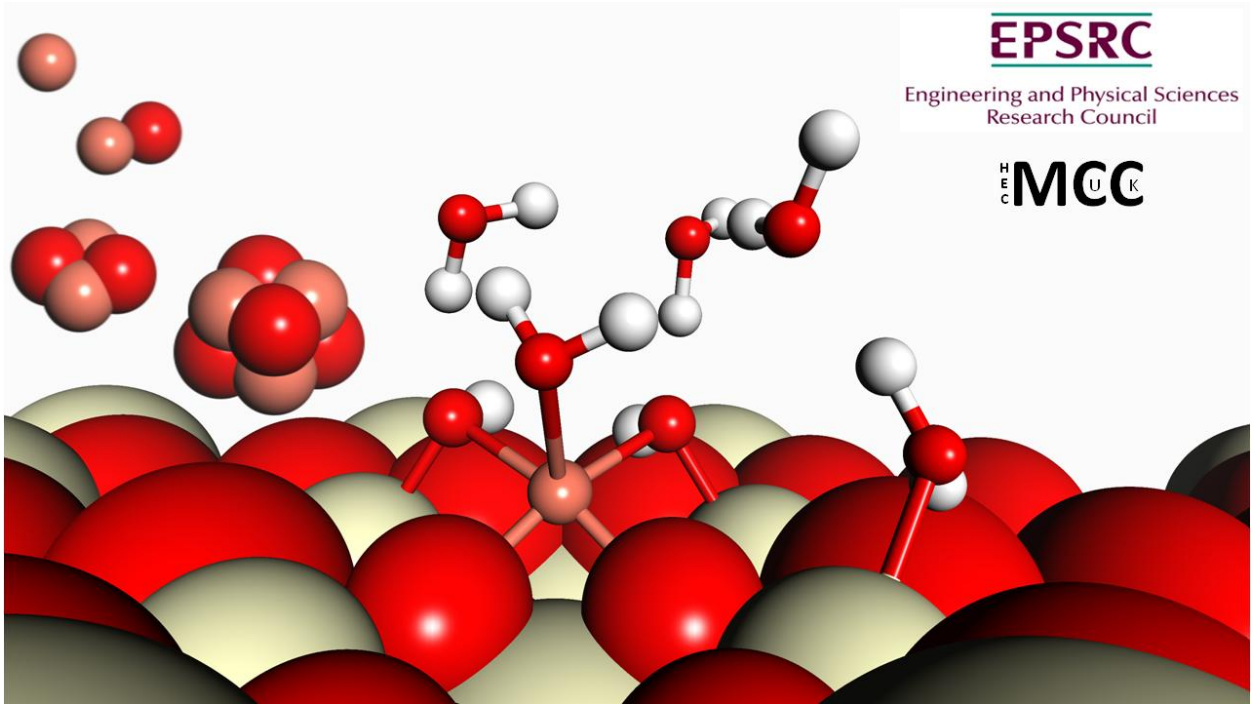
MCC 7th Conference



Daresbury

Conference: Monday 7th July 2025 – Wednesday 9th July 2025
MD Workshop: Thursday 10th July 2025
ML Training: 10th July 2025 – 11th July 2025

Sponsors

The MCC is very grateful for funding from EPSRC (**EP/X035859**).

We are grateful that this event is also sponsored by:

STFC (Daresbury)

Department of Chemistry (UCL)

Organising Committee

Georgia Lomas, Alin-Marin Elena, Thomas Keal, Alexey Sokol, and Scott Woodley

We also would like to thank all members, our theme leaders, and our invited guests, who have supported this event by joining us in-person in Daresbury, albeit one, two, three, or all five days.

Kostya Trachenko
1971 - 2025

Professor Kostya Trachenko, who has died earlier this year aged 54 of cancer, was a theoretical physicist and a theme leader on the MCC Committee. After graduating at Lviv (Physics, 1996) and Cambridge (PhD, 2001), he moved to Queen Mary University of London in 2010. “He developed a mathematical framework explaining how, under certain conditions, liquids behave like solids – such as when falling into water from a height. This insight led to a deeper understanding of how transverse sound waves propagate through liquids, and how these behaviours change with temperature. His work also explained the longstanding mystery of why the heat capacity of liquids often decreases with temperature, unlike in solids. In addition Kostya showed how ultra-thin liquid films behave more like solids when confined to surfaces, offering explanations for phenomena observed in nanotechnology and material science. He is survived by his wife, Carmen, whom he married in 2013, their two children and his mother [1].”

[1] Alessio Zaccone, The Guardian, https://www.researchgate.net/publication/392159035

MCC 7th Conference - Programme

Monday 7rd July			
11:30	**Registration Desk Open**		**Daresbury Laboratory**
12:00	**Lunch**		
Session 1:	Power / Discovery	**Chair** Richard Catlow	
13:30	Suppressing phase transformation and cation migration with Mg and Si doping in Fe–Mn layered oxides for sodium-ion cathodes		Arup Chakraborty Oxford (power-isl)
13:50	Interlayer Metals for Zero-Excess Li Metal Batteries: A Multiscale Approach Combining Machine Learning Potentials and DFT		Neubi Xavier Jr. Surrey (power-cai)
14:10	**Invited:** BioSimDB: Data Tools and Infrastructure for Biomolecular Simulation		James Gebbie PSDI, STFC
14:50	PSDI Thematic Portals		Abraham Nieva de la Hidalga, Cardiff
15:10	REMatch plus SOS: Machine-learning-accelerated structure prediction for supported metal nanoclusters		Yunyu Zhang UCL(discov-cat)
15:30	**Tea**		
Session 2:	Surfaces and Interfaces	**Chair** Thomas Keel	
16:00	Modelling core-electron binding energies at metal oxide surfaces		Johannes Lischner Imperial (lis)
16:20	Modelling of fcc Ruthenium Surfaces and Particles with Hydrogen		Marietjie Ungerer Leeds (react-lee)
16:40	Exploring the possible superconducting mechanism of infinite-layer nickelates		Hangbo Qi UCL (bulk-sok)
17:00	Atomic-Scale Insights into Passivation and Halide Mixing in 3D and 2D Halide Perovskites		Vikram Oxford (nano-isl)
17:20	Atomistic modelling of SiO_2/Ta interfaces		Margherita Buraschi UCL (shl)
17:40	**Poster Session** Starting with 45s-lightning presentations (1 slide per poster) Session will include food and refreshments		
20:00	Session ends		

Tuesday 8th July			
Session 3:	Bulk	**Theme Leader** Alexey Sokol	
9:00	**Invited:** Challenges and opportunities for Computer Modelling in calculating X-ray absorption Near Edge Structure (XANES) – a powerful tool for local structure determination		Gopinathan Sankar UCL
9:40	QM/MM simulations of intrinsic point defects in Sr based materials SrO and $SrTiO_3$		Taifeng Liu Henan (sok)
10:00	QM/MM Investigations of Defects in MgO and Their		Liam Morgan

	Use as a Model System for High-Tc Superconductivity		UCL (cat)
10:20	Computational prediction of $Cd_2Sb_2O_7$ as a candidate TCO		Peter Russell Birmingham (dos)
10:40	Investigating Electron Localisation in Defective Bulk Ceria with Dispersion Corrected DFT		Thomas Hill Cardiff (react-cat)
11:00	**Coffee**		

Session 4:	Reactivity	**Theme Leader** Umberto Terranova	
11:20	**Invited (New Investigator):** Computation for a green future: exploring the catalytic conversion of methane		Matthew Quesne Leeds (que)
12:00	Computational investigation of the Fe_xRh_y alloy structures and the mechanism and selectivity of CO_2 hydrogenation		Shihia Sun UCL (cat)
12:20	Developing Next-Generation Catalysts for Iso-butanol Production Using Machine Learning		K. Agrawal Cardiff (log)
12:40	Amide-Rich NaH as a Highly Active Catalyst for Ammonia Synthesis		Michael D. Higham UCL (cat)
13:00	**Lunch**		
Session 5:	Algorithms	**Chair** Scott Woodley	
14:00	Workflows for QM/MM Simulations of Metal Oxides		Oscar van Vuren Cardiff (log)
14:20	Hybrid QM/MM and Machine Learning for Zeolite Catalysts and Silica Polymorphism		Jamal Abdul Nasir UCL (react-sok)
14:40	First-principles modelling of infrared and Raman spectra		J. M. Skelton Manchester (ske)
15:00	The Crystal Isometry Principle infers chemistry from geometry		Vitaliy A. Kurlin Liverpool (dar)
15:20	**Invited:** TBA		Mark Storr AWE
16:00	**Coffee**		
Session 6:	Kostya Trachenko	**Chair** Richard Catlow	
16:30	Radiation damage: from glasses through solid oxides/ceramics to metals.		Ilian Todorov STFC
16:50	The effect of impurities and irradiation on the glass network – Cs_2O-loaded iron phosphate case study		Cillian Cockrell Bangor (enviro-kos)
17:10	Calculating system properties on-the-fly in DL_POLY 5		H. L. Devereux QMUL (algor-kos)
17:30	TBA		TBA
18:00	session ends		
18:00 – 21:00	**Conference BBQ Dinner**		Daresbury Caterers

Wednesday 9th July			
Session 7:	Bio and Soft Matter	**Chair** Jamieson Christie	
9:00	Nanoscale Non-adiabatic Dynamics Simulation of Charge Generation in Organic Solar Cells		Filip Ivanovic UCL (biosoft-blu)
9:20	Investigating the conductivity of Multi-Heme Cytochromes		A.M. Petho UCL (biosoft-blu)
9:40	Elucidating the effect of doping on the mechanical and chemical stability of hydroxyapatite		Mahdi Tavakol Oxford (biosoft-tan)
10:00	**Coffee**		
Session 8:	Excalibur	**Chair** Jonathan Skelton	
11:00	**Invited:** Integrating quantum computing with HPC		Viv Kendon Strathclyde
11:40	Exploring the thermodynamics of disordered materials with quantum computing		Bruno Camino UCL
12:00	Multiscale workflows and Parallel HDF5 I/O		Rajany KV STFC
12:20	GPU acceleration of the CASINO quantum Monte Carlo program using OpenACC		Neil Drummond Lancaster
12:40	Porting ONETEP and CASINO onto GPUs		Phil Hasnip York
13:00	**Lunch**		
	Reactivity / Bulk	**Chair** David Willock	
14:10	Au and Au_3Cu nanoclusters as catalysts for light-driven CO oxidation		Igor Kowalec Cardiff (react-cat)
14:30	Investigating Hydrogen Interaction with Defective and Doped MgB_2 via Density Functional Theory		Navaratnarajah Kuganathan Nottingham (bulk-lin)
14:50	DFT & Data Mining Assisted Catalyst Discovery of Cu-based Alloys for CO2 Hydrogenation to Alcohols		Z. Lu Cardiff (react-log)
15:10	Localised electron states in Amorphous Alumina		Jack Strand UCL (bulk-shl)
15:30	**MCC General Meeting**		
16:30	Close		

1st reserve	Mechanism of CO_2 Reduction to Methanol with H_2 on an Iron(II)-scorpionate Catalys	Chengxu Zhu Manc'ter (react-que)

Summary of Poster Presentations

1	Design and development of novel composite electrodes for high-energy density supercapacitors	Abeer A. Hdadai Newcastle (power-jad)
2	A Computational Study of CO2 Methanation over Low-index Nickel Surfaces	M. Alotaibi UCL (react-cat)
3	Catalytic Partial Oxidation of Methane using an Yttria-Stabilized Zirconia Catalyst	Erze Gao UCL (react-cat)
4	Investigation of Li-N-H phase space for next generation Li-ion electrolytes	Peter J. Graham Birmingham (power-dos)
5	Computational Modelling of Zeolite-Catalysed DMN Synthesis	Oliver Morris Cardiff (react-cat)
6	Data-driven design of electroactive metal-organic frameworks	Ashna Jose Imperial (discov-wal)
7	Promoting MgH_2 Destabilization Through Reactive Elemental Additives	Navaratnarajah Kuganathan Nottingham (bulk-lin)
8	Germanium Dioxide – A UWBG Material for Optoelectronic Applications	Jacob C. Baggott Birmingham (bulk-dos)
9	A Computational and Theoretical Study on Methanol Synthesis	Matis Ferrini Lincoln (react-arc)
10	Understanding Polarisation at Ferroelectric-perovskite Interface – A molecular Dynamic Perspective	Tingwei Li QMUL (surfin-but)
11	First-principles study of the thermoelectric properties of $Sn(S_{1-x}Se_x)$ alloys	Min Zhang Manchester (power-ske)
12	Structural Characterisation and Analysis of $NaTaO_xC_{l6-2x}$ for Solid¬—state Sodium Batteries	Abigail C. Parsons Birmingham (power-dos)
13	Effect of Nitrogen Incorporation in a-HfO_2	Isaac Mackley UCL (bulk-shl)
14	Vibrational Dynamics and Mechanical Anisotropy in Open Framework Materials: Computational Design Rules for MOF-based Triboelectric Generators	Debayan Mondal Oxford (discov-tan)
15	Carbon Nitride-Supported Metals for the Selective Oxidation of HMF to DFF	Xue Yong Liverpool (react-)
16	Mg Doping in GaN: application of the MACE Potentials	Yuxi Zhang UCL (bulk-smw)
17	Al Doping in ZnO: application of the MACE Potentials	Zhuoran Ji UCL (bulk-smw)
18	Modelling the phase transition of VO_2	Zichao Wang UCL (buk-smw)
19	Quantum Inspired Optimisation of $LiMnO_2$	Selin Kilic UCL (algor-smw)

Talks

BioSimDB: Data Tools and Infrastructure for Biomolecular Simulation

James Gebbie

STFC, Daresbury Lab

BioSimDB is a data infrastructure prototype that has been developed for the biomolecular simulation community in the UK and funded by the Physical Sciences Data Infrastructure (PSDI). In this talk I will introduce the PSDI, the drivers for the programme and its goals along with our work in the biomolecular simulation pathfinder within the PSDI. The biomolecular simulation community (CCPBioSim and HECBioSim) are a large consumer of HPC resources in the UK and they produce a wealth of data in their quest to understand the biological function of protein based systems. As part of this process, researchers often start with experimental structures and perform many operations on the data before eventually running their simulation models. This process is often not fully recorded in publications, which makes reproducing simulation workflows difficult from papers alone.

Historically biomolecular simulators have viewed themselves as the end of the chain of data consumers without other communities consuming their data. The advent of AI and its associated data curation practices has changed this outlook and the battle now is to get data into shape to exploit these tools. Dynamics is increasingly being consumed by the very experimental communities that have historically been the creators of input data for the simulations community, having services where simulation data can be searched and obtained is key to this. In BioSimDB we have been addressing the issues around FAIR-ising biomolecular simulation data without reinventing wheels. In this talk we will look at the drivers for and the issues facing making our data FAIR, we will show our work in the PSDI and the current state of the infrastructure prototype and its associated tools.

Challenges and opportunities for Computer Modelling in calculating X-ray absorption Near Edge Structure (XANES) – a powerful tool for local structure determination

Gopinathan Sankar

[*]School of Chemistry, University College London

Here I will cover examples of what we have been doing on catalytic materials, trying to explain the changes in the XANES through simulating the spectra using local structures and comparing them with experimental data. I hope this will be of some interest to the MCC community.

Computation for a green future: exploring the catalytic conversion of methane

Matthew G Quesne*

*School of Chemistry, University of Leeds, Leeds, UK. e05-react-**que***

Methane emissions now account for ~30 percent of anthropogenic warming, therefore, centralised methane capture and conversion is an extremely promising technology for future green chemistry research. This talk will explore methane conversion by: (1) the photoactive covalant framework material CFT-1[1] and (2) Au supported on ZSM-5.[2]

Materials and Methods: *CFT-1:* Catalyst activity of CTF-1 is compared to: (1) Anatase TiO_2 and (2) g-C_3N_4. The photocatalytic reactions are carried out under irradiation by a 365 nm LED source. *Au-ZSM-5:* H-ZSM-5 with SiO_2/Al_2O_3 ratio of 25 is loaded with Au by deposition–precipitation. Methane oxidation is carried out in an autoclave reactor. Computation calculations for both systems used Density Functional Theory (DFT) in combination with Perdew–Burke–Ernzerhof functional (PBE), as implemented in the Vienna Ab initio simulation (VASP) code.

Results and Discussion *CFT-1:* This study demonstrates that the intrinsic molecular heterojunction in the CTF-1 polymer is highly selective for solar-driven methane transformation towards ethanol. At a GHSV of 2000 mL h^{-1}, the photocatalytic methane-to-ethanol conversion at 65 ℃ represents a very high selectivity of ca. 80 % and a yield of 122.4 mol h^{-1}. The study rationalises a process for methane transformation to ethanol via ethane, without the over-accumulation of strong oxidants, seen in C_3N_4-based catalysts. *Au-ZSM-5:* This study showed that Au nanoparticles supported on ZSM-5 can oxidise CH_4 to methanol and acetic acid with minimal formation of CO_2. In the batch experiments, a maximum oxygenate productivity was observed of 7.3 mol Au^{-1} h^{-1}. This is much higher than that reported for the exemplar Cu-based catalysts tested under similar flow conditions.[3] In contrast to the Cu-zeolite catalysts, for which only C1 products are observed, C2 oxygenates are the major products observed with the Au-ZSM-5 catalyst, demonstrating that the Au catalyst operates by a different mechanism.

References

[1] . J. Xie *et al., Nature*, **639**, 368-374, (2025).
[2]. G. Qi *et al., Nat. Cat.*, **5**, 45-54, (2022).
3. K.T. Dinh et al., J. Am. Chem. Soc, 141, 11641-11650, (2019).

Integrating quantum computing with HPC

Viv Kendon

* *University of Strathclyde, Glasgow*

I will explain how CCP-QC is helping to develop quantum algorithms that can enhance existing classical simulation codes, ready for when quantum hardware is available as an accelerator in HPC facilities. Underneath the current hype, quantum computers are developing steadily towards useful computational devices. Future deployment as quantum accelerators in high performance computing facilities is already beginning for benchmarking and testing. I will explain how quantum computers work, what we can do now, and what still needs to be done to benefit from increased computing power in the future.

Exploring the thermodynamics of disordered materials with quantum computing

Bruno Camino

* *University College London*

Disordered materials — including alloys, solid solutions, and doped systems — are central to technologies such as energy storage, electronics, and catalysis. Accurately predicting their properties remains a significant computational challenge due to the vast number of possible atomic configurations. Classical optimisation methods, such as simulated annealing, often spend computational effort exploring high-energy states that are not thermodynamically relevant.

We present a novel scalable and accurate method that uses quantum annealing to efficiently sample low-energy configurations in disordered systems. This approach incorporates temperature effects and supports large supercells, generating Boltzmann-like distributions that reflect realistic thermodynamic behaviour. A key feature of the approach is the use of chemical potential to continuously tune the composition of materials, ensuring that the resulting energy Hamiltonians are compatible with current quantum annealing hardware.

Recent results we published in *Science Advances* demonstrated the method's effectiveness by predicting band gap bowing in $Al_{1-x}Ga_xN$ and bulk modulus variations in $Ta_{1-x}W_x$, with excellent agreement with experimental data. Ongoing work explores the extension of this framework to broader classes of disordered systems.

Modelling core-electron binding energies at metal oxide surfaces

Johannes Lischner*

[*]Department of Materials, Imperial College London *e05-surfin-**lis***

X-ray photoemission spectroscopy (XPS) measures the binding energies of core levels. As these binding energies are highly sensitive to the local chemical environment, XPS is a powerful technique for gaining insights into the structure of complex materials. However, the analysis of measured XPS spectra can be challenging as it is often difficult to infer the chemical environments from the measured binding energies. To address this problem, my research group has developed accurate parameter-free modelling approaches which can predict absolute core electron binding energies of molecules, solids (both metallic and insulating) and surfaces. These approaches are based on density-functional theory and the Delta-SCF technique and often produce absolute binding energies with errors of less than 0.2 eV. In my talk, I will present a detailed study of core-electron binding energies at SnO2 surfaces which allow insights into recent XPS measurements of these surfaces.

QM/MM simulations of intrinsic point defects in Sr based materials SrO and $SrTiO_3$

Taifeng Liu*, Xingfan Zhang, Richard Catlow, and Alexey Sokol

*Department of Chemistry, University College London e05-bulk-**sok**

Strontium oxide (SrO) belongs to the alkaline-earth oxides family, characterized by its high symmetrical rock salt structure which is suitable for diverse applications including catalysis and microelectronics. It is well-known that the SrO has *p*-type conductivity. In the contrary, its counterpart Strontium titanate $SrTiO_3$ (STO) is a *n* type semiconductor often used for ferroelectric and electronic ceramic devices and also used a photocatalyst for water splitting.

We have investigated the defect chemistry in SrO using QM/MM embedded-cluster approach (JMCA, 2025, 13, 7176). Our analysis suggests that dominant defects in SrO primarily consist of oxygen vacancies, strontium vacancies, and oxygen interstitials, and, in particular, the peroxide-like oxygen interstitial. Confirmation of these findings is evident in the charge carrier and defect concentration results. The hole concentration stems from strontium vacancies with charge -2, and it is the original of the *p*-type conductivity of SrO.

We also explored intrinsic defect calculations in STO using QM/MM approach. For vacancies, we applied ghost atom technique and obtained highly accurate computation results. The other defects like interstitials are going on and will be finished soon.

QM/MM Investigations of Defects in MgO and Their Use as a Model System for High-T_c Superconductivity

Liam Morgan*, C. Richard A. Catlow, Alexey A. Sokol, Thomas W. Keal

*Department of Chemistry, University College London e05-bulk-**cat**

The defect chemistry of magnesium oxide has been extensively studied for many years, with a myriad of applications in catalysis, optoelectronics and many other fields. Both vacancies and substitutionals have been shown to be effective in providing MgO with additional useful properties. Our work employs modern hybrid quantum mechanical – molecular mechanical (QM/MM) techniques, using the ChemShell code[1], to both reassess previously investigated defects and probe others that have been less researched.

Whereas a number of different transition metal ions have been investigated and widely applied as substitutionals in MgO, for example Ni and Co in catalytic systems, research into copper dopants in MgO is more sparse. Currently, the work on such systems is largely focused on nanoparticles, some uses in catalysis or the Jahn-Teller distortion induced by the dopant[2]. Our work focuses instead on the localisation and possible trapping of charge carriers within a $(CuO_6)^{10-}$ unit. Whereas oxygen bound polarons have been observed with a number of doped metal oxides[3], currently, there is no evidence for the influence that copper has, in this regard. Hybrid-DFT within QM/MM has been utilised to track the migration of such holes, with further electromagnetic resonance (EPR) calculations allowing for direct comparison to experimental work. Our work will be used to guide future investigations of hole states and bipolaron formation in superconducting cuprate materials.

Additionally, the optical excitations of F-centres within MgO have been investigated and compared to previous theory of their excited states. It has been reported that excited states of electrons within oxygen vacancies in MgO are hydrogenic in character with distortions from the crystal field environment[4]. Through use of time-dependent DFT (TDDFT) within a QM/MM scheme, these characteristics are reinvestigated.

References:
[1] Y. Lu et al., J. Chem. Theory Comput. 15, 1317 (2019). [2] M. J. Riley, C. J. Noble, and P. L. W. Tregenna-Piggott, The Journal of Chemical Physics 130, 104708 (2009). [3] O. F. Schirmer, physica status solidi c 4, 1179 (2007). [4] C. Jun et al., Eur. Phys. J. B 9, 593 (1999).

Exploring the possible superconducting mechanism of infinite-layer nickelates

Hangbo Qi*, Haiyan Xiao, Liang Qiao, Alexey A. Sokol, Dimitar Pashov, Xiangru Han, Xuelei Sui, Bing Huang, C. Richard A. Catlow

*University College London, Kathleen Lonsdale Materials Chemistry, 20 Gordon Street, WC1H OAJ, London, UK e05-bulk-**sok***

Infinite-layer nickelates have recently attracted significant attention as a promising platform for studying unconventional superconductivity. Despite extensive efforts, the underlying superconducting mechanism in these materials remains unclear yet. In this work, we use density functional theory (DFT) combined with Wannier function analysis to explore how various factors, i.e., polarity mismatch, hydrogen doping, and in-plane strain, affect the electronic properties of $NdNiO_2$. Our results reveal that both $SrTiO_3$ and $LaAlO_3$ substrates induce lattice and electronic reconstructions in the nickelate films, albeit with different degrees of atomic displacement and charge transfer. Upon hydrogen doping, we observe a decrease in the average Ni e_g orbital polarization, distinguishing nickelates from cuprates. As the hydrogen concentration increases, the contribution of the itinerant interstitial *s* (IIS) orbital near the Fermi level gradually diminishes. Furthermore, applying compressive in-plane strain leads to a broadened Ni $3d_{x^2-y^2}$ bandwidth, consistent with enhanced nearest-neighbor hopping extracted from Wannier analysis. However, the influence of hydrogen doping and in-plane strain on the projected orbital character and total density of states is negligible. These findings provide insights into studying the physical properties of nickelates, contributing to the broader understanding of their superconducting mechanism.

Nanoscale Non-adiabatic Dynamics Simulation of Charge Generation in Organic Solar Cells

Filip Ivanovic*,Samuele Giannini, Wei-Tao Peng, *Jochen Blumberger*

*Department *of Physics and Astronomy, UCL* *e05-biosoft-**blu***

Organic solar cells (OSCs) have long been the subject of intensive research, as their easily processible constituent materials suggest a commercially viable alternative to their inorganic counterparts. In OSCs, excitation by light yields a tightly bound electron-hole pair, termed an exciton. Two-component cells, where two species are separated by an interface, constitute OSCs' greatest leap towards commercialisation, with their highest efficiencies exceeding 19%. Although such an interface is crucial for the dissociation of excitons into a sufficient yield of free charges, understanding the exact mechanism by which free charges are generated across the interface remains a formidable theoretical challenge, with contradictory results having been reported in the literature[1].

The complexity of such systems precludes a complete treatment by analytical theories, and instead requires first-principles quantum dynamics simulations, which must also be fast enough to access time scales comparable to experiment.

Here, we use an in-house non-adiabatic molecular dynamics package, termed X-SH[2], to simulate charge generation in an oligothiophene-perylene diimide interface, on experimentally relevant time and length scales. Our use of a DFT-parameterised Hamiltonian, which is updated on-the-fly, bypasses the need for explicit electronic structure calculations during the dynamics.

We elucidate the mechanism of charge generation in such an interface by modelling the excitons and charges with an explicit electronic wavefunction. This allows us to track their locations in real-time, on a microscopic scale that often cannot be resolved with photochemical experiments. We leverage the computational speed and flexibility of X-SH to identify key physical parameters affecting the efficiency of charge generation, and translate this into design rules to guide the synthesis of OSCs with yet higher efficiencies.

[1] A. Armin, W. Li, O. J. Sandberg, Z. Xiao, L. Ding, J. Nelson, D. Neher, K. Vandewal, S. Shoaee, T. Wang, H. Ade, T. Heumüller, C. Brabec, *P. Meredith*, *Adv. Energy Mater*. 2021, **11**, 2003570

[2] WT. Peng, D.Brey, S.Giannini, D.Dell'Angelo, I.Burghardt, *J.Blumberger, J. Phys. Chem. Lett.* 2022, **13**, 7105-7712

Modelling of *fcc* Ruthenium Surfaces and Particles with Hydrogen

Marietjie J. Ungerer*, Nora H. de Leeuw

*School of Chemistry, University of Leeds e05-react-lee

Ruthenium (Ru) is widely used in liquid organic hydrogen carrier frameworks for hydrogen generation and storage.[1] In bulk form, the predominant phase is hexagonal close-packed (*hcp*), but recent research[2,3] shows stable, highly reactive face-centred cubic (*fcc*) nanoparticles can also be produced. Experimental[4] and theoretical[5] studies indicate that in catalytic systems using *hcp* Ru nanoparticles, high concentrations of dissociated hydrogen block active sites, effectively poisoning surfaces. If the more active *fcc* phase is considered, the question remains whether hydrogen poisoning would be equally prevalent. A detailed analysis of hydrogen adsorption, binding, and reaction on the catalytic surface is essential.

Electronic structure techniques based on density functional theory (DFT) with long-range dispersion corrections [D3(BJ)] were used to calculate fundamental catalyst properties, specifically the *fcc* phase of Ru (001), (011), and (111) surfaces. This includes lattice parameters, surface energies, work functions, induced magnetization, electronic structures, chemical bonding, and electron transfer during adsorption of elemental hydrogen (H) and molecular hydrogen (H_2). Surface phase diagrams were generated using surface free energies and hydrogen chemical potential to assess temperature and pressure effects[6,7]. Results showed H_2 dissociation occurred readily, with single H adsorption energies between −0.4 and −0.6 eV.[8] Bader analysis indicated charge transfer from the surface to the adsorbate ranged from 0.17 to 0.27 e^-. The highest surface coverage was observed on Ru (001) and (011) at $\theta = 2.0$, followed by Ru (111) at $\theta = 1.0$. No molecular hydrogen (H_2) formation or surface poisoning was detected. Thermodynamic analysis demonstrated temperature significantly affected H surface coverage.[8]

[1.] K. Müller, et al*; International Journal of Hydrogen Energy 2016, 41, 22097–22103.* [2.] K. Kusada, et al*, Journal of the American Chemical Society 2013, 135, 5493–5496.* [3.] M. Zhao and Y. Xia*, Nature Reviews Materials 2020, 5, 440–459.* [4.] F. Rosowski, et al, *Applied Catalysis A: General 1997, 151, 443–460.* [5.] D.S. Rivera Rocabado, et al*; Catalysts 2022, 12, 331.* [6.] Ungerer, M.J.; De Leeuw, N.H.*, Catalysts 2022, 12, 1287.* [7.] Ungerer, M.J.; De Leeuw, N.H., *Nanomaterials 2023, 13(6), 1118.* [8.] Ungerer, M.J.; De Leeuw, N.H., *Physical Chemistry Chemical Physics 2025,27, 5759-5772.*

Computational investigation of the Fe_xRh_y alloy structures and the mechanism and selectivity of CO_2 hydrogenation

Shijia Sun*, Michael D. Higham, Igor Kowalec, C. Richard A. Catlow

*Chemistry, University College London e05-react-**cat**

CO_2 hydrogenation catalysts are being extensively developed for the synthesis of higher value products, including CO, methane, and oxygenates, such as methanol and ethanol. Rh monometallic catalysts can promote CO_2 hydrogenation to methane, while the addition of Fe to Rh catalysts can facilitate CO_2 conversion and the production of methanol and ethanol, at the expense of CO and methane. However, the mechanistic pathway for CO_2 hydrogenation to various products is still the subject of considerable discussion and debate due to a large number of surface species during the reaction process. Hence, it is essential to understand the key intermediates and elementary processes involved in CO_2 hydrogenation on Fe_xRh_y catalyst, which will help to design unique structures and active sites to regulate selectivity of CO_2 hydrogenation.

The Cluster Expansion (CE) model and the Metropolis Monte-Carlo (MMC) were used to determine the favourable composition of the Fe_xRh_y alloy, with a ratio of 1:2 for Fe/Rh, which is a face-centered cubic bulk structure. Then, the Fe-Rh neighbouring average and the Fe-Rh neighbouring RMSD were considered to obtain the most favourable atomic arrangement of $FeRh_2$ bulk structure, with the lowest mixing energy. The DFT calculations of the pathways of CO_2 hydrogenation were carried out on the (111) surface derived from the face centered cubic $FeRh_2$ bulk structure. DFT results show that the reverse water-gas shift (RWGS) reaction can occur via three different reaction pathways on the $FeRh_2$(111) facet. Furthermore, the alloy surface promotes the formation of HCO via HCOO dissociation or CO hydrogenation, and the resulting HCO species can interact with the CH_2 species to yield ethanol. Meanwhile, methane formation is suppressed by the competing pathways of HCO coupling with CH_2 and CH_2 hydrogenation. However, HCO formation is less likely to occur on the monometallic Rh surface as shown in our previous study, preventing ethanol production, although Rh sites are active for the C-C coupling steps.

REMatch plus SOS: Machine-learning-accelerated structure prediction for supported metal nanoclusters

Yunyu Zhang*, Keith T. Butler, Michael D. Higham, C. Richard A. Catlow

*Department of Chemistry, University College London e05-discov-cat

Predicting stable structures of nanoclusters is crucial yet computationally demanding. This study presents a machine learning-based methodology designed to accelerate the prediction of stable structures in nanoclusters. By integrating local environment descriptors, with dimensionality reduction, kernel-based similarity measure, and outlier detection, we efficiently screen and select promising configurations, thus accelerating identification of global and local minimum structures. The approach is validated through rigorous optimization, demonstrating its capability to identify low-energy structures while significantly reducing computational costs. This method offers a robust framework for structural screening.

Developing Next-Generation Catalysts for Iso-butanol Production Using Machine Learning

K. Agrawal*, R. L. Wingad, M. Sarwar, C. Buda, D. F. Wass, A. J. Logsdail

*Cardiff Catalysis Institute, Cardiff University, UK e05-react-**log**

The Guerbet reaction holds promise for biofuel production by converting small alcohols into fuel-grade branched-chain alcohols. However, it typically requires high temperatures (over 250 °C), making catalysts essential for cost-effective operation. Homogeneous catalysts with transition metals like Ir, Ru, and Mn have shown improved activity, especially in converting ethanol to C4 alcohols.[1] Yet, the detailed reaction mechanism remains unclear due to numerous possible pathways.[2] To address this, machine learning is being used to identify key features and trends, enabling the design of catalysts that efficiently convert ethanol to isobutanol with high selectivity.

A dataset of over 100 unique ligands with six different metals was compiled, and MACCS keys were used to define structural features for training. Principal Component Analysis (PCA) was applied to uncover clusters and geometric patterns. Additional electronic, constitutional, and experimental parameters were added to create a multi-fidelity feature space. This enriched dataset was then analysed using subgroup discovery (SGD)[3] to identify key descriptors influencing iso-butanol production.

The PCA shows that an amine group (-CH_2-NH-CH_2-) gives high iso-butanol selectivity ($S_{i\text{-butanol}}$ >95%). The SGD analysis further confirms the importance of the amine group, validating the PCA observation. The choice of base and the orbital energies of the ligand are also found to drive product selectivity. The study identifies key characteristic features of the catalyst and its components towards iso-butanol production. Insights gained from these models will help develop tailored catalysts towards this process, enabling cheaper and more efficient production of iso-butanol, paving the way towards Net Zero targets.

Acknowledgement

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Workflows for QM/MM Simulations of Metal Oxides

Oscar van Vuren*, Gabriel A. Bramley, Andrew J. Logsdail

*Cardiff Catalysis Institute, Cardiff University e05-algor-log

To effectively model materials, we must ensure that our models are rigorously constructed and accurately describe the physics of a problem. Achieving this goal whilst maintaining computational feasibility is challenging and has led to the development of embedding methods to reduce the cost of simulations whilst maintaining accuracy. One type of embedding method that sees use in the simulation of heterogeneous catalysis is the Quantum Mechanics/Molecular Mechanics (QM/MM) aperiodic embedded cluster method, where a region of chemical interest is simulated at a fully quantum level and the environment surrounding this QM region is simulated by a cheaper classical method.[1,2] The strengths of this method lie in the reduction in cost when compared to a periodic model, which allows for the use of more expensive density functional approximations as well as the aperiodicity removing spurious interactions between periodic images when simulating dilute limit phenomena such as defects.

These models are effective for catalytic reactions on a variety of materials but are complex and challenging to create in a reproducible and standardised manner. We have been engaged in the development of workflows that will enhance the ability of new researchers to use QM/MM methods to simulate novel and complex materials, using knowledge gained from simple systems to generate useful heuristics for future research building on previous work in this field.[3,4]

Having built these workflows, we have deployed them with the QM calculator FHI-aims[5] to calculate neutral and charged oxygen vacancy defect formation energies in bulk and surface MgO, as well as cationic substitution energy of alkali metals in bulk MgO. The electronic structures of these defects have also been investigated to ensure QM/MM models reproduce the electronics of current state of the art periodic models. These data have provided insight into the best methods for setting up and using QM/MM for complex defect simulations, including good heuristics for choice of QM region size and shape. We have also developed methods for the effective choice of MM region sizes, providing users with conservatively-chosen default values to ensure that new users can create effective QM/MM clusters. A major goal of these workflows is to ensure that flexibility is maintained such that researchers experienced in QM/MM methods can use them to increase efficiency without loss of access to all cluster construction settings.

These workflows are generally available to users of the code ChemShell[6] and are seeing use in the simulation of reactivity in zeolites as well as in the development of embedding methods for more complex materials, where we are constructing a model for ternary systems such as perovskites.

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Amide-Rich NaH as a Highly Active Catalyst for Ammonia Synthesis

Michael D. Higham*, Matthew S. Cummings, William I. F. David, C. Richard A. Catlow

*University College London, Department of Chemistry e05-react-**cat**

Ammonia synthesis is also one of the most environmentally challenging industrial processes, with ammonia production being responsible for an estimated 1.8% of global energy consumption. Recent experimental studies have revealed that the $NaNH_2$ catalyst is highly active for ammonia cracking, and may also be a highly active catalyst for ammonia production. It is believed that the active phase resembles a NH_2-rich sodium hydride phase. Hence, in the present work, DFT techniques are applied to model the proposed active phase, and investigate surface reaction processes to determine the likely reaction mechanism.

In the present work, plane-wave DFT calculations using the VASP code. In order to approximate the predominant active phase identified from the experimental results, a model was constructed based on a NaH(100) (2x2) surface facet with NH_2 amide species substituting a fraction of lattice hydride, in order to represent an amide-rich hydride phase. The PBE exchange-correlation functional with a D3 dispersion correction (with Becke-Johnson damping) was applied, along with a 600 eV plane-wave cut-off energy; k-point sampling with performed using a 6x6x1 Monkhorst-Pack grid. Geometric optimization was performed until forces were converged to within 0.01 eVÅ-1. Transition states were obtained using the Nudged Elastic Band (NEB method) and dimer methods.

The calculations reveal that surface hydride/amide vacancies can facilitate N_2 activation, with N_2 adsorbing exothermically with elongation of the N-N bond. Whilst hydrogenation of N_2 requires adjacent H/NH_2 vacancies, dissociation of intermediates such as $NHNH_2$ is facile. NH is readily hydrogenated to regenerate NH_2, the NH_2 species is highly stable, and its hydrogenation to yield ammonia is highly energy-demanding. The calculations demonstrate how H/NH_2 vacancies on the NaH(100) surface can facilitate ammonia synthesis via a Mars-van-Krevelen type mechanism.

Investigating the conductivity of Multi-Heme Cytochromes

A.M. Petho*, J. Blumberger

*Department of Physics and Astronomy, University College London

e05-biosoft-**blu**

Multi-heme cytochromes, a class of proteins with covalently bound heme cofactors, play a crucial part in electrochemical processes within bacteria living in anaerobic environments. The electronic conductivity of these proteins can be magnitudes larger than their non-heme counterparts. However, the mechanism supporting this increased conductivity is not well understood. The main objective of this project is to examine a set of multi-heme cytochrome-gold junctions to establish how the number of heme-cofactors, and certain other properties affect the conductivity. The contribution presented here shows the main results from six systems: current-voltage (I-V) curves along with supporting data. This shows the effects of junction height, protein orientation, and number of contacts on the currents, with each causing up to a magnitude difference for a given voltage. The data we present shows great potential in advancing our knowledge of these fascinating compounds as well as highlighting their potential for the field of bioelectronics.

Computational prediction of Cd2Sb2O7 as a candidate TCO

Peter P. Russell,[1] Romain Claes,[1] Ke Li,[2] Alexander G. Squires,[1] and David O. Scanlon[1]

1 School of Chemistry, University of Birmingham, Edgbaston, Birmingham, B15 2TT

2 Department of Chemistry, University College London, 20 Gordon Street, London, WC1H 0AJ

*e05-bulk-**dos***

Transparent conducting oxides (TCOs) are central to the field of modern optoelectronics, displaying a unique combination of high electrical conductivity and optical transparency, with applications in touch screens, photovoltaics, and more. [1] The industry standard is Sn-doped In_2O_3 (ITO), which displays optical transmission over 90%, with low resistivity and high carrier mobility. However, indium is expensive and has low earth-abundance. Alternative materials that offer comparable properties using different elements are in high demand. [2,3]

Recently, Sb(V) oxides have emerged as promising candidates. [4,5] Sb(V) possesses the $(n-1)d^{10}ns^0\ np^0$ electron configuration common to many TCO cations, including In(III) in In_2O_3, leading to a highly disperse conduction band and a large band gap.[5] The Sb(V) oxides also display three-dimensional connectivity of SbO6 octahedra, creating electron pathways that enable high conductivity. $ZnSb_2O_6$ [4] was computationally predicted and experimentally realised as a TCO, and Sb_2O_5 [5] is predicted to be suitable as well, motivating further exploration of this family of materials.

$Cd_2Sb_2O_7$ contains Sb(V) and was flagged in a high-throughput study as having a relatively large band gap and low electron effective mass, suggesting possible transparency and conductivity. [6] However, there has been no investigation into the material's defect chemistry. For these reasons it was decided to investigate $Cd_2Sb_2O_7$ more thoroughly.

Our results suggest an optical band gap in the transparent range, a high intrinsic electron carrier concentration due to antisite defects, giving high intrinsic mobility and conductivity, a large doping window, and favourable n-type doping using yttrium to further enhance conductivity. This provides further evidence for the potential of Sb(V) oxides as TCOs.

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Investigating Electron Localisation in Defective Bulk Ceria with Dispersion Corrected DFT

Thomas Hill* [a], Igor Kowalec [a], Charles R. A. Catlow [a, b, c]

[a] School of Chemistry, Cardiff University, [b] Department of Chemistry, University College London, [c] Catalysis Hub, Research Complex Harwell

*e05-react-**cat***

Dispersion corrections are widely used in surface studies of ceria but remain uncommon for bulk investigations.[1] We examined the impact of Grimme's D3 dispersion correction on defective bulk ceria, comparing PBE + U (5 eV) with and without D3, and benchmarking against the hybrid PBE0 functional (25% Fock exchange).

The inclusion of D3 with PBE + U = 5 eV led to a 1.4% increase in lattice parameter (0.5% lower than the uncorrected case and 0.5% higher than PBE0) compared to the experimental 0 K value of 5.394 Å.[2] Vacancy formation energies also increased with D3, giving 3.07 eV in a 96-atom cell (11% higher than without D3). 3.69 eV was obtained for the primitive cell, matching PBE0 but still below the experimental value of 4.2 ± 0.2 eV.[1] Energy differences across varying CeO_{2-x} compositions showed that D3 significantly affects systems with lower x, suggesting long-range vacancy interactions are absent in PBE + U.

Full relaxation of varying CeO_{2-x} compositions showed increase in cell size in defective ceria. Higher concentrations (larger x) resulted in more expansion of the cell. Results obtained for the expansion coefficient for x against the lattice parameter were 15% lower than experiment.[3] Likely differences in temperature (0K in this study 800K experimentally) were responsible for the large discrepancy.

Electron localization favoured configurations close to the vacancy but avoided adjacent Ce^{3+} sites. Nearest-neighbour pairs (NN) of Ce^{3+} increased energy due to repulsion, while distant pairs also incurred energy penalties. The lowest energy configuration (3.00 eV) was a nearest-neighbour next-nearest-neighbour (NN*) arrangement, where the second Ce^{3+} is near the vacancy but not adjacent to the first. The electron density favouring sites near the vacancy give reason to the reactivity of vacancy sites in ceria.

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Au and Au_3Cu nanoclusters as catalysts for light-driven CO oxidation

Igor Kowalec*, Thomas Hill, C. Richard A. Catlow

[a] Cardiff Catalysis Institute, School of Chemistry, Cardiff University, Cardiff, CF24 4HQ, United Kingdom

*e05-react-**cat***

Plasmonic nanomaterials hold promise for advancing clean energy, environmental remediation, and sustainable chemical production through efficient, light-driven catalysis. [1] In plasmonic catalytic processes, after the Localized Surface Plasmon Resonance (LSPR) is triggered, the non-radiative decay of the plasmon results in the generation of energetic “hot” electrons and holes, which can be transferred to adsorbed molecules, facilitating catalytic processes on the surface of the nanoparticles (NPs). This study explores gold and gold-copper alloy NPs as plasmonic catalysts for low-temperature CO oxidation, a reaction critical for air purification in closed environments such as the International Space Station. [2]

Using CAM-B3LYP in DFT structural optimization and in the Tamm-Dancoff approximation for excited-state analysis, we investigate the adsorption and electronic behaviour of CO, CO_2, O, and O_2 on spherical Au_{55} and cubic Au_{63} NPs. The results reveal shape-dependent charge localization upon excitation, in alignment with larger-scale simulations. [3] Complementary experimental work uses an Au_3Cu/TiO_2 catalyst characterized by TEM-EDX, guiding a machine-learned force field (MACE-MP) screening of 3:1 Au:Cu alloy nanostructures (Figure 1) for further adsorption studies, laying foundations for rational plasmonic catalyst design.

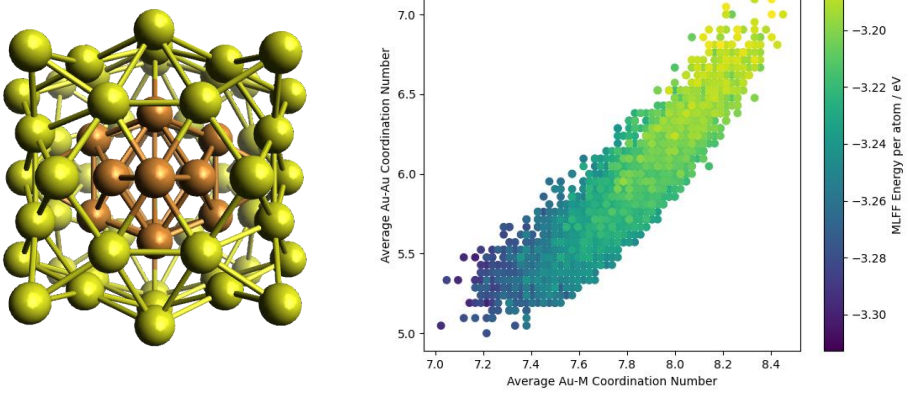

Figure 1. A 63-atom Au_3Cu NP and MLFF configuration screening of Au_3Cu 55-atom alloy NPs.

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Hybrid QM/MM and Machine Learning for Zeolite Catalysts and Silica Polymorphism

Jamal Abdul Nasir*, Alexey A. Sokol, Jingcheng Guan, You Lu, Thomas W. Keal, Andrew M. Beale, Alin Marin Elena, C. Richard A. Catlow

*Department of Chemistry, University College London e05-react-sok

Zeolites and silica polymorphs are central to a range of industrial applications, from emissions abatement to high-pressure structural materials. Here, we present a study employing hybrid QM/MM calculations and machine-learned interatomic potentials (ML-IP) to address silica and zeolite-based chemistry.

First, we investigate the NH_3-SCR mechanism over Cu-chabazite (CHA) [1] and Fe-BEA [2] zeolites. Our hybrid QM/MM simulations [3], supported by DRIFTS experiments with modulation excitation spectroscopy, reveal that water- and ammonia-solvated Cu sites promote the formation of key nitrate intermediates, while also inhibiting reduction steps in the catalytic cycle. Additionally, we extend this approach to Fe-BEA, finding a more exothermic formation of crucial intermediates when framework Fe^{3+}/Fe^{2+} is present, indicating enhanced redox properties compared to Al-based frameworks. The synergy in bimetallic Cu–Fe systems offers further insight into optimizing zeolite-based catalysts for efficient NO_x reduction. In parallel, we apply the MACE-MP-0 machine-learned potential to model the framework energetics of siliceous zeolites and explore high-pressure phase transitions in silica [4]. Unlike classical potentials that require re-parameterization, the MACE approach accurately describes structures with varying coordination of Si. Our simulations reproduce the metastability of microporous phases relative to α-quartz and capture the compression behaviour of quartz, coesite, and stishovite. The predicted transition pressures (~3.5 GPa for quartz→coesite and ~9 GPa for coesite→stishovite) align closely with the experiment, demonstrating the reliability and versatility of this ML-based methodology.

Together, we show how multi-scale computational tools can provide robust insight into both the catalytic and structural properties of silica-based materials, enabling the design of more effective zeolite catalysts and enhancing our understanding of silica polymorph stability under extreme conditions.

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Investigating Hydrogen Interaction with Defective and Doped MgB_2 via Density Functional Theory

Navaratnarajah Kuganathan*, Martin Dornheim, David M. Grant and Sanliang Ling

*Advanced Materials Research Group, University of Nottingham e05-bulk-lin

The $LiBH_4$ + MgH_2 system holds considerable promise for solid-state hydrogen storage; however, the slow rehydrogenation of MgB_2 remains a key limitation [1,2]. In this study, we employ density functional theory (DFT) simulations to examine the energetics of hydrogen in pure, defective, and doped MgB_2 in equilibrium with molecular hydrogen. Our results identify the B-Frenkel defect as the most thermodynamically favorable defect mechanism in MgB_2 [3]. The computed hydrogen solution energy, which includes both defect formation and the incorporation of hydrogen into the lattice, indicates limited hydrogen solubility at moderate temperatures, particularly at interstitial sites [3]. However, hydrogen incorporation is significantly enhanced in the presence of a pre-existing boron vacancy, especially when oxygen is doped onto the B site [3]. Furthermore, lithium doping at the Mg site facilitates hydrogen uptake through the formation of strong Li–H bonds, suggesting reduced activation barriers for hydrogen dissociation and diffusion. Both oxygen and fluorine dopants at the B site also promote increased hydrogen solubility [3]. These findings provide valuable insights into defect and doping strategies to improve the hydrogen storage performance of the $LiBH_4$ + MgH_2 system, supporting ongoing efforts to optimize its practical application.

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First-principles modelling of infrared and Raman spectra

J. M. Skelton*, C. Umeh, A. R. Pallipurath

*Department of Chemistry, University of Manchester e05-algor-**ske**

In this contribution, we will present a recent update to our open-source Phonopy-Spectroscopy code to implement a comprehensive workflow for predicting infrared (IR) and Raman spectra using density-functional theory (DFT).

Phonon frequencies and eigenvectors are obtained within the harmonic approximation. IR spectra are modelled by computing the mode dipole oscillator strengths and infrared dielectric function, providing access to absorption, reflectance and transmission spectra. Raman spectra are obtained by computing the polarizability derivatives numerically from either the high-frequency dielectric constant or the energy-dependent dielectric function, the latter of which allows for partial inclusion of resonance effects. Finally, linewidths can optionally be obtained from perturbative three-phonon interactions.

The code can perform IR and Raman simulations on both single crystals and powders, accounting for the orientation of the crystal and the polarisation of the incident and detected radiation. This allows for the simulation of a wide variety of measurements, ranging from routine laboratory characterization to the more sophisticated experiments possible with high-end or custom instrumentation.

Simulations can be performed through a command-line interface or with a Python API for more flexibility. The code has also been designed in such a way as to “decouple” the preparatory DFT calculations and spectrum modelling, to better facilitate collaboration between theory and experimental groups.

We have benchmarked the code against single-crystal IR and Raman measurements on orthorhombic SnS and SnSe. Simulated IR reflectance spectra and dielectric functions, obtained at very modest computational cost, are found to agree well with experiments. While the Raman-active modes and polarisation behaviour are correctly predicted, the narrow electronic bandgaps of SnS and SnSe relative to typical Raman laser wavelengths mean that careful modelling of the dielectric functions is required to reproduce the band intensities.

The Crystal Isometry Principle infers chemistry from geometry

Vitaliy A. Kurlin*, Daniel E. Widdowson

*Materials Innovation Factory, University of Liverpool, UK e05-algor-abc

Structures of solid crystalline materials (periodic crystals) are determined in a rigid form and hence keep all their properties under rigid motion within the same ambient environment. However, crystal structures that have different rigid shapes can substantially differ by properties and hence should be reliably distinguished, for example, polymorphs with different solubility. Conventional representations based on reduced cells discontinuously change under almost any perturbation, which led to the accumulation of near-duplicates in major databases of experimental structures [1].

This ambiguity was resolved by generically complete and continuous invariants that distinguish all non-duplicate periodic crystals in major databases within a few minutes on a modest desktop [2]. Now any dataset of experimental or simulated crystals can be visualized on maps with continuous and analytically defined invariant coordinates [3]. These invariants can be inverted to any generic periodic structure in 3 dimensions, uniquely under any distance-preserving transformation.

Inspired by Richard Feynman's hint in Fig.1-7 of his first lecture on physics, the resulting Crystal Isometry Principle says that any real periodic material is uniquely determined by precise enough geometry of only atomic centers without chemical elements, under the same ambient conditions such as room temperature and normal pressure.

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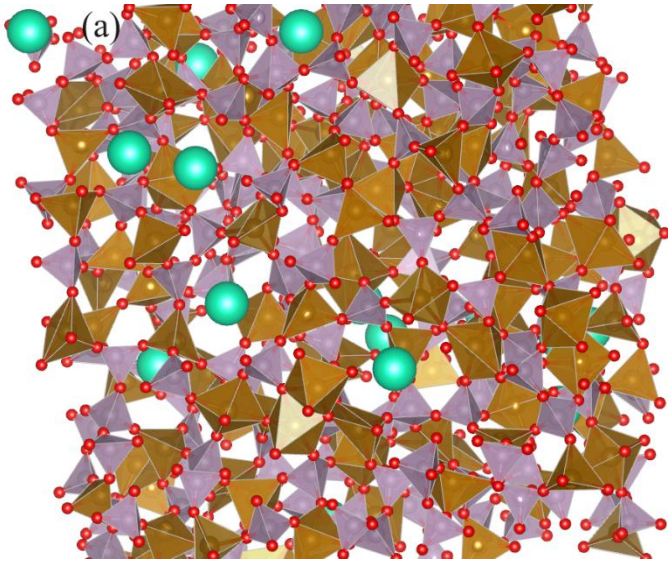
The effect of impurities and irradiation on the glass network – Cs_2O-loaded iron phosphate case study

Cillian Cockrell*, Kitheri Joseph, Maulik Patel, Robin Grimes, Kostya Trachenko

*Nuclear Futures Institute, Bangor University *e05-enviro-**kos***

The use of iron phosphate glass as a wasteform is contingent both on its response to the addition of waste products and on its evolution in response to radiation emitted by these waste products. Both of these factors present significant alterations to the glass network and therefore our understanding of the resulting glassy state. We perform molecular dynamics simulations to study the effect of caesium, a nuclear decay product, on the glass network of a vitreous wasteform, both in pristine glass and after high-energy nuclear recoil cascades. Specifically, we simulate overlapping 70 keV cascades and examine the structural and topological effects that caesium has on the iron phosphate glasses before and after these cascades. We find that the glass network is substantially altered by the presence of caesium as a potent network modifier (though we hesitate to assign to it this label) and that radiation cascades produce qualitatively different effects from those in pure iron phosphate glasses. Overlapping cascades produce minimal effects on the mobility of caesium at low loading – radiation damage is much more severe in pure iron phosphate glasses. At higher loading, the glass network accommodates caesium atoms less well, particularly after irradiation. We explain this in terms of caesium's role as an “excluded” network modifier in comparison to iron, which is tightly incorporated into the glass network, as can be seen in Fig. (a). The network-disrupting effect of the caesium anticipates the similar effect of radiation damage, causing the microscopic structure to resist alterations from radiation cascades. However, at large enough concentrations, the space demanded by caesium atoms may cause them to aggregate and be expelled from the glass phase entirely.



Atomic-Scale Insights into Passivation and Halide Mixing in 3D and 2D Halide Perovskites

Vikram*, M. Saiful Islam

*Department of Materials, University of Oxford e05-nano-isl

Some of the key factors for achieving high efficiency and long-term operational stability in metal-halide perovskite optoelectronic devices require precise control over film growth and effective defect passivation strategies. Recently, amino-silane-treated perovskite solar cells have demonstrated exceptional stability.[1] However, the atomic-scale interactions of these silane molecules at perovskite surfaces remain poorly understood. Separately, to enhance efficiency in quasi-2D perovskites, a recent study achieved vertical crystallization by using methylammonium chloride (MACl) additives during precursor processing.[2] However, the absence of detectable I/Cl halide mixing in 3D perovskites raise fundamental questions about such halide mixing in 2D perovskites.

Our density functional theory and ab initio molecular dynamics simulations reveal atomic-scale insights into these performance improvements, shedding light on the mechanisms of silane passivation in 3D perovskites and halide mixing in 2D perovskites. These findings align with the experimental results and offer a deeper structural and mechanistic understanding at the atomic level.

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Calculating system properties on-the-fly in DL_POLY 5

H. L. Devereux*, C Cockrell, AM Elena, Ian Bush, Aidan BG Chalk, Jim Madge, Ivan Scivetti, JS Wilkins, IT Todorov, W Smith, K Trachenko

**School of Physical and Chemical Sciences, Queen Mary University of London, Mile End Road, London, E1 4NS, UK*

*e05-algor-**kos***

Performing calculations on-the-fly can confer performance advantages by eliminating the need to save data or perform input/output operations at runtime. Such algorithms operate by sequentially updating their state as new data arrive. This paradigm, also termed “online algorithms”, is ubiquitous in the analysis of big data streams. In molecular dynamics the system trajectory, mapping atomic configurations through simulation time, is an example of such a data stream where analysis after the fact remains a common paradigm. Such an approach in large scale systems, e.g. reaching 1 Billion atoms (length scales of 1 μm) is infeasible.

By casting standard analysis of system trajectories as online algorithms, we can eliminate trajectory storage. In DL_POLY, we have implemented a general framework for calculating arbitrary correlation functions of simulation data on-the-fly with no trajectory storage. We support commonly used system properties such as atom and rigid body velocities, rigid body orientational velocities, stress, heat flux, as well as k-space currents, densities and stresses. Using these we are able to calculate for example the viscosity and thermal-conductivity, elastic constants, and velocity auto-correlation functions.

Our implementation in DL_POLY is designed to be highly extensible to new simulation properties, and allowing a user to combine any components of supported correlatable quantities into user defined correlation in the CONTROL file.

Atomistic modelling of SiO_2/Ta interfaces

Margherita Buraschi*, Alexander Shluger

*Department of Physics and Astronomy, University College London e05-surfin-**shl***

In recent years, resistive random-access memory (ReRAM) devices have emerged as promising candidates for advanced memory technologies [1]. These devices offer high scalability, high endurance and fast programming speed [1]. Typically, ReRAMs consist of a wide-gap metal oxide sandwiched between two metallic electrodes. Application of a small voltage induces the formation and destruction of conductive oxygen vacancy filaments in the oxide, enabling reversible switching between high and low resistive states [1]. As oxygen vacancies are central to the working principle of ReRAM devices, a complete understanding of the formation of such defects at the metal/oxide interface is essential. In this project, we investigate the interface between silicon dioxide (SiO_2) and a tantalum (Ta) film. Tantalum (and its sub-oxides) have shown a strong ability to promote the formation of oxygen vacancies at the metal/oxide interface by extracting oxygen from the oxide [2]. Indeed, thermodynamic analysis show that the formation of interstitial oxygen is energetically more favorable in Ta than in SiO_2. To better understand this exchange, we examine various defects at the SiO_2/Ta interface. Models consisting of Ta_n clusters of various sizes on SiO_2 surfaces are also studied. Beyond oxygen exchange, observations suggest the formation of silicides at the SiO_2/Ta interface, a phenomenon previously noted in other systems, such as HfO_2/SiO_2 and ZrO_2/SiO_2 [3].

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Elucidating the effect of doping on the mechanical and chemical stability of hydroxyapatite

Mahdi Tavakol*, Jin-Chong Tan, Alexander M. Korsunsky

*Department of Engineering Science, University of Oxford e05-biosoft-**tan***

Hydroxyapatite (HAp) as the primary mineral components of many mineralized tissues performs various critical roles most important of which are load transfer and structural support. For the effective support of these roles, various chemical and mechanical properties are required which has changed with human evolution. For the human teeth which is the focus of the current study, the importance of the chemical stability superseded the mechanical strength with the change in the human food source from the hard nuts and meat to the prevalent fast food which creates an acidic environment in the oral cavity. To reinforce the shift in the significance of the chemical stability to the mechanical one, various strategies have been suggested with the ionic doping being the most important investigated in the current study.

Here, the effect of doping with various concentrations of Magnesium, Fluoride and Carbonate ions was investigated through molecular dynamics to identify the most effective candidate for tailoring the required chemical and mechanical stability balance. Mechanical stability was studied through the uniaxial compression test *de novo* while the Thermodynamic Integration (TI) was opted for the chemical stability. The MD compression results revealed a deterioration in mechanical stability with the increase in the Mg^{2+} ion concentration, while the other two candidates showed minimal impact on this parameter. Similarly, doping with F^- and CO_3^{2-} ions didn't have any noticeable effect on either the chemical stability or the free energy of the surface. However, both the free energy and chemical stability parameters obtained through TI revealed a favorable change with Mg^{2+} substitution. The latter was more favorable than the former which implies that the water solvation decreases the favorable role of Mg even though it does not completely cancel it out. Notably, inside the HAp a higher chemical stability is expected in the absence of the solvation layer. The kinetics of doping was studied through a combination of Steered Molecular Dynamics (SMD) and conventional MD simulation. SMD used for the first stage of doping, ion removal, showed that passive doping is just possible in the surface in line with the findings from conventional MD simulations.

DFT & Data Mining Assisted Catalyst Discovery of Cu-based Alloys for CO_2 Hydrogenation to Alcohols

Z. Lu*, I. Kowalec, H. I. Rivera Arrieta, C. R. A. Catlow, A. Logsdail

*Cardiff Catalysis Institute, School of Chemistry, Cardiff University, Cardiff, CF24 4HQ, UK

Direct methanol synthesis from CO_2 is important to deliver a circular economy,[1] for which Cu-based catalysts are promising candidates.[2] Some Cu-based catalysts can achieve high methanol selectivity, but the inability of maintaining adequate CO_2 conversion rate and stability still hinders their industrial application.[2] One mechanistic bottleneck here is the slow reaction of formate, HCOO, to subsequent intermediates.[3] Cu-based alloys possess unique electronic properties which can introduce reactivity, such as C-O scission ability, leading to easier conversion of HCOO. Density functional theory (DFT) studies on dilute alloy models, such as single-atom alloys (SAAs), have been greatly helpful in understanding alloying effects in catalysis.[4] Data-mining methods, such as subgroup discovery (SGD)[5], can help to identify local patterns of outstanding materials. Here, DFT and SGD were used to investigate four elementary reactions of formate on a series of Cu SAAs. The traits of alloys facilitating each reaction were captured, and analyses on their physical meaning showed electron donating and oxygen affinity are key properties determining the reactivity of alloy catalysts. Extrapolation to further alloy materials were performed based on these findings. Promising alloy catalysts, such as CuSc alloy, were proposed for further investigation.

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Suppressing phase transformation and cation migration with Mg and Si doping in Fe–Mn layered oxides for sodium-ion cathodes

Arup Chakraborty and Saiful Islam

*Department of Materials, University of Oxford, Parks Road, Oxford, UK

e05-power-**isl**

Na-ion batteries have received considerable attention for large-scale energy storage due to the abundance of sodium. The Fe-Mn-based layered oxide cathode material, $Na_xFe_{0.5}Mn_{0.5}O_2$, is a promising cathode because of inexpensive elements such as Fe. However, this system suffers from a structural phase transformation (P2 to O2) at high charge states and irreversible cation migration, leading to fast capacity fading with cycling. In this study, we investigate whether doping into $Na_xFe_{0.5}Mn_{0.5}O_2$ can effectively suppress this phase transformation and cation migration, focusing on the atomic-scale effects of Mg^{2+} and Si^{4+} substitution, using ab initio simulation techniques. [1] These dopants are contrasting species in terms of bonding character from divalent ionic (Mg) to tetravalent covalent (Si). Our study indicates that Mg-doping delayed Fe migration into the Na-layers in line with the experiment. In contrast, Si-doping stabilised the P2 phase over the entire charging range and suppresses Fe migration with no O–O dimer formation, suggesting that the Si-doped system should be a promising Na-ion cathode material.

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Interlayer Metals for Zero-Excess Li Metal Batteries: A Multiscale Approach Combining Machine Learning Potentials and DFT

Neubi F. Xavier Jr.*, Qiong Cai

* *School of Chemistry & Chemical Eng., University of Surrey, UK* *e05-power-cai*

The concept of anode-free or zero-excess Li metal batteries (ZELMBs), has attracted a lot of interest as it simplifies the battery manufacturing process, improves safety, and could bring significant cost reduction. However, the Li stripping/plating occurring on the surface of the commonly adopted Cu current collector, are one of the main disadvantages of ZELMBs [1]. In this work, we present an efficient computational workflow to screen the performance of interlayer metals at the anode side, aiming to obtain stable deposition/stripping process of Li during operation. We performed density functional theory (DFT) calculations to investigate Li deposition on 19 interlayer metals, including those forming solid solutions with Li (Ca, Mg, Zn, Al, Ag, Au, Pt, Pb, Tl, In, Y, Tm, Sb, Pd), those forming intermetallic alloys with Li (Sn, Si, Ge), and those non-alloying with Li (Cu, Ni). We evaluated the adsorption energy (E_{ads}), substitutional energy (E_{subst}), and Li diffusion barriers. It was observed that formation of an alloy layer increased the binding strength of Li on Ca, Mg, Tl, Zn, and Sb. Conversely, lithiated sites in the Al, Sb, Ag, Au, and Pt alloys surfaces reduced the energy barrier for Li diffusion. We derived a relationship between the Li deposition overpotential and diffusion barriers revealing that Mg–Li alloys with low Li content (<11.25 at. % Li), as well as Li–In, Li–Tl, Li–Pb, Li–Tm, and Li–Y alloys with high Li atomic concentration (>98 at. Li), offer the best performance for efficient Li deposition. We conducted large-scale molecular dynamics simulations using machine learning interatomic potentials (MLIPs), fine-tuned with DFT-based data, for the Li-Cu, Li-Zn, Li-Mg, and Li-Bi systems, considering varying amounts of Li metal, and we studied the crystallization and alloying of Li on those different interfaces. For the Li-Cu interface, we observed that with low amounts of Li metal, Li aggregates are formed, leaving empty spaces on the Cu surface. Solid solution formation was observed at the Li-Zn and Li-Mg interfaces, with a larger presence of hcp lattice of Li in the former and bcc lattice of Li in the latter. Finally, we predicted that Li preferentially forms Li_3Bi intermetallic alloys at the Li-Bi interface, based on the morphology and coordination analysis.

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Localised electron states in Amorphous Alumina

J. Strand*, A. Shluger*

*Department of Physics and Astronomy, UCL e05-bulk-**shl**

Electron traps significantly impact the performance and reliability of oxides in nanoscale technologies. They contribute to current leakage via trap-assisted tunneling and may degrade the oxide by lowering activation energies for oxygen vacancy formation, potentially leading to dielectric breakdown. In this work, density functional theory is used to investigate electron trapping defects in amorphous alumina models and finds that electrons can self-trap with trapping energies up to 1.0 eV, similar to behavior in other amorphous oxides. Traps localise either on single Al ions or form "two-center" states, where electrons are shared between closely spaced Al ions in a bond-like manner. Most traps occur on low-coordinated Al sites. We also examine localised electron states in oxygen-deficient alumina, using two methods to introduce vacancies: (1) the *pick-and-relax* method, where an oxygen atom is removed from a stoichiometric model followed by structural relaxation; and (2) the *in-melt* method, involving quenching a substoichiometric melt from high temperature via ab initio molecular dynamics. Vacancies produced from the melt exhibit behavior similar to self-trapped electrons, with electrons shared between Al ions, frequently trapping on 3-coordinated Al sites. We also used the in-melt method positively charged cells, analogous to creating +2 charged oxygen vacancies. In these cases, there is initially no distinct point-defect (‘global’ vacancy). However, a 'local' vacancy emerges upon neutralization and perturbation of atomic positions, which enables the formation of Al–Al bonds.

The results show that electrons can self-trap in amorphous alumina, impacting the reliability of alumina-based dielectrics. Vacancy states are qualitatively similar to self-trapped electrons, and the ability of vacancies to switch between global and local character highlights the complex nature of oxygen deficiency in amorphous oxides, with implications for their use in nanoscale electronic devices.

PSDI Thematic Portals

Abraham Nieva de la Hidalga,* Arif Mehmood, Arif Mehmood, Richard Catlow

*UK Catalysis Hub, Cardiff University *e05-discov-**cat***

The Physical Sciences Data Infrastructure (PSDI) is set to become the largest UK data infrastructure for the physical sciences, supporting diverse research communities in accessing and sharing data resources. Thematic portals are designed to provide customized access points, enabling communities to showcase the research outputs generated using PSDI's resources and services. The database, services, code, and deployed portal demonstrate the benefits of community portals, highlighting their adaptability and ease of customization and deployment for diverse research communities.

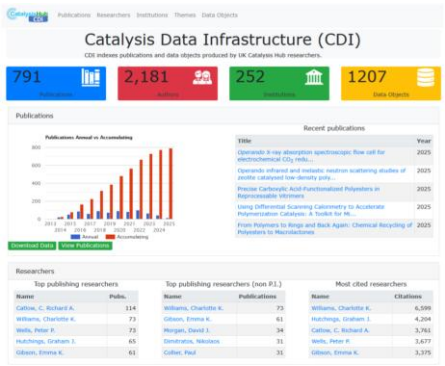

Figure 1 Current view of the UK Catalysis Hub CDI

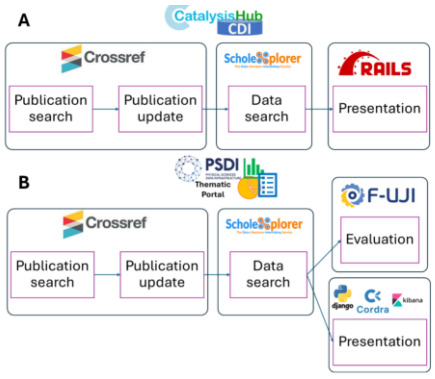

Figure 2 Comparisons of the applications supporting the Catalysis Data Infrastructure (A) and the ones used to integrate PSDI thematic portals (B).

In this talk we explain how the UK Catalysis Hub Catalysis Data Infrastructure (CDI) application has been adapted to serve as the basis for thematic portals. The CDI was designed to catalogue references to research outputs, maintains links between them and promotes publishing and sharing of data. The proposal is to create persistent relationships between the different types of data and publications while also monitoring how well they comply with FAIR data principles (findability, accessibility, interoperability, and reuse). After introducing the CDI, we will discuss and demonstrate how this tool could be customised to support other research communities, using the MCC publications database as a working example. The main features we highlight are: Low maintenance (most tasks are automated); Cheap setup (automated deployment, easy configuration); Try before you acquire (test deployment for evaluation); and Support/guidance for further customization.

Standby Talks

Mechanism of CO_2 Reduction to Methanol with H_2 on an Iron(II)-scorpionate Catalyst

Chengxu Zhu*, Carmine D'Agostino, Sam P. de Visser

*Manchester Institute of Biotechnology, The University of Manchester

e05-react-**que**

CO_2 utilization is an important process in the chemical industry with great environmental power. In this work we show how CO_2 and H_2 can be reacted to form methanol on an iron(II) center and highlight the bottlenecks for the reaction and what structural features of the catalyst are essential for efficient turnover. The calculations predict the reactions to proceed through three successive reaction cycles that start with heterolytic cleavage of H_2 followed by sequential hydride and proton transfer processes. The H_2 splitting process is an endergonic process and hence high pressures will be needed to overcome this step and trigger the hydrogenation reaction. Moreover, H_2 cleavage into a hydride and proton requires a metal to bind hydride and a nearby source to bind the proton, such as an amide or pyrazolyl group, which the scorpionate ligand used here facilitates. As such the computations highlight the non-innocence of the ligand scaffold through proton shuttle from H_2 to substrate as an important step in the reaction mechanism.

Posters

Design and development of novel composite electrodes for high-energy density supercapacitors

Abeer A. Hdadai*, Shayan Seyedin and James A. Dawson

*Chemistry – School of Natural and Environmental Sciences and School of Engineering, Newcastle University

e05-power-**jad**

A comprehensive understanding of the charge and discharge mechanisms in supercapacitors is critical for enhancing the efficiency of electrode materials. To achieve this, the integration of experimental techniques with advanced simulation methods has proven highly effective.

This poster presents the early-stage development of a $NiCo_2S_4$/reduced graphene oxide (rGO) electrode. The bulk and surface structures of the individual components are initially examined to assess their intrinsic properties and formation energies. Subsequently, a composite $NiCo_2S_4$/rGO structure is constructed and analyzed to investigate interfacial interactions and structural compatibility between the constituents.

Density functional theory calculations are employed to identify the most energetically favorable configurations, analyze charge distribution, and assess the overall stability of the hybrid system in the latest stage of the project. Furthermore, the influence of rGO incorporation on the electronic conductivity and surface reactivity of $NiCo_2S_4$ is explored in detail.

Structural data are obtained through atomistic modeling using VASP on the ARCHER2 supercomputing platform. Ultimately, this study aims to determine the optimal configuration for maximizing the performance of the proposed electrode material.

A Computational Study of CO_2 Methanation over Low-index Nickel Surfaces

M. Alotaibi*, M. Higham, S. Sun, J. Abdul Nasir, A. Sokol and C.R. Catlow

*Department of Chemistry, University College London e05-react-**cat**

Carbon dioxide (CO_2) emissions are widely accepted as a significant risk for climate change. Technologies like CO_2 methanation are economically and environmentally beneficial as they provide a means to produce combustible liquid fuels that do not rely on fossil fuel extraction. Not only can we potentially remove CO_2 from the atmosphere, but also burn less oil and gas in the first place, contributing to "closing the loop" and providing a synthetic carbon cycle, paving the way toward a circular economy for fuel production. To mitigate this emission, nickel-based catalysts are considered an effective solution for converting CO_2 into methane, benefiting industrial applications such as power–to–gas (PtG) technology. In addition to its efficiency in the CO_2 methanation reaction, nickel metal is a low–cost industrial alternative to noble metals such as Rh and Ru. Since nickel-based catalysts present challenges, such as the sintering of nickel particles on supported catalysts, further research is necessary. To understand the precise role of support materials (whether to inhibit sintering, maximise Ni surface area, or provide active sites at the metal/support interface, etc.), we must first understand the reaction mechanism that proceeds on unsupported Ni surfaces. In this study, plane–wave DFT calculations, as implemented within the Vienna Ab initio Simulation Package (VASP), were performed to investigate CO_2 methanation on low-index nickel surfaces, namely Ni(111), Ni(110), and Ni(100). Surface energies for each low–index face were calculated, and it was found that the Ni(111) surface is the most stable. In addition, the adsorption of CO_2, H_2, CO, and O on the surface slab models was examined to obtain adsorption energies and explore CO_2 activation on the Ni surfaces. Subsequently, CO_2 and H_2 dissociation on Ni(111) surface was investigated, which are key initial elementary processes associated with CO_2 methanation and for co–adsorbed intermediate species. Furthermore, transition state searches using the Nudged Elastic Band (NEB) and dimer techniques were performed to obtain activation barriers for the key elementary processes. Therefore, this work is a prerequisite for future studies investigating CO_2 methanation processes on model-supported Ni particles. Comparing the calculated results, such as activation barriers for key processes and adsorption energies of key reactants between supported and unsupported Ni systems, provides deeper insights into the role of the support, facilitating more rational catalyst design.

Catalytic Partial Oxidation of Methane using an Yttria-Stabilized Zirconia Catalyst

Erze Gao*

*Chemistry, UCL e05-react-**cat***

This project is about discovering the catalytic cycle for Catalytic Partial Oxidation of Methane (CPOM) via using Yttria-Stabilized Zirconia (YSZ). The partial oxidation of methane is an important industrial process for upgrading natural gas into syngas, a mixture of CO and H_2 which is widely used in the industrial field. The reaction of CPOM occurs on the (111) plane of YSZ, in which the methane is physisorbed and oxidized into formaldehyde, before decomposing into syngas and other by-products. The surface model in this project is large symmetric slab model, which consists of 4*4 supercell with 96 ZrO_2 formula units. The thickness, width and vacuum separation of the slab are respectively 16.36Å, 14.6 Å and ~15Å. The concentration of Y_2O_3 is 14.3 mol% on the surface, which fits the experimental observation of 10-14 mol%.

The surface was firstly pre-oxidized via the removal of one lattice oxygen and absorption of the oxygen molecule in the atmosphere. Thus, the pre-oxidized 'YSZ+O' model was prepared for further reaction. Then the research discussed the process of methane absorption and oxidation into surface formaldehyde on the YSZ, with various routes and reactions being analyzed. To be specific, the results mainly indicated a possible method which led to the decomposition of surface formaldehyde through H-Zr interaction. Proper structures were discovered with lower potential energy compared with the initial structure, which suggested that CO may be desorbed firstly. When the CO was abstracted at first, the remaining hydrogen could form a surface hydroxyl and an isolated proton located at the surface vacancy. The electronic structures are simulated based on the Density Functional Theory (DFT) via VASP, to achieve its configuration and polarization if possible. Besides, other routes such as abstracting H_2 initially were also discussed, with higher energy barrier in the end.

Investigation of Li-N-H phase space for next generation Li-ion electrolytes

Peter J. Graham*, Oskar G. Soulas, Alexander G Squires, David O. Scanlon

*School of Chemistry, University of Birmingham e05-power-**dos**

Lithium-ion batteries (LIBs) are one of the most versatile forms of energy storage, being present in devices we use daily such as our phones and laptops to much larger devices such as electric vehicles and even electric grid storage. One of the most significant problems with LIBs comes from electrolytes which traditionally consist of a lithium salt dissolved in an organic electrolyte. However, the organic solvents used are quite volatile and can cause batteries to ignite at elevated temperatures.[1] This is where solid electrolytes come in as a desirable alternative as they have a much higher thermal stability making all solid-state batteries (ASB) much safer.[2] As well due to wider electrochemical stability window of solid electrolytes they become compatible with lithium metal anodes something that traditional electrolyte can't do.[3] This will allow all ASBs to surpass traditional LIBs in terms of energy density. In order for a material to become a solid electrolyte it must first meet a few criteria: it must have an ionic conductivity of at least 0.1 mS cm^{-1}, it must be electronically insulating (possessing an electric conductivity lower than 1 pS cm^{-1}) and must possess a wide electrochemical stability window.[4]

Lithium Imide is a potential solid-state electrolyte which has been reported to possess an ionic conductivity of 1 mS cm^{-1} at 30 ^{o}C as well as a wide electrochemical stability window of 5 V.[5] Combined with its very high gravimetric capacity and relative ease of synthesis it becomes a very promising solid-electrolyte candidate.[6] However there are still many unknowns relating to how this material functions as solid electrolyte and it is the goal of my PhD to elucidate it's behavior through the use of ab-initio methods such as Density Functional Theory (DFT) and Machine Learnt Potentials (MLP).

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Computational Modelling of Zeolite-Catalysed DMN Synthesis

Oliver Morris*, Igor Kowalec, Marc Pera-Titus, C. Richard. A. Catlow

* Cardiff Catalysis Institute, School of Chemistry, Cardiff University, Cardiff, CF24 4HQ, United Kingdom

*e05-react-**cat***

Zeolites, porous tetrahedral aluminosilicates, are used in ion exchange processes, separations, and as catalysts for a wide range of industrial processes, predominantly in the petrochemical industry.[1] Their unique ability to undergo doping, a substitution of a Si atom for another, heterometallic atom, allows for a change in physicochemical properties.

Dimethylnaphthalenes (DMNs) have been synthesized via the multi-step alkenylation of xylene since the 1990s however high yields throughout are required to be commercially viable and uses a Na-K alloy catalyst which is combustible in moist air.[2] 2,7-DMN has been noted to be a precursor to many polycyclic aromatic compounds (PACs), notably coronene,[3] which is used in the synthesis of graphene - a promising candidate in energy storage.[4,5]

Periodic Density Functional Theory (DFT) has been used to study zeolites and, more recently, Quantum Mechanics/Molecular Mechanics (QM/MM) embedding has been introduced to the zeolite field. The latter method allows for pore interactions to be investigated at a high level of theory, while, at a lower level of detail, still incorporating the long-range and electrostatic interactions the rest of the zeolite has on the site of interest.

In this work, DFT has been used to parametrize for further investigations using QM/MM embedding to explore a novel synthesis scheme (Figure 1) proposed by experimental collaborators in the group of Prof. Marc Pera-Titus at Cardiff University. This single-step process is cheaper, safer, and allows for direct synthesis of 2,7-DMN without the need for isomerization.

Figure 1. Proposed novel synthesis scheme to produce 2,7-DMN using an acid zeolite.

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Data-driven design of electroactive metal-organic frameworks

Ashna Jose*, Aron Walsh

*Department of Materials, Imperial College London e05-discov-**wal***

Metal-organic frameworks (MOFs) are versatile materials with tunable crystal structures, morphologies, and chemistries, offering diverse physical and chemical properties. Though typically electrically insulating, specific combinations of organic and inorganic components can impart electrical conductivity and redox activity in MOFs. The virtually limitless chemical space of MOFs however, presents a significant challenge in identifying the most optimal material for specific applications. Although Density Functional Theory (DFT) offers valuable insights into the fundamental chemistry and physics of these materials, and is used to study MOFs extensively, the computational expense that comes with DFT is a major obstacle in the identification and discovery of novel electroactive MOFs. To tackle these challenges, we compare several approaches. We employ the semi-empirical extended tight binding approach (xTB) to compute electronic band gaps of a large dataset of MOFs. The results are compared to a transformer model (MOFTransformer) that is fine-tuned to predict electronic properties. Further, we train more interpretable classical machine learning models to provide chemical insights that can be exploited to accelerate the design and discovery of functional MOFs.

Promoting MgH_2 Destabilization Through Reactive Elemental Additives

Navaratnarajah Kuganathan*, Martin Dornheim, David M. Grant and Sanliang Ling

*Advanced Materials Research Group, University of Nottingham e05-bulk-lin

Magnesium hydride (MgH_2) is a promising candidate for hydrogen storage, but its high thermodynamic stability presents significant challenges [1]. One effective strategy to overcome this limitation involves alloying MgH_2 to reduce its hydrogen desorption temperature and reaction enthalpy [2]. Although previous studies have examined specific alloying elements and their corresponding dehydrogenation enthalpies, a comprehensive investigation into the formation of alloys with a broader range of additives, encompassing varied stoichiometries and possible interactions with MgH_2, including alloy formation, remains limited [3]. In this study, we employ density functional theory (DFT) to systematically explore the formation of Mg_xA_y alloys (where x, y = 1, 2, or 3 and A includes elements from Group II, III, IV, transition, noble, and post-transition metals) and to calculate the dehydrogenation enthalpies for hydrogen release from MgH_2 through various reaction pathways. Our results reveal that Rh and Pd strongly tend to form multiple alloy types, significantly destabilizing MgH_2 and yielding exothermic dehydrogenation enthalpies. We identify several elements comprising the most thermodynamically stable alloy types with favorable dehydrogenation properties: Ag and Ga for MgA; Ge, Ni, and Sn for Mg_2A; B, Co, Cu, and Zn for MgA_2; Al for MgA_3; and Ru for Mg_3A_2. Additionally, we assess the stability and hydrogen release characteristics of several ternary Mg–A–H hydride systems.

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Germanium Dioxide – A UWBG Material for Optoelectronic Applications

Jacob C. Baggott*, Romain Claes, Alexander G. Squires, David O. Scanlon

*School of Chemistry, University of Birmingham e05-bulk-**dos**

For materials to be optically transparent, it must possess an optical band gap greater than approximately 3.1 eV[1]. Due to such a wide band gap, these materials are considered insulators. Conversely, due to their overlapping valence and conduction bands, conductive materials, such as metals, are assumed to be opaque. Usually, these two properties are regarded as mutually exclusive. However, transparent conducting oxides (TCOs) defy this notion, uniquely combining optical transparency and electrical conductivity in a single material. It is this combination of properties that makes TCOs indispensable in optoelectronic applications, such as solar cells, smart windows, touch screens, and flat panel displays[1].

Currently, the field relies heavily on a narrow set of post-transition metal oxides (e.g. ZnO, SnO_2, In_2O_3, Ga_2O_3)[1-3], limiting the diversity and performance of devices. Expanding the library of WBG oxides presenting favourable electronic properties for technological applications remains of critical importance.

Building on recent work on antimony oxides[4, 5], we turned our attention to exploring other underutilized families of oxides, beginning with Ge(IV) oxides, specifically rutile GeO_2. Despite various experimental and computational studies[6-9], some disagreements remain regarding the true potential of GeO_2, particularly concerning its optical properties. By using state-of-the-art computational approaches, we investigated these optical properties but also the defect chemistry of GeO_2, demonstrating the potential of this material for optoelectronic applications**.**

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A Computational and Theoretical Study on Methanol Synthesis

Matis Ferrini, Arunabhiram Chutia, Stewart F. Parker

School of Natural Sciences@University of Lincoln; ISIS@STFC *e05-react-**arc***

Methanol, a versatile molecule, is produced globally at an estimated 98 million tons per year.[1] The reaction uses syngas (H_2/CO/CO_2) as the starting materials and operates over a Cu/ZnO/Al_2O_3 catalyst at mild reaction conditions.[2] While the reaction pathways are well characterised, there are uncertainties regarding the complex states and interactions of the reactants. In this work, we performed Density Functional Theory (DFT) based quantum chemical calculations using the Vienna Ab-initio Simulation Package (VASP) to understand the interactions of H atoms on the Cu(111) surface. We used the non-local vdW-DF2 by Lee et al.[3] exchange-correlation functional, along with the projector augmented wave (PAW) method. The surface was modelled using a 4 x 4 supercell with 5 atomic layers and a theoretical lattice constant 3.746 Å. A 3 x 3 x 1 k-point grid was used, and the plane-wave cutoff energy was set to 475 eV.

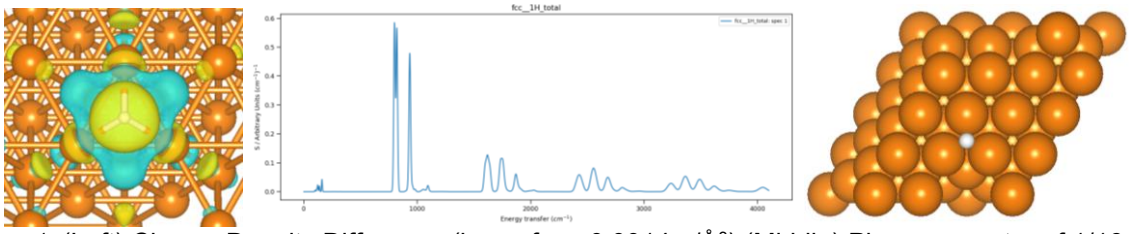

Figure 1: (Left) Charge Density Difference (isosurface 0.0014 e/Å^3) (Middle) Phonon spectra of 1/16 FCC coverage. (Right) 4x4 Cu(111) model with 1/16 coverage.

As expected, the results show that H adsorption is more stable on the FCC site. As coverage increases, the adsorption energy per H atom becomes more negative, indicating increasing stability. Energy barriers for the diffusion of adsorbed H atoms between sites were also investigated using the Climbing Image Nudged Elastic Band (CI-NEB) method. Additionally, phonon spectra were computed to analyse the vibrational properties, which will be compared with the INS experiments to be performed at the ISIS pulsed neutron and muon source. In Figure 1, the charge density difference of H atom in the FCC site (Left) shows how the three Cu atoms lose electron density (cyan) as they donate their electrons to the H atom (Yellow), thus making the H atom negatively charged on the surface. Phonon Spectra (Middle) and 1/16 model (Right), in Figure 1, show the phonon modes of the H atom in the FCC site. The signal ~935 cm^{-1} refers to the out of plane mode and, due to the interaction with the threefold Cu atom site, it is more energetic than the in-plane mode (~817 cm^{-1}). With these theoretical results, along with the preliminary experimental data, this project aims to further evaluate the interactions and conversion of CO_2 to methanol.

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Understanding Polarisation at Ferroelectric-perovskite Interface – A molecular Dynamic Perspective

Tingwei Li[1*], Keith Butler[2], Joe Briscoe[1]

[*1] School of Engineering and Materials Science, Queen Mary University of London, London, UK
[2] Department of Chemistry, University College London, London, UK

e05-surfin-but

There has been widespread research into the effect of ferroelectric polarisation on perovskite solar cells. This was based on the principle that the electric field associated with ferroelectric polarisation could influence carrier dynamics and reduce non-radiative recombination. We have successfully constructed appropriate interface models of $BaTiO_3$ (BTO) and $CH_3NH_3PbI_3$ (MAPI) and preliminarily demonstrated that, in addition to the direct influence of the electric field generated by the spontaneous polarisation in BTO on the CH_3NH_3 (MA) dynamics in MAPI, there was also potential to stabilize the orientation and polarisation of MA molecules in MAPI. Thus, we aimed to explore the effects of different BTO polarisation directions on the dynamics of MA molecules in MAPI within large-scale molecular dynamics simulations.

To achieve this, we employed machine learning interatomic potential (MLIP)-based molecular dynamics (MD) simulations to explore the dynamics of MA molecules and carrier transport within the BTO-MAPI interface. More specifically, we sampled data from ab initio molecular dynamics (AIMD) trajectories calculated by using ABACUS. Then the training data was used to train a machine-learning potential model, enabling molecular dynamics simulations on the longer time-scale and the larger length-scale. In addition, we used CP2K to generate training data for the dipole moments and polarizabilities of the interface structures. This data was used to train a machine-learning model capable of predicting the real-time changes in dipole moments and polarizabilities of the interface in MD simulations, thereby quantitatively illustrating the effect of the polarisation electric field in ferroelectric materials on the polarisation of MAPI.

First-principles study of the thermoelectric properties of $Sn(S_{1-x}Se_x)$ alloys

Min Zhang*, Joseph M. Flitcroft, Jonathan M. Skelton

* *Department of Chemistry, University of Manchester* *e05-power-**ske***

Thermoelectric power is an efficient and flexible technology for improving energy efficiency, but widespread applications require cost-effective and sustainable materials with a high figure of merit, *zT*. Alloying is a common strategy for optimising *zT*, but predicting and understanding its impact on the materials properties is made challenging by the structural disorder.

In this work, we develop a practical and rigorous approach to predicting the *zT* of alloys, by combining electrical-transport and lattice thermal-conductivity calculations with approximate models for the electron and phonon scattering, and apply it to study the $Sn(S_{1-x}Se_x)$ alloy with $x \simeq 0.8$. We compare predictions obtained by averaging over all possible configurations in a 32-atom supercell with those obtained using the special quasi-random structure (SQS) method with a series of supercell sizes. It was found that the 32-atom SQS results are similar to the averaging over all configurations in the 32-atom supercell and the 128-atom SQS achieves an optimal balance between convergence of thermoelectric properties and computational efficiency. Our results indicate that $Sn(S_{0.2}Se_{0.8})$ alloys with n-type doping are able to obtain better thermoelectric performance compared to the typically applied p-type doping in experiments and n-doped $Sn(S_{0.2}Se_{0.8})$ may have a comparable *zT* to SnSe due to lower lattice thermal conductivity.

Structural Characterisation and Analysis of $NaTaO_xCl_{6-2x}$ for Solid—state Sodium Batteries

Abigail C. Parsons*, Alexander G. Squires, David O. Scanlon

School of Chemistry, University of Birmingham, Edgbaston, Birmingham, United Kingdom

e05-power-dos

As the demand for efficient and sustainable energy storage solutions grows, next-generation sodium-ion batteries have emerged as a promising alternative to lithium-ion batteries. With sodium's abundance and wide geographical distribution, sodium ion batteries offer advantages in cost, sustainability, and economic viability.[1] Solid-state sodium batteries employ solid electrolytes and potentially offer further advantages including superior safety, higher energy density and prolonged lifespan.[2,3]

Recent studies have focused on investigating sodium conducting halide-based solid electrolytes, such as $NaTaCl_6$, which has a modest ionic conductivity of 6.2×10^{-5} S cm^{-1} at 25℃.[4] To improve upon the properties of ordered low-ionic conductivity halides, extensive ball milling has been used to disrupt the ordering, successfully increasing ionic conductivity by two orders of magnitude.[5] An alternative approach to improving ionic conductivity is to explore related chemistries, as demonstrated by studies exploring $NaTaOCl_4$, a new sodium metal oxyhalide catholyte material. [6,7] This work builds upon previous studies of $NaTaOCl_4$ by exploring a series of $NaTaO_xCl_{6-2x}$ compositions (x = 0.5, 1), with a primary focus on how varying oxygen content influences structural and electrochemical properties. The phases $NaTaO_{0.5}Cl_5$ and $NaTaOCl_4$, exhibit excellent ionic conductivities of 3.8 mS cm^{-1} and 1.5 mS cm^{-1}, respectively. However, instability against sodium metal currently limits the application of this material as a standalone solid electrolyte. Understanding the atomistic origins of the performance of this material is challenging given its amorphous nature.

This research establishes a computational workflow to characterise common structural motifs present in $NaTaO_xCl_{6-2x}$ and identify low-energy crystal structures via *ab initio* random structure searching. Understanding the material's structure and how oxygen content influences the degree of amorphization will enable a deeper evaluation of its performance as a solid electrolyte contributing to the development of safer and more efficient sodium-ion battery technologies. Furthermore, this study paves the way for investigation of related glassy and amorphous ion conductors and the derivation of common materials design rules in this chemical and structural space.

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Effect of Nitrogen Incorporation in a-HfO_2

Isaac Mackley, J strand, A L Shluger

Department of Physics and Astronomy, UCL *e05-bulk-shl*

The nitridation of HfO_2 has demonstrated improvements in leakage current, bias temperature instability, and mobility of films. However, experimental studies have shown that the improvements are only at low nitrogen concentrations and that at higher concentrations performance is degraded. This study aims to investigate the mechanisms underlying the negative aspects of nitridation by examining different nitrogen species within hafnia, including N, N_2, and NH. We explore the charge states, geometries, charge transition levels (CTLs), and mobility of these species, highlighting their impact on the material's electronic behaviour. Our results show that while molecular nitrogen (N_2) is thermodynamically more stable, its higher CTLs make it a less desirable defect, potentially detrimental to device performance. The study also examines defect reactions and interactions, revealing the formation of N_2 as energetically favourable, with the largest energy gains occurring when two N interstitials combine. These findings offer insights into the defect landscape of HfO_2 and provide guidelines for mitigating the negative effects of nitrogen incorporation in semiconductor devices, particularly in relation to leakage currents and electron tunnelling. To carry out this computational work, CP2K is used to perform Density Functional Theory calculations. Non-local functionals are used to provide more accurate values for the band gap allowing for better analysis of the electronic structure of defects.

Vibrational Dynamics and Mechanical Anisotropy in Open Framework Materials: Computational Design Rules for MOF-based Triboelectric Generators

Debayan Mondal*, Jin-Chong Tan

*Multifunctional Materials and Composites (MMC) Laboratory, Department of Engineering Science, University of Oxford e05-discov-**tan***

This computational study will employ periodic density functional theory (DFT) to elucidate the role of vibrational dynamics and elastic properties in governing contact electrification (CE) of metal-organic frameworks (MOFs) for triboelectric energy harvesting. Using the *ab initio* CRYSTAL23 DFT code with the *PBEsol0-3c* composite method[1], we will perform Γ-point phonon calculations on MOFs with a large open framework structure (e.g., ZIF-71, ZIF-72, MAF-5) to study the low-frequency vibrational modes linked to charge trapping sites and interfacial electron transfer. Concurrently, the elastic tensor calculations (C_{ijkl} coefficients) will be performed to quantify the anisotropic mechanical response (Young's and shear moduli, Poisson's ratio, bulk modulus and linear compressibility[2]), correlating framework flexibility with adhesion energy at MOF- polymer interfaces. By integrating vibrational spectra and elastic moduli with defect engineering strategies, akin to our recent methodology reported in ref[3], we will establish basic structure-property relationships to predict triboelectric polarity and elucidate energy conversion efficiency mechanisms, providing first-principles guidelines for designing high-performance MOF-based triboelectric generators.

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Carbon Nitride-Supported Metals for the Selective Oxidation of HMF to DFF

Xue Yong*, Weiwei Xu

*Department of Electrical Engineering and Electronics, University of Liverpool e05-react-**xue***

Biomass is a renewable feedstock with immense potential for producing non-food chemicals. For example, biomass-derived 5-hydroxymethylfurfural (HMF) can be electrochemically oxidized into 2,5-diformylfuran (DFF), a key intermediate used in the production of polymers, coatings, and fine chemicals. However, achieving selective oxidation of HMF to DFF presents significant challenges in catalyst design and optimization.

Graphitic carbon nitride (g-C_3N_4), a versatile and sustainable material, has gained attention as a promising support for metal catalysts due to its exceptional stability, unique electronic properties, and tunable surface functionalities. While g-C_3N_4 itself is not active for the oxidation of HMF, its ability to facilitate charge transfer and enhance catalytic activity makes it an excellent candidate for supporting single-atom catalysts (SACs). Using systematic density functional theory (DFT) calculations, we identified Ti/Pt-g-C_3N_4 as a promising candidate for HMF oxidation and elucidated the underlying reaction mechanisms.

Mg Doping in GaN via MACE Potentials

Yuxi Zhang*, Scott Woodley, Alexey Sokol, Alin Marin Elena

University college London* *e05-bulk-smw***

Materials Studied: This project investigates magnesium (Mg) doped gallium nitride (GaN), a wide-bandgap semiconductor crucial for optoelectronics such as LEDs and power devices. We study bulk wurtzite GaN supercells with varying levels of Mg substitution and native defects, focusing particularly on nitrogen vacancies as charge compensators.

Importance: Achieving efficient p-type GaN is a longstanding challenge that limits device performance. Mg is the primary acceptor dopant, but compensation by native defects and defect clustering reduce the achievable hole concentration. A better atomic-scale understanding of these phenomena could directly benefit materials design for next-generation electronic and optoelectronic applications.

Methodology: We employ machine-learned interatomic potentials (specifically the MACE model within the Janus Core framework, trained on DFT data) to perform large-scale atomistic simulations. This approach allows us to efficiently model realistic supercells and explore a wide range of Mg doping and defect configurations beyond the reach of conventional DFT.

Key steps include: Generate and optimize GaN supercells with various Mg and N-vacancy configurations; Statistically sample both random and clustered arrangements to capture disorder; Analyze energetics, structural changes, and defect associations.

Goals: Our main goal is to systematically assess the energetics and structural impact of Mg doping and charge compensation in GaN. Specifically, we aim to: Quantify formation energies for a broad set of doped/defected structures; Identify favorable dopant and defect arrangements; Provide insights relevant to improving p-type doping efficiency in GaN

Notes

Delegates @2024 MCC Conference: Daresbury Laboratory

Arunabhiram Chutia (arc)	
School of Chemistry, University of Lincoln	
Matis Ferrini	25807163@students.lincoln.ac.uk
Anuradha Pallipurath (arp)	
School of Chemical and Process Engineering, University of Leeds	
Anuradha Pallipurath Namrata Ramesh	a.r.pallipurath@leeds.ac.uk namrata.ramesh@gmail.com
Jochen Blumberger (blu)	
Department of Physics, University College London	
Ewen Jones Filip Ivanovic Oillie Russell Andras Petho	ucapthj@ucl.ac.uk ucanfiv@ucl.ac.uk ucapoga@ucl.ac.uk andras.petho.21@ucl.ac.uk
QiongCai (cai)	
University of Surrey	
Neubi Xavier Matthias Golomb	n.xavier@surrey.ac.uk m.golomb@surrey.ac.uk
C. Richard A. Catlow	
Chemsitry, UCL and Cardiff University	
R. Catlow Erze Gao Oliver Morris Shijia Sun Yunyu Zhang Alec Desmoutier Mohammed Alotaibi Abraham Nieva de la Hidalga Thomas Hill Igor Kowalec Michael Higham	catlowr@cardiff.ac.uk erze.gao.19@ucl.ac.uk morriso7@cardiff.ac.uk uccaunb@ucl.ac.uk yunyu.zhang.23@ucl.ac.uk ddesmoutier@gmail.com mohammed.alotaibi.20@ucl.ac.uk nievadelahidalgaa@cardiff.ac.uk hillt8@cardiff.ac.uk kowaleci@cardiff.ac.uk m.higham@ucl.ac.uk

David Scanlon (dos)	
Department of Chemistry, University of Birmingham	
Jacob Baggot Peter Russell Abigail Parsons	jzb492@student.bham.ac.uk ppr466@student.bham.ac.uk axp137@student.bham.ac.uk
Saiful Islam (isl)	
Department of Materials, University of Oxford	
Vikram Arup Chakraborty	dr.vikram@materials.ox.ac.uk arup.chakraborty@materials.ox.ac.uk
James Dawson (jad)	
University of Newcastle	
Daniel Cole	daniel.cole@ncl.ac.uk
Jamieson Christie (jam)	
University of Loughborough	
Jamieson Christie	j.k.christie@lboro.ac.uk
John Buckeridge (jbu)	
London South Bank University	
John Buckeridge	j.buckeridge@lsbu.ac.uk
Juliana Morbec (jul)	
University of Keel	
Juliana Morbec	jmmorbec@gmail.com
Kostya Trachenko (kos)	
Department of Physics, Queen Mary University of London	
Harvey Devereux	h.devereux@qmul.ac.uk
Nora H. de Leeuw (lee)	
School of Chemistry, University of Leeds	
Marietjie Ungerer	m.j.ungerer@leeds.ac.uk

Sanliang Ling (lin)	
Faculty of Engineering, University of Nottingham	
Navaratnarajah Kuganathan	navaratnarajah.kuganathan@nottingham.ac.uk
Johannes Lischner (lis)	
Imperial College	
Johannes Lischner	j.lischner@imperial.ac.uk
Andrew J. Logsdail (log)	
Cardiff Catalysis Institute, School of Chemistry, Cardiff University	
Kushagra Agrawal Zhongwei Lu Oscar van Vuren	chaudharia@cardiff.ac.uk luz30@cardiff.ac.uk vanvureno@cardiff.ac.uk
Keith McKenna (mck)	
Department of Physics, University of York	
Thomas Lynch	wlk510@york.ac.uk
Thomas Keal	
Scientific Computing Department, STFC	
Thomas Keal Andrew Duff Thomas Durrant Elliot Kasoar Rajany RV Joseph Thacker Ilian Todorov Alin Elena You Lu	thomas.keal@stfc.ac.uk andrew.duff@stfc.ac.uk thomas.durrant@stfc.ac.uk elliot.kasoar@stfc.ac.uk rajany.kv@stfc.ac.uk joseph.thacker@stfc.ac.uk ilian.todorov@stfc.ac.uk alin-marin.elena@stfc.ac.uk you.lu@stfc.ac.uk
Anh Thi Van Phan (pha)	
University of Surrey	
Linquan Gong	l.gong@surrey.ac.uk
Matthew Quesne	
Department of Chemistry, University of Leeds	
Matthew Quesne Chengxu Zhu	M.G.Quesne@leeds.ac.uk zhuchengxu77@163.com

Alexander L. Shluger (shl)	
Department of Physics and Astronomy, University College London	
Isaac Mackley Jack Strand Margherita Buraschi	zcapijm@ucl.ac.uk jack.strand.14@ucl.ac.uk m.buraschi@ucl.ac.uk
Jonathan Skelton (ske)	
Department of Chemistry, University of Manchester	
Jonathan Skelton Min Zhang Zhiwu Wang Joseph Flitcroft	jonathan.skelton@manchester.ac.uk min.zhang-19@postgrad.manchester.ac.uk zhiwu.wang@postgrad.manchester.ac.uk joseph.flitcroft@manchester.ac.uk
Scott M Woodley (smw)	
Department of Chemistry, University College London	
Scott M Woodley Bruno Camino Karen Stoneham Selin Kilic Zichoa Wang Yuxi Zhang Zhuoran Ji	scott.woodley@ucl.ac.uk b.camino@ucl.ac.uk k.stoneham@ucl.ac.uk kowaleci@cardiff.ac.uk asd190102@163.com yuxi.zhang.19@ucl.ac.uk zhuoran.ji.20@ucl.ac.uk
Alexey Sokol (sok)	
Department of Chemistry, University College London	
Alexey Sokol Jamal Abdul Nasir Jingcheng Guan Taifeng Liu Liam Morgan Hangbo Qi	a.sokol@ucl.ac.uk jamal.nasir.18@ucl.ac.uk jingcheng.guan@ucl.ac.uk tfliu@vip.henu.edu.cn uccalmo@ucl.ac.uk uccahq0@ucl.ac.uk
Jin-Chong Tan (tan)	
Department of Engineering Science, University of Oxford	
Mehdi Tavakol Debayan Mondal	mahdi.tavakol@eng.ox.ac.uk debayan.mondal@eng.ox.ac.uk

Umberto Terranova (ter)	
School of Postgraduate Medicine and Allied Health, University of Buckingham	
Umberto Terranova	umberto.terranova@buckingham.ac.uk
Aron Walsh (wal)	
Department of Materials, Imperial College London	
Ashna Jose	a.jose@imperial.ac.uk
David Willock (wil)	
Department of Chemistry, University of Cardiff	
David Willock	willockdj@cardiff.ac.uk
Xue Yong (xue)	
University of Liverpool	
Xue Yong Joshua Griffiths	xue.yong@liverpool.ac.uk j.e.griffiths@liverpool.ac.uk
Invited Speakers / Registered from Unknown Group	
Vitaliy Kurlin (University of Liverpool) Sofia Oliveira Oliver Henrich Viv Kendon Neil Drummond Phil Hasnip Mark Storr	vitaliy.kurlin@gmail.com sofia.oliveira@bristol.ac.uk oliver.henrich@strath.ac.uk viv.kendon@strath.ac.uk n.drummond@lancaster.ac.uk phil.hasnip@york.ac.uk Mark.Storr@awe.co.uk