to a top management journal in September 2022. Based on this study, we will continue exploring the different roles of VC types (government VCs and private VCs) in influencing the environmental behaviours of focal firms. For the Emerging Opportunities Fund (EOF) project, our current plan is to perform extensive computational analysis to strengthen the argument before journal submission. We expect to submit the paper before the end of this year. A new colleague will also contribute her skills to the EOF project. Lastly, a new project on current industrial sustainability has been started, and ecosystem trends enabled and empowered by advanced digital technologies.

We have published one paper (reported previously) on sustainable shipping fuels as part of our shared project with Prof Epaminondas Mastorakos (IRP 4) and Dr Li Chin Law, which is an input calculation into the related changes likely to occur in the chemical sector in Singapore as bunkering changes over the next decades.

Professor Steve Evans, PI
University of Cambridge

Update on work package BB.1

Business model innovation potentials

Dr Lemy MARTIN (Research Fellow, NTU) continues to work with **Prof S. VISWANATHAN (PI, NTU)** on business model innovations for adopting sustainable innovations and technologies. The previous work continues researching business model innovations related to solar energy adoption. The manuscript is being revised by adding numerical studies to extend the analytical results for the general model with feed-in tariffs, including an example of a hybrid pricing scheme for third-party ownership models. A new research project on business models for remanufactured products is being explored, focusing on leasing remanufactured items versus selling new items.

Update on work package BB.2

Policy formulation, customer and industry perceptions

Dr Abhiruchi GADGIL (Research Fellow, NTU) continues to work with **Prof S. VISWANATHAN (PI, NTU)** to conclude the survey study and revise the paper for re-submission based on reviews. The study's main contributions include cross-sectoral similarities and differences in behaviours towards clean technology adoption. Another theoretical contribution from the survey study is a newly adapted Belief-Action-Outcome framework to delineate the different parameters and possible correlations related to clean technology adoption.

Update on work package BB.3

Venture capital policy and firm pollution in the chemical and energy-intensive industries

Dr Michelle Xiaomin FAN (Research Fellow, NUS) continues to work with **Prof Kenneth HUANG (PI, NUS)** to analyse how the strengthening institutions for VC investment influence the pollution behaviour of firms in China's chemical and energy-intensive sectors. They find that strengthening formal institutions for VC investment associated with a major, top-down VC policy reform results in increased intensity of air and water pollution emissions produced by the target firms. Nevertheless, VC firms' prior experience in the energy-related industry and local government's monitoring and environmental protection measures could mitigate such adverse impacts on the environment.

Based on the last version of the manuscript, they have received extensive comments and suggestions from several friendly reviewers and participants from two research seminar presentations at Stanford University and INSEAD, respectively (see "Other activities and achievements" in this chapter section). They have made substantial revisions based on these constructive comments and suggestions. Specifically, the study extends prior literature on coalition theory for the theoretical part. It links it to institutional theory to advance our understanding of how members in the dominant coalition could jointly influence and shape the coalition's performance and outcome under the constraints of an institutional environment. For the empirical part, they collect more data and perform supplementary analyses to provide more robust and comprehensive analyses for the research. These supplementary analyses include subsample analyses and examining more moderators on the characteristics of VC firms and local governments. The manuscript is being prepared for submission to a top-tier journal like *Management Science*.

In addition to the above study, they continue to develop and collect data for new studies. For example, they seek to investigate the heterogeneous roles of different VC firms in influencing the focal firms' green innovation and pollution emissions. Specifically, government VC firms and private VC firms hold different institutional logics; the former holds state-dominant logic and the latter holds market-dominant logic. Therefore, they would share different preferences regarding environmental behaviours when investing in the focal firms.

Update on work package BB.4

Future roadmap for industrial decarbonisation, including international comparisons

Under the supervision of **Prof Steve EVANS (PI, Cambridge), Ms Can CUI (PhD student, CAM)** has conducted several literature reviews and analyses on current industrial sustainability and ecosystem trends enabled and empowered by advanced digital technologies. The ecosystem model, orchestration, organisation transformation, and the relationship between ecosystem strategy and sustainability have been initially analysed. Design Research Methodology (DRM) has been proposed due to the novelty of research on companies' transformation strategy towards sustainability using ecosystem orchestration. DRM is chosen as it can build up practical tools that could provide an instructive guideline for companies to orchestrate an ecosystem to design transformation strategies to respond to the ever-changing external environment, disruptive trends, and achieve sustainability. Preliminary company interviews have been initiated and industrial insights and current pain points have been obtained. A few expert interviews have also been conducted with some initial insights that have helped in understanding innovation and change management.

Emerging Opportunities Fund

The team also worked closely with C4T Emerging Opportunities Fund 06 - Carbon reduction strategies of top chemical companies, see page 148 for further updates.

Other activities and achievements

Prof Kenneth HUANG (PI, NUS) presented a joint work done with **Dr Michelle Xiaomin FAN (Research Fellow, NUS)** and Prof Jiaxing YOU (Xiamen University) titled "Selling the Cow and Drinking the Milk Too? An Examination of Venture Capital Investments and Firms' Environmental Performance" at the Management Science and Engineering Research Seminar in Stanford University on 25 July 2022. The same work was presented at INSEAD Strategy and Entrepreneurship Research Seminar, INSEAD, in Singapore on 6 September 2022.

Prof Huang has been appointed as the Senior Editor of *Management and Organization Review*. *Management and Organization Review* is a premier journal published by Cambridge University Press for ground-breaking insights about management and organisations in China and global comparative contexts.

Prof Huang received the following awards:

- Academy of Management Annual Conference Best Paper, 2022
- Strategic Management Society Annual Conference Best Paper Prize (Nominee), 2022

Prof S. VISWANATHAN (PI, NTU) participated as a speaker/panellist at several industry conferences on topics related to Corporate Sustainability and Supply Chain Resiliency. These are listed below:

- ISCA PAIB Conference, Panel Discussion on "Financial Reporting Implications – Environmental, Social & Governance in Focus" held on 25 August 2022
- Singapore Environment Council (SEC) Annual Conference Day, Panel Discussion on "What are the upstream innovative design processes to reduce the environmental footprint of packaging material" held on 23 August 2022

Dr Abhiruchi GADGIL (Research Fellow, NTU) was a mentor for EB Impact's Sustainability Exchange (Jan-July 2022). Her team won the challenge and has been awarded funding of ~SGD40K for testing their idea in Singapore. The media coverage for the team is below:
https://www.todayonline.com/singapore/vending-machine-reusable-containers-sustainability-youth-ocbc-1916056

IRP JPS

THE J-PARK SIMULATOR

IRP JPS is an overarching research activity, with the ultimate purpose to show how research coming from each IRP affects the CO_2 output in Singapore and in particular the operations on Jurong Island. The research uses the latest ideas from Semantic Web technologies and Industry 4.0 to integrate real-time data, knowledge, models and tools to fulfil objectives such as simulation and optimisation in cross-domain and multi-level scenarios. One of the focuses is to create superstructures of models contained within the developed ontologies for industrial parks to provide an accurate and fast-to-evaluate approximation of computationally expensive mathematical models for process industry plants in high dimensions.

IRP JPS Principal Investigators:

Professor Markus KRAFT
University of Cambridge

Professor Raymond LAU Wai Man
Nanyang Technological University

Professor Iftekhar KARIMI
National University of Singapore

OVERVIEW

Over the past six months, the J-Park Simulator (JPS) has progressed on several fronts. We have developed new functionalities and increased its autonomy and connectivity to the physical world. For instance, we have dramatically improved the JPS infrastructure by augmenting the cloning tool, access agent framework, and agent routing. We introduced a significantly faster and more robust approach for cloning Resource Description Framework (RDF) data from one triple store to another. We also improved the Access Agent and StoreRouter infrastructure and extended the Store Client and Access Agent frameworks to support the querying of relational databases.

Additionally, we have investigated the applicability of the JPS to build a Universal Digital Twin to foster interoperability in broad smart city contexts. To develop a proof of concept, the impact of potential flood scenarios across different domains is investigated for a city in the UK as data are readily and publicly available. This work can easily be extended and applied to other regions where data are available, particularly Singapore. This work combines geospatial city data with building metadata and transient information, such as river water levels and flood warnings. As part of this work, we are instantiating the ontologies in the knowledge graph using publicly available data from various Application Programming Interfaces (APIs). We have also developed an ontology called 'OntoHeatNetwork' to represent (municipal) district heating networks including the heat and power generation, heat distribution, market conditions, and relevant weather and calendar effects. This ontology comprises the concepts needed to optimise a (municipal) district heating network.

Moreover, as part of an attempt to digitalise and automate the CARES Laboratory, we have implemented a Radio-Frequency Identification (RFID) based tracking system that tracks the chemical bottles going in and out of an explosive precursor cabinet located in the laboratory and developed a weather station agent to periodically retrieve weather measurement data from a weather station near the National University of Singapore (NUS). We have also created a 3D building model of the CARES Laboratory using Building Information Modelling (BIM) software. The BIM model augments the existing Geographic Information System (GIS) models by containing building information and representation of their materials, floor layouts, furniture, equipment, and attributes. We can now visualise the CARES Laboratory at various spatial scales (laboratory, building, district) across multiple domains (chemistry, building management, and energy management). Users can also interact via a web application with laboratory equipment such as the fridge and fume hoods to view their status and information.

Professor Markus Kraft, PI
University of Cambridge

Update on work package JPS.1

Big data — sensors and data modelling

As part of an attempt to digitalise and automate the CARES Laboratory, **Dr Ghulam MAJAL (Software Developer, CARES)**, **Mr Arkadiusz CHADZYNSKI (Senior Research Fellow, CARES)**, **Mr Wilson ANG (Software Developer, CARES)**, and **Mr Simon RIHM (PhD Student, CAM)** have made progress in several areas including the implementation of a Radio-Frequency Identification (RFID) based tracking system that tracks the chemical bottles going in and out of an explosive precursor cabinet located in the laboratory, the development of a weather station agent to periodically retrieve weather measurement data and development of an ontology for describing assets in the laboratory. For instance, Mr Ang has implemented an RFID-based system (shown in Figure 5.1) consisting of an antenna placed at the entrance of the cabinet and a reader. The reader receives information from the antenna and several RFID label tags affixed to the chemical bottles in the cabinet. Mr Ang has also developed an RFID agent that extends the functionalities of the Application Programming Interface (API) provided by the RFID reader vendor. The API allows the RFID reader to send the data received from the antenna as an Extensible Markup Language (XML) string to a specified endpoint. The RFID agent receives and parses the XML string to retrieve the RFID tag ID number, timestamps, the in/out status and various other details regarding the RFID system and saves this information in a PostgreSQL database. The RFID agent also provides an API interface for users to retrieve historical data. In addition, he has developed an RFID Update agent, which periodically retrieves the in/out status data from the RFID agent, converts this data into time-series format and stores it in the knowledge graph. Mr Ang is developing an RFID Query agent that will periodically query the knowledge graph for the latest in/out status of each chemical bottle, determine the period of time that the bottle has been out of the cabinet and send an email to inform the relevant laboratory personnel if the bottle has been removed for a long period.

Figure 5.1: Smart meter prototype to measure the electricity consumption of a fridge in the CARES Laboratory, and the temperature and humidity of the surroundings.

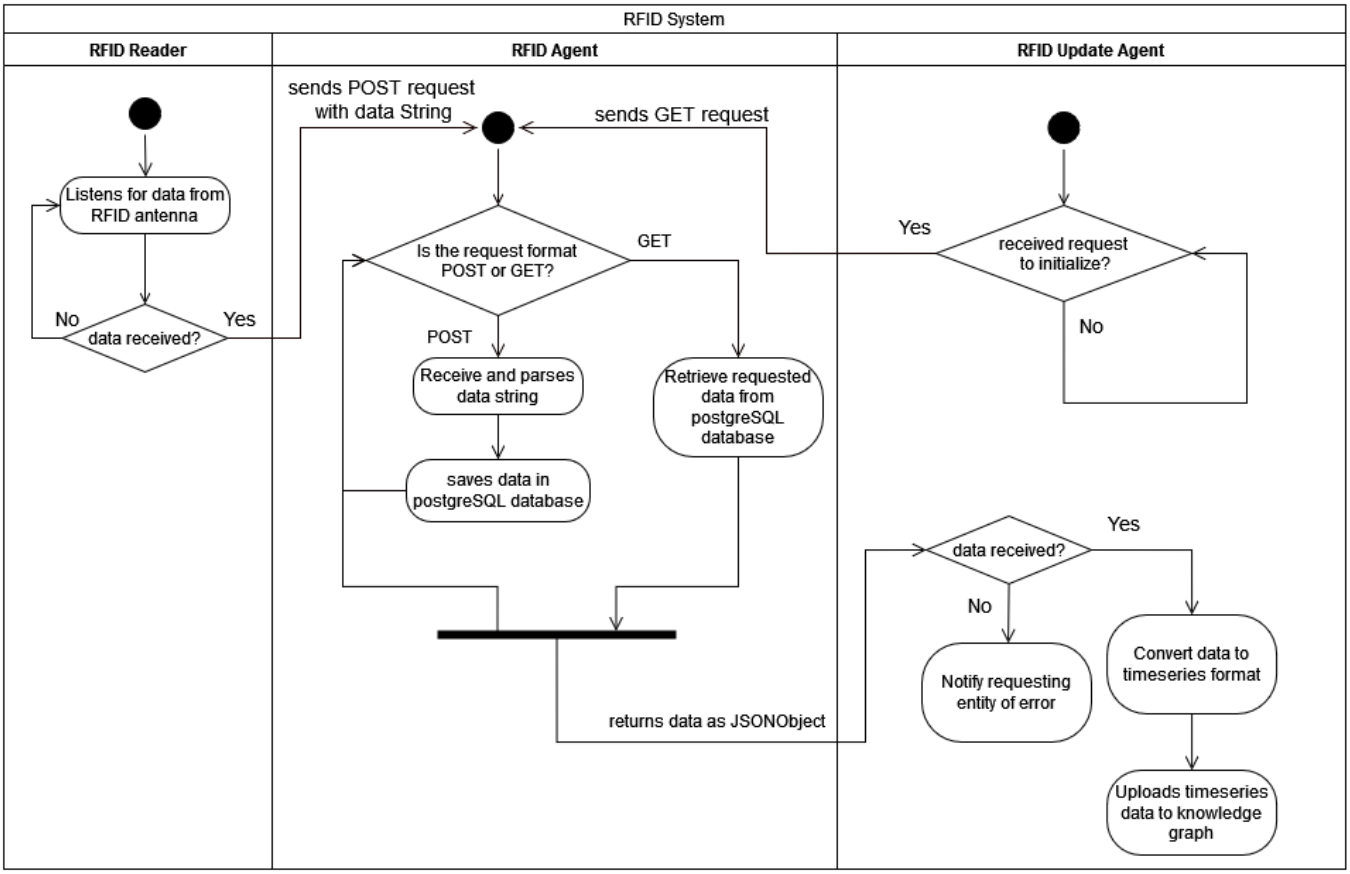


Figure 5.2: An activity diagram of the Radio-Frequency Identification (RFID) agent and RFID Update agent.

Dr Majal has developed a 'NUSDavisWeatherStation' agent to periodically retrieve weather measurement data from a weather station near the National University of Singapore (NUS) (Figure 5.3). The weather station transmits measurement data to a website called 'WeatherLink', and the data is retrieved periodically by the NUSDavisWeatherStation agent using an API. Dr Majal is also conducting a literature review on open-source Gaussian plume packages to estimate the dispersion of pollutants in the environment with a relatively inexpensive method. The primary objective of this literature review is to identify open-source models that can be incorporated in The World Avatar (TWA) with relative ease.

Figure 5.3: A weather station located in the vicinity of the National University of Singapore (NUS).

Mr Rihm is currently developing an ontology to describe critical assets in the CARES Laboratory within the knowledge graph. Using the knowledge graph, the instantiated assets will also be linked to information about chemicals and chemical reactions, as well as information obtained from the CARES Laboratory Building Management System – which collects real-time data using a variety of sensors. Figure 5.4 illustrates a diagram of various concepts, ontologies, aspects, domains and scales and their relationships to create a chemistry laboratory's digital twin.

As part of this digitalisation and automation work, Mr Ang, together with **Dr Casper LINDBERG (Research Fellow, CARES)** and Mr Chadzynski, has set up a Linux (Ubuntu) server for hosting parts of the knowledge graph related to the CARES Laboratory.

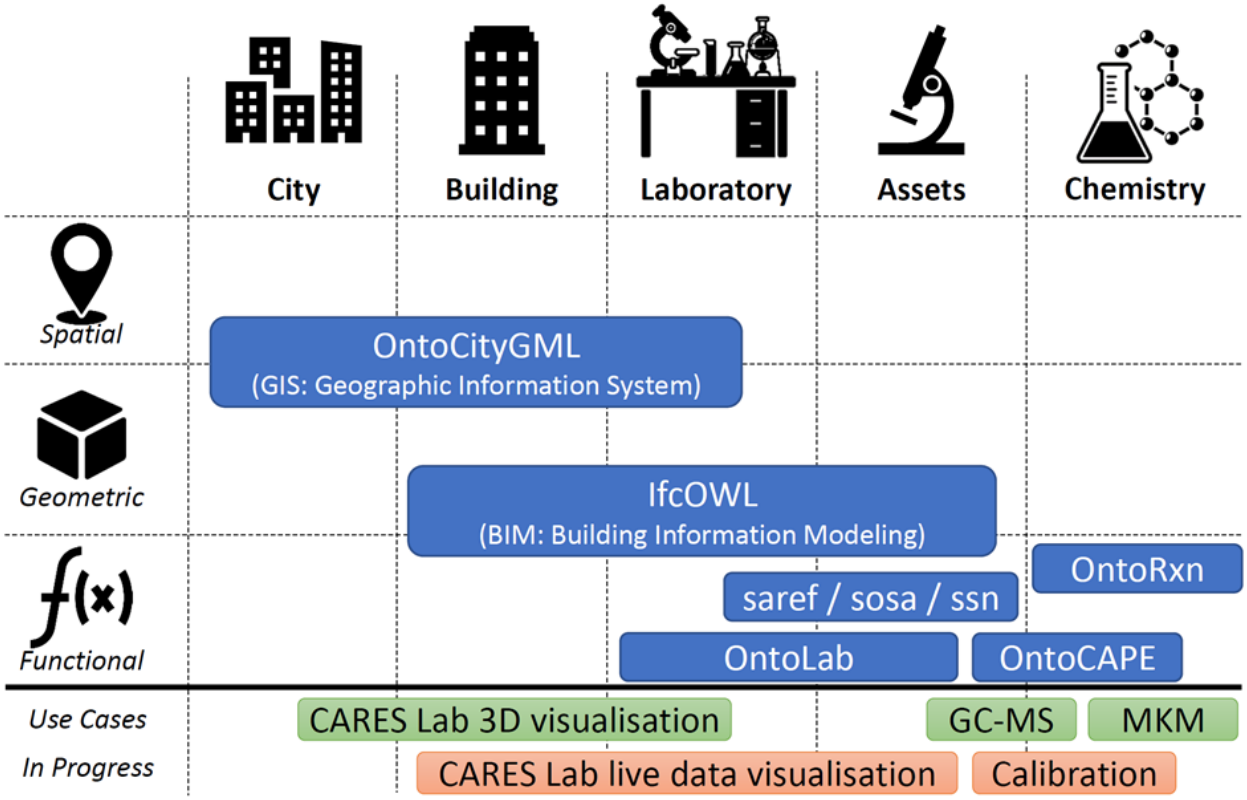


Figure 5.4: A representation of concepts and a selection of use cases across different scales and domains to facilitate a chemistry laboratory's digital twin. Existing ontologies such as 'OntoCityGML' and 'IfcOWL' are used to describe building information, and the 'OntoCAPE' ontology to describe the chemistry domain. It also requires extending existing ontologies with other existing ontologies such as 'saref', 'sosa', or 'ssn' for adequate representation of analytical devices and the creation of new ontologies e.g., 'OntoLab' to represent experimental setups and 'OntoRxn' to represent the processes taking place in them.

Update on work package JPS.2

Surrogate models, superstructure and architecture development

Dr Sebastian MOSBACH (Senior Research Fellow, CARES), **Dr Casper LINDBERG (Research Fellow, CARES)**, **Dr Jethro AKROYD (Senior Research Fellow, CARES)**, **Dr Feroz FARAZI (Research Associate, CAM)** and **Mr Arkadiusz CHADZYNSKI (Senior Research Fellow, CARES)** have been improving The World Avatar (TWA) infrastructure which includes augmenting the cloning tool, access agent framework, and agent routing. A significantly faster and more robust approach was introduced for cloning Resource Description Framework (RDF) data from one triple store to another. The new approach addresses several challenges when cloning large triple stores using multiple SPARQL operations and reduces the risk of source data corruption by ensuring that operations on the source store are read-only.

Dr Lindberg also improved the Access Agent and StoreRouter infrastructure. A cache was introduced to the StoreRouter to reduce the need to query the ontokgrouter triple store for frequently used SPARQL endpoints (Figure 5.5). A new Docker-compose stack was also created to permit much simpler deployment of a local Access Agent and associated triple stores in a developer's local development or testing environment. Furthermore, configuration variables can now be easily passed to the agent when deployed using Docker.

In addition, Dr Lindberg has developed an AgentRouter class to locate agents by querying the OntoAgent ontology and then direct requests to the intended agent based on the same cached router framework employed in the StoreRouter. The new AgentRouter is incorporated into the JPSAgent framework, thereby permitting agents extending the JPSAgent framework to easily send requests to other agents.

Ms Mehal AGARWAL (Software Developer, CARES) is currently working closely with Dr Lindberg on extending the Store Client and Access Agent frameworks to support the querying of relational databases (Figure 5.6). Ms Agarwal

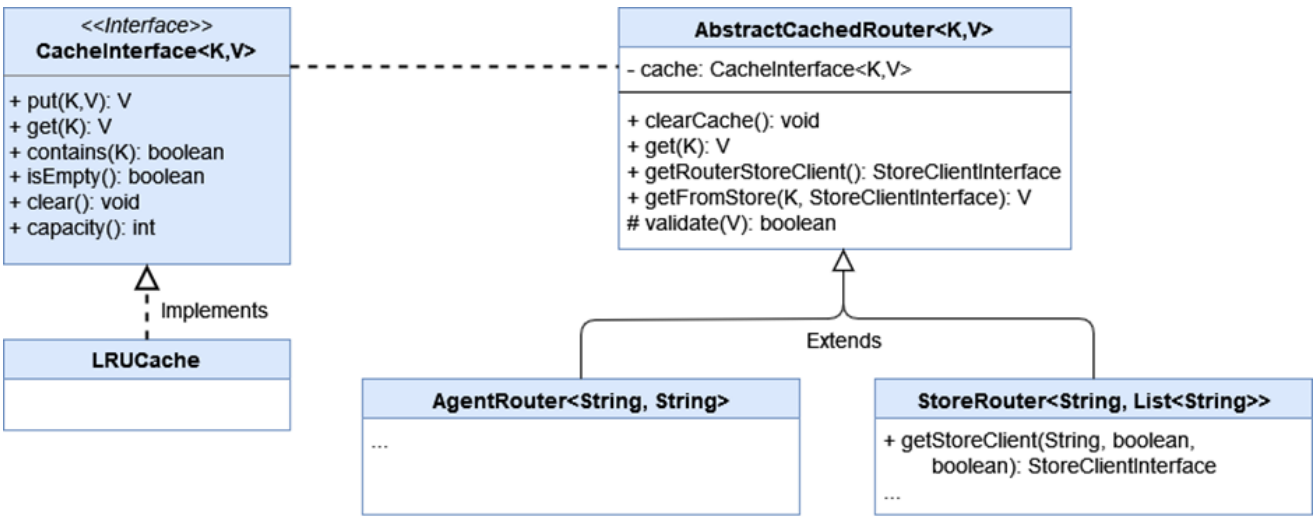


Figure 5.5: UML diagram of the AgentRouter and StoreRouter implementing the AbstractCachedRouter class with a Least Recently Used (LRU) cache.

has created the RemoteRDBStoreClient to connect, query and update relational databases and refactored how the time-series classes connect to the PostgreSQL database. The next steps include integrating general relational database access with the Access Agent.

Mr Chadzynski is also involved in training, supporting and providing guidance, especially to new team members, concerning documentation, questions on software design, agent development, and non-functional requirements such as performance and scalability. **Ms Srishti GANGULY (Project Engineer, CARES)** is involved in testing, deploying, and maintaining the various projects in TWA. Ms Ganguly has also been working towards migrating the projects in TWA from Java 8 to Java 11.

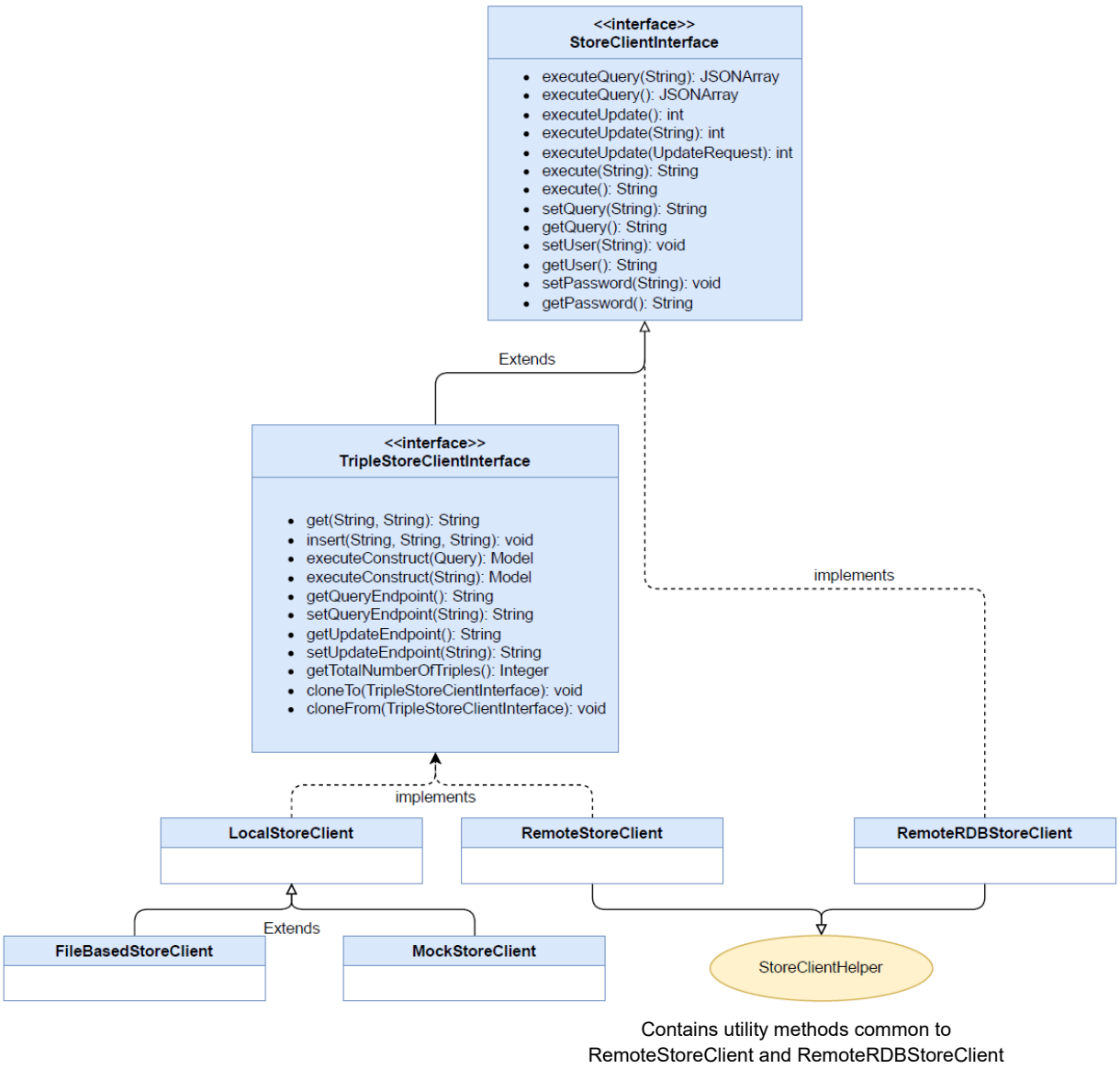


Figure 5.6: UML diagram of the StoreClientInterface and its implementing classes. The RemoteRDBStoreClient directly implements the general StoreClientInterface for relational database access. StoreClientInterface is extended by the TripleStoreClientInterface, which is implemented by various StoreClient classes to provide access to local and remote triple stores.

Update on work package JPS.3
Implementation

Dr Jethro AKROYD (Senior Research Fellow, CARES), **Dr Sebastian MOSBACH (Senior Research Fellow, CARES)** and **Dr Feroz FARAZI (Research Associate, CAM)** have reviewed and analysed semantic modelling of materials to enable the development of material passports. Examples of ontologies that have been reviewed include: the European Materials Modelling Ontology (EMMO), an ontology developed for describing materials, including material models and characteristics, physical and chemical properties and engineering aspects covering components, systems and processes; the Materials and Molecules Basic Ontology (MAMBO) that focuses on the representation of nanomaterials, molecular materials, organic and polymeric materials and their different properties. A material modelling ontology is being developed to describe the transformation of materials from raw materials extraction and processing to the assembly of all components for creating a product. A suite of Product Life Cycle (PLC) ontologies is also being developed to use in combination with the Basic Formal Ontology (BFO) for representing the product life cycle phases from design to end of life.

In addition, Dr Akroyd, Dr Mosbach and Dr Farazi have been working to adopt the technology-agnostic 'Findable, Accessible, Interoperable, and Reusable (FAIR)' Data Principles in publishing digital resources. These measurable principles can guide data owners, allowing machines to find and use their data automatically. Dr Akroyd, Dr Mosbach and Dr Farazi are analysing the first published FAIR Data Principles to understand how data owners can prepare their data to be FAIR principle compliant and how to objectively evaluate its metrics.

Mr Markus HOFMEISTER (PhD Student, CAM), in collaboration with Dr Akroyd and Dr Mosbach, has investigated the applicability of The World Avatar (TWA) to build a Universal Digital Twin to foster interoperability in broad smart city contexts. To develop a proof of concept, the impact of potential flood scenarios across different domains is investigated for a city in the UK as data are readily and publicly available. This work can easily be extended and applied to other regions where data are available, particularly Singapore. This work combines geospatial city data, such as information about the

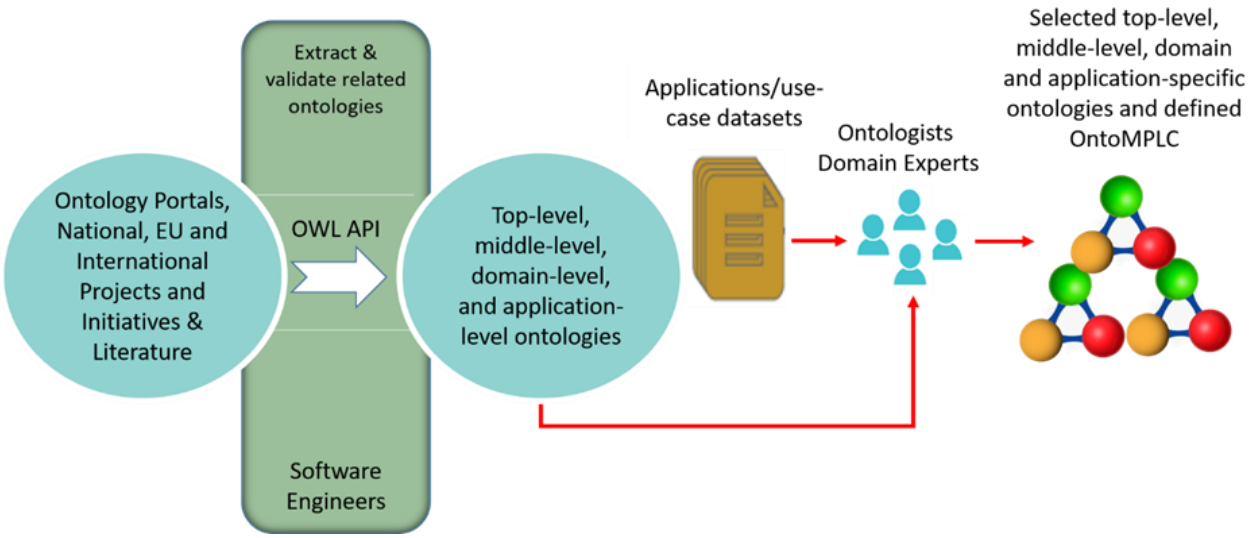


Figure 5.7: Approach for defining an ontology for materials and products lifecycle (OntoMPLC) to describe material models, attributes and different aspects of products starting from raw materials collection to its end of life.

built environment and land use, with building metadata, such as usage and near real-time environmental observation data, and transient information, such as river water levels and flood warnings. The ontologies developed have been instantiated in the knowledge graph using publicly available data from various Application Programming Interfaces (APIs).

Ms Mehal AGARWAL (Software Developer, CARES) has supported Mr Hofmeister in the instantiation task by developing a Building Matching agent to query the knowledge graph for equivalent building instances (instantiated using two different ontologies) using the Unique Property Reference Numbers (UPRNs) as identifiers. The Building Matching agent then creates a relationship between the instances and instantiates the area-weighted centroid property for each building using their Level of Detail 0 (LOD0) footprint. Mr Hofmeister is developing computational agents to estimate the impact of potential flooding events, for example, with regard to the number of affected people and property value at risk. The agents use the 'Derived Information' framework to manage data provenance and allow for information cascading. The main feature provided by this framework is the ability to update quantities calculated by agents acting on the knowledge graph when the inputs to the calculations are found to be out-of-date. A new Docker stack is also being developed to host relevant agents and data in a decentralised manner. Mr Hofmeister presented this work at the International Conference on Evolving Cities 2022, held 13 - 15 July in Southampton, UK. In addition, he and **Ms Huay Yi TAI (Software Developer, CARES)** have conducted a literature review to identify available ontologies for describing sewage networks in the knowledge graph.

Dr Hansong XUE (Research Fellow, CARES), in close collaboration with Mr Hofmeister and Dr Mosbach, has developed an ontology called 'OntoHeatNetwork' to represent (municipal) district heating networks including the heat and power generation, heat distribution, market conditions, and relevant weather and calendar effects. This ontology comprises of the concepts that are required for the optimisation of the operation of a district heating network. Dr Xue is consolidating and instantiating the available data using the OntoHeatNetwork ontology in the knowledge graph. **Mr Magnus MUELLER (Research Intern, CARES)** will use the instantiated time-series data in the knowledge graph to develop a time-series forecasting agent.

Figure 5.8: Visualisation of an example of an instantiated flood warning and the buildings potentially at risk of flooding.

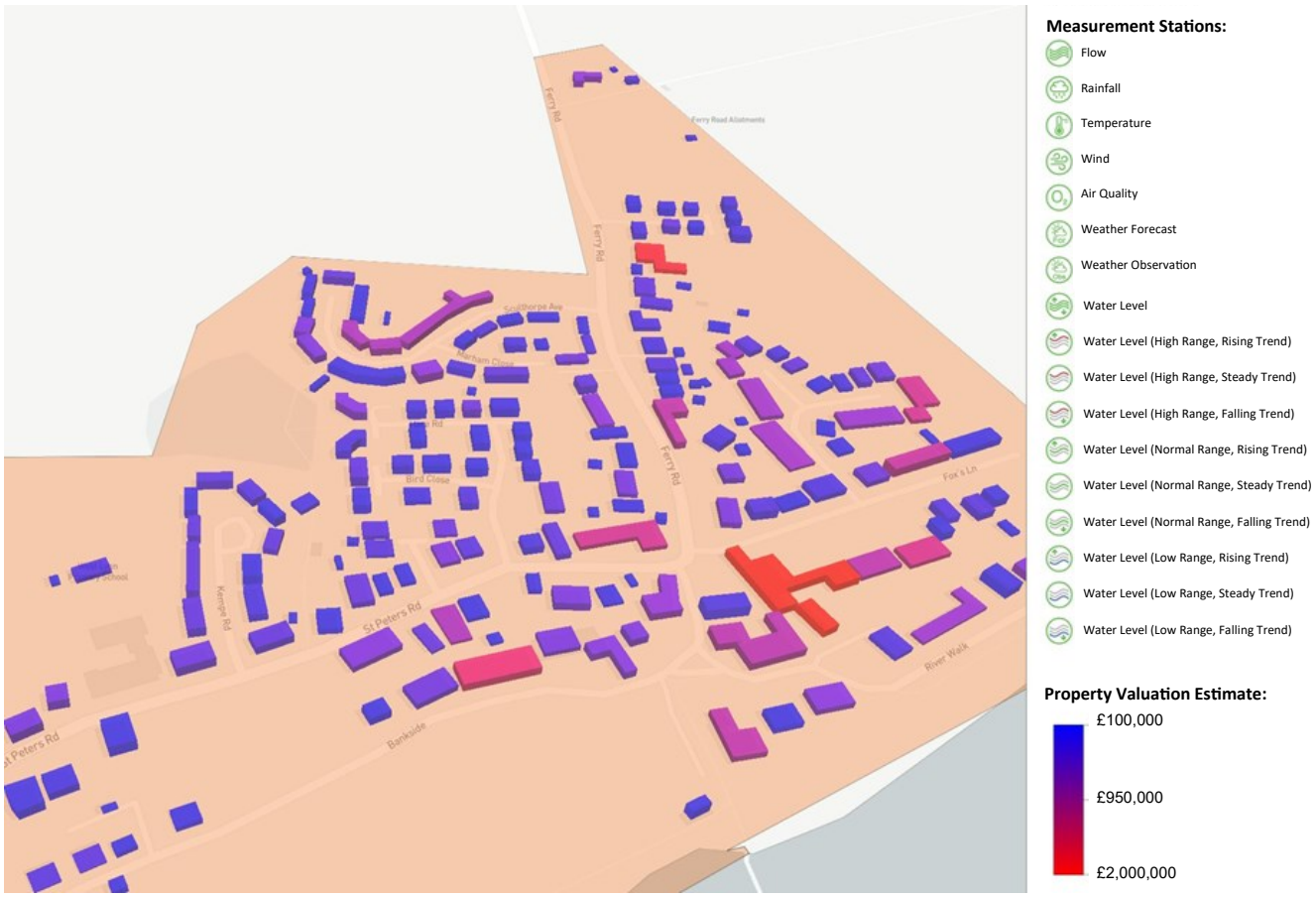


Figure 5.9: Visualisation of an example of an instantiated flood warning and its associated economic value at risk based on property value estimates.

Mr John ATHERTON (PhD Student, CAM) has developed a framework for querying, processing, and cleaning several live datasets from the Balancing Mechanism Reporting Service (BMRS). He has used these data to conduct multiple investigations into the British energy market and wind energy curtailment. The former investigation investigates the increased stresses faced by the British overall energy market and energy imbalance market, and particularly the energy imbalance market's inconsistent behaviour with the stated policy objectives. The latter investigation addresses whether price/subsidy policy or transmission constraints play a greater role in wind energy curtailment. New data on recent infrastructure expansions indicate that transmission constraints dominate British wind farm curtailment.

Ms Wanni XIE (Non-C4T PhD Student, CAM) has been working on developing the knowledge graph-based digital twin of the UK and its application in studying the decarbonisation of the UK power system by replacing the carbon-intensive power plants with Small Modular Nuclear Reactors (SMRs). A two-step algorithm has been proposed. The first step of the algorithm determines, for a given carbon tax amount, the capacity of the carbon-intensive power plants to be replaced by SMRs, and the locations and number of SMR units to be installed. The algorithm recommends different designs by minimising the total cost, including the potential carbon emission cost, the SMR capital investment and the site risk cost, which is defined as the monetary value of the casualties if a reactor failure occurs in the neighbourhood of the SMR site. The second step of the algorithm is an Optimal Power Flow (OPF) simulation that determines the output power of each generator based on the design given in the first step. This step establishes which generators are demoted from the base load to become the back-up load and which generators are shut off and phased out.

Update on work package JPS.4

Model analysis and visualisation

Mr Hou Yee QUEK (Research Associate, CARES) has been working on the visualisation of the digital twin of the CARES Laboratory. He has created a 3D building model of the CARES Laboratory using Revit, a Building Information Modelling (BIM) software, as shown in Figure 5.10. The BIM model augments the existing Geographic Information System (GIS) models by containing building information and representation of their materials, floor layouts, furniture, equipment, and their attributes. GIS models, on the other hand, contain geospatial information and have fewer details, making them more suitable for modelling and visualising at a larger scale, e.g., neighbourhoods. The CesiumJS library was used for visualisation as it can visualise 3D models and incorporate geospatial data. This facilitates the visualisation at various spatial scales (laboratory, building, district) across multiple domains such as chemistry, building management, and energy management in the digital twin of the CARES Laboratory.

Figure 5.10: The CARES Laboratory's detailed 3D Building Information Modelling (BIM) model.

Mr Quek has also developed a preliminary workflow to convert BIM models and extract their data into the '3D Tiles Next' data format. The converted '3D Tiles Next' tiles are loaded into a web application developed using the CesiumJS library for visualisation as shown in Figure 5.11 and Figure 5.12. Users can interact via the web application with different laboratory equipment such as the fridge and fume hoods to view their status and information. This web application is expected to enable laboratory users to have access, monitor the status, and interact with their ongoing experiments, even when they are not in the laboratory. The next steps involve developing an ontology to provide more links between the BIM models and the data from other domains, e.g., real-time measurement data from the CARES Laboratory Building Management System, to enable a more comprehensive and dynamic interaction experience in the web application and also to improve browser performance. In addition, Mr Quek has developed an agent to extract and store time-series data, such as utility consumption data, into the knowledge graph and relational database.

Mr Xiaochi ZHOU (PhD Student, CAM) and **Ms Shaocong ZHANG (Software Developer, CARES)** have been working on prototyping a new design to improve the robustness and response time of the knowledge graph-based Marie Question Answering system. The current design answers questions posed in natural language sentences by parsing the sentences with Natural Language Processing (NLP) tools and constructing

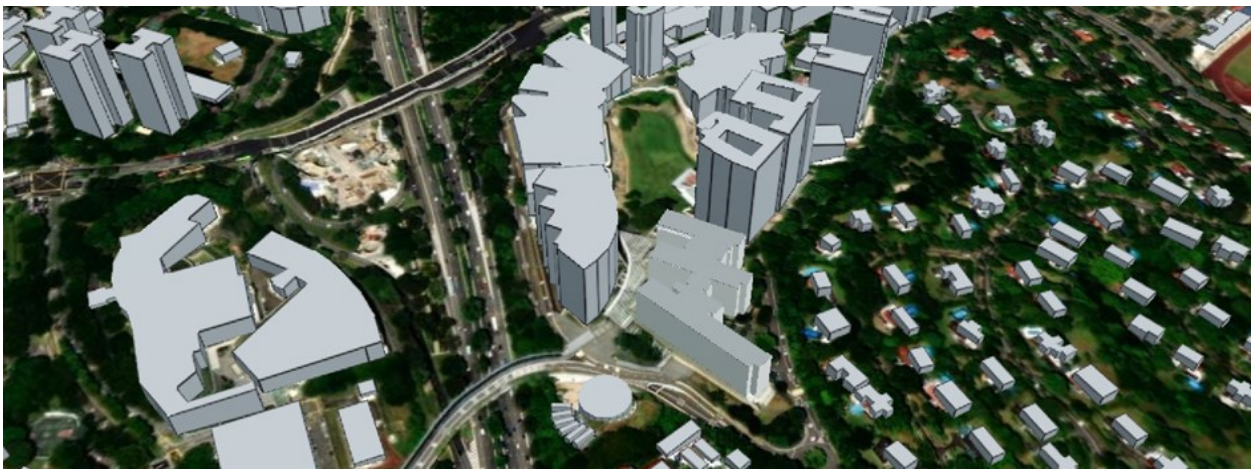


Figure 5.11: Visualisation of the 3D Building Information Modelling (BIM) model of the CREATE Tower and its surrounding buildings in Geographic Information System (GIS).

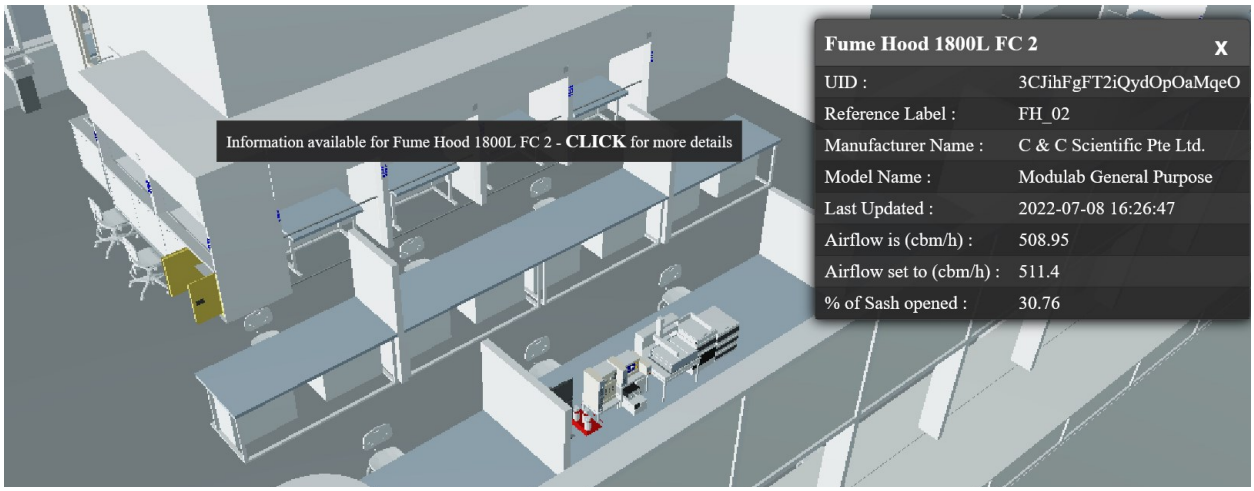


Figure 5.12: Visualisation of the information of laboratory equipment (Walk-in Fume Hood) in the CARES Laboratory.

formal representation (e.g., SPARQL query) of the sentences (i.e., a semantic parsing method) to query and retrieve information from the knowledge graph. Unlike the current design, the new design follows an information retrieval method that ranks a set of candidate answers by their likelihood of being the correct answer. This new design uses graph embedding algorithms and hence requires the embedding of entities and relations present within the question to represent them in a vector space. The successful embedding of entities and relations ensures that for each triple that is valid, any one of the components can be derived through the arithmetic operations between the other two components.

A relation mapping model is built upon the Bidirectional Encoder Representations from Transformers (BERT) pre-trained NLP model, which is the state-of-the-art multi-purpose model for NLP tasks. The model takes a question and maps it to an explicit or implicit relation in the training set. An entity linking model is also trained to identify and map the entities within the question to possible candidates within the knowledge graph. Using the relation mapping and entity linking models, a scoring function will retrieve a subgraph and measure the score of each candidate. The candidate with the highest score or a score higher than a certain threshold value will be returned.

Ms Srishti GANGULY (Project Engineer, CARES) has enhanced and instantiated over 6,000 CityGML models of different structures on Jurong Island into the knowledge graph. This consisted of approximately 5,000 Building models in CityGML Level of Detail 2 (LOD2) and 1,000 CityFurniture models in LOD1.

Scientific output

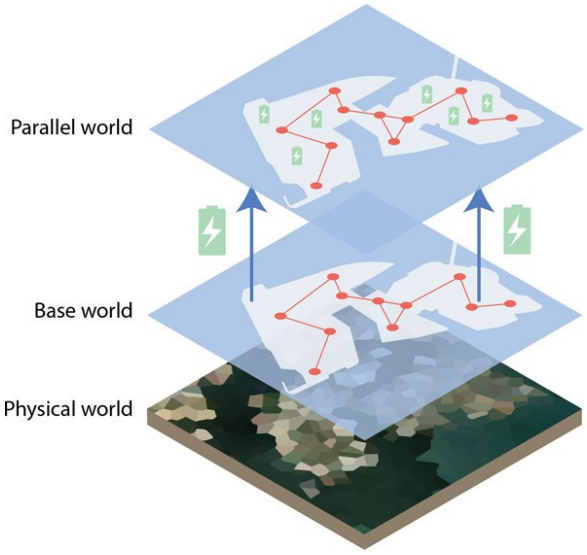
The following are the CREATE-acknowledged publications generated by IRP JPS during the reporting period, excluding those already featured in the Scientific Highlights section on page 14.

Embedding Energy Storage Systems into a Dynamic Knowledge Graph

Laura Ong, Gourab Karmakar, John Atherton, Xiaochi Zhou, Mei Qi Lim, Arkadiusz Chadzynski, Lanyu Li, Xiaonan Wang, and Markus Kraft, *Industrial & Engineering Chemistry Research*

DOI: 10.1021/acs.iecr.1c03838

Abstract: This Article illustrates how a dynamic knowledge graph approach in the context of The World Avatar (TWA) project can support the decarbonization of energy systems by leveraging the existing energy storage system (ESS) selection framework to assist in the selection and optimal placement of the ESS. TWA is a dynamic knowledge graph based on the Semantic Web and its associated technologies, with intelligent agents operating on it. The agents act autonomously to update and extend TWA, and thus it evolves in time. TWA also provides the ability to consider different scenarios, referred to as parallel worlds, allowing for scenario analysis without mutual interference. A use case—the addition of a battery energy storage system to the Singapore Jurong Island electrical network—is introduced to demonstrate the application of this approach. The domain ontology, OntoPowSys, was extended to describe and instantiate the relevant ESSs considered in the use case. This extension is described in the Article using the description logic syntax. The Article also outlines the details of how the various agents involved in the use case are being integrated into TWA and how the parallel world framework can facilitate scenario analysis by considering different scenarios without affecting the real-world representation.



Question answering system for chemistry—A semantic agent extension

Xiaochi Zhou, Daniel Nurkowski, Angiras Menon, Jethro Akroyd, Sebastian Mosbach, and Markus Kraft, *Digital Chemical Engineering*

DOI: 10.1016/j.dche.2022.100032

Abstract: This paper introduces an extension of a previously developed question answering (QA) system for chemistry, operating on a knowledge graph (KG) called Marie. This extension enables the automatic invocation of semantic agents to answer questions when static data is absent from the KG. The agents are semantically described using the agent ontology, OntoAgent, to enable automated agent discovery and invocation. The natural language processing (NLP) models of the QA system need to be trained in order to interpret questions to be answered by new agents. For this purpose, we extend OntoAgent so that it becomes possible to automatically create training material for the NLP models. We evaluate the extended QA system with two example chemistry-related agents and an evaluation question set. The evaluation result shows that the extension allows the QA system to discover the suitable agent and to invoke the agent by automatically constructing requests from the semantic agent description, thereby increasing the range of questions the QA system can answer.

Other activities and achievements

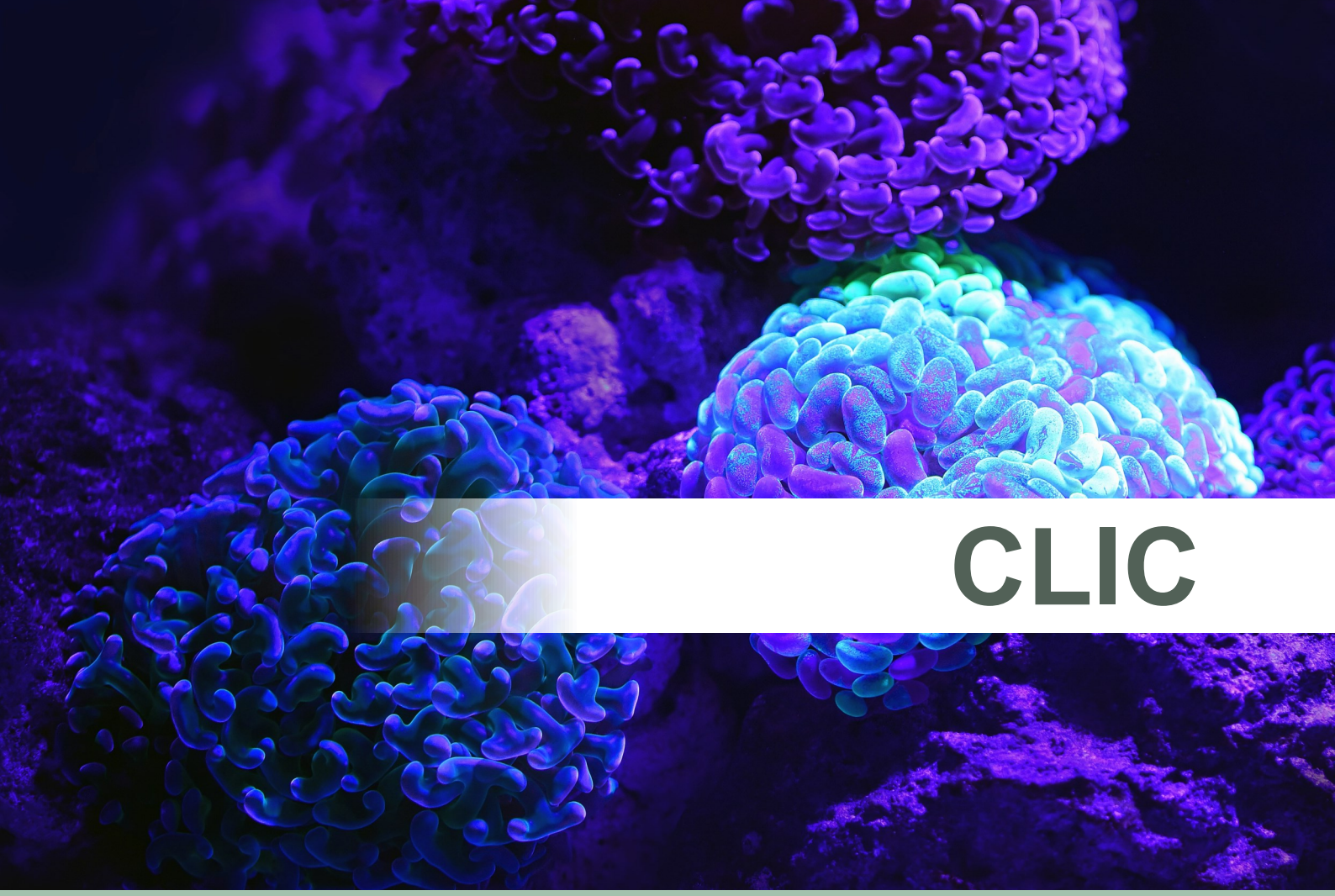
Dr Casper LINDBERG (Research Fellow, CARES) was recently awarded the Intra-CREATE seed collaboration grant with Asst Prof Hung Dinh NGUYEN (NTU) for the project 'Knowledge Graph-Driven P2P Energy Trading and Real-time Network Operation for High Renewables'. See page 138 for more details.

Prof Markus KRAFT (PI, CAM) co-edited a ground-breaking new book called *Intelligent Decarbonisation,* released on 23 April 2022. The book outlines how to maximise the economic and environmental use of digital technologies with AI to curb humanity's CO_2 emissions while minimising the inherent threats they pose.

Prof Kraft presented The World Avatar (TWA) project at a series of talks this reporting period:

- "The World Avatar Project - From Platform to Knowledge Graph: Evolution of Laboratory Automation" was presented at the Symposium on Data Science Powered High-Throughput Experimentation on 23 May 2022. The talk explored how TWA could bring about autonomous discovery in chemical sciences by orchestrating resources in a self-driving lab.
- A keynote talk was presented at InnovFest x Elevating Founders (Asia's leading innovation conference) on 1 June 2022. Prof Kraft spoke about the technology behind TWA and how knowledge graphs can lead to interoperability between diverse software and datasets.
- "The World Avatar Project – A Universal Digital Twin" was presented at the International Center for Computational Logic, Technische Universität Dresden on 15 June 2022. The talk explained the architecture behind TWA and how it can make data-driven decisions to optimise any complex system.
- "Universal Digital Twin – The Impact of Heat Pumps on Social Inequality" was presented at the Energy Visions Seminar on 8 July 2022. Prof Kraft was recognised as a distinguished lecturer and his talk emphasised how TWA revealed that using heat pumps for domestic heating would exacerbate fuel poverty and social inequality in the UK.
- "The World Avatar project – A Universal Digital Twin" was presented at a number of universities in the United States: University of California, Berkeley, on 4 August 2022; Stanford University on 8 August 2022; and Massachusetts Institute of Technology on 10 August 2022.
- Prof Kraft gave a talk at the United Nations Development Programme on 18 August 2022 where he spoke about how TWA can align with their Sustainable Development Goals (SDGs) and Human Development Index (HDI).

TWA and CARES was listed by the World Economic Forum (WEF) under its selection of Global Use Cases as part of their Global Digital Twin Cities initiative. The initiative offers a potential solution for policymakers worldwide to improve city governance and create a conducive urban ecosystem for industries and people. TWA was recognised for its ability to represent and simulate the world's behaviour using an ecosystem of autonomous computational agents that update in real-time and for its current practical use cases. Read more on the CARES website: https://bit.ly/WEFlisting



CLIC

CENTRE FOR LIFELONG LEARNING AND INDIVIDUALISED COGNITION

CLIC is a flagship programme in the Science of Learning initiative to harness advancements in neuroscience to develop training programmes for lifelong flexible learning. The research team has made significant headway since the commencement of the program. Investigators in the four different workgroups have worked continuously and collaboratively to ensure an optimal and cohesive task battery, data collection, organisation and analysis.

Professor Annabel CHEN Shen-Hsing
Nanyang Technological University

Professor Zoe KOURTZI
University of Cambridge

OVERVIEW

The Centre for Lifelong Learning and Individualised Cognition (CLIC) aims to improve lifelong learning and cognitive agility through innovative cross-disciplinary research in the science of learning. CLIC intends to translate its findings into an integrated model of learning that can be applied in day-to-day life. Our CLIC researchers have been diligently working since the last report toward completion of our main studies with adults and adolescents. We have optimised processes for recruiting participants, performing data quality checks and data analysis. We have also conducted data analysis workshops, commenced writing for research publication and implemented novel data collection methodologies.

Data collection between the **Cognition** and **Social Workgroup** has been on-going smoothly since the last report with close to 200 completed datasets. Chiefly, the Social Workgroup has been responsible for delivering the demographic, personality, and social variable questionnaires. Together, the Cognition, Social and School teams have collaborated to construct a pre-processing and data analysis framework for the WP0.1 study. Preliminary data analysis of both the adult and adolescent data has yielded encouraging findings which will be presented during the Scientific Advisory Committee meeting in late October 2022.

The Cognition Workgroup has also launched the Hybrid Remote Guided Method for supervised web-based cognitive testing, which allows participants to complete the testing battery online. This method affords participants with greater flexibility in their timing and location of testing, whilst maintaining high quality data through one-to-one online supervision. The Cognition Workgroup has been leading the RGT setup in close collaboration with the Social Workgroup to ensure a smooth transition in testing methodologies.

The **School Workgroup** team has presented several workshops over the course of the year, including during the 2022 Reinventing Pedagogy International Conference, the Annual Meeting of the International Society of Learning Sciences, and at the Academy of Singapore Teachers. These workshops cover topics that aim to close the gap between educators and neuroscience, cognition, and teaching practices. Another milestone by the School Workgroup was the mass data collection done in local secondary school. Packaged as a Brain Camp, the team administered the cognitive test battery over a two-day session, which was alternated with engaging activities to improve student's motivation. This was a substantial undertaking which involved the planning, training, and coordination between CLIC's staff, student helpers, and teachers.

The **Neuroimaging Workgroup** has completed Pilot 3 of their study and this Structure Learning training paradigm showed good fidelity without confounding components of cognitive flexibility. Thus, a decision was made to implement this training paradigm as the intervention design for the main study of WP0.2 to test if structure learning training improves cognitive flexibility. The workgroup has also finalised the MRI protocols for the team and the development of analyses

pipeline for the different MRI modalities. Data collection for the workgroup was unfortunately stalled by several unexpected software and hardware issues, resulting in time consuming troubleshooting. The team worked closely with Prof John Suckling, Prof Balázs Gulyás, and the Cognitive Neuroimaging Centre (CoNiC) imaging team to mitigate these technical issues and are currently steadily recruiting participants.

A 2-week data analysis bootcamp was jointly organised by Prof Henriette Hendriks and Assoc Prof Victoria Leong, which was hosted at the University of Cambridge in June 2022. The bootcamp allowed the Research Fellows to present daily data updates and engage in deep discussions with the CLIC PIs, staff and collaborators. Team members were able to join in on the hybrid event virtually from Singapore. A second 1-week data analysis bootcamp was also held in August, this time over Zoom, to discuss further findings of the study as well as to prepare for the upcoming SAC meeting. These bootcamps focused on hypothesis testing through data analyses and proved fruitful in consolidating findings across the different workgroups and test batteries, toward the aims of the programme.

Finally, CLIC has also made significant efforts to increase its social media footprint through our profiles on LinkedIn, Twitter and Instagram. We hope the social media posts on these platforms help to broadcast the relevance of cognitive flexibility skills and engage CLIC's work to a non-academic audience.

Professor Annabel Chen Shen-Hsing
Director of CLIC, NTU

Professor Zoe Kourtzi
Director of CLIC, CAMBRIDGE

Update on Cognition Workgroup

Assessing Cognitive Flexibility, Other Executive Functions and Learning in Healthy Young Adults

The Cognition Workgroup has been continuously working with the Social Workgroup on the large-scale data collection for the WP0.1 Adult Study (target N=400 adult participants). As of 29 September 2022, the Adult Study has completed data collection for 190 participants. The Cognition Workgroup collaborated closely with the School and Social Workgroups to establish the pre-processing and data analysis framework of the CLIC WP0.1 Studies.

Dr Ke TONG (Research Fellow, NTU) worked with Dr Aleya MARZUKI (Non-CLIC PI, CAM), Dr Christelle LANGLEY (Non-CLIC PI, CAM), and Dr Anahita TALWAR (University College London) to build the computational modelling pipelines of the cognitive flexibility tasks. Interim analyses suggested that the computational modelling parameters was promising in revealing key hypothesised relationships between cognitive flexibility and structure learning (Figure 6.1). Dr Tong and research staff from the other CLIC Workgroups presented the interim analyses over two Cambridge-NTU data analysis bootcamps.

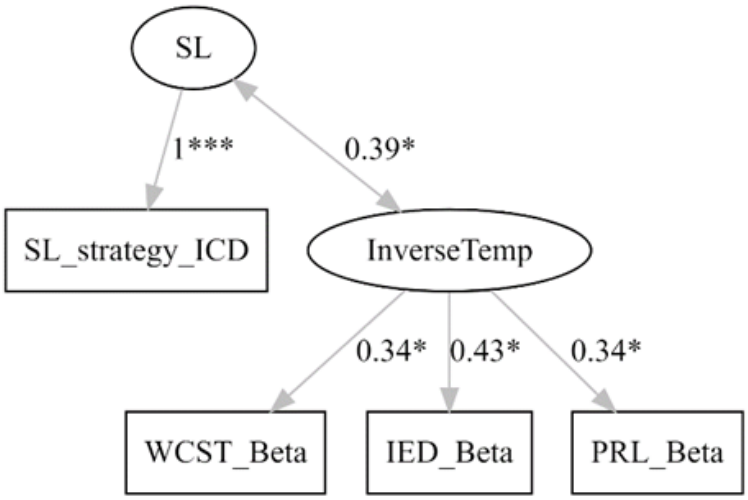


Figure 6.1. Interim analyses showed the Inverse Temperature factor scores from cognitive flexibility (CF) tasks are positively correlated with structure learning (SL) strategy ICD, suggesting that participants who adopted a strategy closer to maximising in the SL task (higher strategy ICD) showed more deterministic responding (higher β) on CF tasks. Inverse Temperature factor scores are estimated from reinforcement learning models from the three key cognitive flexibility tasks, i.e., Wisconsin Card Sorting Task (WCST), Intra-Extra Dimensional set shifting (IED), and Probabilistic Reversal Learning (PRL). The model shown was estimated on the WP0.1 adult sample (N = 173).

Mr Kean Mun LEE (Research Assistant, NTU), with supervision from **Assoc Prof Victoria LEONG (PI & Deputy Director, NTU)** and **Prof Henriëtte HENDRIKS (PI & Deputy Director, CAM)**, and assistance from **Mr Timothy LEE (Research Associate, NTU)**, Dr Chie TAKAHASHI (Non-CLIC PI, CAM) and Dr Lorena Santamaria COVARRUBIAS (Non-CLIC PI, CAM), has set up a data depository to efficiently share anonymised data amongst CLIC collaborators. The new data depository allows information to be easily downloaded from the Microsoft Teams application or accessed from Windows Explorer in the OneDrive folder. The improved

data accessibility through the OneDrive folder allows MATLAB and R scripts to read and analyse data with ease.

The Cognition Workgroup spearheaded a Hybrid Remote Guided Method for Supervised Web-Based Cognitive Testing (Figure 6.2) to facilitate participant recruitment and balance demographic distributions. This is a methodology adapted from Assoc Prof Leong's Remote Guided Testing (RGT) implemented during the COVID-19 lockdown. **Ms Natalie Philyra HOO (Research Assistant, NTU)**, **Ms Restria FAUZIANA (Research Associate, NTU)** and Mr Lee conducted multiple pilot sessions of the Hybrid Remote Guided Testing Method to rectify potential challenges. Ms Hoo is the lead coordinator for the Hybrid Remote Guided Testing and her role includes drafting the IRB amendment for the new mode of testing, creating the SOP and digitising hardcopy materials used in in-person testing sessions to ease the administration of cognitive tasks online, screening and scheduling potential participants.

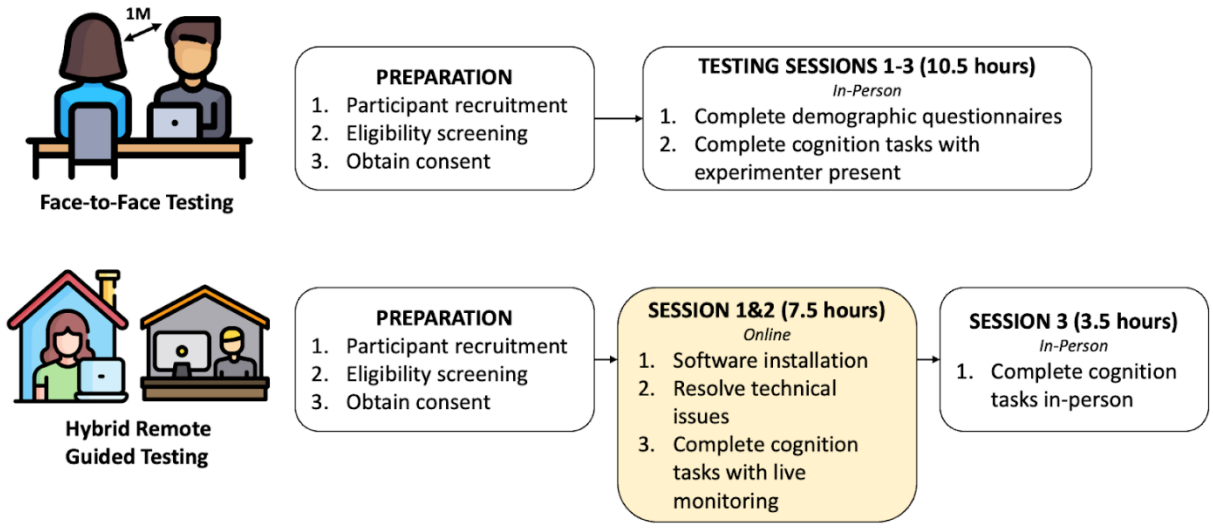


Figure 6.2. Parallel study schedules for Face-to-Face Testing and Hybrid Remote Guided Testing. The Face-to-Face Testing schedule includes three in-lab testing sessions (10.5 hours in total). The Hybrid Remote Guided Testing schedule includes two Remote Guided Testing sessions (7.5 hours in total) and one 3.5-hour in-lab testing session.

Workgroup	Title	Submission Links
WP0.1 Cognition	Assessing Cognitive Flexibility, Other Executive Functions and Learning in Healthy Young Adults	Pre-registration link: https://osf.io/6rc9h

Update on Schools Workgroup

Translation to Education: Assessing Cognitive Flexibility, Other Executive Functions and Learning in Healthy Adolescents

The School Workgroup has been continuously working with the Cognition and Social Workgroup to implement large-scale data collection in schools and ideal-condition study for the WP0.1 Adolescent Study. The School Workgroup conducted a mass two-day data collection camp involving over 200 Secondary One students (whole cohort) in May 2022. As of 30 September 2022, the Adolescent Study has completed data collection for 247 participants (190 in school setting and 57 in ideal condition setting). The School Workgroup has worked closely with the Cognition and Social Workgroups to establish the preprocessing and data analysis framework of the CLIC WP0.1 Study. In addition, the School Workgroup continues to meet with schools to share and update on the research progress and to explore the impact of educational neuroscience research in practice.

Since the last report and under the guidance of **Prof David HUNG (PI, NTU/NIE)**, **Dr Chew Lee TEO (Co-PI, NTU)**, and **Dr Peter SEOW (Co-PI, NTU/NIE)**, **Dr Nastassja LOPES FISCHER (Research Fellow, NTU)** has liaised closely with the Adults' Working Groups (i.e., Cognition and Social teams) to ensure that data collection procedures, coding and analysis are properly aligned across the Working Groups. In particular, she coordinated the mass two-day data collection in May 2022 and the 1:1 testing. Dr Lopes Fischer performed the analysis of multilingualism and social variables data using the adolescent sample in collaboration with the Social Workgroup. She presented the key findings from the analysis of these variables over two data analysis "bootcamps" held in June-July in Cambridge and August-September in Singapore. Finally, Dr Lopes Fischer led the coordinating and compiling of key findings and project milestones for CLIC's Progress Report which will be presented to the Scientific Advisory Committee (SAC) in October 2022.

Research Fellow **Dr Ryutaro UCHIYAMA (NTU)** has been leading the analysis of the adolescent data in CLIC and has also been playing a significant role in the analysis of the adult cognition data. Dr Uchiyama presented key findings from the adolescent data as well as the creativity data in both adults and adolescents during the two data analysis bootcamps. He has been working with the School Team Research Assistants to code the creativity tasks and has used new methods like semantic network analysis to study the relationship between cognitive flexibility and creativity.

Ms Yingqi CHUI (Research Assistant, NTU) and **Ms Phillis FU (Research Associate, NTU)** are also involved in data coding and analyses. They set the standard for the scoring of both the adult and adolescent responses and continue to test adolescent participants in laboratory settings, scored responses on the Woodcock-Johnson Tasks and coded the Alternate Uses Task which was presented at the analysis bootcamp. **Mr Timothy LEE (Research Associate, NTU)** was also involved in data processing and analysis of data collected from the WP0.1 Studies. Mr Lee contributed heavily to developing code for preprocessing the Adolescent WP0.1 dataset. He was also actively assisting with data analysis, contributing information on data pre-processing, and following-up on further work uncovered at the workshops at the first bootcamp.

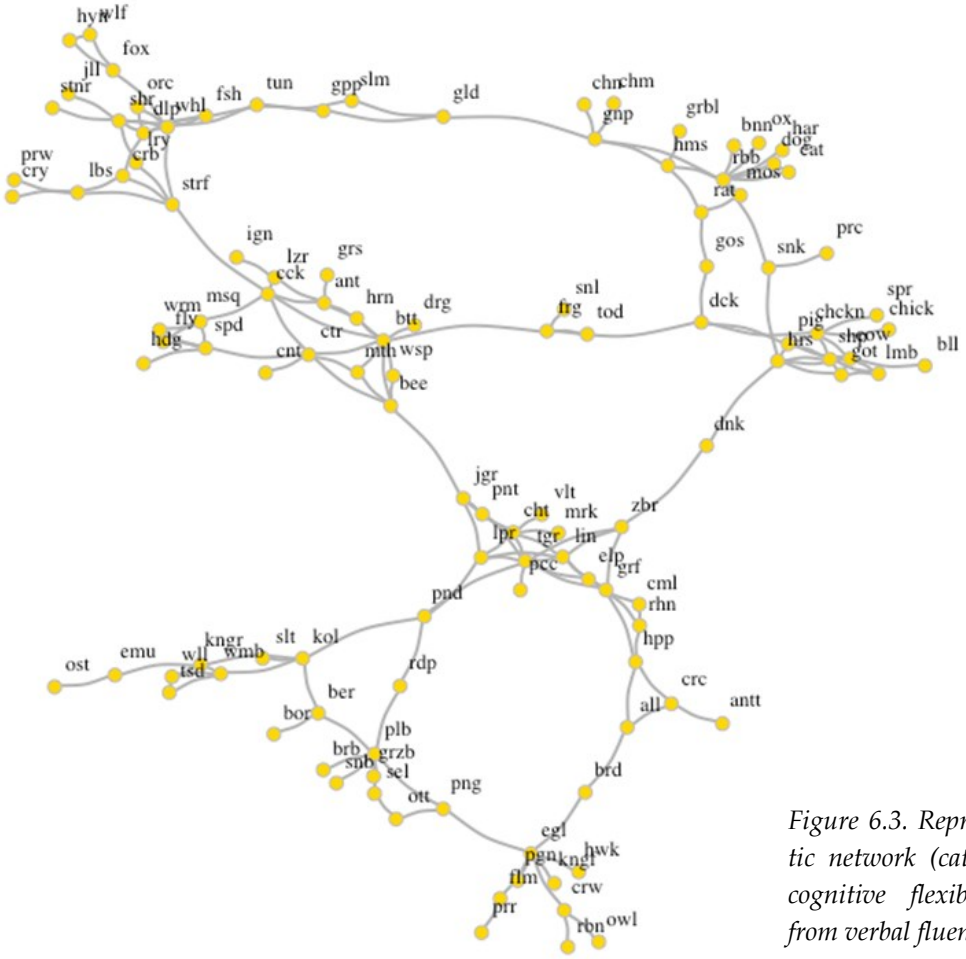


Figure 6.3. Representation of the semantic network (category: animals) of high cognitive flexibility adults, estimated from verbal fluency data.

The School Workgroup under the guidance of Prof Hung, Dr Teo, **Prof Michelle ELLEFSON (PI, CAM)** and **Dr Peter SEOW (Co-PI, NTU/ NIE)** developed and implemented a novel approach to conduct mass data collection in a school. This 2-day data collection was packaged into a "Brain Camp", which brought neuropsychological assessment to an ecological classroom setting. The mass data collection camp received strong support from many key Research Fellows and Research Assistants from the other work groups. There were multiple complexities in this mass data collection, ranging from logistical, technological and digital requirements, coordination with the school's schedule and space demands, student assistants and teachers' training, as well as making sure students and participants were motivated and rested to complete the task battery. Apart from the task battery, students were engaged in tinkering and craft activities such as the neuron building activity (Figure 6.4 and 6.5).

Figure 6.4. A student's neuron model from the Make-a-Neuron engagement activity

Figure 6.5. Brain Camp featured on CLIC's social media accounts

The School Workgroup continues to work closely with the Cognition Workgroup to understand educational neuroscience. Dr Lopes Fischer, under the guidance of Assoc Prof Leong, has been involved in establishing new computational modelling methods to analyse infants' electroencephalographic (EEG) signals and unveil the neural processes behind new language pattern assimilation. She has co-authored two journal papers (DOI: 10.1002/hbm.25759 and DOI: 10.1002/hbm.26035).

Dr Teo, Dr Lopes Fischer, and Assoc Prof Leong, collaborated with Research Scientist Dr Katherine YUAN from the Centre for Research in Practice and Pedagogy (CRPP) to conduct a pre-conference workshop titled "Exploring the Bridge between Educational Neuroscience and Learning Sciences in Knowledge Building Classrooms" at the International Society of the Learning Sciences (ISLS) Annual Meeting 2022. Ms Chui and Ms Fu also supported the discussion and activities during the virtual workshop.

In early June 2022, the School Workgroup participated in the Redesigning Pedagogy International Conference (RPIC) hosted by the National Institute of Education, Singapore. For the full details outlining the School Workgroup's outreach activities, see "Other activities and achievements" in this chapter section.

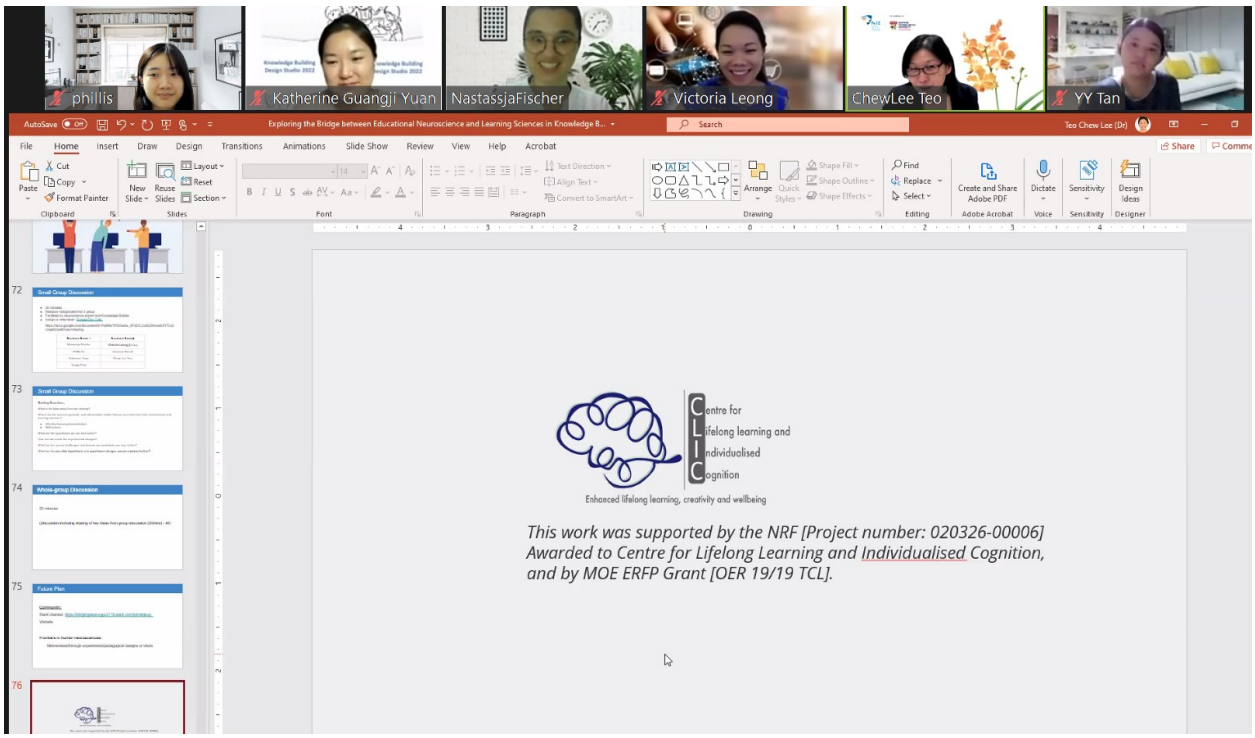


Figure 6.6. International Society of Learning Sciences (ISLS) conference workshop.

Workgroup	**Title**	**Submission Links**
WP0.1 Translation to Education	Assessing Cognitive Flexibility, Other Executive Functions and Learning in Healthy Adolescents	Pre-registration link: https://osf.io/md4tv/

Update on Social Workgroup

Cognitive Flexibility and Its Association with Linguistic Preferences, Decision-Making, Tolerance of Uncertainty and Perceived Social Support

Social Workgroup

The Social Workgroup is particularly focused on the constructs pertaining to multilingualism, tolerance of uncertainty, perceived social support, social decision-making, and, recently, career development. Since April 2022, the Social Workgroup has been continuously collecting data from (online) questionnaires and (lab-based) cognition data for WP0.1. At the time of writing, N=190 adult participants have fully completed the WP0.1 Study (both the online and face-to-face portions). Research Assistants with the Social Workgroup are also assisting the Neuroimaging Workgroup with data collection for WP0.2. **Dr Shengchuang FENG (Research Fellow, NTU)**, **Dr Nadhilla Velda MELIA (Research Fellow, NTU)** and **Ms Emma Yoke Loo SAM (PhD Student, IGP-CRADLE)** have been reviewing the quality of the incoming data by checking participants' attention while completing the online surveys, tracking any missing data, and monitoring participants' "straight-lining" behaviour. The team also monitors the online Qualtrics survey platform closely for any issues. Finally, the Social Workgroup has prepared and submitted pre-registrations for the socio-cognitive hypotheses pertaining to the socio-cognitive variables in the Adult Study.

Dr Feng, Dr Melia, and Ms Sam have also been conducting some initial data analyses of the social data; some preliminary and encouraging findings will be presented at the upcoming SAC meeting at the end of October. The Research Fellows presented some of these preliminary results in the 2nd Joint Cambridge-NTU Data Analysis Bootcamp held on 31 August 2022 to 9 September 2022. The discussions from the Social Workgroup were led by **Assoc Prof Georgios CHRISTOPOULOS (PI, NTU)**.

The Social Workgroup also leads and organises data collection for the adult population (online and face to face studies). Assoc Prof Christopoulos supervised the Research Fellows on their analysis of the relationship between social factors and variables of interest, including cognitive flexibility and structure learning. Assoc Prof Christopoulos, along with **Prof Annabel CHEN (PI & Director, NTU)** also received non-CLIC funding for Ms Sam who currently examines the relationship between cognitive flexibility and career choices. Finally, Assoc Prof Christopoulos and his team have provided measurements for social factors in WP0.2 and the adolescent data.

Prof Henriëtte HENDRIKS (PI & Deputy Director, CAM) works closely with the data and communications team, overseeing the presence of the project on social media, and issues related to data protection. She was also involved with the organisation of the first analysis bootcamp held in Cambridge for the CLIC project. During both the June and September bootcamps, she co-led the sessions on social factors, and language and maths.

Prof Hendriks has continued to be involved in the day-to-day running of the Social Workgroup and attends the weekly Social Workgroup meetings on Tuesdays, along with the larger WP0.1 meeting on Fridays. She has been involved in the writing of the pre-registration documents and IRB documentation related to the Social and Cognition group data collection. She has also been closely involved in the preliminary analyses related to multilingualism, and the reporting for the SAC meeting in October 2022. She was integral to the discussion to move data collection online and coordinating this between the Cognition and Social Workgroups (in collaboration with Assoc Prof Leong).

Dr Feng has contributed to the analysis of social decision-making variables and the testing of related hypotheses. He has submitted a pre-registration titled "Social Decision-Making and

Its Association with Cognitive Flexibility in Healthy Young Adults" (DOI: 10.17605/OSF.IO/JB38T; Feng et al., 2022) and has continued data cleaning and analysis of social decision-making questionnaires and tasks in the adult sample, including reliabilities of scales, descriptive statistics, correlations, exploratory factor analysis, and regression analysis. He is collaborating with the School Workgroup to conduct data cleaning and basic analysis of social decision-making questionnaires and tasks in the adolescent sample, including preparing R scripts and conducting analyses of reliabilities of questionnaires, descriptive statistics, and correlations. The preliminary results from the above analysis were presented in the 2nd Joint Cambridge-NTU Data Analysis Bootcamp. He is also preparing a poster titled "Cooperativeness and Cognitive Flexibility" to be presented during the CLIC SAC review meeting.

Dr Melia has been conducting data quality checks, data pre-processing, and preliminary data analysis from the socio-cognitive questionnaires. Specifically, she is responsible for analysing the social variables of tolerance of uncertainty, perceived social support, and multilingualism. She is conducting analyses such as descriptive statistics, reliabilities, correlations, exploratory and confirmatory factor analyses, and the moderating effects of these variables on the relationship between cognitive flexibility and structure learning in preparation for the report and presentation for the SAC meeting. She is also collaborating with Dr Lopes Fischer from the School Workgroup to pre-process and analyse the multilingualism data. She presented the preliminary results from these analyses at the 2nd Joint Cambridge-NTU Data Analysis Bootcamp. She has also prepared a pre-registration for these analyses titled "Tolerance of Uncertainty, Perceived Social Support, and Their Association with Structure Learning and Cognitive Flexibility in Healthy Young Adults" (DOI: 10.17605/OSF.IO/SCJMP; Melia et al., 2022).

Ms Sam has contributed to the general logistical and administrative planning for WP0.1 (e.g., pre-registration and data analyses). She is also assisting with the participant recruitment for the WP0.1 study, such as determining the demographic characteristics of targeted populations. She is also responsible for the cognitive flexibility and career transition/adaptation sub-study and prepared a pre-registration titled "Cognitive and Social Aspects of Career Transition and Adaptation" (DOI: 10.17605/OSF.IO/N352U; Sam et al., 2022) under WP0.1. Specifically, for the report and presentation to the SAC, she examined how cognitive flexibility was associated with well-established career variables, such as career adaptability, career exploration, and perceived employability.

Ms Hui Shan YAP (Research Assistant, NTU) contributed to the general logistical and administrative planning for WP0.1 and WP0.2, including setting up multiple survey forms for different studies and designing ways to allow for questionnaires to be administered online, as well as drafting and updating relevant SOPs used for the data collection of social measures. She is also involved in the social measures data collection for WP0.1 Adults and WP0.2.

Ms Li Ling LEE (Research Assistant, NTU) contributed to the general logistical and administration for WP0.1. She is a part of the recruitment team where she is in-charge of broadcasting advertisements on various social platforms (e.g., emails, SONA, and telegram). As the lead coordinator for Part 2 of the study, she holds various tasks such as drafting data collection SOPs, scheduling testing session, and managing study compensation in the data collection process. Ms Lee is also responsible for drafting IRB amendments and documents to ensure that the study protocol is up to date with IRB guidelines.

Ms Yuan Ni CHAN (Research Assistant, NTU) assisted in the general logistical and administrative planning for WP0.1. This includes setting up the online survey forms, designing the decision-making games, and updating the relevant SOP and manuals. She is also currently involved in the data collection process for the WP0.1 Adults and WP0.2.

Ms Jia Ying PEI (Research Assistant, NTU) is part of the recruitment team for WP0.1 where she handles the logistics and administrative planning, including screening and recruiting eligible

adult participants into the study. She is involved in the data collection for WP0.1 Adults and the training and management of NTU's undergraduate student assistants to administer various computerised and cognitive tasks to assist with the data collection process. In addition, Ms Pei is the data management lead of the Social Workgroup and ensures that the social group adheres to the data compliance guidelines.

References

Feng, S., Christopoulos, G. I., Hendriks, H., Melia, N., Sam, Y. L., Yap, H., … Chen, S. A. (2022, July 25). Social Decision-Making and Its Association with Cognitive Flexibility in Healthy Young Adults. https://doi.org/10.17605/OSF.IO/JB38T

Melia, N., Christopoulos, G. I., Hendriks, H., Feng, S., Sam, Y. L., Yap, H., … Chen, S. A. (2022, July 29). Tolerance of Uncertainty, Perceived Social Support, and Their Association with Structure Learning and Cognitive Flexibility in Healthy Young Adults. https://doi.org/10.17605/OSF.IO/SCJMP

Sam, Y. L., Christopoulos, G. I., Chen, S. A., Feng, S., Melani, I., Melia, N., … Robbins, T. W. (2022, May 24). Cognitive and Social Aspects of Career Transition and Adaptation. https://doi.org/10.17605/OSF.IO/N352U

Workgroup	**Title**	**Submission Links**
WP0.1 Social	Cognitive Flexibility and Its Association with Linguistic Preferences, Decision-Making, Tolerance of Uncertainty and Perceived Social Support	Pre-registration link: https://osf.io/ay9gr

Update on Neuroimaging Workgroup

Neuroimaging: Structure Learning Training and Cognitive Flexibility

The Neuroimaging Workgroup ran and completed Pilot 3 to trial a new intervention design that includes two training stages, (Stage 1 Training at 80/20 contingency and Stage 2 Training at 70/30 contingency) and two test points (test of generalisation and test of reversal). Data analyses for Pilot 3 was completed by **Dr Xiaoqin CHENG (Research Fellow, NTU)**. Based on the results from Pilot 3 and upon consultations with **Prof Annabel CHEN (PI & Director, NTU), Assoc Prof Victoria Leong (PI & Deputy Director, NTU), Prof Zoe KOURTZI (PI & Director, CAM)**, **Prof Barbara SAHAKIAN (Senior Scientific Advisor, CAM)**, and **Prof Trevor ROBBINS (Senior Scientific Advisor, CAM)**, a unanimous decision was made to use the same intervention design implemented in Pilot 3 as the intervention design in WP0.2 Main Study.

Apart from Pilot 3, **Dr Chia-Lun LIU (Research Fellow, NTU)** and Dr Cheng completed MRI pilots in May in preparation for WP0.2 Main Study. In total, the full CLIC MRI protocol was run on 11 volunteers and data quality checks were performed on the acquired data. The MRI sequence protocols were then optimised to balance between data quality (signal-to-noise ratio) and practicability (total scanning time). Even though all MRI sequences run in CLIC are motion-sensitive, the different MRI modalities were ordered to prioritise protocols that are most susceptible to motions to ensure data quality.

Dr Liu and Dr Cheng then proceeded to finalise the MRI protocols for multiparameter parameter mapping (MPM), resting-state functional magnetic resonance imaging (rsfMRI), and magnetic resonance spectroscopy (MRS) through several consultation with CLIC's imaging leads, **Prof John SUCKLING (Imaging Lead, CAM)** and **Prof Balázs GULYÁS (Imaging Lead, CAM)**, alongside critical inputs from Prof Chen and Prof Kourtzi**.** These discussions also contributed to the development of analyses pipelines for the different MRI modalities. For instance, considering the huge impact that physiological noise has on resting-state brain activity, the Neuroimaging team will hence acquire respiratory data and use that

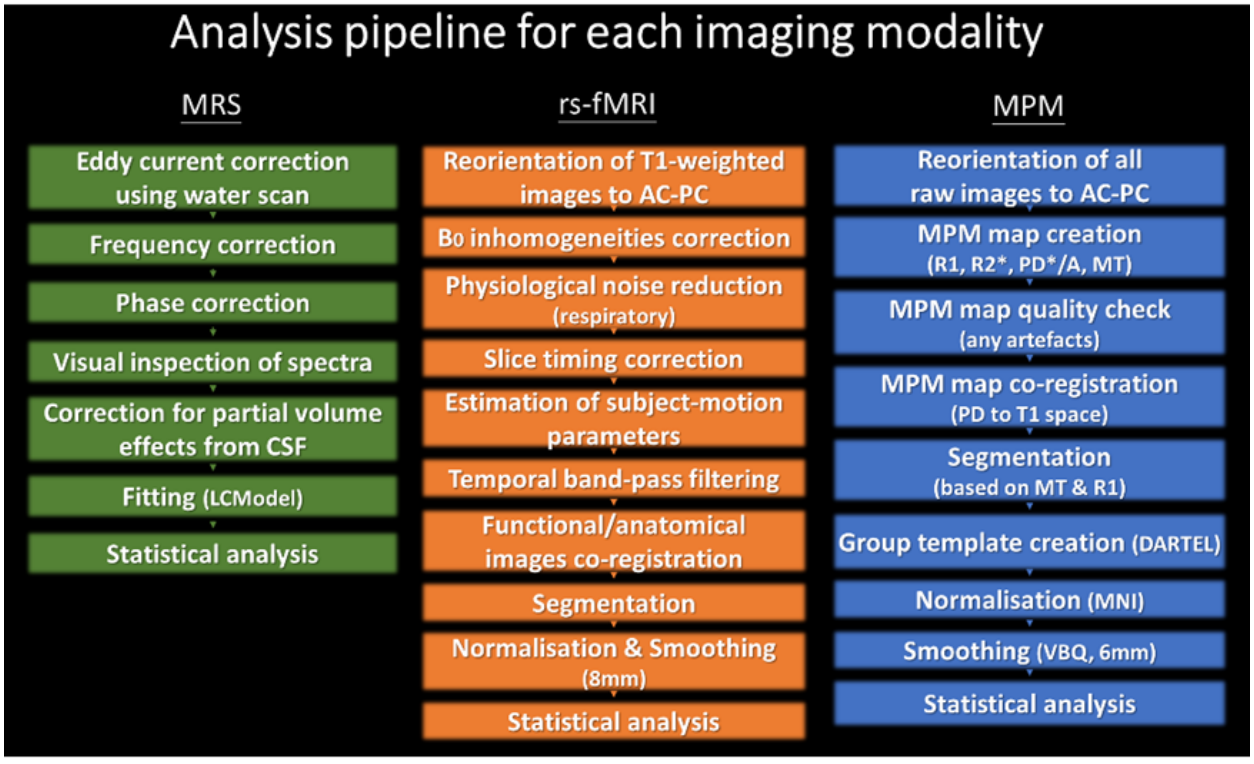


Figure 6.7. Finalised analysis pipeline for MRS, rs-fMRI and MPM.

to directly regress the influence of physiological activities on resting-state brain activity instead of indirectly removing these influences via independent component analysis. The finalised analysis pipelines for each MRI modalities can be found in Figure 6.7.

Prior to the start of data collection for WP0.2 Main Study, operational planning was extensively undertaken to accommodate multiple longitudinal behavioural sessions and pre- and post-intervention MRI scans. Recruitment, scheduling and data collection workflow for WP0.2 Main Study were planned and implemented with the help of **Ms Jia Li TEO (Research Assistant, NTU)** and **Ms Janet TAN (Student Assistant, NTU)**. Participants were recruited in batches to facilitate data analyses of baseline sessions for subsequent matching and group assignment of participants into Control or Training group. The entire study needed to span a minimum of two months for each participant, and due to the longitudinal nature of WP0.2 Main Study, experimenters handled participants at different phases of the study. Hence, an optimal rate of recruitment needs to be determined to ensure sufficient manpower and to avoid overscheduling experimenters with several concurrently ongoing participants. At present, the entire recruitment workflow is managed by Ms Tan upon her onboarding as a full-time Research Assistant in August 2022.

Ms Teo and Dr Cheng also conducted a four-month planning of data collection (June 2022 to September 2022) of the Main Study. The Neuroimaging Workgroup had multiple meetings with the NTU Cognitive Neuroimaging Centre (CoNiC) helmed by Prof Gulyás to coordinate pre - and post-MRI scans for participants in the Main Study. With the help of CoNiC, designated scan days were assigned to and pre-booked by CLIC to facilitate scheduling of participants in the Main Study. Concurrently, Dr Liu, Dr Cheng, **Ms Winlynn CHOO (Research Associate, NTU)** and Ms Teo also organised and trained the student Research Assistants under the Neuroimaging Workgroup in task administration of CLIC's cognitive battery as well as in the standard workflow of the MRI sessions (Figure 6.8). Each student Research Assistant went through up to 20 hours of training, including both hands-on and theory sessions to better understand the different tasks

Figure 6.8. MRI training and simulations to familiarise the student Research Assistants with the workflow of the MRI session. Full-time research staff in the Neuroimaging Workgroup acted in the capacity of mock participants so that student Research Assistants could practise what to do during informed consent, mock scanner session and participant preparation in the scanner suite prior to the actual scan.

and the importance for tasks to be administered in a consistent manner. They were then assigned different roles (e.g., recruitment, operations and/or data collection) and were delegated further training for their specific assigned roles.

In anticipation of heavy computational needs once data collection for WP0.2 Main Study starts, Ms Choo completed the set-up of the Ubuntu Analyses Server (CLIN40003) for the Neuroimaging Workgroup in July 2022. At present, all installations of essential MRI analyses software have been completed and software updates are maintained by Ms Choo. Concurrently, she is also in-charge of maintaining the REDCap web server and ensures that any updates to the backend is successfully brought to production.

The Neuroimaging Workgroup formally began data collection for WP0.2 Main Study in the first week of June after incorporating critical insights from Pilot 3, MRI pilots, and discussions sessions with the PIs. However, the MRI data collection was stalled by unexpected technical issues (software and hardware) with the Siemens 3T MRI Scanner, in spite of extensive piloting of the MRI scanning protocol. Despite extensive piloting, several scan-related issues emerged — specific absorption rate of the participant would exceed even at the start of a scan, the scanner bed would become stuck, and issues occurred with the MRI head coil. Efforts were made by CoNiC and the Neuroimaging Workgroup to resolve the scanner issues and extensive discussions of the scanner issues were had with CoNiC and Siemens to pinpoint underlying reasons. Scan sequence order was adjusted for a smoother MRI data collection. With the help of CoNiC, the team also contacted Siemens professionals who checked the WP0.2 MRI protocol. In addition, the team also paused MRI data collection for two weeks from 6 - 20 August to make provisions for a software upgrade that would help mitigate the scanner issues going forward.

Analyses of behavioural and MRI data for WP0.2 Main Study was carried out by Dr Cheng and Dr Liu with assistance from Ms Choo and Ms Tan. Ms Choo ensures the smooth running of analyses pipelines on the Ubuntu server and has been instrumental in the navigation and resolution of various network complexities. Ms Tan assisted Dr Cheng with data collation of cognitive flexibility tasks ran during the post-training phase of the study. However, due to technical issues, the sample size used in the current analyses is rather limited. In addition, as all analyses were done amidst ongoing data collection, these analyses are preliminary and will likely change with an increase in sample size and more time for analyses once data collection is completed.

For the behavioural data, 33 participants (15 Control, 18 Training) from Pilot 3 and the Main Study were analysed together to enhance the power of the analyses. Initial analyses demonstrated that strategy use during structure learning training, in particular the use of Maximising strategy, is critical for success in the test of reversal where the exhibition of reversal is considered a trademark of cognitive flexibility, after structure learning training. For the MRI data, initial analyses of 19 participants (8 Control, 11 Training) showed promising trends of a relationship between strategy use and changes in functional connectivity after structure learning training. As data is coming in at a consistent rate, Ms Choo is currently working on implementing a bash script that can automate the sorting and renaming of MRI data into their respective folders grouped by modality in accordance with the Brain Imaging Data Structure (BIDS) format. The implementation of this bash script will substantially streamline MRI data pre-processing.

As of 30 September 2022, the Neuroimaging Workgroup has completed data collection for 26 participants (14 Control, 12 Training). At present, 22 participants (11 Control, 11 Training) are still in the midst and at varying phases of the study.

The Neuroimaging Workgroup also welcomed a new **Research Associate**, **Ms Min HONG,** who officially commenced duty on 29 September 2022. She has started receiving relevant training with the support of current team members and is expected to assist with data collection in October 2022. Ms Hong has had ample experience with participant preparation for MRI scans as a Master's student in Peking University. She is also familiar with the administration and analyses for several tasks, such as Task Set Switching, Stroop

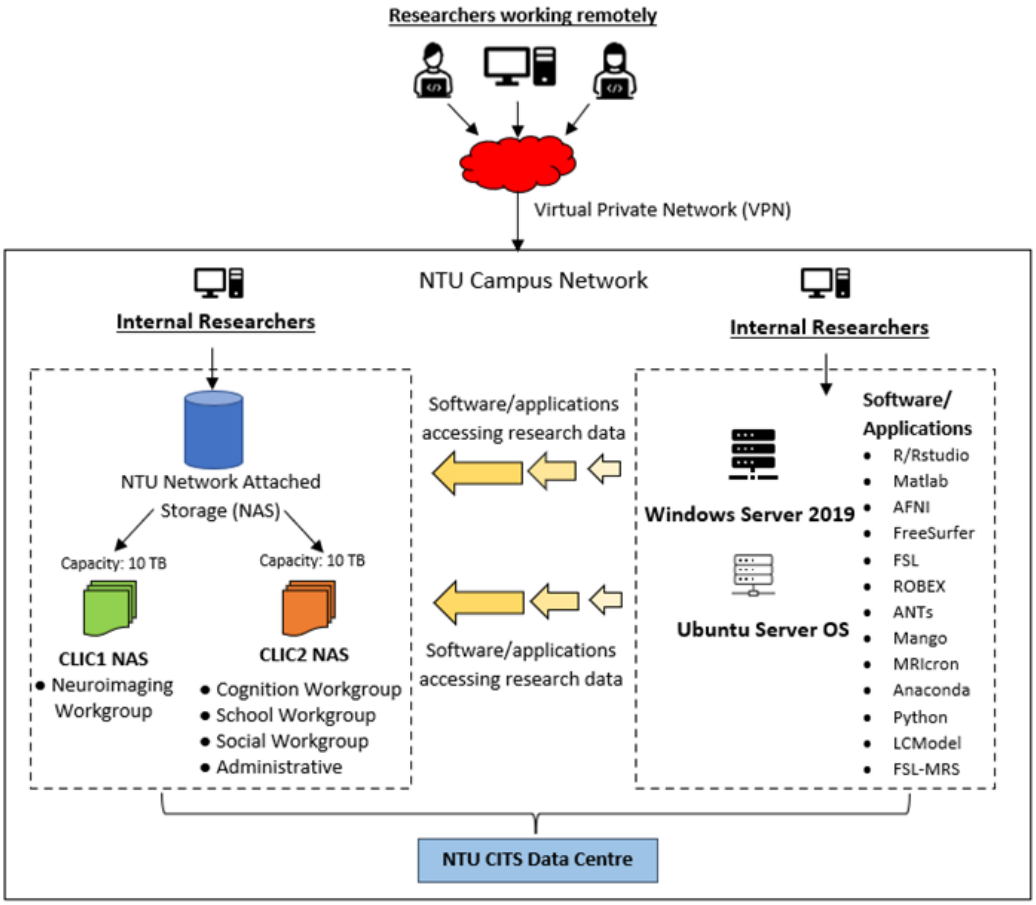


Figure 6.9. CLIC data storage and server computing infrastructure.

Task and Raven's Advanced Progressive Matrices, all of which are within CLIC's cognitive task battery.

Dr Sheng Hung CHUNG (Research Engineer, NTU) oversees the overall maintenance of CLIC data storage and server computing infrastructure as exemplified in Figure 6.9. The Network Attach Storage (NAS) provided by NTU CITS (Centre for IT Services) is a dedicated file storage that enables multiple researchers/workstations to retrieve data from a centralised disk via a dedicated secured shared drive. CLIC researchers within the NTU campus (internal) may access CLIC resources via NTU Local Area Network/ Wireless connection. Researchers who are off-campus (working remotely) are required to access to NTU Virtual Private Network (VPN) that allows the users to access NTU systems and resources remotely.

CLIC is currently being provisioned with 20 terabytes (TB) capacity of CLIC NAS storage and which is distributed into 10 TB each for two storage clusters to cater for different research Workgroups (Neuroimaging, Cognition, School, and Social) and administrative tasks. In addition, two computing servers (Windows Server 2019 and Linux Server – Ubuntu Server OS) have been set up with various computing software for researchers to perform analysis and computations. The following software/applications are setup and configured in the computing servers which include R/Rstudio, Matlab, AFNI, FreeSurfer, FSL, ROBEX, ANTs, Mango, MRIcron, Anaconda, Python, LCModel and FSL-MRS.

Workgroup	Title	Submission Links
WP0.2 Neuroimaging	Structure Learning Training and Cognitive Flexibility	Pre-registration link: *Pending Submission*

Pre-registered Studies

The following are the pre-registered studies generated by CLIC during the reporting period.

Cognitive and Social Aspects of Career Transition and Adaptation
Yoke Loo Sam, George I Christopoulos, Annabel Chen, Shengchuang Feng, Irene Melani, Nadhilla Melia, Henriëtte Hendriks, Ke Tong, Victoria Leong, Barbara Sahakian, Zoe Kourtzi, and Trevor William Robbins, *OSF Registries*

DOI: 10.17605/OSF.IO/N352U

Description: The present pre-registration primarily focuses on addressing the specific hypotheses regarding the relationship between cognitive flexibility, career development resources, behaviours, and outcomes, which might not be described in the main studies (DOI: 10.17605/OSF.IO/6RC9H and DOI: 10.17605/OSF.IO/AY9GR) in detail. This project aims to utilise both experimental cognitive tasks and psychometric assessments to: 1) assess the degree to which cognitive flexibility contributes to aspects of career adaptability over and above working memory and inhibitory control. 2) examine the interrelationship between cognitive flexibility, career adaptability, and related career beliefs, behaviours, and outcomes.

Tolerance of Uncertainty, Perceived Social Support, and Their Association with Structure Learning and Cognitive Flexibility in Healthy Young Adults
Nadhilla Melia, George I Christopoulos, Henriëtte Hendriks, Shengchuang Feng, Yoke Loo Sam, Yap Hui Shan, Ke Tong, Ryutaro Uchiyama, Xiaoqin Cheng, Victoria Leong, Barbara Sahakian, Trevor William Robbins, Zoe Kourtzi, and Annabel Chen, *OSF Registries*

DOI: 10.17605/OSF.IO/SCJMP

Description: The present pre-registration seeks to further elaborate on certain hypotheses and analyses related to tolerance of uncertainty (TOU) and perceived social support (PSS) that have been described, but without details in the main studies This study plans to generate specific scores for TOU and PSS, examine possible latent construct(s) for these variables, evaluate their associations with cognitive flexibility, and assess their interactions with the association between cognitive flexibility and structure learning. These analyses will help to identify potential factors that may influence the effectiveness of cognitive flexibility training.

Social Decision-Making and Its Association with Cognitive Flexibility in Healthy Young Adults
Shengchuang Feng, George I Christopoulos, Henriëtte Hendriks, Nadhilla Melia, Yoke Loo Sam, Yap Hui Shan, Ke Tong, Ryutaro Uchiyama, Xiaoqin Cheng, Victoria Leong, Barbara Sahakian, Trevor William Robbins, Zoe Kourtzi, and Annabel Chen, *OSF Registries*

DOI: 10.17605/OSF.IO/JB38T

Description: The present pre-registration seeks to further elaborate on certain hypotheses and analyses related to social decision-making that have been described, but without details in the main studies. This study plans to generate specific scores for each social decision-making variable, examine possible latent construct(s) for these variables, evaluate their associations with cognitive flexibility, and assess their interactions with the association between cognitive flexibility and structure learning. These analyses will better our understanding of the components of social decision-making, unveil the connection between social decision-making and cognitive flexibility, and help to identify potential factors that may influence the effectiveness of cognitive flexibility training.

Other activities and achievements

Prof Trevor ROBBINS (Senior Scientific Advisor CAM) received the William James Fellow Award from the Association of Psychological Science (APS) in May 2022.

Prof Robbins was also awarded a 3-year grant ($600K, Co-PI) by the Foundation for OCD Research (FFOR) on "Neural circuitry underlying loss of control over goal-directed behavior in obsessive-compulsive disorder" which began on 1 June 2022.

School Workgroup

The School Workgroup conducted a pre-conference workshop titled "Exploring the Bridge between Educational Neuroscience and Learning Sciences in Knowledge Building Classrooms" at the 2nd International Society of the Learning Sciences (ISLS) Annual Meeting 2022 held from 6-10 June 2022.

There was strong participation from the School Workgroup at the Reinventing Pedagogy International Conference hosted by the National Institute of Education Singapore from 30 May – 1 June 2022.

- The School Panel consisting of **Prof David HUNG (PI, NTU/NIE)**, **Dr Chew Lee TEO (Co-PI, NTU)**, **Dr Peter SEOW (Co-PI, NTU/NIE)**, **Dr Nastassja LOPES FISCHER (Research Fellow, NTU)**, **Dr Ryutaro UCHIYAMA (Research Fellow, NTU)**, **Mr Timothy LEE (Research Associate, NTU)**, and **Ms Yingqi CHUI (Research Assistant, NTU)** co-authored a conference paper titled "Investigation of cognitive flexibility and learning in adolescents".
- Dr Lopes Fischer, Mr Lee and Dr Teo presented a workshop session titled "Developing Applied Knowledge in the Role of Cognitive Flexibility, Working Memory and Inhibitory Control in Students' Learning and its Pedagogical Implications" to connect researchers and teachers in neuroscience, cognition, and teaching practice.
- Dr Uchiyama presented a talk titled "Investigation of cognitive flexibility and learning in adolescents" together with the other members of the School Team.
- Dr Uchiyama also led another talk with Dr Teo, Prof Hung, and Mr Lee titled "A neurodevelopmental learning map for education research"

The team has been involved in several engagements with Singapore educators to connect CLIC's research in psychology and neuroscience with educational policy and the experience of teachers. In this reporting period., the team has engaged with the Singapore Examinations and Assessment Board (SEAB) on the assessment of 21st Century Competencies. In addition, there are continued engagements with schools on the connection between CLIC's research in executive function, teaching, and learning.

Cognition Workgroup

Cambridge Cognition recently interviewed **Assoc Prof Victoria LEONG (PI & Deputy Director, NTU)** on her development of the new experiences with Remote Guided Testing method, which benchmarked how CLIC has incorporated CANTAB™ tasks into its testing protocols. In the email interview, the Cognition Workgroup has outlined the methodology, alongside results and implications of the study published in the Journal of Medical Internet Research in January 2022. A blog post of the interview can be found on the Cambridge Cognition website: https://www.cambridgecognition.com/blog/entry/developing-method-experimenter-led-online-cognitive-assessments

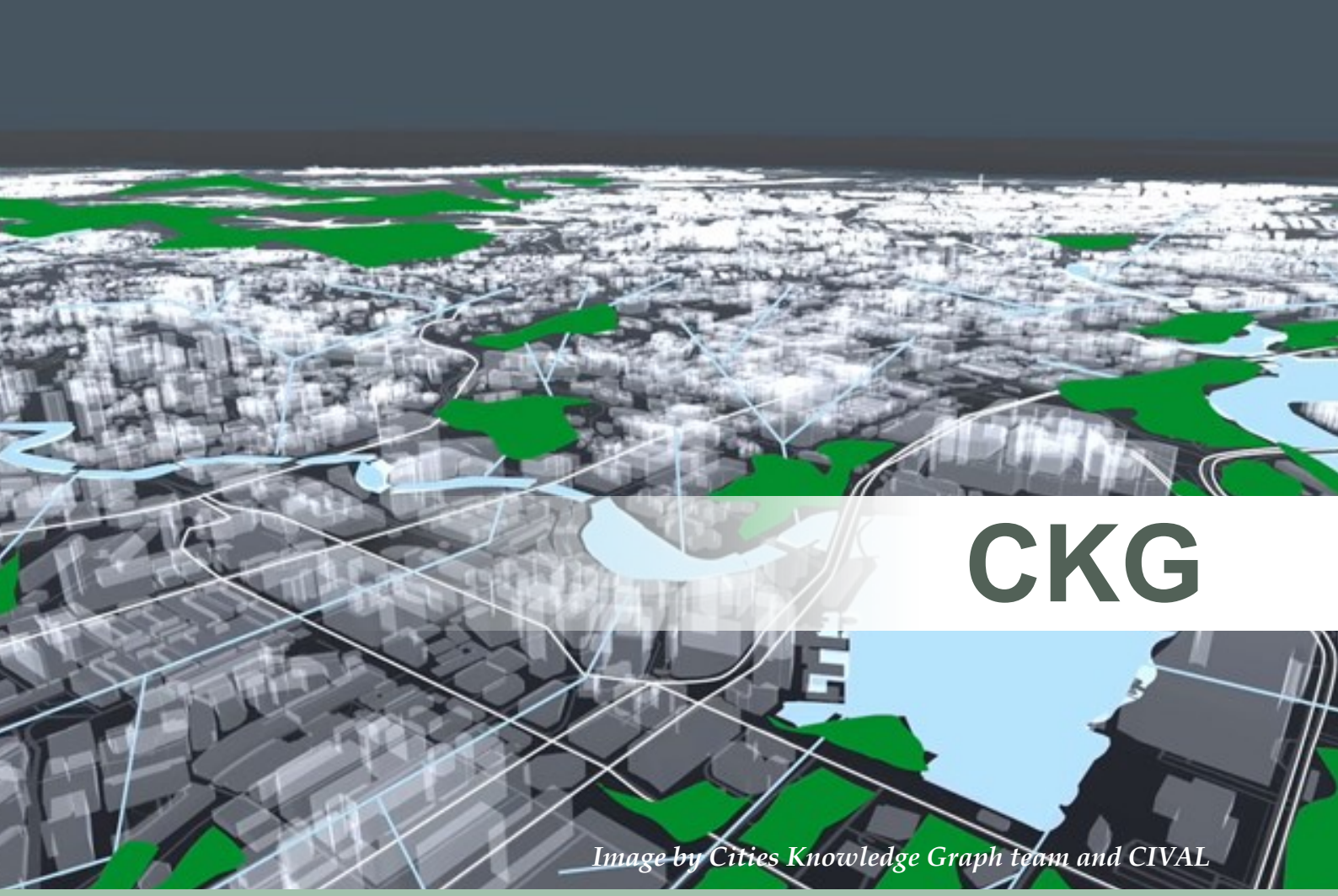


Image by Cities Knowledge Graph team and CIVAL

CITIES KNOWLEDGE GRAPH

Cities Knowledge Graph (CKG) aims to transform city planning related data, information and knowledge into a semantic and extensible platform – a knowledge graph. The proposed CKG would be similar to a knowledge management system for urban planning, integrating information from various sources and domains, evaluating planning proposals against visions and targets set for future urban development, and supporting policymakers and planners by mapping interesting planning directions. It further ties together existing 3D geo-databases, as well as novel analysis, simulation and visualisation tools developed by CARES and SEC, creating an unprecedented knowledge graph for city planning.

CKG Principal Investigators:

Professor Markus KRAFT
University of Cambridge

Professor Stephen CAIRNS
(April 2020 – June 2022, moved from SEC)
ETH Zürich

Professor Martin RAUBAL
(July 2022 – present)
ETH Zürich

OVERVIEW

Cities Knowledge Graph (CKG) is an Intra-CREATE Thematic Grant project in the 'Cities' thematic area. The project brings together expertise from Cambridge CARES, the host institution of the project, and SEC (the Singapore-ETH Centre, established by ETH Zürich-the Swiss Federal Institute of Technology Zürich) and its Future Cities Laboratory Global programme. The team is led by Principal Investigators from the University of Cambridge (Prof Dr Markus Kraft) and ETH Zürich (Prof Dr Martin Raubal). Dr Pieter Herthogs (Senior Researcher, SEC) is Co-Investigator and Project Leader. Dr Aurel von Richthofen (Team Leader Cities, Arup Germany), Prof Dr Stephen Cairns (ETH) and Prof Dr Franziska Sielker (TU Vienna) are Co-Investigators of the project.

Over the past six months, we have developed the knowledge graph architecture further by introducing an Object Graph Mapper module that is analogous to the Object Relational Mapper engines commonly used with object-oriented programming languages. The Object Graph Mapper module facilitates the interaction with structured data in the knowledge graph. We have also developed a proof-of-concept Graph Inference agent that uses graph-based inference algorithms to make new conclusions and inferences based on existing data. This proof of concept demonstrates the application of three broad classes of graph algorithms: pathfinding, centrality detection and community detection on a subgraph consisting of Singapore plots data.

Furthermore, we have developed and implemented several new features for the various autonomous agents, including the City Export Agent, the City Information Agent, the City Energy Analyst Agent, and the Thematic Surface Discovery Agent. For instance, the City Export Agent can now export building data from the knowledge graph in different Levels of Detail – in footprint and geometry modes, and the tiling functionality can be executed automatically after the export is completed. The City Information Agent has been extended with plot filtering functionality for land uses and buildable space quantities. We have also prepared a preprint entitled 'Semantic 3D City Interfaces – intelligent interactions on Dynamic Geospatial Knowledge Graphs' to describe the work.

To demonstrate the potential applications and extensibility of the project, we are developing a Programmatic Plot Finder demonstrator to enable querying for plots that allow particular combinations of programmes or uses, a particular amount of buildable space, or a combination of both. This reduces the manual effort required to go through regulatory documentation of each plot considered for potential development.

We are continuing to develop various ontologies to include more concepts for city planning. For instance, a new preliminary ontology called 'OntoBuildableSpace' has been created using the OntoZoning ontology (for land-use planning concepts and regulations in Singapore) and the Units of Measure ontology (to formalise measurable characteristics of buildable space and built form on a plot).

Professor Markus Kraft, PI
University of Cambridge

Professor Martin Raubal, PI
ETH Zürich

Update on work package 1

Developing master-planning ontologies

To automate allowable Gross Floor Area (GFA) calculations, **Ms Ayda GRIŠIŪTĖ (Researcher, SEC)** and **Ms Heidi SILVENNOINEN (Researcher, SEC)** have developed a semantic spatial policy model. The first step toward representing building volume and land use regulations in Singapore was to create a script for calculating the allowed GFA of plots in the Singapore River Valley area based on the Urban Redevelopment Authority's (URA) land use planning regulations. Allowable GFA is calculated by cross-referencing planning regulations for each plot and extracting the applicable number of storeys, setbacks, allowed site coverage, and Gross Plot Ratio (GPR) values. The process starts with plot data from Singapore's Masterplan 2019 dataset. Since the allowed built form is influenced by many regulations set by URA in various mediums, this GFA calculation script only served as a proof of concept for representing a small subset of these complex regulations in a machine-readable way. The generated GFA dataset, which quantifies regulatory data and allowable built form for the Singapore River Valley area, can be queried together with zoning data in the knowledge graph for more detailed programme-based plot finding. The dataset provides a more differentiated view of allowed buildable space, with more than half of the plots included in the automated calculation process having multiple allowable GFAs. This increased differentiation could inform planners about efficient and feasible use combinations. The team, led by Ms Grišiūtė, has translated this work into a conference paper submitted for the CAADFutures 2023 conference, which will be held July 2023 in Delft, The Netherlands.

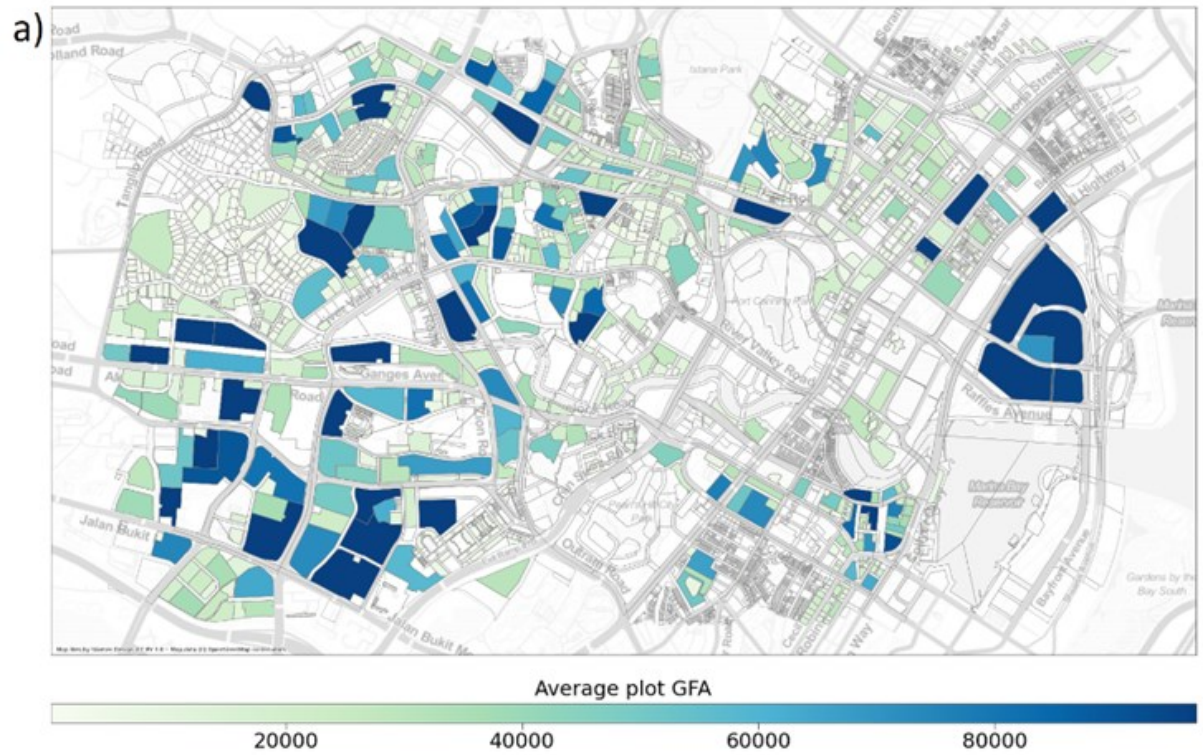


Figure 7.1 (a): The main generated GFA dataset characteristics. A gradient map displaying average allowable GFA values for plots in the Singapore River Valley area.

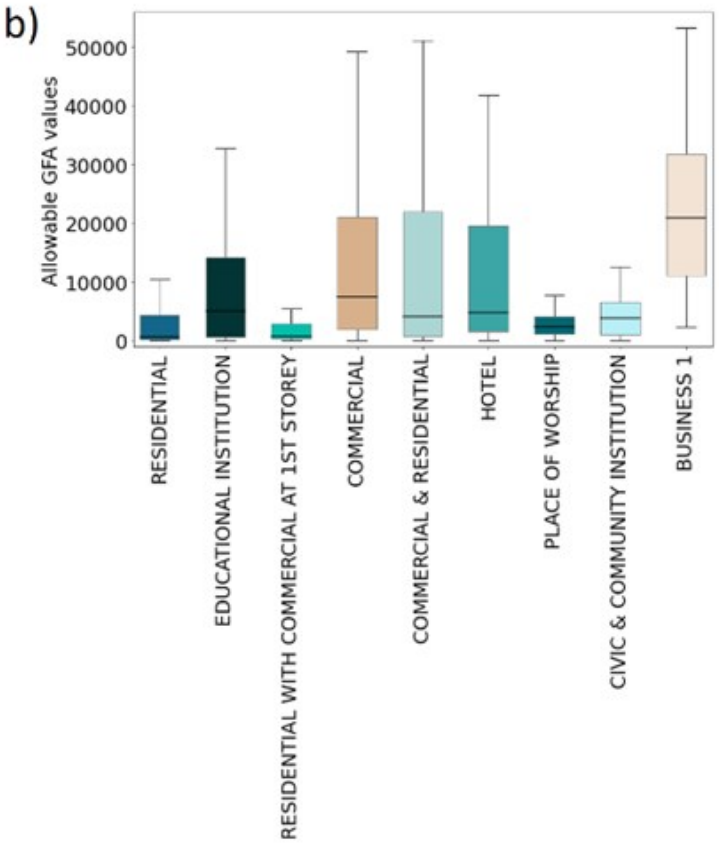


Figure 7.1 (b): The main generated GFA dataset characteristics. b) Distribution profile of allowable GFA values for individual zoning types.

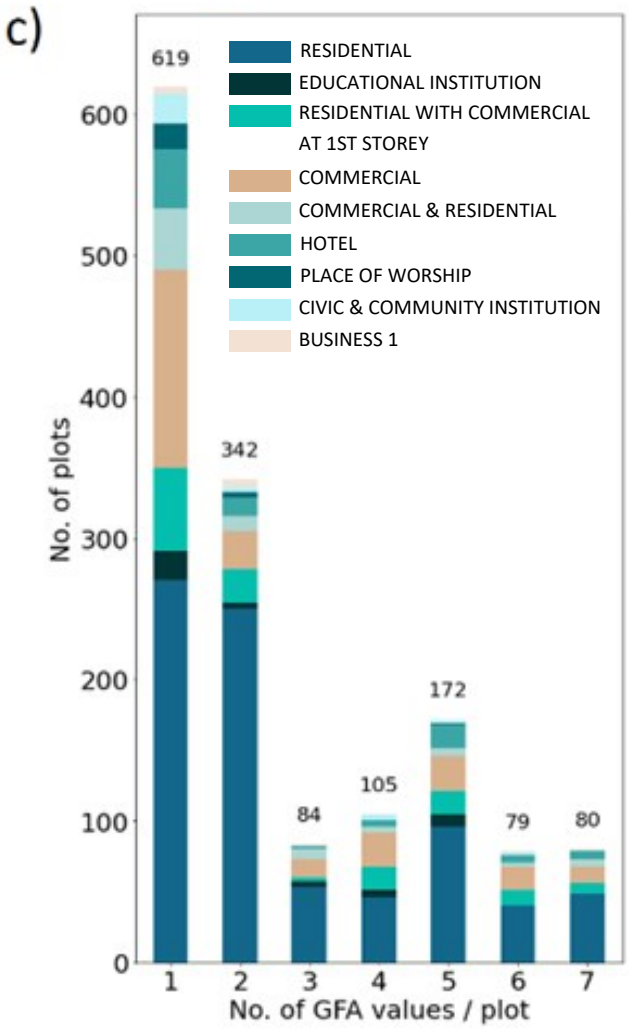


Figure 7.1 (c): The main generated GFA dataset characteristics. Bar chart illustrating how many plots have a certain number of allowable GFA values, thereby providing a more distinctive view of buildable spaces per plot.

Ms Grišiūtė is developing an ontology called 'OntoBuildableSpace', using the existing OntoZoning ontology (for land-use planning concepts and regulations in Singapore) and the Units of Measure ontology (to formalise measurable characteristics of buildable space on a plot). She has also developed an automated workflow to, as a proof of concept, semantically represent MATSim transport simulation outputs using existing ontologies (iCity Transport Planning Suite of Ontologies). This work consisted of two main parts: data mapping and automated triple generation. Concepts in the native MATSim output were mapped to transport ontology classes, and this mapping was used as a schema to generate semantic triples, which are stored in the knowledge graph. This automated semantic enrichment and the examples of resulting multi-domain urban indicators have been described in a conference paper and presented at the eCAADe 2022 international conference, which was held September 13-16 in Ghent, Belgium.

Ms Silvennoinen has continued to improve the OntoZoning ontology to increase its compatibility with the GFA calculations, inferencing, and urban energy modelling. This work has been preprinted and submitted to a journal.

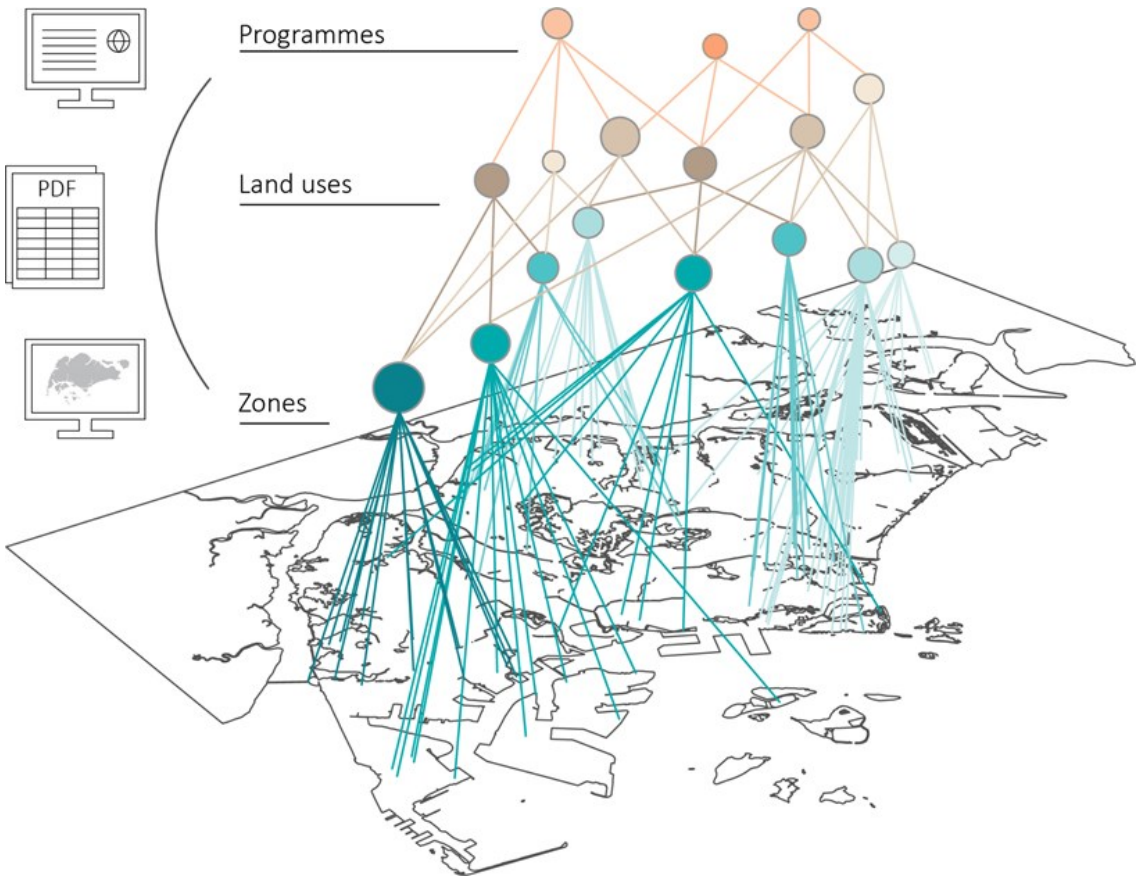


Figure 7.2: Graphical abstract for the preprint entitled 'Multi-criteria site selection using an ontology: the OntoZoning ontology of zones, land uses and programmes for Singapore', illustrating how diverse data are represented in a knowledge graph, enabling various types of geospatial queries to be performed.

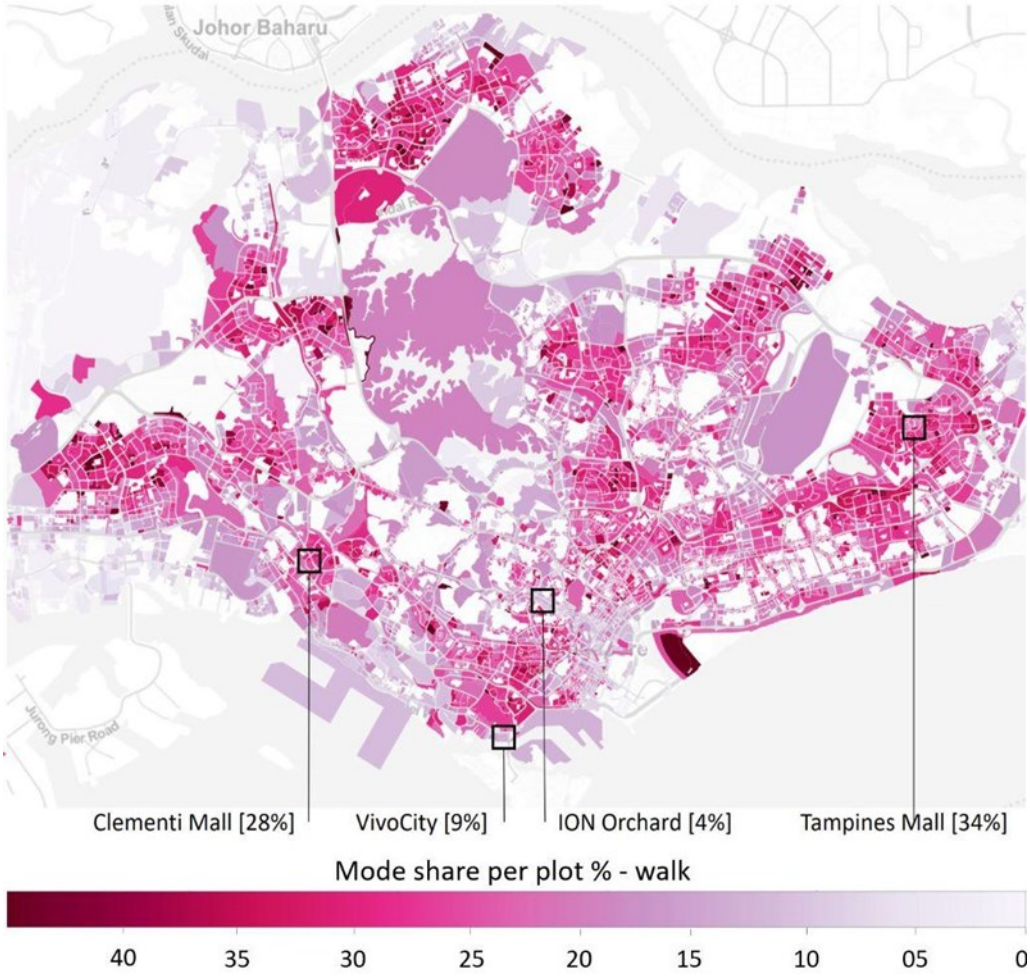


Figure 7.3: Map of plots in Singapore representing the share of simulated trips for a particular transport mode (walking) based on combining available transportation and land use data. Trips are assigned to plots based on the shortest distance between the trip destination coordinate on the road and a plot. The data and derived evaluation shown is part of a proof-of-concept exploration and are not validated.

Mr Yi-Kai TSAI (Software Developer, CARES) has extended the OntoUBEMMP ontology, which links two domains: Urban Building Energy Modelling (UBEM) and Master Planning (MP). The extensions include additional concepts for energy generating devices such as solar and photovoltaic thermal collectors, concepts to describe energy supply and consumption, and provision for representing different time resolutions.

Update on work package 2

Developing the knowledge graph's architecture

Mr Arkadiusz CHADZYNSKI (Senior Research Fellow, CARES), in collaboration with **Ms Mehal AGARWAL (Software Developer, CARES)** and Mr Jefferson CHUA (Non-C4T student, CAM), has developed an architecture that is comparable to the architectures following the Model-View-Controller (MVC) design pattern, at a high level. This architecture allows mapping the semantically stored objects to models in Java by using an Object Graph Mapper (OGM) engine developed for this purpose. It is analogous to the Object Relational Mapper (ORM) engines commonly used with object-oriented programming languages. The agent could be compared to the controller in the MVC – the agents operate on instances of the objects in Java that have been mapped to appropriate graphs. While agents perform intelligent operations on the instances, objects instantiation, the persistence of updates, additions and deletions are handled by the OGM engine automatically. Ms Agarwal has also prepared and released the OGM module as a library for ease of use.

Mr Chadzynski and **Ms Huay Yi TAI (Software Developer, CARES)** have designed and developed a proof of concept for utilising graph inference algorithms. In conjunction with ontology-based inference algorithms, graph inference algorithms are used to make new conclusions and inferences based on existing data. Graph-based algorithms consider only the graph structure, i.e., the result of interconnections of objects described using ontologies, and disregard any semantics of the relationships. The proof of concept demonstrates the application of three broad classes of graph algorithms: pathfinding, centrality detection, and community detection. Pathfinding algorithms analyse the graph structures to identify a path between nodes that satisfies certain criteria, e.g., the shortest path. Centrality detection algo-

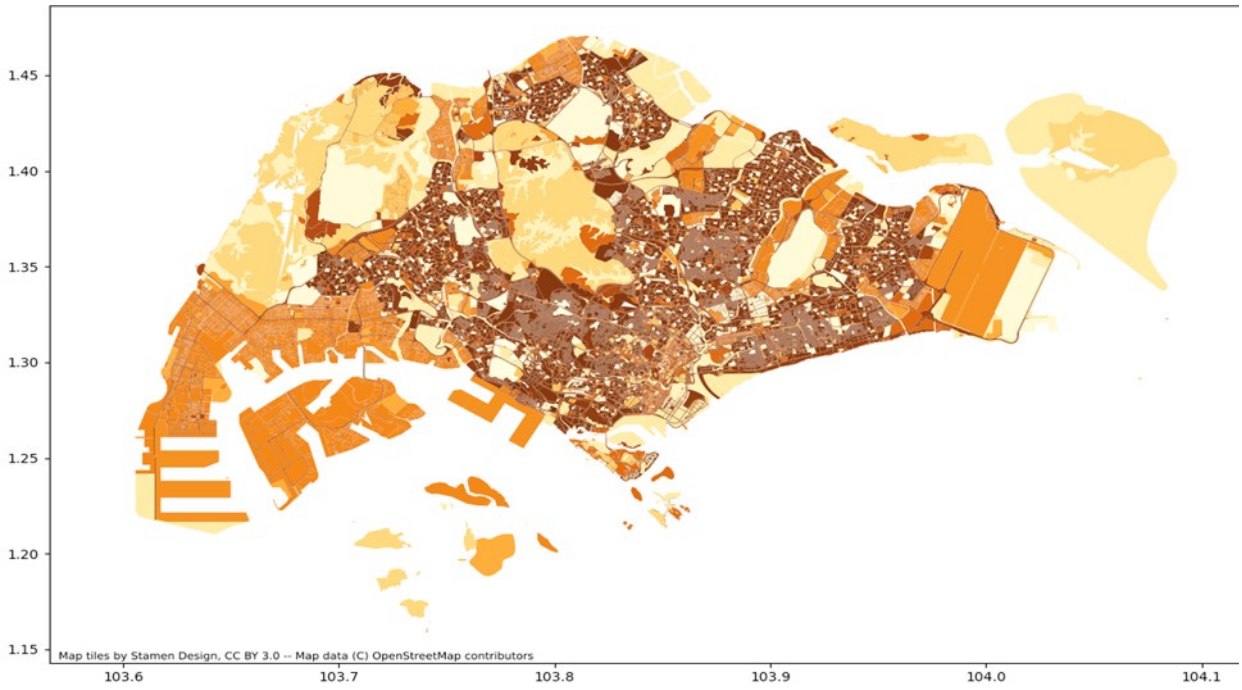


Figure 7.4: Result of applying the community detection algorithm on a subgraph consisting of Singapore plots data that are generally described as 2D generic city objects using the OntoCityGML ontology and its functionally described as plots with zones using the OntoZoning ontology. Plots of the same colour belong to the same cluster. The colour gradient represents sequential numbers, with clusters having the lowest sequential number in light yellow and clusters having the highest sequential number in dark brown.

rithms analyse the graphs to understand the roles of nodes within a graph and their importance within the overall structure of the relationships. Community detection algorithms identify groups of nodes within a graph.

Mr Chadzynski and Ms Tai have developed a Graph Inference agent that incorporates examples of algorithms that belong to each of the above-mentioned graph inference classes. Mr Chadzynski has also developed an ontology called 'OntoInfer' to describe the basic information of the agent, the inference algorithms and the tasks that implement them within the knowledge graph. This allows the agent to query the knowledge graph based on the received input request and information about the selected algorithm to identify which task to instantiate. In addition, he has developed an owlconverter utility that allows uploading ontologies from files to triple stores, such as Blazegraph. The Graph Inference agent was used to detect the shortest epistemic distance between pairs of plots described using the OntoZoning and OntoCityGML ontologies in the knowledge graph. Ms Tai has also supported this work by integrating the AccessAgent functionality into the Graph Inference Agent and developing the relevant unit tests.

Ms Tai, together with Mr Chadzynski, Ms Agarwal, **Ms Shiying LI (Software Engineer, SEC)** and **Ms Srishti GANGULY (Project Engineer, CARES)**, has also prepared the latest release for the CKG project. This included upgrading the City Import Agent, City Export Agent, City Information Agent, Distance Agent, and Thematic Surface Discovery Agent to work with newer versions of the JPS Base Library, unit testing, bug fixes, and deployment.

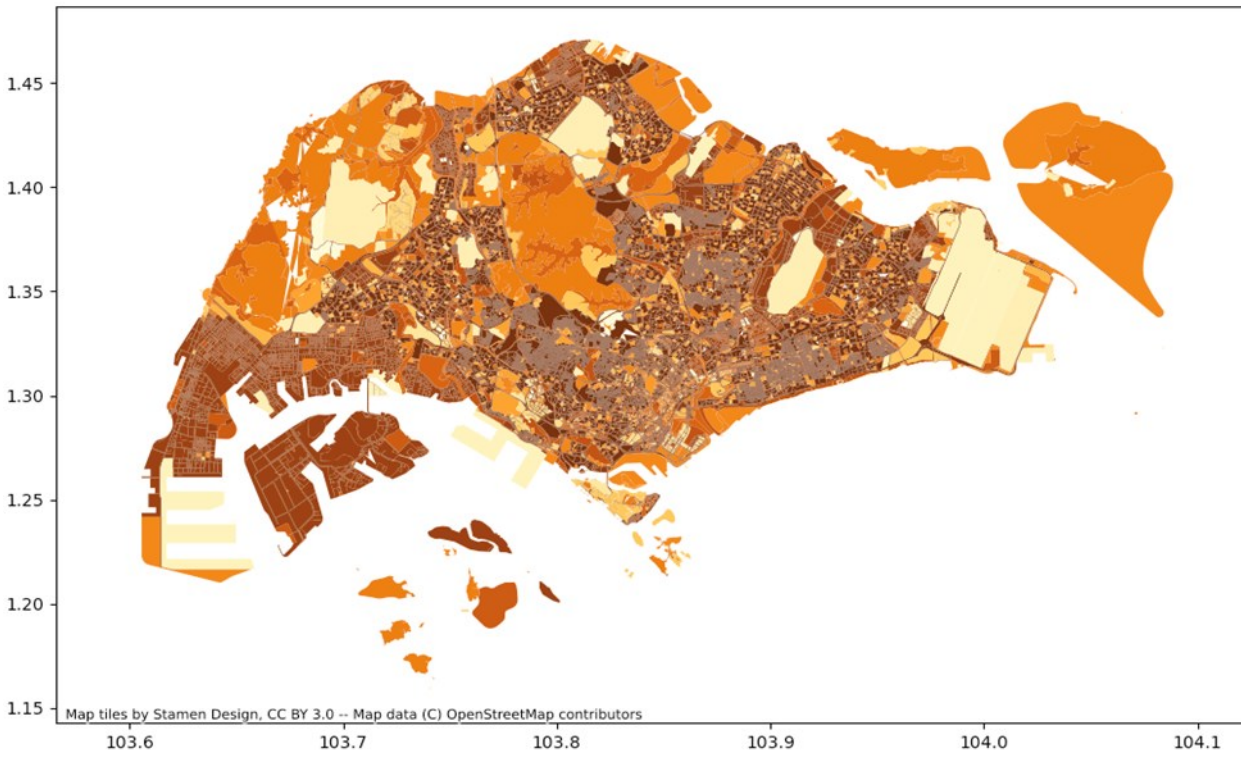


Figure 7.5: Result of applying the centrality detection algorithm on a subgraph consisting of Singapore plots data that are generally described as 2D generic city objects using the OntoCityGML ontology and its functionally described as plots with zones using the OntoZoning ontology. The colour gradient represents the PageRank score of the zones, with plots having the lowest PageRank score of the zones in light yellow and plots having the highest PageRank score of the zones in dark brown.

Update on work package 3

Developing agents to operate software and integrate data

Dr Emily LLOYD (Research Fellow, CARES) has performed a sensitivity analysis of the input variables for the City Energy Analyst (CEA) to evaluate which of the inputs impact the output data most. In this analysis, each variable is varied by -10%, -5%, 5% and 10% while keeping other variables at default values. The corresponding percentage changes in output electricity demand, heating demand, and photovoltaics (PV) supply were recorded. Figure 7.6 displays the results of the analysis, which highlight that the key variables are the setpoint/setback temperatures of the heating system (Ths_set_C/ Ths_setb_C), the heated/cooled floor area (Hs_{ag}), the thermal transmittance of windows (U_{win}), the fraction of gross floor area with electrical demands (E_s), the peak specific electrical load due to appliances/lighting (Ea_{Wm2}/ El_{Wm2}), and the efficiency of installed PV panels (PV_n). Dr Lloyd and **Mr Markus HOFMEISTER (PhD Student, CAM and co-sponsored by CMCL)** have translated this work

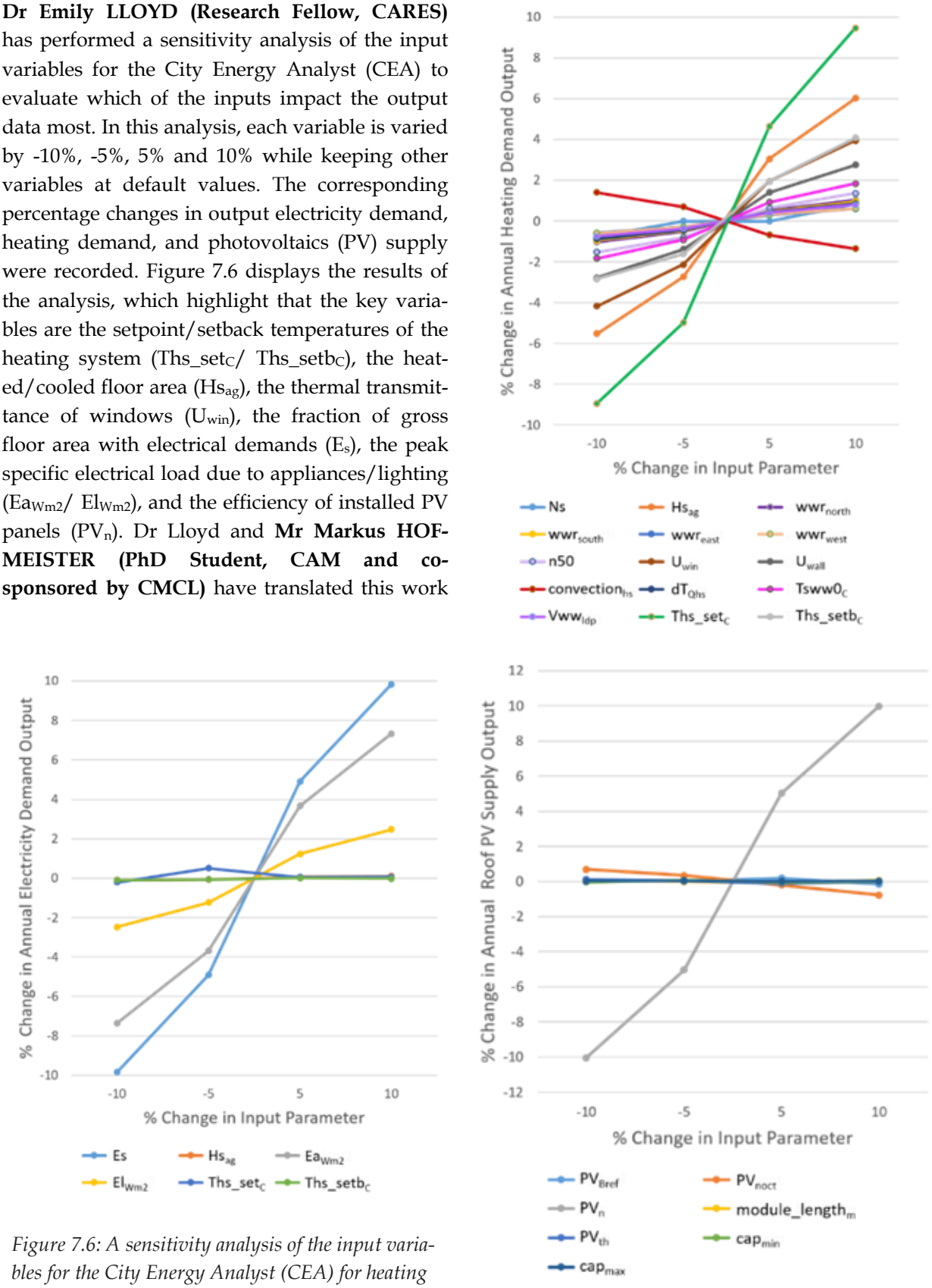


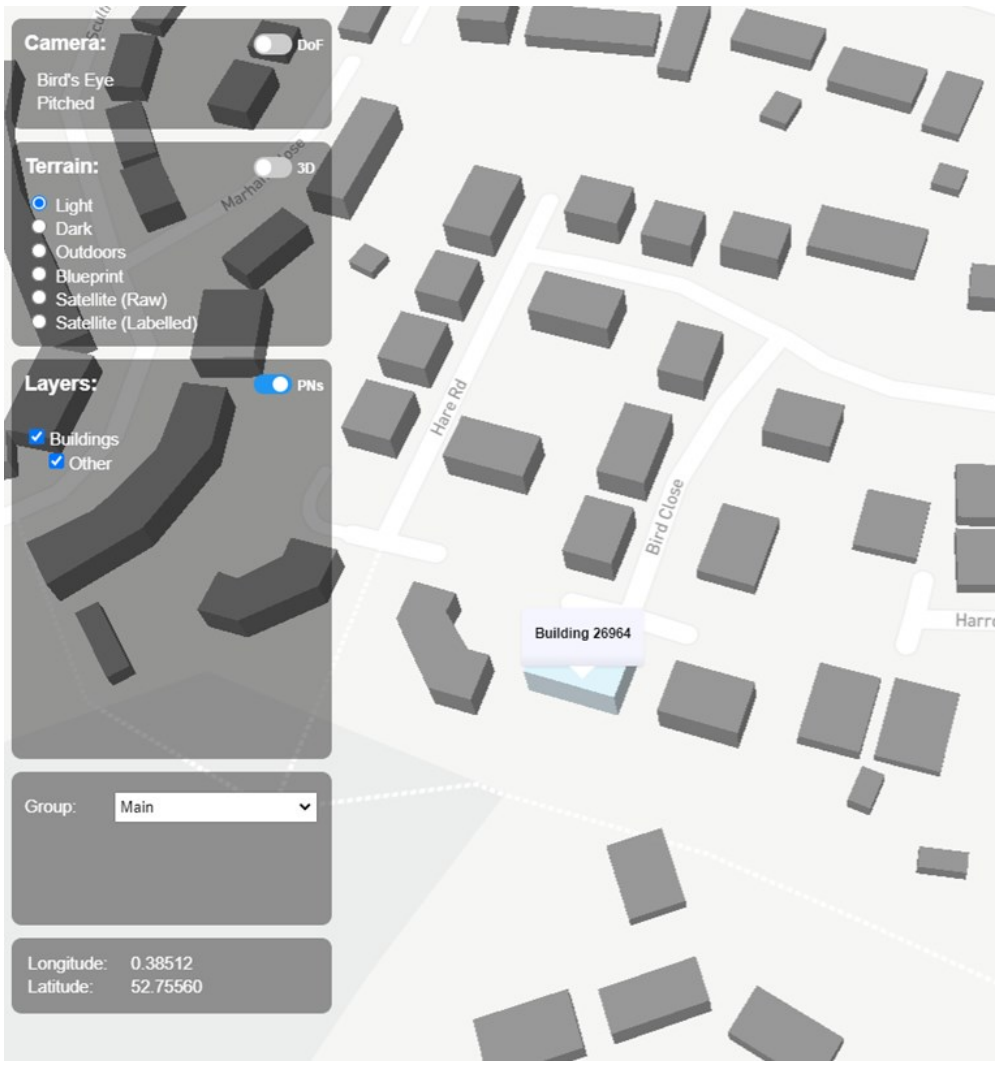
Figure 7.6: A sensitivity analysis of the input variables for the City Energy Analyst (CEA) for heating demand (top right), electricity demand (left), and photovoltaics (PV) supply (bottom right).

into a conference paper for the International Conference on Applied Energy 2022, held August 2022 in Bochum, Germany.

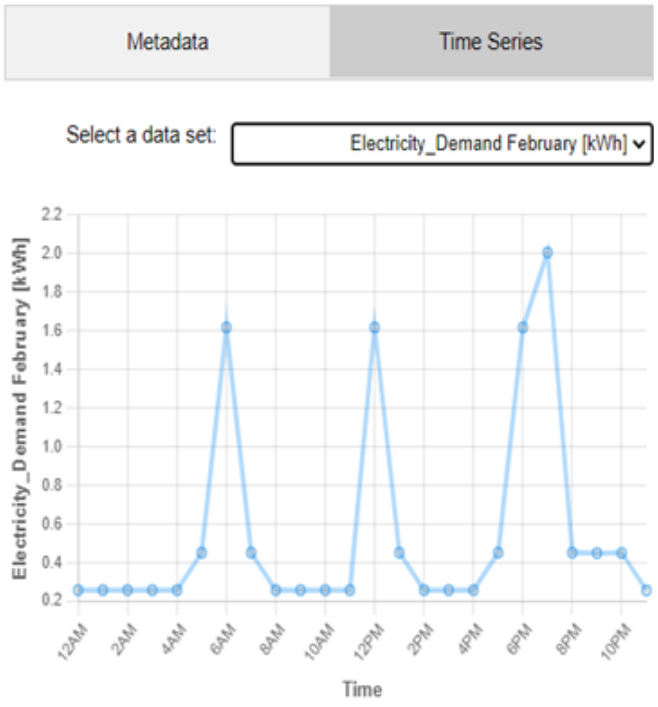
Dr Lloyd, in collaboration with **Mr Arkadiusz CHADZYNSKI (Senior Research Fellow, CARES)** and **Ms Ayda GRIŠIŪTĖ (Researcher, SEC)**, has also developed visualisations for the output data of the CEA agent using the City Information Agent (CIA) and web-based front-end 3DCityDB-Web-Map-Client. The CIA retrieves the energy data (via the CEA agent) and other attributes of the selected building from the knowledge graph upon receiving a POST request with the respective city object ID. Any time series energy data is summed to return an annual value. The data returned is displayed in a pop-up window, as shown in Figure 7.7.

Dr Lloyd and Mr Hofmeister have also used the Digital Twin Visualisation Framework (co-developed by CMCL) to visualise the output data of the CEA agent for buildings in Kings Lynn, United Kingdom, where building data are readily and publicly available. The time series data for electricity demands, heating demands, and potential PV electricity supply if PV panels were installed can be displayed by selecting the desired dataset from the dropdown menu, as shown in Figure 7.8.

Figure 7.7: Visualisation for the output data of the City Energy Analyst (CEA) agent using the City Information Agent (CIA) and web-based front-end 3DCityDB-Web-Map-Client.



Building 26964



Time	Electricity_Demand February [kWh]
00:00:00	0.258
01:00:00	0.258
02:00:00	0.258
03:00:00	0.258
04:00:00	0.258
05:00:00	0.452
06:00:00	1.618
07:00:00	0.452
08:00:00	0.258

Figure 7.8: Digital Twin Visualisation Framework used to visualise the output data of the City Energy Analyst (CEA) agent for a building in Kings Lynn, United Kingdom. Dataset for electricity demand for an average day in February is shown.

Time	PV_Supply_roof_and_walls February [kWh]
00:00:00	0
01:00:00	0
02:00:00	0
03:00:00	0
04:00:00	0
05:00:00	0
06:00:00	0
07:00:00	0
08:00:00	0.903571428571427

Time	Electricity_Demand with PV February [kWh]
00:00:00	0.258
01:00:00	0.258
02:00:00	0.258
03:00:00	0.258
04:00:00	0.258
05:00:00	0.452
06:00:00	1.618
07:00:00	0.452
08:00:00	-0.645571428571427

Figure 7.8 (continued): Digital Twin Visualisation Framework used to visualise the output data of the City Energy Analyst (CEA) agent for a building in Kings Lynn, United Kingdom. Two datasets for an average day in February are shown: potential PV power generation (left), and electricity demand if PV panels were installed (right). The shaded region indicates a period of electricity surplus.

Ms Shiying LI (Software Engineer, SEC) has implemented several new features for the 3D City Database Importer/Exporter tool. The new features include the ability of the City Export Agent to export building data from the knowledge graph in different Level of Detail (LOD) – in footprint and geometry modes and the integration of the tiling approach for the visualisation of large city models into the City Export Agent such that it can be executed automatically after the export is completed. All newly implemented features are tested for compatibility with the graphical user interface managed by Gradle (a dependency management system) and the agent system managed by Maven.

Ms Grišiūtė has extended the CIA to perform plot filtering operations based on user input for desired land uses and buildable space quantities. More specifically, the CIA generates SPARQL queries using user inputs from the 3DCityDB-Web-Map-Client and retrieves from the knowledge graph plots that meet the specified land use type and further filters out plots that do not meet the specified allowable GFA value.

Mr Yi-Kai TSAI (Software Developer, CARES) has enhanced the CEA agent to work with buildings having multiple ground geometries, as CEA only accepts a single building footprint geometry as input. With advice from **Dr Jingya YAN (Research Fellow, CARES)**, Mr Tsai has modified the CEA agent to initially query for all ground geometries of the building and then merge all the ground geometries into a single geometry. The boundary of the resultant merged geometry becomes the building's new footprint, which is passed to the CEA agent as an input.

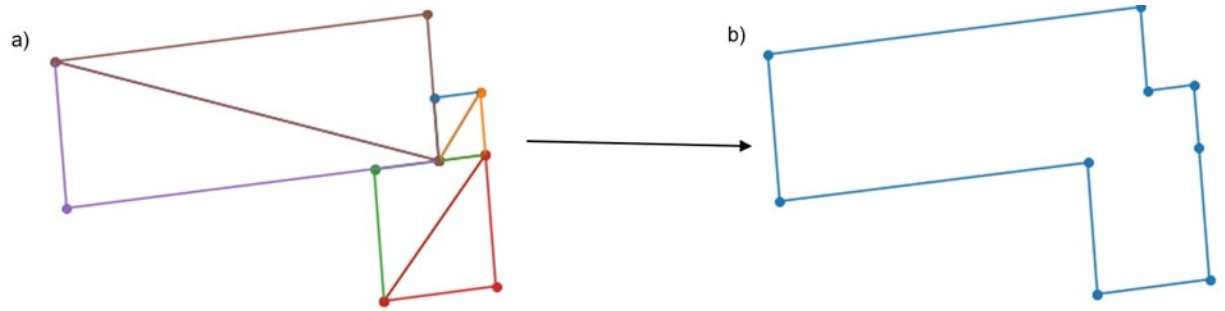


Figure 7.9: Example of how the CEA agent generates a single building footprint geometry from multiple ground geometries: a) multiple ground geometries of a building and b) resultant merged geometry, i.e. building's new footprint.

Ms Mehal AGARWAL (Software Developer, CARES), in collaboration with Mr Chadzynski and Mr Jefferson CHUA (Non-C4T student, CAM), has evaluated, validated, and documented the performance of the Thematic Surface Discovery Agent (TSDA) using the Semantic 3D City Database of Berlin comprising of 12 districts (consisting of 539,274 buildings, 9,558,218 surface geometries and 2,936,408 thematic surfaces). The process resulted in the report of the accuracy with which the TSDA identifies the bottom-level thematic geometry types (roof, wall, and ground) linked to the corresponding thematic surfaces. As part of this process, the bottom-level thematic geometry types identified by the TSDA was compared with the ground truth associated with those surface geometries. Ms Agarwal also tuned the parameters used by the TSDA to improve its accuracy in classifying roof surfaces and refactored the code to improve the agent's efficiency. The TSDA is reported to have a minimum of 99.8% accuracy and a maximum of 0.07% inaccuracy in identifying the bottom-level thematic geometry types for the Semantic 3D City Database of Berlin.

The TSDA was also applied to Level of Detail 1 (LOD1) building models of Pirmasens, Germany, to transform them into LOD2 building models. LOD1 models are represented as solids and do

not contain any thematic surface information. The classification executed by TSDA allows retrieval of information applicable to the individual thematic surfaces instead of the whole solid. Berlin and Pirmasens have been selected as data are readily and publicly available for these regions. This work can easily be extended and applied to other regions where such building data are publicly available, particularly Singapore. Extending and applying the TSDA to Singapore data is in progress.

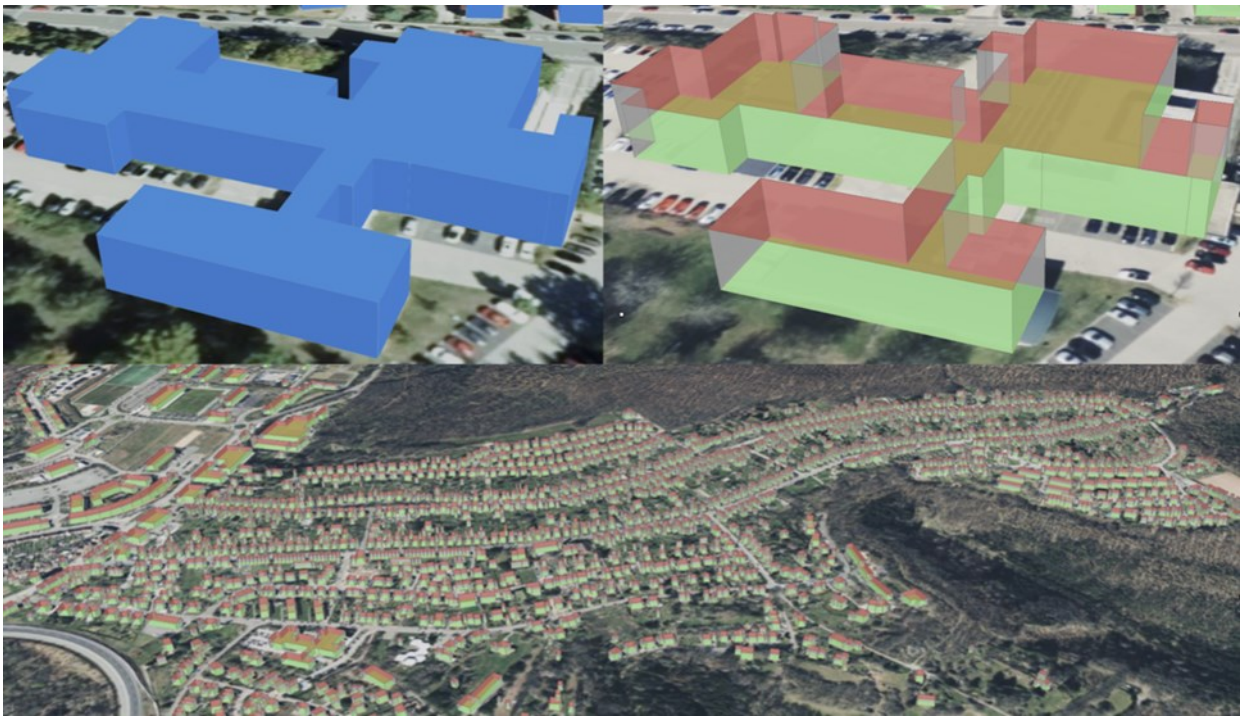


Figure 7.10: Upgrade of building models from Level of Detail 1 (LOD1) to LOD2 by the Thematic Surface Discovery Agent (TSDA) for Pirmasens, Germany. Top left displays an example of a LOD1 model of a building (before applying the TSDA), and top right displays a LOD2 model of the same building (after applying the TSDA). TSDA discovers walls (grey), ground (green), and roof (red) surfaces from the set of surface geometries in a LOD1 model and assigns them to the appropriate thematic surface categories.

The team, led by Mr Chadzynski, has prepared a preprint entitled 'Semantic 3D City Interfaces – intelligent interactions on Dynamic Geospatial Knowledge Graphs' to describe the work done. The preprint presents a system architecture and a set of interfaces that can be used to build scalable information systems capable of large city modelling based on dynamic knowledge graphs to avoid pitfalls of Web 2.0 applications while incorporating artificial and human intelligence during the knowledge enhancement processes. The preprint first describes a geospatial processor, an SQL2SPARQL transformer, and a geospatial tile ordering functionality that have been integrated into the City Export Agent to visualise and interact with city models on an augmented web-based front-end 3DCityDB-Web-Map-Client. The preprint then presents the TSDA that upgrades the building model's Level of Detail and the CIA that assists with retrieving contextual information and provides data concerning city regulations. The CEA agent that allows automation of building energy analyses was also described in the preprint. Lastly, the preprint describes the Distance Agent that autonomously calculates the physical distance between two data instances in the knowledge graph by tracking external interactions with the representation via the 3DCityDB-Web-Map-Client.

Figure 7.11: Graphical abstract of the preprint entitled "Semantic 3D City Interfaces – intelligent interactions on Dynamic Geospatial Knowledge Graphs". The preprint describes the work done in the project in the context of GeoWeb 2.5 systems and how the dynamic knowledge graph and the agents can lower the risks of deliberate misinformation.

Update on work package 4

Developing interfaces and planning libraries for the CKG

Ms Shiying LI (Software Engineer, SEC) has extended the 3D City Database Importer/ Exporter tool to import the Singapore plot dataset as genericCityObject models into the knowledge graph and to export the data as Level of Detail 1 (LOD1) footprint data in Keyhole Markup Language (KML) file for visualisation using the 3DCityDB-Web-Map-Client. The City Export Agent automatically applies the tiling process to the exported KML files. The 3DCityDB-Web-Map -Client has been extended to display the plot data, as shown in Figure 7.12.

Figure 7.12: Visualisation of Singapore plot data with the 3DCityDB-Web-Map-Client.

Ms Srishti GANGULY (Project Engineer, CARES) has extended the 3D City Database Importer/Exporter tool to handle CityGML's CityFurniture models. This extension allows more variety of geometric objects to be represented in the knowledge graph.

Dr Jingya YAN (Research Fellow, CARES) is cleaning Singapore LOD3 building data before instantiating them in the knowledge graph. This data processing task includes identifying geospatial relationships between building components and splitting and/or merging building components based on their geospatial relationships. Dr Yan has already processed approximately 60,000 Singapore LOD3 building models.

In areas where the vertical geo-referencing of the data is not relatively flat, it is necessary to add a terrain model for visualisation. For example, Maptiler terrain data, an open terrain data model at a global scale, is loaded for the visualisation of LOD2 building models in Pirmasens, Germany, as shown in Figure 7.14. **Ms Huay Yi TAI (Software Developer, CARES)** has supported Dr Yan in preparing the tiled visualisation.

Figure 7.13: Visualisation of LOD3 building models in the National University of Singapore UTown, Singapore.

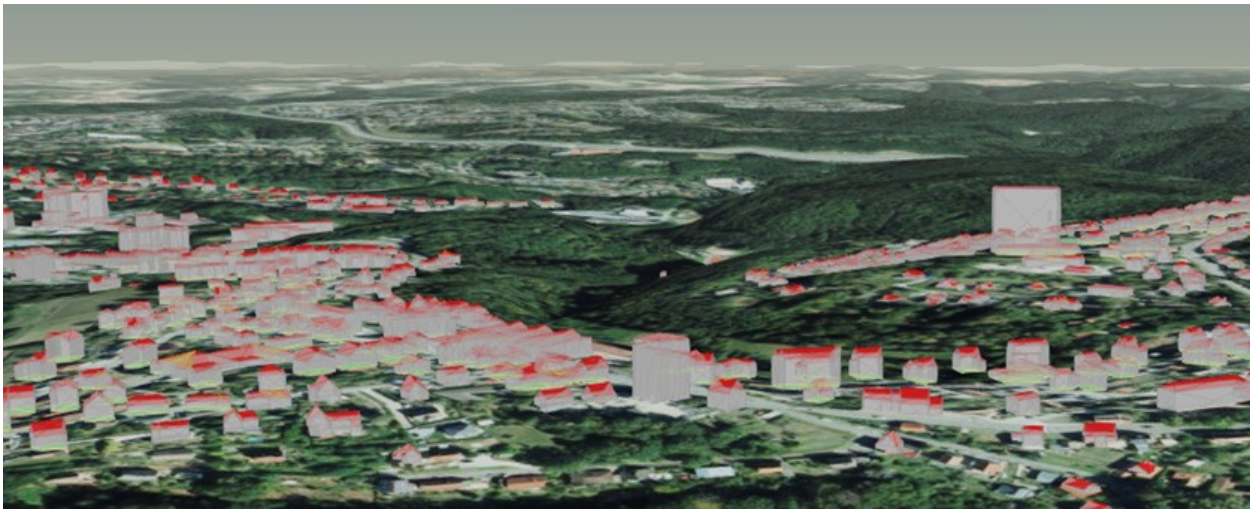


Figure 7.14: Visualisation of LOD2 building models with terrain model in Pirmasens, Germany.

Update on work package 5

Developing Design Informatics Functions

Dr Pieter HERTHOGS (Senior Researcher, SEC) is developing an ontological framework for design goals and their evaluation, structuring nine goal types into three interrelated hierarchical levels. It is a mid-level, domain agnostic ontology defined in relation to top-level ontology Basic Formal Ontology (BFO).

Ms Ayda GRIŠIŪTĖ (Researcher, SEC) has developed a Strengths, Weaknesses, Opportunities, and Threats (SWOT) analysis framework for automated multi-domain evaluations of Sustainable Development Goals (SDG) related planning targets for four key growth areas in Singapore. The framework allows the automated derivation of planning metrics from the knowledge graph. It integrates the metrics into a SWOT framework for the multi-domain evaluation of on-site solar energy potential across plots in the key growth areas. This work aims to form the basis for a more comprehensive planning support tool that is based on the knowledge graph and a versatile SWOT framework without making it location-specific. Ms Grišiūtė has prepared and submitted a conference paper describing this SWOT analysis framework and has presented it at the annual International Conference of the Association for Computer-Aided Architectural Design Research in Asia 2022 (CAADRIA2022), which was held 9-15 April in Sydney, Australia.

The work mentioned above will become the underlying technology for the second demonstrator: a suitable site selector with multi-domain and multi-criteria evaluation that can rank plots in Singapore based on a SWOT analysis of set goals.

Update on work package 6

Demonstrators: horizontal and vertical use cases

This work package aims to develop three demonstrators to show the project's potential applications and extensibility. Each demonstrator is conceived as an incremental extension of the previous work and focuses on one city planning meta-practice (i.e., planning action or use case). The city planning meta-practices include: Representation (Demonstrator 1: programmatic plot finder), Evaluation (Demonstrator 2: suitable site selector), and Projection (Demonstrator 3: future scenario builder). Together, these demonstrators examine different use cases related to land use planning city-wide, per planning area or plot.

In the current state, the CKG team led by **Dr Pieter HERTHOGS (Senior Researcher, SEC)** and with main inputs by **Ms Shiying LI (Software Engineer, SEC)**, **Ms Ayda GRIŠIŪTĖ (Researcher, SEC),** and **Ms Heidi SILVENNOINEN (Researcher, SEC)**, has developed the Programmatic Plot Finder demonstrator to enable querying for plots that allow particular combinations of programmes or uses, a particular amount of buildable space, or a combination of both, i.e., particular amounts of buildable space for particular combinations of programmes or uses. For instance, a user may search for plots that allow at least 200 square metres of programme type 'Flat' and at least 300 square metres of programme type 'Nursing Home'. Previously, such searches required users to manually review regulatory documentation for each plot considered for potential development.

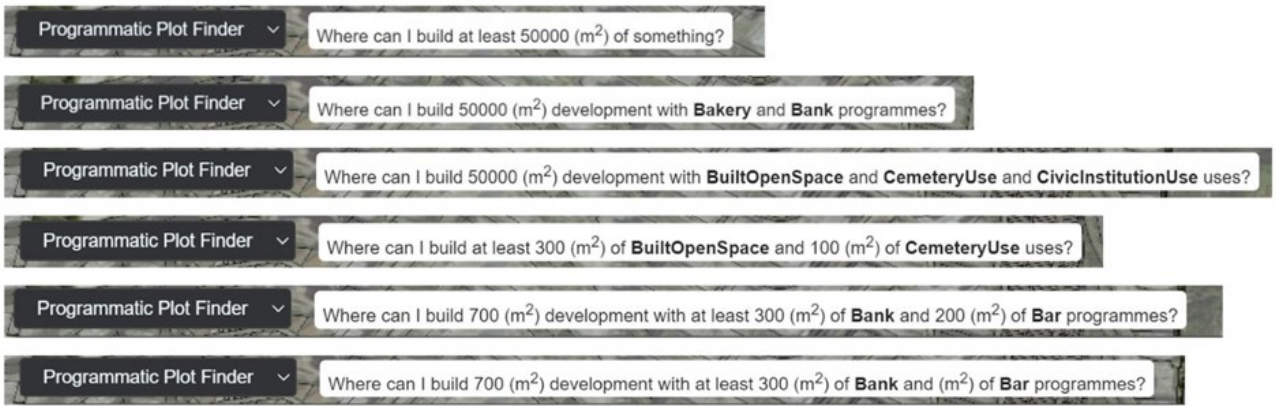
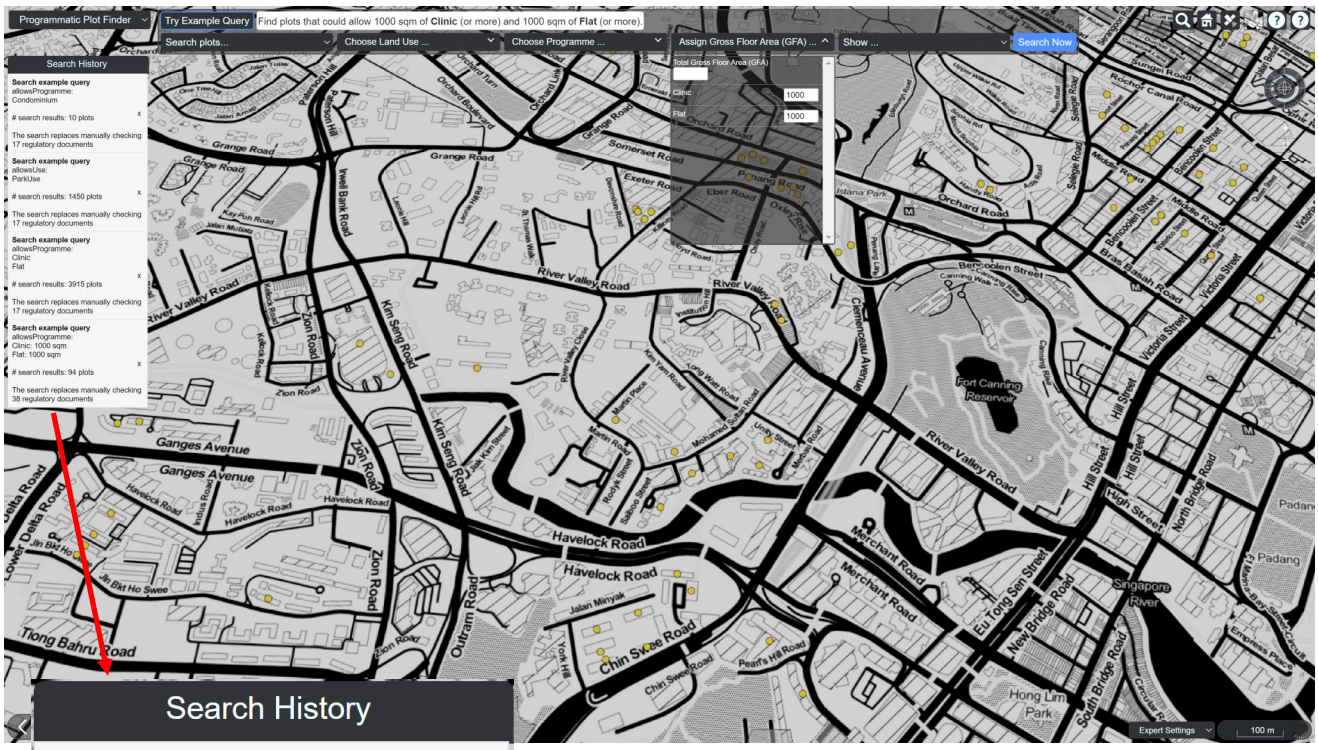


Figure 7.15: Examples of potential human-readable queries that a user can formulate.

The information regarding the plots is retrieved from the knowledge graph using relevant queries constructed by the City Information Agent (CIA). The results are processed using the CIA's filtering functionality described in Work Package 3. The resulting plots that meet all land use and buildable space requirements are visualised on the 3DCityDB-Web-Map-Client.

From the dropdown menus in the 3DCityDB-Web-Map-Client, the user can select combinations of programmes or uses that should be allowed on a plot and provide their corresponding required amounts. These inputs are also translated into human-readable sentences, such as 'Where can I build at least 200 m² of Clinic?'. The resulting plots are highlighted in two levels of detail: using map pins when zoomed out and using coloured plot polygons when zoomed in. The results of every query input are also summarised in an information box that displays an overview of the initial input and valid results.



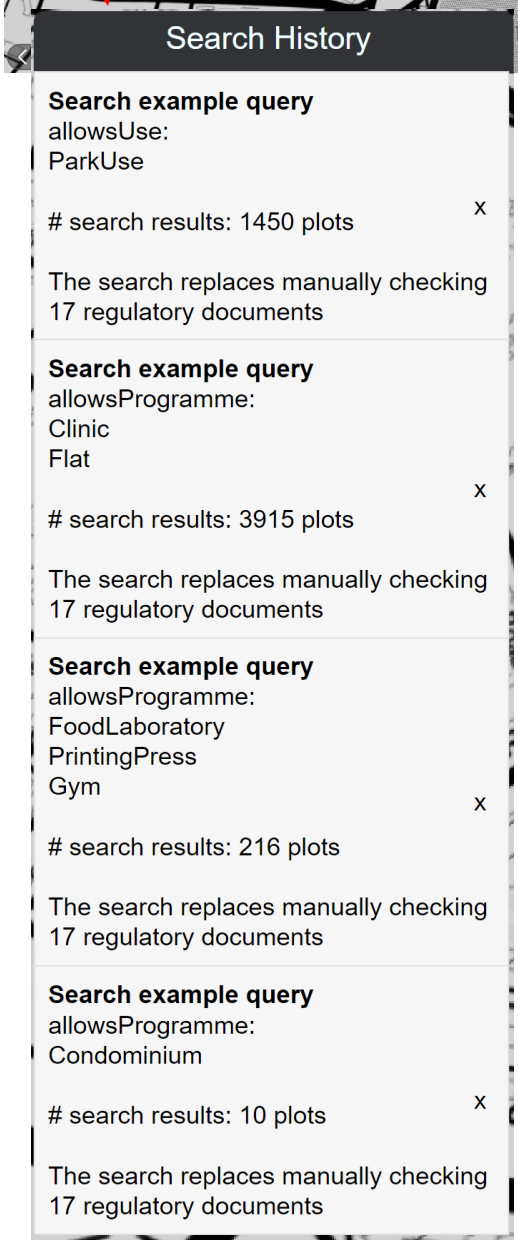


Figure 7.16: Visualisation of the current Programmatic Plot Finder demonstrator user interface that allows searching for plots with a particular combination. In this example, the user searches for plots that allow a combination of at least 1,000 square metres of programme type 'Clinic' and at least 1,000 square metres of programme type 'Flat'. The query results, i.e., plots, are displayed with yellow pins. The search history has been enlarged to the image on the left.

Other activities and achievements

As part of the stakeholder engagement strategy, we have continued outreach activities towards academia, industries of urban project consultants and developers, and government agencies. Further exchanges took place with the Smart City Projects Office (Smart Nation and Digital Government Office) and Urban Redevelopment Authority's (URA) Design & Planning Lab in May 2022.

Dr Pieter HERTHOGS (Co-PI, SEC) is part of a Future Cities Laboratory Global (FCLG) team developing a professional urban design and planning course for Singapore's government agencies at the request of URA's Design & Planning Lab (July 2022, ongoing). The course is structured along the four meta-practices of City Planning defined in CKG's literature review on Semantic City Planning Systems (von Richthofen et al., 2022). Dr Herthogs also presented a seminar on the CKG project and the importance of Knowledge Representation at SEC in August 2022.

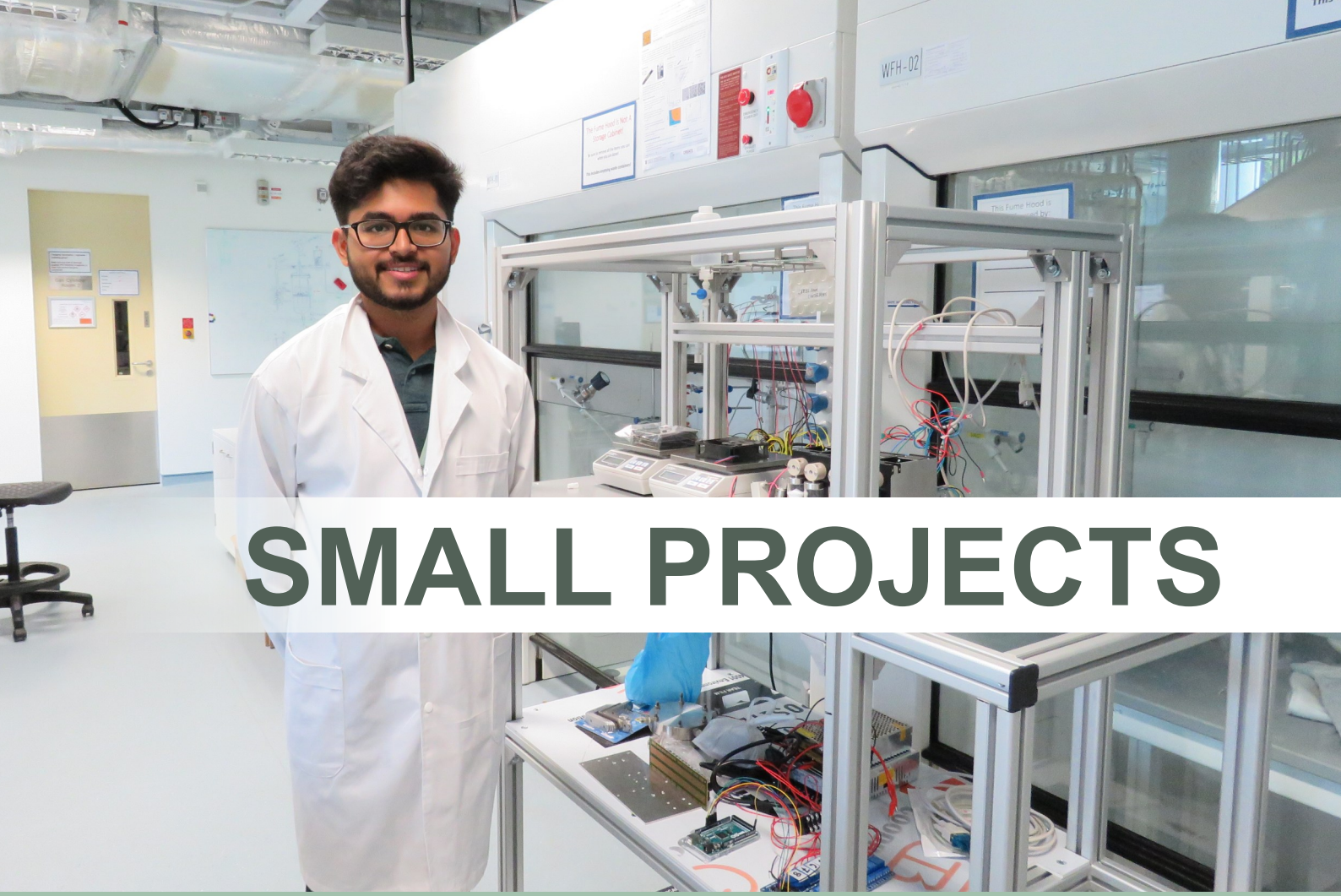
Dr Aurel VON RICHTOFEN (Co-PI, Arup Germany) presented the CKG project at the RISE Cities Responsible Leaders Round Table by BMW Foundation in Barcelona in May 2022. Dr von Richthofen also gave a keynote lecture on 'Digitalisierung Für/Und Nachhaltige Städte' at the 30 Years Arup Germany celebration in June 2022. In addition, Dr von Richthofen gave a talk on 'City Planning with Knowledge Graphs' at the Urban Twin Conference, HafenCity Universität Hamburg in June 2022 and was a panellist at the International Public Policy Observatory (IPPO) Cities event #7 (July 2022) on the topic 'Urban Intelligence: How are Cities Using Evidence and Data to Improve Decision-Making?'. Subsequently, Dr von Richthofen wrote an input piece for the event that can be found here: https://covidandsociety.com/intelligent-data-ecosystem-urban-planning/

Dr Herthogs and Dr von Richthofen met with ESRI directors in September 2022 to exchange information about the company's recent ArcGIS Knowledge product and CKG.

A book chapter about the CKG project by **Prof Dr Stephen CAIRNS (Co-PI, ETH)**, Dr Herthogs, **Prof Dr Markus KRAFT (PI, CARES)**, and Dr von Richthofen was published in Future Cities Laboratory - Indicia 03 (Cairns et al., 2022).

Prof Cairns officially stepped down as the Future Cities Laboratory co-director and the CKG Principal Investigator in June 2022. Prof Cairns remains involved in the CKG project as a Co-Investigator. In July 2022, the CKG team officially welcomed **Prof Dr Martin RAUBAL (PI, SEC)** as the new SEC Principal Investigator of the project. Prof Raubal is a Professor of Geoinformation Engineering at the Institute of Cartography and Geoinformation, ETH Zurich. He is also the Principal Investigator of the Digital Underground project and Cluster Leader of the Future Resilient Systems cluster's Distributed Cognition for Social Resilience at SEC.

The CKG project's industry collaboration with Takenaka Corporation, under the form of a research visit by Mr Genki UNNO (Visiting Researcher, Takenaka Corporation), will continue for its second year until August 2023. During the second year, we will explore a practical collaboration on a Takenaka case study, applying the plot search and site selection features developed as part of our demonstrators. Mr Unno, advised by Dr Herthogs, is developing a thematic evaluation framework for Smart City projects and visions, specifically introducing criteria for context adaptation. Mr Unno presented the evaluation framework for the first theme – smart context-adaptive mobility – at the 2022 International Conference on Transport and Smart Cities in Changsha, China, in September 2022.



SMALL PROJECTS

OTHER CARES-FUNDED PROJECTS

In addition to C4T and CLIC, CARES hosts a number of other projects. These give our researchers an opportunity to explore new areas, develop technologies for commercialisation or build relationships with new industry partners or public sector collaborators. The smaller projects are also often a good opportunity for interns (such as Mr Aman SINGHAL, pictured above during his time working on the RINGs project in 2019) to have a novel experience of research and technology development not easily available during their undergraduate degrees.

The current CARES small projects include An Accelerated Manufacturing Platform for Engineered Nanomaterials (AMPLE) supported by the Central Gap Fund, Knowledge Graph Driven P2P Energy Trading and Real-time Network Operation for High Renewables (an Intra-CREATE seed collaboration grant), and Pharmaceutical Innovation Programme Singapore (PIPS) as funded by a private-public partnership. CARES is now collaborating with the Singapore-ETH Centre on Cooling Singapore 2.0 and an update on this work is included.

This section also includes updates on the ten projects under the C4T Emerging Opportunities Fund, which was created to support exciting new ideas that have arisen since the start of C4T Phase 2.

An Accelerated Manufacturing Platform for Engineered Nanomaterials (AMPLE)

Central Gap Fund

The AMPLE project began in June 2022 and is a direct result of C4T's IRP1 research in WP1 in Phase 1 and Phase 2. The SGD$6.5M funded project seeks to translate annular flow microreactor technology to 100 kg/day scale production, utilising machine learning and an agile product development methodology. By pushing the technology to a TRL7/8 level, the team will overcome the scale-up "gap" for industrialisation of new material technologies.

The key deliverables of Phase 1 (the first year) of AMPLE are 1) commissioning of the pilot plant in C4T's lab and 2) securing of industry partners for trials in Phase 2. Recent work has focused on procurement, hiring, technical and business development. The project implements an agile methodology for project management, and is utilising a digital, modular plant concept to overcome the challenges of current supply chain and logistics.

Ms Satya KENCHA (Quality Control Scientist, AMPLE) is developing testing protocols for evaluation of nanoparticle samples. Ms Kencha has completed coursework in quality control to upgrade her skills in quality control methods, and has used tools like "Cause & Effect diagrams" and "Flow charts" to fix the operational challenges in operation of the flow reactor, inconsistencies in analysis and conduct consistent synthesis of ZnO nanoparticles at different conditions. In future, she plans to develop quality control tools and methods for large scale synthesis of ZnO nanoparticles and antibacterial testing of ZnO coated films.

Figure 8.1: 7 Quality Management Tools used by Ms Kencha

Dr Mikhail KOVALEV (Technical Development Manager, AMPLE) works with industry partners to create new products using materials generated in the project. He has produced agreed -upon product specifications with two industry partners for trials, and has begun work on developing the products, including antimicrobial ZnO coatings for outdoor applications. He has also been responsible for procurement of major equipment related to the characterisation of new products developed in the project, including a scanning electron microscope.

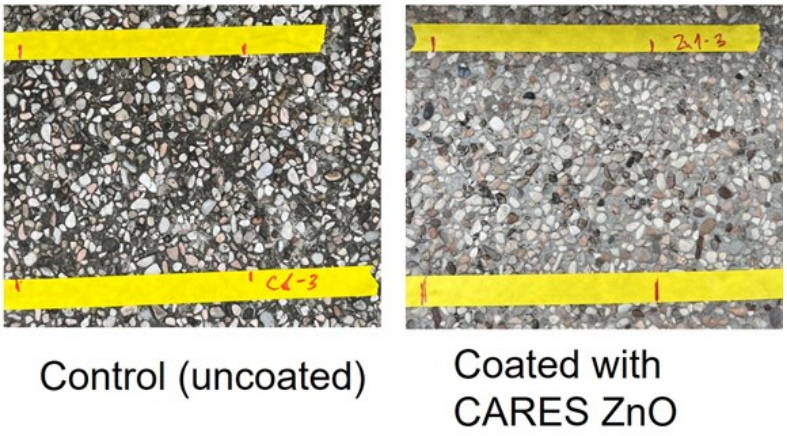


Figure 8.2: Prolonged antimicrobial effect for outdoor coating with ZnO (6 months trial)

Ms Faye NG (Automation Engineer, AMPLE) is developing the automated control of the 100 kg/ day pilot process. She has completed induction in the areas of materials science, chemistry, reactor operation, biological testing and marketing. She has designed the modular architecture for components of the process and done procurement for the automation equipment and software. She has also helped to reorganise the particle technology room, where the process will be located.

Figure 8.3: Images of the laboratory following an initial clean-out

Dr Nicholas JOSE (Project Lead), **Dr Mikhail KOVALEV (Technical Development Manager, AMPLE)**, and **Prof Alexei LAPKIN (PI, CAM)** presented the topic "The AMPLE Story: Filling the sale-up gap for new material technologies" at the CREATE Webinar on 26 August 2022. There were 46 attendees from various academic institutions and industries.

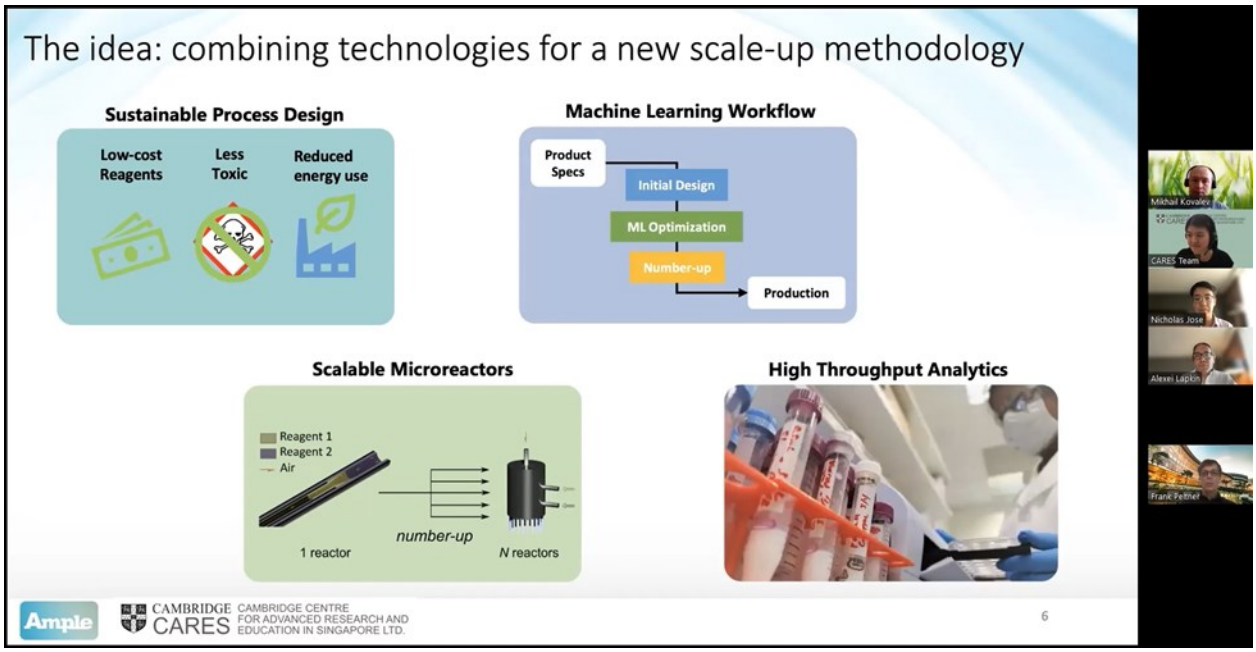


Figure 8.4: Screen capture of the webinar

Knowledge Graph Driven P2P Energy Trading and Real-time Network Operation for High Renewables

Intra-CREATE seed collaboration grant

Dr Casper LINDBERG (PI, CARES) was recently awarded the Intra-CREATE seed collaboration grant with **Asst Prof Hung Dinh NGUYEN (PI, NTU)** for project commencement in October 2022. The project aims to develop an energy market framework that leverages the benefits of a dynamic knowledge graph for efficient peer-to-peer (P2P) energy trading between prosumers, while satisfying the stable and secure operation of the distribution grid.

Data2Knowledge in the Digital Manufacture of Pharmaceuticals

With funding from Pharma Innovation Programme Singapore (PIPS), via A*STAR

Data2Knowledge in the Digital Manufacture of Pharmaceuticals is a project funded under the Pharma Innovation Programme Singapore (PIPS) programme and led by Professors Alexei Lapkin and Markus Kraft. This project commenced in December 2020 and ended in September 2022. The project aimed to develop a complete digital framework for automated experiments within a digital laboratory.

In this reporting period, Mr Jiaru BAI (Non-C4T PhD Student, CAM) and **Dr Sebastian MOSBACH (Senior Research Fellow, CAM)** have been working on assembling the developed agents into one framework to close the loop in silico and also deploying the agents in the laboratory to run experiments on actual equipment. The experimentation aspect was assisted by **Dr Dogancan KARAN (Research Fellow, CARES)** and Dr Connor TAYLOR (Postdoctoral Researcher, CAM).

As this project uses technologies developed as part of The World Avatar (TWA) project, all the agents have been upgraded to leverage the latest best practices in creating agents. As a breakdown and re-implementation of the control codes from the PIPS C4 project, five agents were developed: DoE Agent, VapourtecSchedule Agent, Vapourtec Agent, HPLC Agent, and HPLCPostPro Agent.

Figure 8.5 depicts selected concepts abstracted for this project and the agents that operate on the knowledge graph using these concepts as their input/output signature(s). The iterative workflow begins with the DoE Agent recommending the following experiments to be conducted. The VapourtecSchedule Agent will then schedule the experiment, assigning the reaction to the hardware it deems most appropriate. Consequently, the Vapourtec Agent controlling the hardware configures itself in response to the request and

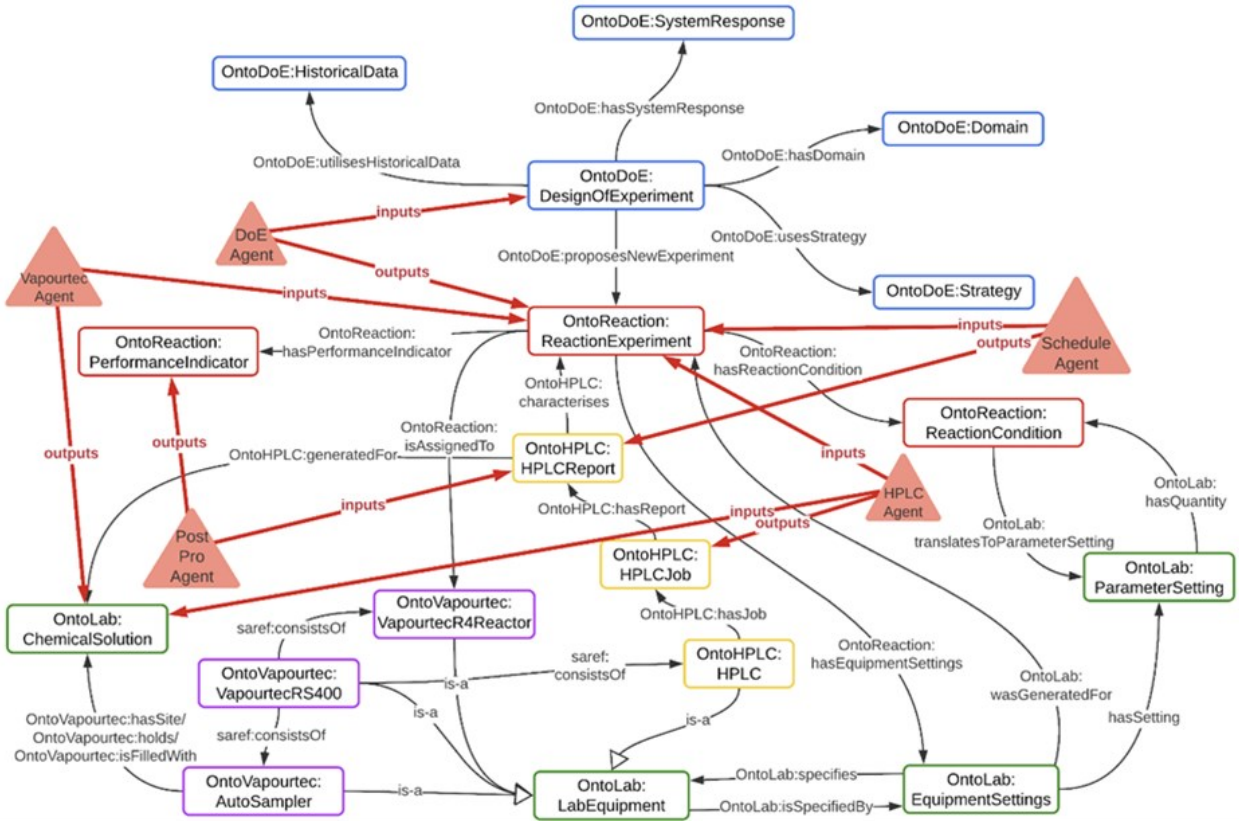


Figure 8.5: Semantically integrated system with agents operating on the knowledge graph

begins executing the experiment. The information is subsequently sent to an HPLC Agent, who monitors the creation of a new high-performance liquid chromatography (HPLC) analysis report. Finally, in the post-processing phase, a list of performance indicators is computed. The findings are collected to determine whether to commence the next cycle.

The system was constructed and deployed for test runs in both the Singapore and Cambridge laboratories. The chemistry published in the PIPS C4 project, in particular, was optimised in the Singapore laboratory. Using the same optimisation configuration, the system was, as a demonstration, left unsupervised for 15 continuous reactions. In this demonstration, no prior experiment data was supplied. After stating the desired chemical reaction, the system automatically begins using randomly generated experimental conditions. Given the many optimisation factors, the results thus far have primarily explored the chemical space. Nonetheless, the demonstration serves as a proof of concept for the dynamic knowledge graph-based approach to closed-loop optimisation.

Development of Multi-Step Processes in Pharma

With funding from Pharma Innovation Programme Singapore (PIPS), via A*STAR

Development of Multi-Step Processes in Pharma was funded under the Pharma Innovation Programme Singapore (PIPS) programme and led by Professor Alexei Lapkin. The stream successfully closed on 14 June 2022. The project has submitted its final report to A*STAR within this reporting period. The final report indicates that all milestones and deliverables that were planned were achieved. This project has developed a new software package which is being transferred to the companies co-funding the PIPS programme: GSK, Pfizer, Syngenta and MSD.

Digital Workflow and Continuous Processing in Pharmaceuticals Manufacturing

With funding from Pfizer as part of the Pharma Innovation Programme Singapore (PIPS)

Digital Workflow and Continuous Processing in Pharmaceuticals Manufacturing is funded under the Pharma Innovation Programme Singapore (PIPS) programme and led by Professor Alexei Lapkin. This is a two-year project which commenced in January 2021.

Transformation of manufacturing in the pharmaceutical industry to new emerging business models (on demand, customisation, sustainable manufacturing, etc.) is heavily dependent on the development of supporting technologies, such as a novel manufacturing paradigm of fully continuous processes and digital tools for support of R&D and manufacturing.

A number of current challenges in the supporting technologies are interlinked. Thus, development of effective flow processes and the use of continuous flow technology in manufacturing requires innovation in process modelling, reactor technology/reactor manufacturing, process data monitoring and knowledge management. This requirement spans the areas of synthesis, process engineering, process control, data science and artificial intelligence.

Dr Dogancan KARAN (Research Fellow, CARES) is currently working on an industrially relevant multistep batch-to-continuous process transformation for the synthesis of an active pharmaceutical ingredient (API). Earlier in the project, Dr Karan developed first principal models to optimise the process parameters for flow reactor development for the first step of the process. As a follow-up, the same reaction has been optimised by a fully automated machine learning -enabled robotic platform. The platform utilises a range of supervised and unsupervised learning methods to represent and optimise the chemical systems. An optimisation campaign of 60 experiments has been conducted for multiple competing objectives (process yield vs process cost) to identify the most desirable process parameters. 75% reaction yield with minimum process cost has been identified. Future work aims to expand the optimisation space to further increase the yield by using different process metrics. In the next 6 months, Dr Karan will be working on developing a new fully automated platform which will enable the optimisation of the second step via machine learning.

Dr Guoying CHEN (Research Fellow, CARES) reports that machine learning applications in process development and optimisation have been well adopted in the pharmaceutical industry. Using the development of synthetic processes, Dr Chen applied his machine learning algorithms to the first step of the reaction which he has studied using the TSEMO algorithm that optimises process parameters.

The process contains two-step reactions. Dr Chen has optimised the first step reaction with heterogeneous catalyst using algorithms. While different objectives have been achieved by his current system, his main aim was yield and cost. In his initial optimisation campaign, 60 experiments were automated when one type of catalyst was applied. Dr Chen expects that the optimal and practical processes could be achieved further by optimising catalyst, cost, yield, and space-time yield.

The second step reaction relies on new system development. Although excellent results have been achieved by manually performed flow reaction, a more robust flow system is needed for the process optimisation campaign using his algorithms. It has been challenging to find the right hardware, assemble, and automate the system due to the complexity in chemistry of this step. Dr Chen will be continuing his efforts in this area.

Furthermore, Dr Chen expects that he could combine the two-step reactions into one process by using the same solvents. His multi-step optimisation platform could be utilised in this scenario. Combining the information of step-wised study and multi-step optimisation, a practical flow process could be developed for industry and lower the cost of drug manufacturing.

Other activities and achievements

The PIPS Pfizer project is now in the planning stage for a three-month internship by CARES post-docs to Pfizer to assist in technology transfer of the developed methods to Pfizer.

Following the completion of PIPS D2K (C12) project, the team in Cambridge is engaging with a large number of companies to create a user-friendly environment for researchers to interact with agents and instruments available via a knowledge graph.

Cooling Singapore 2.0

In collaboration with the Singapore-ETH Centre

Cooling Singapore 2.0 aims to construct a Digital Urban Climate Twin for Singapore. This platform will bring together several computational models (environmental, land surface, industrial, traffic, building and energy) as well as climate models to investigate ways to reduce Singapore's urban heat and mitigate its effects. The Digital Urban Climate Twin will also allow researchers to trial various scenarios and predict the impact they may have on urban heating.

CARES' contribution to Cooling Singapore 2.0 is to evaluate anthropogenic heat emissions from Industry in Jurong Island, Singapore by developing computational energy models using The World Avatar (TWA) Knowledge Graph. Ultimately, these energy models will be fed into the Digital Urban Climate Twin. CARES will also develop scenarios to investigate the impact of the anthropogenic heat emissions from Industry in Jurong Island on Singapore.

Dr Hansong XUE (Research Fellow, CARES) and **Ms Srishti GANGULY (Project Engineer, CARES)** have developed an ontology called OntoChemPlant to represent the processes, business activity and economic performance of plants on Jurong Island. This ontology comprises the concepts required to estimate the heat emissions for the major industrial heat emitters on Jurong Island. Using the OntoChemPlant ontology, Ms Ganguly has consolidated and instantiated the data in TWA knowledge graph. She has also linked the instances with their corresponding CityGML models in the knowledge graph. The CityGML models provide geospatial information regarding the infrastructures, which is useful in assigning the estimated heat emissions to their

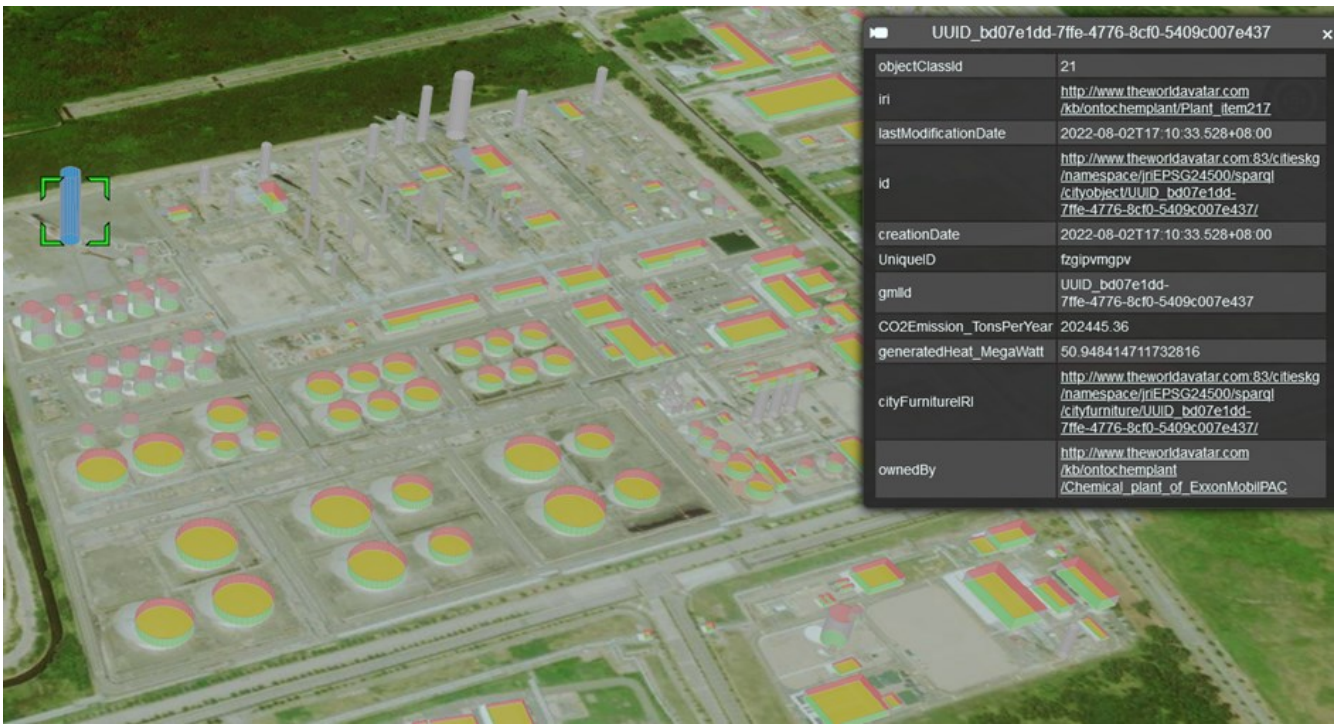


Figure 8.6: 3D visualisation of the infrastructures on Jurong Island, Singapore. Main infrastructures are represented in CityGML Level of Detail 2 (LOD2) (multi-coloured), while other infrastructures are represented in LOD1 (grey). The pop-up window displays a subset of the information about the selected object available in the knowledge graph.

corresponding locations and heights. Dr Xue has developed a Heat Emission agent that performs cross-domain queries on the knowledge graph to estimate the heat emissions for different heat emitting structures in a given geographic area. The heat emission values are estimated using the carbon dioxide emission values, the carbon emission index of the fuel used, and the plant's thermal efficiency. In this work, Dr Xue has assumed that the major industrial heat emitters on Jurong Island produce energy by combusting natural gas with a 50% thermal efficiency and that the emitted heat is distributed evenly among relevant heat emitting structures. This agent also updates the knowledge graph with the estimated heat emission values. Currently, 214 anthropogenic heat emission points have been instantiated with their centroid's information and estimated heat emission values in the knowledge graph.

In close collaboration with **Mr Arkadiusz CHADZYNSKI (Senior Research Fellow, CARES)**, Ms Ganguly has also developed an OntoChemPlant agent (OCPA). This agent retrieves information related to the various structures on Jurong Island from the knowledge graph after obtaining a POST request with the respective city object IDs to process and visualise the desired information, as shown in Figure 8.6.

C4T Emerging Opportunities Fund

1) Brown carbon laser characterisation and light-absorbing property

Prof Markus KRAFT and Dr Yichen ZONG

The purpose of this research is to investigate brown carbon (BrC, light-absorbing organic carbonaceous species) from combustion emissions. BrC is a major source of air pollution in Southeast Asia and a factor of climate change. The project's experimental work is carried out in partnership with researchers from the Department of Environmental Engineering in NUS. The major experiments have been finished in the last few months.

A wide range of fuel and fuel additives were combusted in a compression ignition engine, and the soot emissions were characterised in real-time by a combination of a soot-particle aerosol mass spectrometer (SP-AMS) and a seven-wavelength aethalometer. A paper covering both BC and BrC emissions from PODE3–diesel blended fuels is currently under review.

2) Chemical farming

Assoc Prof YAN Ning and Prof Alexei LAPKIN

The aim of the project is to identify nanoalloy catalysts for amination reactions. While there have been no technical updates since the previous reporting, a joint paper has been submitted by **Prof Alexei LAPKIN (PI, CAM)** and **Assoc Prof Ning YAN's (PI, NUS)** team on the development of an automated search route to prepare renewable organonitrogen compounds.

3) Impact of Singapore's shipping activities on urban air quality

Prof Markus KRAFT and Ms Mei Qi LIM

The initial motivation of this work is to evaluate the impact of emissions from shipping activities on air quality in Singapore and to demonstrate the knowledge graph technology in handling a cross-domain application. The generation and dispersion of the pollutants is influenced by an extremely diverse range of geospatial factors such that the data required to conduct a simulation of the dispersion of pollutants are highly heterogeneous, resulting in a demand for data interoperability. For instance, geospatial information is typically described in raster and vector data types, normally stored in relational databases. Over the past six months, we have worked on using the knowledge graph to store semantically multiple heterogeneous geospatial data from different domains (in raster and vector data types) related to pollution generation and dispersion modelling.

We are investigating the use of ontology-based data access (OBDA) to semantically search, access, and manipulate structured geospatial data types such as elevation data, land cover data, and terrain data. As shown in Figure 8.7, the OBDA solution consists of three main modules: the input data, the mapping, and the ontological representation. The input data consists of the raster and vector data for elevation, wind speed and building geometries, which are conventionally stored in relational databases. A rigorous mapping scheme was established to represent the query logic and entities in the relational database to relate to the virtual knowledge graph. An ontology for describing the raster data is created based on the raster files' metadata structure. Another ontology for describing geographic polygons called 'OntoGeospatialPolygon' is also created to facilitate more efficient interaction between the raster dataset and the vector dataset, which is characterised by polygons. The ontologies provide a logical abstraction independent of how and where the data is being physically stored.

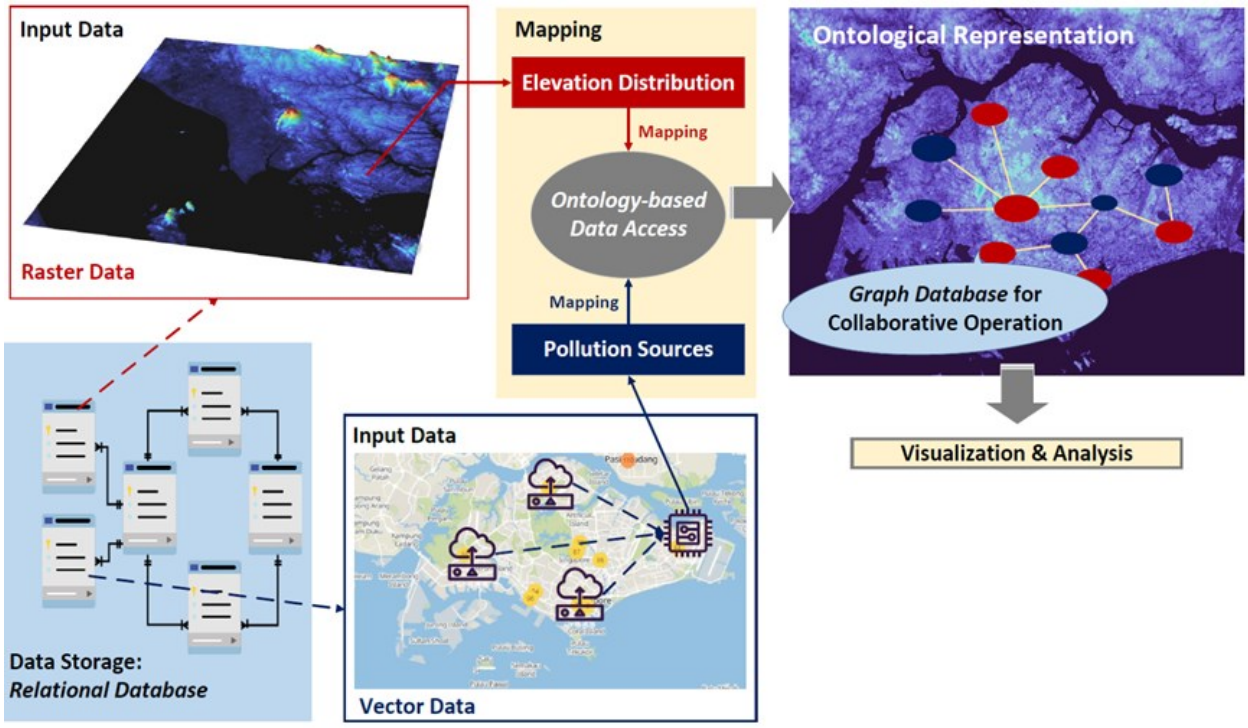


Figure 8.7: Schematic diagram for using ontology-based data access (OBDA) to semantically search, access, and manipulate structured geospatial data types such as raster and vector datasets stored in relational databases.

4) Ignition systems for natural gas engines

Prof Epaminondas MASTORAKOS and Dr B HARIKRISHNAN

Dr B HARIKRISHNAN (Research Fellow, CARES) has been developing the solver isrnConverge in collaboration with **Dr Savvas GKANTONAS (Research Associate, CAM)**. They achieved this by integrating the code for the incompletely stirred reactor network (ISRN) with CONVERGE CFD using its user-defined function APIs. As a next phase, Dr Harikrishnan was able to run the ISRN solver on flow snapshots (or mixing-field) from adaptive mesh refinement (AMR) based CFD simulations. As a result, computationally inexpensive ISRN's kinetic post-processing and the high fidelity CFD results obtained by advanced AMR capabilities of CONVERGE are combined.

The post-processing step requires time-averaged quantities, such as flow variables and mixing-field, from the CFD simulation. Large-eddy simulations (LES), including non-reacting and reacting flows, are performed to obtain these variables. Currently, the reacting flow is modelled using a chemical reduction technique called flamelet-generated manifold (FGM). FGM models premixed and partially premixed flames by solving the transport equation for a much smaller number of control variables.

The ISRN solver can take in fully converged or flow snapshots from the CFD simulation. ISRN is solved till convergence in the mixture fraction space for a wide range of operating conditions. This flexibility comes with high predictive power and low computational cost for complex geometries and fuels/fuel blends with detailed chemistry.

The current focus is to tackle complex engineering applications by scaling the number of ISR cells. In future, the solver will also integrate conditional moment closure (CMC) and doubly-conditioned moment closure (DCMC) with CONVERGE.

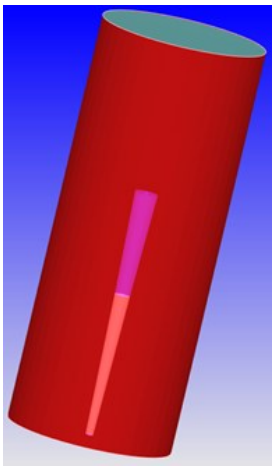


Figure 8.8: Geometry for Sandia Flame D simulation.

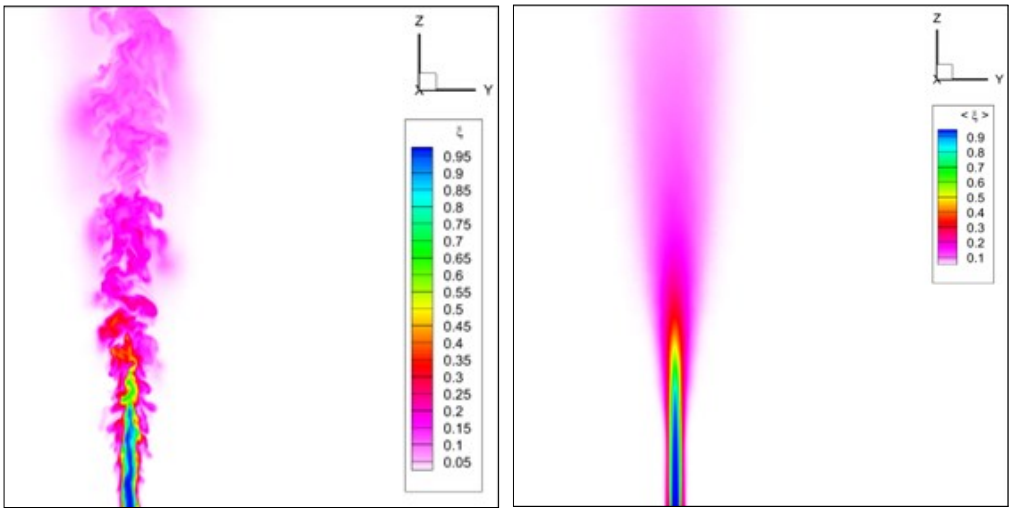


Figure 8.9: (left) Instantaneous and (right) Time-averaged mixture fraction.

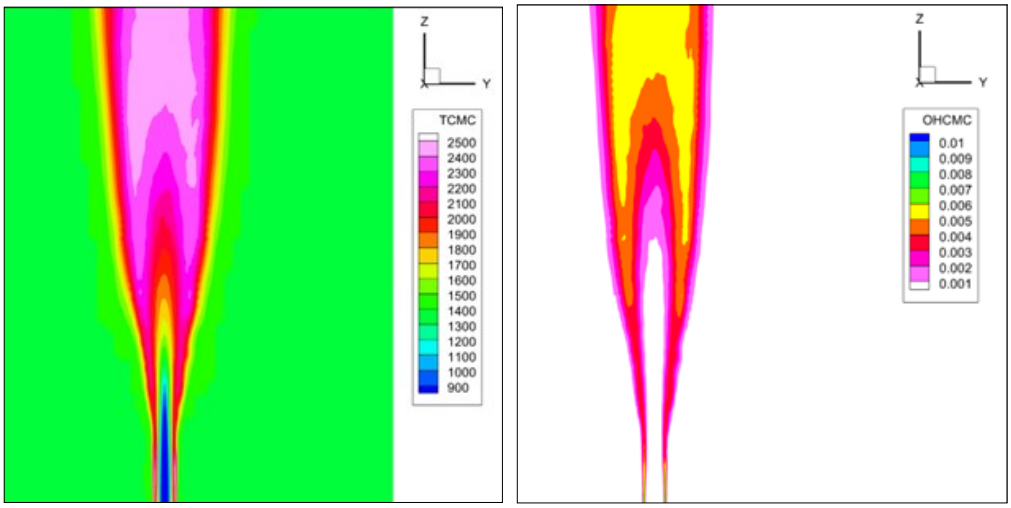


Figure 8.10: (left) Unconditional temperature and (right) OH mass fraction.

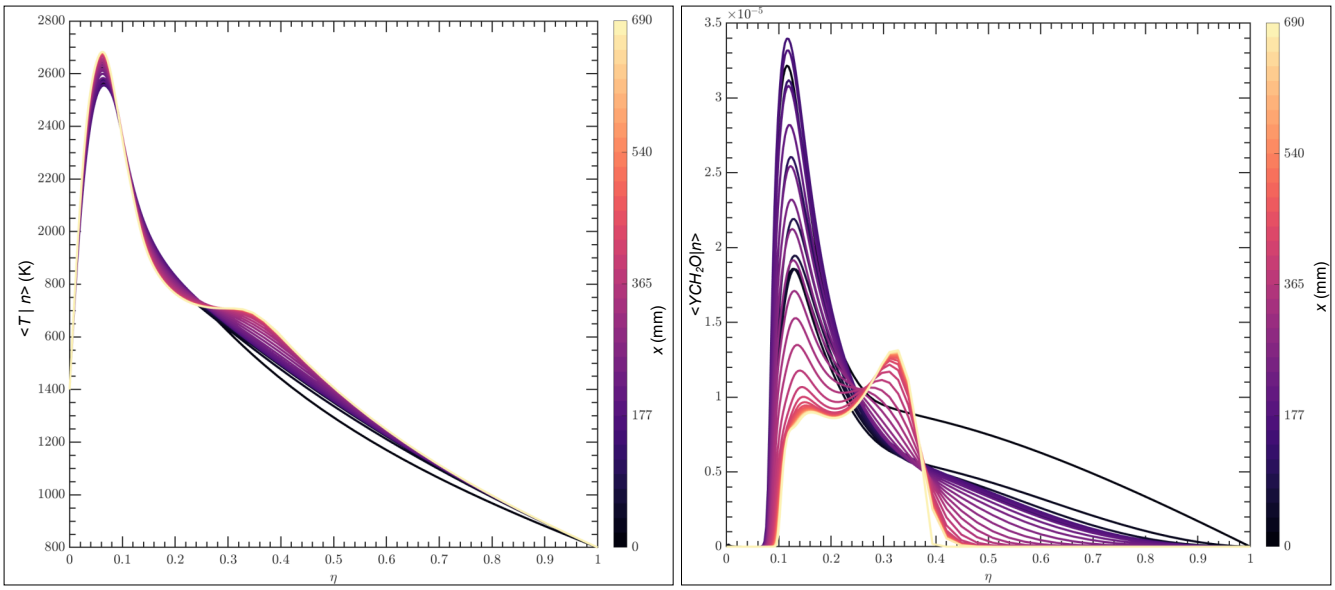


Figure 8.11: (left) Conditional temperature and (right) CH_2O mass fraction of the ISRs.

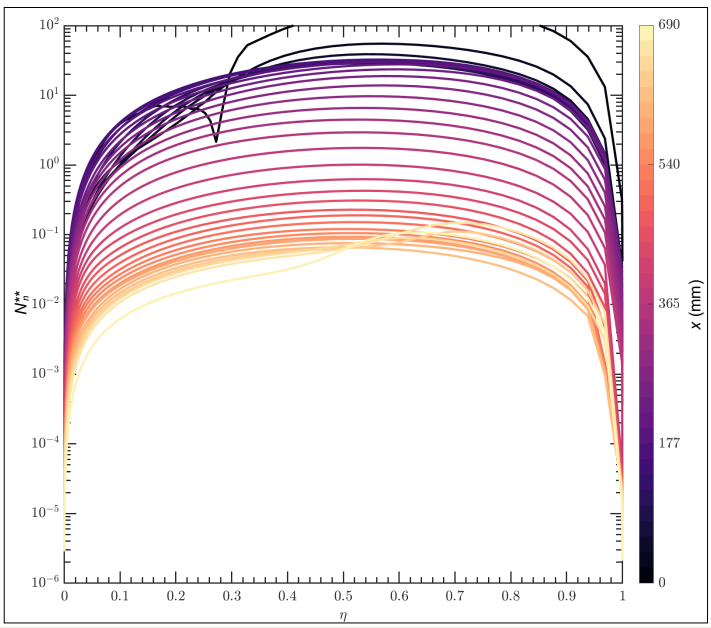


Figure 8.12: Core-averaged scalar dissipation rate.

5) Future marine economy

Prof Epaminondas MASTORAKOS, Prof Steve EVANS and Dr Li Chin LAW

Dr Li Chin LAW's (Research Engineer, CARES) work started with the lifecycle assessment and comparison between 22 pathways for alternative, low-carbon marine fuels. Over the past six months, selected marine fuels were analysed further from a fleet-level perspective, quantifying their potential for various types of ship. In this context, four performance indexes: (i) cargo attainment rate, CAR (i.e., percentage of cargo that can be carried as compared to the reference ship powered by HFO); (ii) specific energy, ES (iii) specific cost, CS and (iv) carbon intensity indicators, CII (i.e., Tank-to-Wake (TTW) energy, TTW cost, and TTW CO_2 emission per ship's deadweight and voyage distance) are defined as ship performance indexes. These indexes allow comparisons between different ships to be done numerically regardless of the ship size and distance travelled. The data is interpreted in various ways. For example, a normalised scoring system is created by allocating equal weight to each of the indexes (CAR, ES, CS, CII) for quantification of the decarbonisation potential of various fuels using the normalised score (Figure 8.13). If CAR is fixed, the maximum distance travelled by the ship can be estimated (Figure 8.14). The analysis has implications for the performance of alternative fuels for the shipping sector and is useful for many stakeholders in this area.

Figure 8.13: Normalised score for ships powered by various alternative fuels

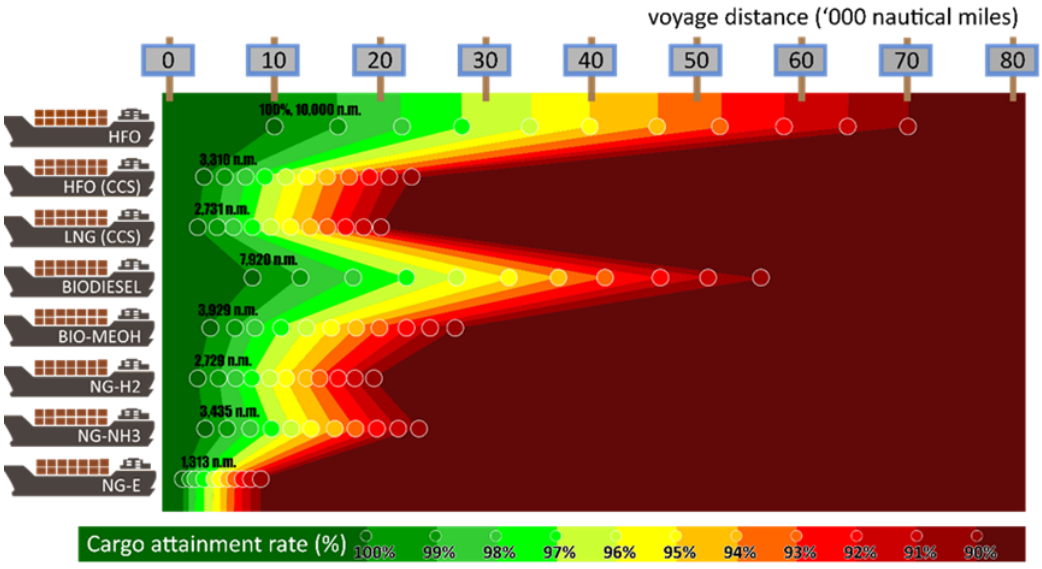


Figure 8.14: Chart of cargo attainment rate for various alternatives against voyage distance travelled

6) Carbon reduction strategies of top chemical companies

Prof S. VISWANATHAN, Dr Abhiruchi GADGIL and Dr Yan WANG

Dr Abhiruchi GADGIL (Research Fellow, NTU) has collected data from the subscribed Trucost datasets for Scope 1, 2, and 3 emissions, financial data, Paris alignment status, carbon earnings at risk, ESG scores, and several other minor datasets, for the sectors of Oil and Gas, Cement, Chemical Manufacturing and Steel sector. This data collection concluded at the end of July 2022. For the oil and gas sector, we have started analysing and formulating paper ideas based on establishing correlations of various parameters like emissions, climate goals, and investments in low-carbon energy to actual emission reduction likelihood, which is aligned toward the Paris agreements' goal of 1.5 degrees. Besides the Trucost datasets, extensive data collection is also being done for other relevant factors, including, Research and Development budgets for high emitting sectors, internal carbon tax status, etc. Oil and Gas manufacturing is currently a geopolitical issue, with several conflicting requirements like the oil crisis due to the Russia-Ukraine war in some parts of the world and the need for the planet to adopt clean energy. Several stress tests will also be done to understand the effect of such stress scenarios and the possible strategies of the countries and organisations. An overarching understanding of the hard-to-abate sector will also be attempted in the upcoming studies triangulating the analysis from studies of all the different sectors mentioned above. We expect to submit a paper on the oil and gas insights by the beginning of next year.

For the EOF project relating to the carbon tax, we focus on an economic mechanism called the Internal carbon tax, which companies increasingly implement to realise their emission reduction targets. Using game theory and optimisation methods, we model the scenario in which the firm implements the internal carbon tax to meet its science-based emission targets. The BUs make carbon abatement investment and production decisions in response to the tax. We find that the firm can achieve the first-best solution using a uniform carbon tax for all its BUs. However, BUs in emission-intensive and trade-exposed sectors lose business and are forced to close prematurely. In such instances, we recommend that the firm sacrifice short-term profits for the long-term benefit of the firm. Using computational experiments, we show that the EITE BUs stay in business and remain profitable in the long run with the adoption of non-uniform carbon, in which a regulatory carbon tax affects all the firms in the market in the future.

We have formulated, analysed, and derived the above insights. However, the current plan is to perform extensive computational analysis to strengthen the argument before further journal submission. We expect to submit the paper before the end of this year.

7) Decarbonisation of Singapore by 2050

Asst Prof Paul LIU, Prof Markus KRAFT and Dr Hui Ling TAN

Dr Hui Ling TAN (Research Fellow, NTU) has been focusing on analysing the potential strategies to decarbonise Singapore, aiming for net-zero emissions by 2050. As shown in Figure 8.15, industry, power, and transport are expected to be the major contributors to primary greenhouse gases (GHG) emissions in 2050. The analysis suggests that improving energy efficiency and electrification of the industry sector, adopting solar power, replacing the existing fuel mix in the power sector with clean and low-carbon renewable energy sources, and electrification of vehicles are helpful in reducing carbon emissions. However, a negative emission method, that is, bioenergy with carbon capture and storage (BECCS) is demonstrated to be essential for Singapore to achieve net-zero emissions by 2050.

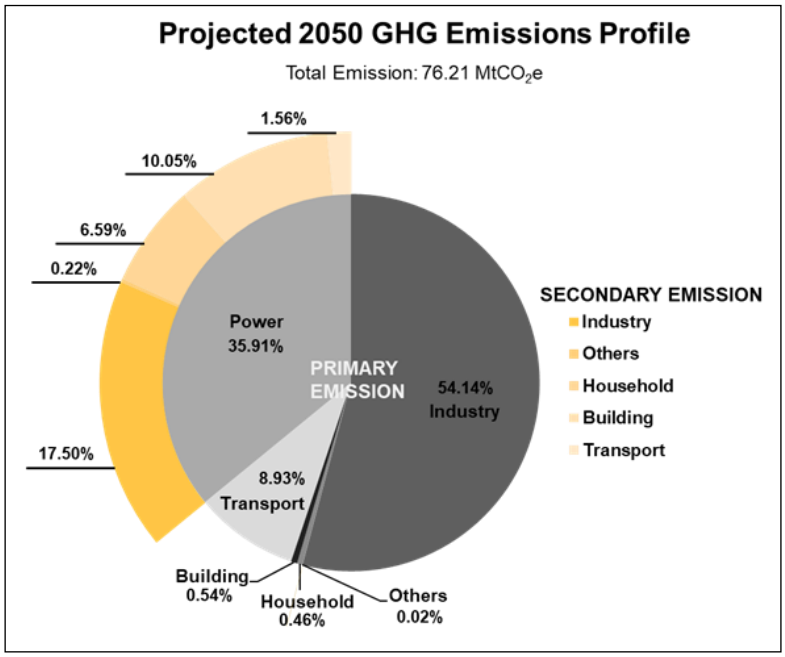


Figure 8.15: Singapore's projected GHG emissions profile in 2050 based on 2019 data provided by the Energy Market Agency (EMA).

8) Reaction Pathways of Formic Acid Conversion

Asst Prof Paul LIU and Dr Mingwu TAN

Dr Mingwu TAN's (Research Fellow, NTU) main research interest lies in the study of CO_2 hydrogenation reactions. Recently, he has been focusing on the exploration of developing a new 2D nanomaterial with the main purpose to provide a novel catalyst to convert CO_2 to high value added higher carbon alcohols (C2+). In his research, he achieved the synthesis of MoS_2 supported Rh catalyst via a simple hydrothermal method by adjusting the composition and pH value of the solvent for the hydrothermal reaction. He successfully synthesised various thickness levels of MoS_2 with the layer number of MoS_2 controlled from 2 to 20. The results show more in-plane sulfur vacancy and edge sulfur vacancy were exposed as the layer number decreased, enhancing interaction between Rh and MoS_2 support. More importantly, increased sulfur vacancy favourably anchored Rh species in the form of single atoms, increasing the utilisation rate of Rh. The prepared synthesis was used in CO_2 hydrogenation in the aqueous phase with H_2O as the solvent. The results show CO_2 can dissociate using Rh/MoS_2 catalyst to yield surface-bound CO and CH3 under the aqueous phase, thus enabling the hydrogenation of CO_2 to methanol and ethanol.

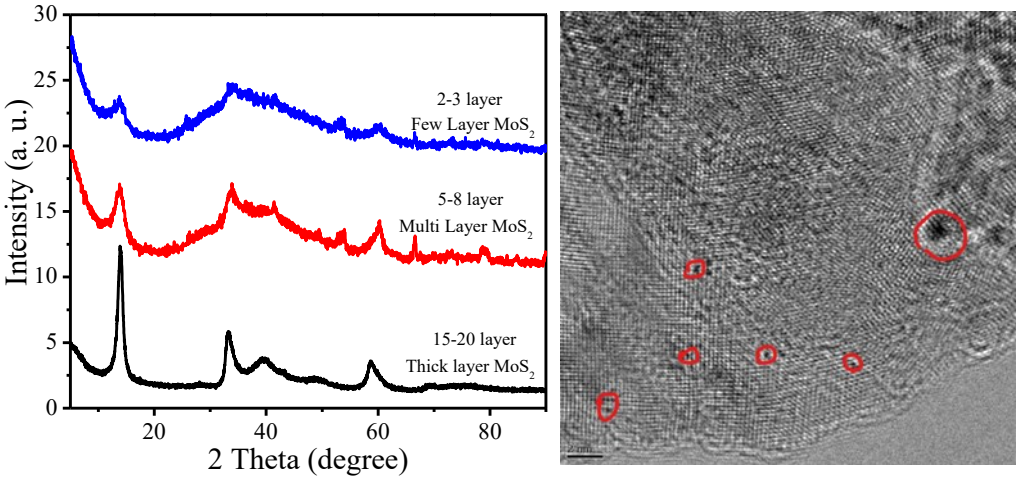


Figure 8.16: (left) XRD patterns of the prepared MoS_2 (right) TEM image of the prepared Rh/ MoS_2

9) Electrified Chemical Production

Prof Jason XU Zhichuan, Dr Adrian FISHER and Dr Yubo CHEN

Dr Yubo CHEN (Research Fellow, NTU) has been focused on developing efficient catalysts for oxygen evolution, a sluggish anodic reaction that provides essential electrons and protons for various electrochemical processes, such as hydrogen generation. He reports that the oxygen evolution reaction (OER) can be efficiently catalysed by cobalt tetrahedra, which is stabilised over the surface of a Swedenborgite-type $YBCo_4O_7$ material. The surface of $YBaCo_4O_7$ possesses strong resilience towards structural amorphisation during OER, which originates from its distinctive structural evolution toward electrochemical oxidation. The bulk of $YBaCo_4O_7$ composes of corner-sharing only CoO_4 tetrahedra, which can flexibly alter their positions to accommodate the insertion of interstitial oxygen ions and mediate stress during the electrochemical oxidation. The density functional theory calculations demonstrate that the OER is efficiently catalysed by a binuclear active site of dual corner-shared cobalt tetrahedra, which have a coordination number switching between 3 and 4 during the reaction. Dr Chen expects that the reported active structural motif of dual corner-shared cobalt tetrahedra in this study could enable further development of compounds for catalysing the OER.

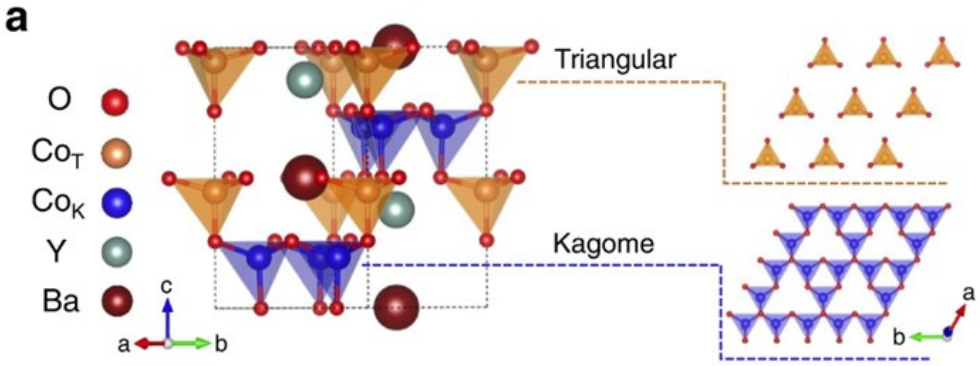


Figure 8.17: Crystal structure of $YBaCo_4O_7$ and the arrangements of corner-shared CoO_4 tetrahedra in the Triangular layer and the Kagome layer.

10) Electrolysis for renewable carbon utilisation

Prof Xin WANG Xin, Dr Xiaogang LI and Dr Hongwei ZHANG

Dr Hongwei ZHANG (Research Fellow, NTU) focused his research on the development of novel electrocatalysts in CO_2RR and ORR applications. He recently published a review titled "Tailoring of active sites from single to dual atom sites for highly efficient electrocatalysis". Single atom catalysts (SACs) have been attracting extensive attention in electrocatalysis because of its unique structure and extreme atom utilisation. However, its activity and practical applications may be limited by the low metal loading and unified single sites induced scaling relations. Tailoring active sites at the atomic level is a promising approach to overcome the existing limits of SACs. Five tailoring strategies were evaluated towards improving the electrocatalytic activity on SACs, namely strain engineering, spin-state tuning engineering, axial functionalisation engineering, ligand engineering, and porosity engineering. From the angle of inducing electron redistribution and optimising adsorption nature of active centres, Dr Zhang summarised the synergistic effect from adjacent atoms and recent advances of tailoring strategies on active sites with binuclear configuration. This includes simple, homo-nuclear, and hetero-nuclear dual atom catalysts (DACs). The review concludes with a summary and perspective of achieving efficient and sustainable electrocatalysis.

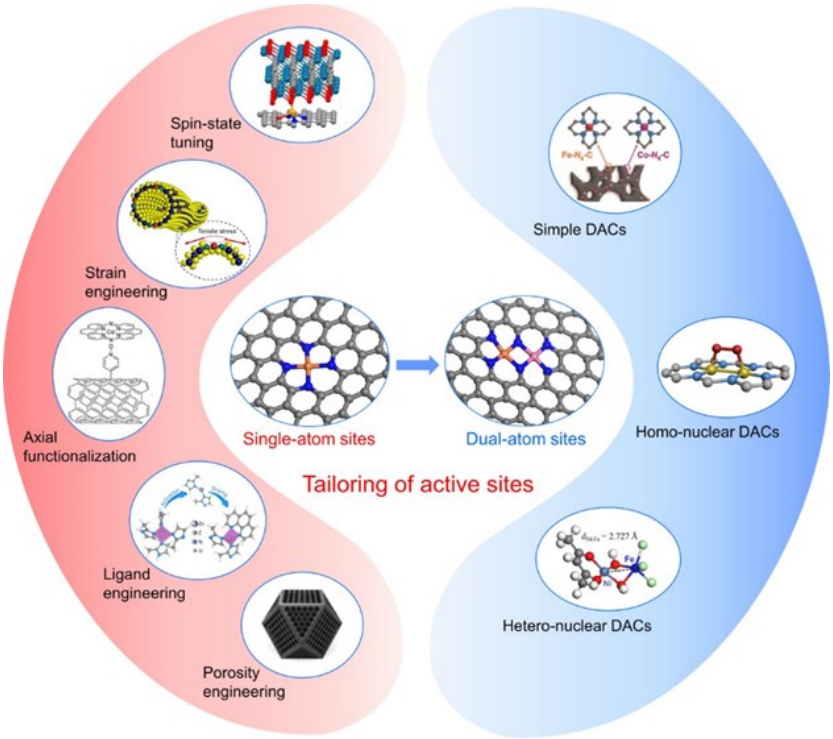


Figure 8.18: Tailoring of active sites from single to dual atom sites for highly efficient electrolysis.

Dr Xiaogang LI (Research Fellow, NTU) has been focusing on the design of electrocatalysts for H_2O_2 production. For electrochemical oxygen reduction toward H_2O_2 production, the adsorption strength of OOH* on the catalytic sites is crucial for catalytic efficiency. Based on previous work, the isolated CoN_4 sites with relatively strong OOH* adsorption could act as a promising candidate for H_2O_2 production, which could achieve high activity and selectivity simultaneously if OOH* adsorption is optimised.

Here, a molecule confinement strategy is developed for efficient H_2O_2 generation. Metal-phthalocyanine (MPc) molecules were introduced on isolated CoN_4 sites to modulate the OOH* adsorption towards high-efficiency H_2O_2 production. Through π-π interaction, the MPc molecule could be confined on the carbon support with CoN_4 sites. In the confined space between the two single-atom sites from MPc and CoN_4 sites, OOH* adsorption could be modulated to facilitate the 2e⁻ ORR pathway. Experimental results show that CoPc could endow the CoN_4 sites with enhanced selectivity of over 85% for H_2O_2 in a wide potential range and a higher onset potential of 0.855V, compared to bare CoN_4 sites with lower selectivity and onset potential of 0.844V. Related DFT calculations are ongoing in collaboration with **Asst Prof Tej CHOKSI (Co-PI, NTU)** and a manuscript is being prepared.

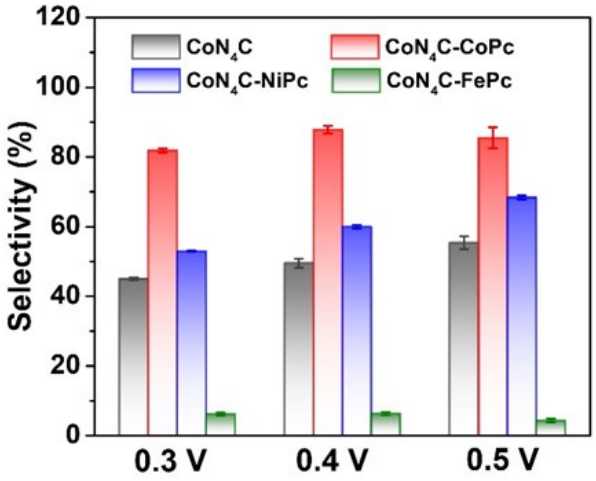


Figure 8.19: Catalysts selectivity for H_2O_2 production at applied potential.

PUBLICATIONS

ALL C4T PUBLICATIONS WITH CREATE ACKNOWLEDGEMENT

The following list includes all the C4T publications from the beginning of Phase 2 (November 2018). Those in bold are new for this reporting period. For a full record of Phase 1 publications (April 2013—October 2018) please visit our Publications page on the CARES website: www.cares.cam.ac.uk/publications/

C4T joint IRP publications

IRP 1 and IRP 3

- **Liu, Guanyu, Peace Adesina, Noushin Nasiri, Haojing Wang, Yuan Sheng, Shuyang Wu, Markus Kraft, Alexei A. Lapkin, Joel W. Ager, and Rong Xu. 2022. 'Elucidating Reaction Pathways of the CO 2 Electroreduction via Tailorable Tortuosities and Oxidation States of Cu Nanostructures'. *Advanced Functional Materials* 32 (36): 2204993. https://doi.org/10.1002/adfm.202204993.**

IRP 2 and IRP 3

- **Sun, Libo, Vikas Reddu, Shibo Xi, Chencheng Dai, Yuan Sheng, Tan Su, Adrian C. Fisher, and Xin Wang. 2022. 'Cobalt Quaterpyridine Complexes for Highly Efficient Heterogeneous CO 2 Reduction in Aqueous Media'. *Advanced Energy Materials* 12 (34): 2202108. https://doi.org/10.1002/aenm.202202108.**

IRP 3 and IRP JPS

- **Rihm, Simon D., Jethro Akroyd, and Markus Kraft. "Modelling a Detailed Kinetic Mechanism for Electrocatalytic Reduction of CO2." *Proceedings of the Combustion Institute*, 2022, S1540748922001249. https://doi.org/10.1016/j.proci.2022.07.096.**

C4T IRP 1: Sustainable reaction engineering

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