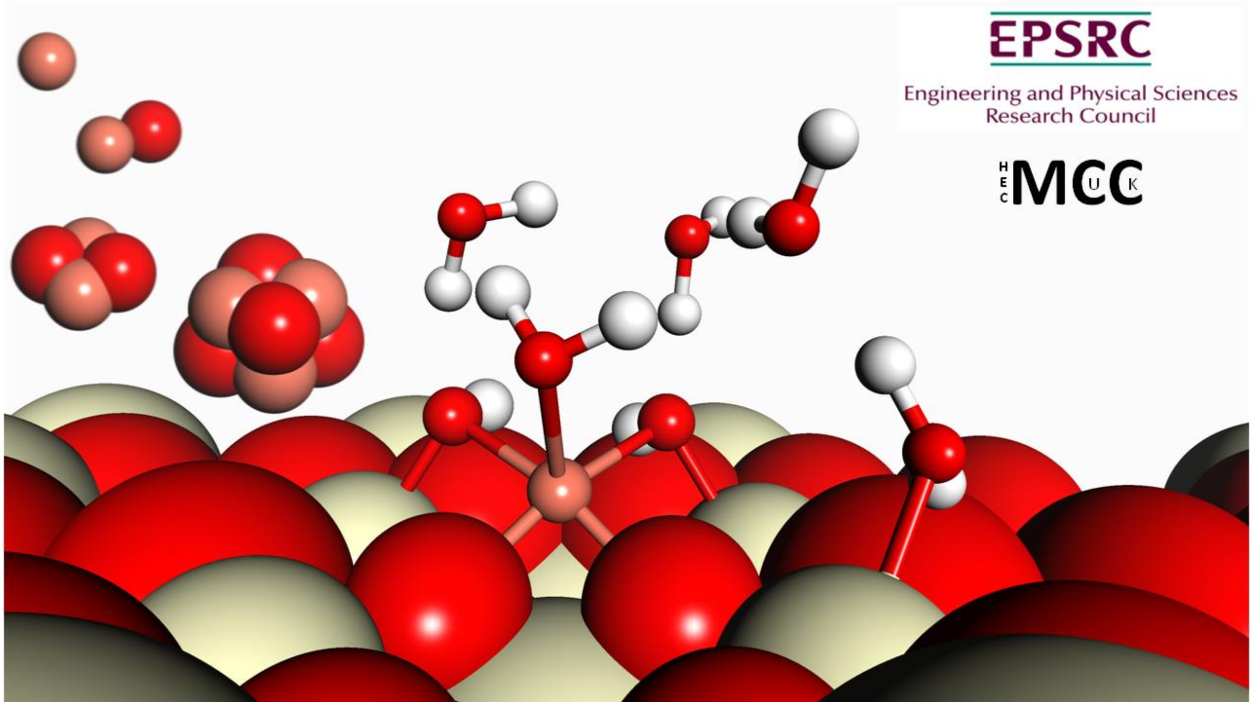
MCC 5th Conference



Daresbury Laboratory

Wednesday 28th June – Friday 30th June, 2023

Sponsors

The MCC is very grateful for funding from EPSRC.

We are grateful that this event is also sponsored by:

STFC, Daresbury Laboratory

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, particularly colleagues who have helped us complete our recent EPSRC proposal during 2022, our theme leaders and all of you, including our invited guests, who have joined us in Daresbury in-person or online.

MCC 5th Conference - Programme

Wednesday 28th June			
12:30	**Registration Desk Open**	**Daresbury Laboratory**	
12:30	**Lunch**		
Session 1:	Low Dimensional Materials	**Chair** Umberto Terranova	
13:45	**Start – Welcome Address**		
13:50	**Invited:** Disorder?		Neil Allan Bristol
14:20	Predicting the Atomic Structures of Nanoclusters for Compounds with 1:3 Stoichiometry		Dong-Gi Kang UCL (smw)
14:40	Multiscale modelling of the electrical conductivity of carbon nanotube films		Thomas R Durrant UCL (shl)
15:00	**Invited:** ONETEP, with some exemplar applications thrown in.		Chris Skylaris Southampton
15:30	**Tea**		
Session 2:	Reactivity	**Theme Leader** David Willock	
16:10	**Invited:** Current Challenges in Catalytic Science		G Hutchings Cardiff
16:50	Mechanism of NH$_3$ Synthesis on Fe$_3$Mo$_3$N		M. D. Higham UCL (cat)
17:10	The Electronic Structure and Catalytic Activity of a Ru3 Cluster Embedded on Nitrogen-doped Graphene		Yao Zhao Oxford (mcg)
17:30	Computational investigation of NiOOH, from bulk to catalysis		Ruotao Yang UCL (cat)
17:50	**Poster Session** Starting with 24 45s-lightning presentations (1 slide / poster)		
20:00	Session ends		

Thursday 29th June			
Session 3:	Bulk	**Chair** Chris Lorenz	
9:10	**Invited:** Amorphous ZnO: Structure, Stability and Charge Trapping		David Mora-Fonz HTU, Mexico
9:40	Ab-Initio Molecular Dynamics Simulations of Substoichiometric Amorphous Alumina		Jack Strand UCL (shl)
10:00	Unravelling Hydrogen Bonding Behaviours and their Impact on Proton Conductivity in Supramolecular Cages		Lei Lei Nottingham (lin)
10:20	Investigation of unintentional aggregation of impurity-related defects within 4H-SiC		N. Smith UCL (shl)
10:40	Simulation of correlation driven metal-insulator transitions in real solids using deep neural networks		H. Sutterud Imperial (fou)
11:00	**Coffee**		

Session 4:	Surfaces and Interfaces	**Theme Leader** Matthew Watkins	
11:30	The effect of ceria surface composition on scavenging reactive oxygen species		Khoa Minh Ta Huddersfield (mol)
11:50	Modelling the effect of H in the formation, stabilisation and evolution of voids in polycrystalline Cu		Vasileios Fotopoulos UCL (shl)
12:10	Simulating Hydrogenation in Magnesium Through the Application of Machine Learning Models		Oliver M. Morrison Nottingham (lin)
12:30	Study of Hydroxyl Nests in Zeolites		Alec Desmoutier UCL (cat)
12:50	**Lunch**		
Session 5:	Power	**Theme Leader** Lucy Whalley	
13:50	Bulk and Surface Contributions to Ionisation Potentials of Metal Oxides		Xingfan Zhang UCL (sok)
14:10	Bismuth oxychalcogenide materials for thermoelectric applications		Joseph M. Flitcroft Manchester (ske)
14:30	Relating the formation energies for oxygen vacancies defects to the structural properties of tungsten oxides		R Kerr Lancaster (mur)
14:50	Controlling the heat transport in thermoelectric materials		Jonathan M. Skelton Manchester (ske)
15:10	**Coffee**		
Session 6:	Environment and Algorithms	**Chair** Scott Woodley	
15:40	**Invited:** The Baskerville Tier-2 Service		Jon Wakelin Birmingham
16:00	**Invited:** The GW4 Tier-2 Service		Steven Chapman Bristol
16:30	Oxygen-functionalised graphene: structure, properties, and potential for phosphate sensing		Natalia Martsinovich Sheffield (enviro-nat)
16:50	An Efficient Model for sp-Lone Pair Cations		Woongkyu Jee UCL (algor-smw)
17:10	**Invited:** Fundamental Principles of a Virtual Tokamak Fusion Reactor		Sergei L. Dudarev UKAEA
17:40	session ends		
18:00 – 21:00	**Conference BBQ**		

Friday 30th June		
Session 7:	Materials Discovery	**Chair** Marco Molinari
9:00	Optimisation of decision sequences from selection of elements to chemical formulae of functional materials	A. Vasylenko Liverpool (dar)
9:20	Computational Modelling of Wide Band Gap Transparent Conducting Oxide Sb_2O_5	Ke Li UCL (dos)
9:40	A computational study characterising the intrinsic and extrinsic doping profile of ZnS	S. Aggarwal UCL (dos)
10:00	Organosulfide-Halide Perovskites incorporating Zwitterions	Santanu Saha Oxford (fil)
10:20	**Coffee**	
Session 8a:	Biomaterials and Soft Matter	**Chair** Richard Catlow
10:50	**Invited:** Prediction of critical micelle concentrations in surfactant mixtures by DPD simulations	Guadalupe Jiménez-Serratos, STFC
11:20	Structural and dynamic properties of poly(styrene-co-maleic anhydride) at the interface of conjugated polymers	Christian D. Lorenz KCL (lor)
11:40	Iron-sulfur peptides for an efficient electron transfer to the hydrogenase enzyme	Umberto Terranova Buckingham (react-ter)
Session 8b:	MCC General Meeting	
12:00	computational simulations to understand molecular crystals and crystallisation - New Member Code acc	Aurora Cruz-Cabeza Durham
12:20	Generating a Machine-Learned Equation of State for Fluid Properties - New Member Code mul	Erich Muller Imperial College
12:40	ARCHER2 update	William Lucas EPCC
12:50	YOUNG update	Heather Kelly UCL

13:00	Lunch	
14:00	MCC Main Meeting	
16:30	Close	

Reserve Talk	How bulk and surface properties of X_4SiC_3 (X=Ti,V,Nb,Zr) tune reactivity: A computational study	Matthew Quesne Cardiff (react-cat)

Talks

Disorder?

Neil Allan*

[*] *School of Chemistry, University of Bristol*

TBA

Predicting the Atomic Structures of Nanoclusters for Compounds with 1:3 Stoichiometry

Dong-Gi Kang*, Alexey A Sokol, Alin-Marin Elena, Scott M Woodley

*Department of Chemistry, UCL e05-nano-**smw**

Aluminium halides and hydrides are important compounds in multiple areas of materials chemistry. Our interest is in predicting the atomic structure of their stoichiometric, non-charged clusters. However, the study of nanoclusters, using molecular dynamics (MD) or Monte Carlo (MC) simulations of largish sized particles composed of many atoms, is limited by the exponentially increasing computer cost as each configuration to evaluate and the number of possible configurations increases rapidly. Consequently, a cheaper method to evaluate the configurations along with developing clever optimization methods for generating them are required. Here, we concentrate on the former and explore whether interatomic potentials (IP) and/or machine learning (ML) potentials can be developed and employed to model compounds/systems of interest and thus address some of the challenges of structure prediction of nanoclusters. Note that we employ an initial search over the IP/ML energy landscape for plausible structures can be refined at a later stage using an appropriate electronic structure approach.

In the first part of this talk, results for developing suitable IPs for ionic materials that have a high stoichiometric ratio, viz. Ti_3N_4, $Mg_xSi_yO_z$, AlX_3 (X=H, F, Cl), will be presented.

The second half will be about the trend in the structural characteristics analysed as a function of size (number of stoichiometric units) for the predicted ground state atomic configurations (PBEsol local minima, LM) of XF_3 clusters. Results are obtained using the KLMC genetic algorithm developed in our group. For more challenging compounds to model, we are training ML potentials.

1. *S. M. Woodley, J. Phys. Chem. C 2013, 117, 45, 24003–24014;* 2. *T. Krahl, E. Kemnitz Catal. Sci. Technol., 2017,7, 773-796*
3. *A. M. Escatllar, T. Lazaukas, S. M. Woodley, S. T. Bromely, ACS Earth Space Chem. 2019, 3, 2390−2403*
4. *V. S. Bhadram, H. Liu, T. Li, Vitali. B. Prakapenka, R. Hrubiak, S. Lany, T. A. Strobel., Phys. Rev. Materials 2, 011602(R*
5. *V. L. Deringer, A. P. Bartók, N. Bernstein, D. M. Wilkins, M. Ceriotti, and G. Csányi, Chem. Rev. 2021, 121, 16, 10073–10141.*

Multiscale modelling of the electrical conductivity of carbon nanotube films

Thomas R. Durrant*, Yvelin Giret, David Z. Gao, Alexander L. Shluger

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

Carbon nanotubes (CNTs) are a fascinating low dimensional material with very high electrical conductivity. CNTs hold significant technological promise due to their extremely high electrical conductivities. This is because metallic CNTs behave as ballistic conductors, which enables highly energy efficient electrical devices to be constructed from them. In many practical applications, CNTs are deposited in a disorganized film. One such example application is the use of a CNT film as resistance changing element in non-volatile computer memory. In order to model the electrical conductivity of such films, models of the electrical conductivity of individual CNT junctions must be combined with larger-scale models of their geometric structure.

In this project, a multiscale model of the electrical conductivity of CNT films was developed through the following steps: **(a)** A large library of CNT-CNT junction structures was simulated using density functional tight binding (DFTB) in combination with the nonequilibrium Green's function (NEFG) approach. The intrinsic electrical conductivity of individual CNTs is high and the primary source of electrical resistance within CNT films is anticipated to occur at CNT-CNT junctions where electrons must tunnel from one CNT to another. The results were used to generate an atomistic conductivity model. **(b)** Mesoscopic models of nanoscale CNT films were generated using course-grained molecular dynamics (CGMD). A library of CNT films was generated. **(c)** The atomistic conductivity model developed in (a) was used in combination with larger-scale molecular dynamics (MD) calculations in order to parameterize electrical conductivities between the mesoscopic segments employed in (b). **(d)** The structural models generated in (b) were then combined with the electrical parameters established in (c) in order to produce a large-scale electrical model of the conductivity of a nanoscale CNT film, using the method of nodal analysis.

Using this approach, the electrical conductivity of realistic CNT films can be evaluated. Our results demonstrate that depositing CNT films in layers is one approach that can be used to tailor the conductivity of the resultant films.

ONETEP, with some exemplar applications thrown in

Chris Skylaris

University of Southampton

TBA

Current Challenges in Catalytic Science

Graham Hutchings

University of Cardiff

TBA

Mechanism of NH_3 Synthesis on Fe_3Mo_3N

M. D. Higham*, Constantinos D. Zeinalipour-Yazdi, Justin S. J. Hargreaves, C. Richard A. Catlow

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

Plane-wave Density Functional Theory (DFT) techniques are applied to investigate two distinct reaction mechanisms for ammonia (NH_3) synthesis over a thin-film model $Fe_3Mo_3N(111)$ surface, namely an associative Mars van Krevelen mechanism and the conventional, dissociative, Langmuir Hinshelwood mechanism. The η-carbide structured Fe_3Mo_3N is experimentally reported to be highly active for NH_3 synthesis, in common with the isostructural Co_3Mo_3N. Experimental studies revealed that lattice N is much more active and easily removed in Co_3Mo_3N, in contrast to Fe_3Mo_3N, and computational studies showed that the presence of surface lattice N vacancies can enhance N_2 activation and NH_3 synthesis over Co_3Mo_3N. Hence, the present work aims to provide a detailed complementary investigation of the Fe_3Mo_3N system in order to elucidate the similarities and differences between the Fe and Co systems in terms of N vacancy formation behaviour and surface reaction mechanism.

Plane-wave DFT as implemented within the VASP code was applied to obtain N vacancy formation energies and obtain optimised geometries for all intermediates involved in the MvK and LH mechanisms, and to identify transition states for all elementary processes. The revised PBE functional was used throughout, along with a 650 eV plane-wave cut-off and a Monkhorst-Pack k-point sampling mesh of density (4×4×1). All structures were optimised until atomic forces were converged to within 0.01 $eVÅ^{-1}$. Transition states were identified using the CI-NEB and Dimer methods, and were confirmed via vibrational analysis.

The calculations reveal that whilst surface N vacancy formation is more energy-demanding for Fe_3Mo_3N compared to Co_3Mo_3N, the vacancy formation energies are comparable, implying that it is highly likely that surface lattice N vacancies are present on Fe_3Mo_3N, and that loss of lattice N is limited by bulk N diffusion. It was also found that N_2 activation was enhanced on the Fe_3Mo_3N surface compared to Co_3Mo_3N. The calculations will inform future studies considering Cr promotion of Fe_3Mo_3N and the development of quaternary $Fe_{3-x}Co_xMo_3N$ catalysts with an optimal Fe:Co ratio.

The Electronic Structure and Catalytic Activity of a Ru3 Cluster Embedded on Nitrogen-doped Graphene

Yao Zhao*, John E. McGrady

*Department of Chemistry, University of Oxford e05-react-mcg

Pyrolysis has been a popular synthetic technique in producing single-atom catalysts (SACs) embedded on graphene-based materials. A slight variation, triple-atom catalysts composed of three Ru atoms embedded on N-doped graphene was synthesised in a confined pyrolysis of Ru3(CO)12 and reported to catalyse alcohol oxidation 10 times more efficiently compared to its SAC analogue.[1] Here, we build the models of Ru3/N-graphene and analyse their electronic structures and orbital interactions between the cluster and N-graphene. Significant orbital interactions with localised p defect states on the N-graphene model led to the transfer of two electrons from the cluster to the conduction band of the surface. The surface therefore acts as a buffer, removing and restoring electrons to the active site during the catalytic cycle. Our initial exploration of the cycle indicates a potential role for Ru=O species in the C-H bond cleavage step of alcohol oxidation.

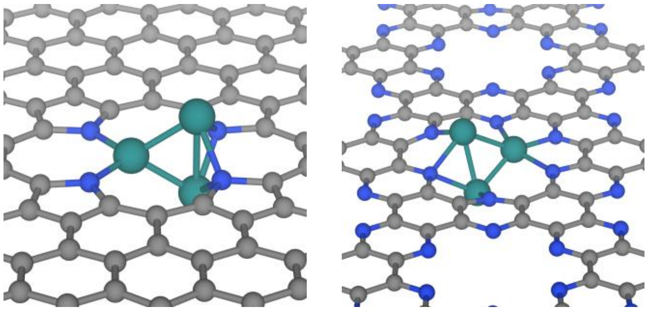


Figure 1: Ru_3 cluster embedded on (a) N-graphene and (b) g-C_2N

1 *J. Am. Chem. Soc.* 2017, 139, 29, 9795–9798

Computational investigation of NiOOH, from bulk to catalysis

Ruotao Yang*, Alexey A. Sokol, C. Richard A. Catlow

*Department of Chemistry, University College London

e05-react-**cat**

Mitigating environmental and ecological damage caused by human emissions are amongst the grand challenges of the 21st century. Therefore, the transition from conventional fossil energy to renewable energy, including solar and wind energy is an increasingly important topic. Power-to-gas (P2G), a process of converting renewable energy into hydrogen gas via electrolyser, is considered to be promising due to its carbon-free nature. However, to produce green hydrogen, the sluggish anodic reaction becomes the bottleneck, which reduces the efficiency of the electrolyser. To address such issues, a cheaper and more stable catalyst is desired. NiOOH (nickel oxyhydroxide) shows high suitability as an anode material for several electrocatalytic oxidation reactions. In addition, NiOOH is considered a promising electrode material for rechargeable batteries due to its proton-exchange capacity. However, limited knowledge of the bulk and surface structures of NiOOH, due to the lack of experimental evidence, caused by the complexity of the electrochemical environment, hinders a deeper understanding of its structure and reactivity.

To gain a deeper insight into the NiOOH bulk structure, we employ Ab Initio Molecular Dynamics (AIMD) based on density functional theory (DFT) to simulate its bulk structure. The initial configuration was obtained by removing a layer of protons in bulk $Ni(OH)_2$ to mimic the deprotonation process. The MD calculation showed a noticeable proton transfer, in which the proton layer immigrated to the neighbouring oxygen layer. The structure generated by AIMD was then optimized by DFT using with PBE+U functional. The as-obtained structure has a lower energy level, indicating that a mixture of $Ni(OH)_2$ and NiO_2 layer in the bulk system is favoured over a stoichiometric NiOOH layer. Furthermore, the electrostatic analysis suggested such proton transfer was caused by strong O-H interaction between the oxygen and hydrogen layers. Based on these results, we further cleaved its surface to investigate its catalytic activity in heterogeneous catalysis. We introduced the SO_4^{-2} anions on its surface as an adsorbate and a Ni-bulk system beneath the NiOOH surface to generate an SO_4^{-2}-NiOOH@Ni heterostructure. We show that the Ni-bulk system can stabilize the adsorbate and further promote the reactivity, which agrees with the experimental data.

Amorphous ZnO: Structure, Stability and Charge Trapping

David Mora Fonz

HTU, Mexico

TBA

Ab-Initio Molecular Dynamics Simulations of Substoichiometric Amorphous Alumina

J. Strand*, A. Shluger

*Department of Physics and Astronomy, University College London

e05-bulk-***shl***

Oxygen vacancies play an important role in the application of oxides in technology. The notion of a vacancy is well defined in the case of crystals, where a defect is simply any break from crystallographic order. In amorphous materials, where there is no long-range order, defects are less well defined. Amorphous oxides are widely used in technology, yet there has not been a great deal of focus on this ambiguity. Often, vacancies are modelled by deleting oxygen atoms from a stoichiometric amorphous model, previously produced using the melt-and-quench technique, and then relaxing the system geometry (‘post-quench’ method). However, this may not be the appropriate way to model substoichiometry in amorphous oxides. Here, we use an alternative approach where we produce substoichiometric amorphous oxide models by quenching from an oxygen-deficient melt (‘in-melt’ method). We use ab-initio molecular dynamics (AIMD) with a hybrid DFT functional to investigate the structure of substoichiometric alumina. The models produced are periodic and pseudo-amorphous, with 357 atoms in the unit cell. We find that there are vacancy-like point defects in the amorphous oxide, characterized by undercoordinated Al ions and formation of Al-Al dimers. In subsequent simulations, we take post-quench vacancies and anneal them with ab-initio MD at 1000K. The resulting configurations are similar to those produced with the in-melt method, suggesting that use of ab-initio MD allows for a fuller exploration of configuration space and therefore more stable ‘vacancy’ structures.

Overall, the results suggest that, while vacancies may be ill-defined in amorphous oxides, oxygen deficiency leads to the formation of point-defects with similar properties to crystallographic vacancies. Further, it is possible that AIMD is necessary to find the most stable configurations of these defects.

Unravelling Hydrogen Bonding Behaviours and their Impact on Proton Conductivity in Supramolecular Cages

Lei Lei, Sanliang Ling*

**Advanced Materials Research Group, Faculty of Engineering, University of Nottingham*

*e05-bulk-**lin***

Proton-exchange membrane (PEM) fuel cells hold promise as a solution for achieving a zero-emission economy. Recently, molecular cages have emerged as potential PEM candidates for stable and efficient fuel cells through structural design. However, due to their complex structures, understanding the proton transport dynamics and mechanisms using experimental or simulation methods has proven challenging. In this study, we employed *ab initio* molecular dynamics simulations to investigate these complex systems over extended timescales, spanning hundreds of picoseconds, to unravel these intricate systems. Additionally, we developed efficient python code to analyse the vast trajectory data generated. Our results highlight a significant disparity in the interaction of water with two molecular cages composed of the same functional groups, attributed to their distinct geometries. Notably, Cage 1, characterized by a spacious cavity, exhibits an average reduction of one hydrogen bond per water molecule. Consequently, Cage 1 displays faster water dynamics, a higher diffusion coefficient, and significantly diminished diffusion anisotropy. Our investigation underscores the critical role of hydrogen bonding behaviours between water molecules and functional groups within supramolecular cages, influencing water diffusion mechanisms and dynamics. Furthermore, we emphasize that precise control of hydrogen bonding behaviour through rational arrangement and passivation of strong electronegative atoms can further enhance proton conductivity within supramolecular cages.

Investigation of unintentional aggregation of impurity-related defects within 4H-SiC.

N. Smith*, A. Shluger

*Department of Physics and Astronomy, UCL

*e05-bulk-**shl***

To create p-doped regions within 4H-SiC MOSFET devices, 4H-SiC is bombarded with high energy Al^+ ions from gaseous AlH_3, during ion implantation. However, recent experimental studies have found shallow electron and hole donor states affecting channel mobility, whose densities are directly proportional to the Al-implantation dose even after post oxidation anneal in NO atmospheres. This suggests that Al-related defects present after the creation of the $(11\bar{2}0)$-4H-SiC/SiO_2 interface could be unintentionally interacting via aggregation with N- and O-related passivation defects at the interface.

We investigate the possibility of Al-O and Al-N complex formation within SiC devices, as well as whether such complexes can be correlated with the experimentally found mobility reducing states. In accordance with the charge compensation and site-competition theories, the $Al_{Si}N_CAl_{Si}$ and $Al_{Si}O_CAl_{Si}$ complexes have been examined using density functional theory (DFT) calculations. For each complex, four different geometry configurations corresponding to impurity substitution at different hexagonal (HEX) and cubic (CUB) lattice sites, derived using the Site-Occupancy Disorder package, were studied. Incorporation and binding energies, and charge transition levels (CTL) of these complexes in four different charge states were calculated in bulk 4H-SiC using DFT with the PBE0 functional.

Within bulk 4H-SiC, negative binding energies were obtained for all configurations, indicating that, if Al_{Si} and N_C/O_C are separately present within 4H-SiC, it is more energetically favourable for them to form aggregates. Negative incorporation energies were also obtained for all eight configurations when the impurity chemical potential sources were taken to be AlH_3, NH_3, and NO, in correspondence to the device creation process. Within the 4H-SiC bandgap, the -1/0 charge transition is found to be the only stable transition for both complexes. The -1/0 charge transition levels of three out of four $Al_{Si}N_CAl_{Si}$ geometry configurations were found to occur within the activation range of the I_D-DLTS identified trap state. Using NH_3, rather than NO, for the chemical potential of N resulted in even lower incorporation energies for the $Al_{Si}N_CAl_{Si}$ configurations, implying the formation of the $Al_{Si}N_CAl_{Si}$ complex is easier under anneal in NH_3. This aligns extremely well with experimental measurements which find the density of the shallow hole donor interface state to be higher after annealing with NH_3 than with NO. For the $Al_{Si}O_CAl_{Si}$ complex, the total and incorporation energies of each charge state were found to be lowest for the $Al_{Si(HEX)}O_{C(HEX)}Al_{Si(CUB)}$ configuration. The -1/0 transition level of this configuration was found to be at E_c-0.004 eV, within the activation energy range of the shallow electron donor state observed experimentally. These results demonstrate how Al-O and Al-N complexes, can be responsible for channel mobility degradation in SiC devices.

Simulation of correlation driven metal-insulator transitions in real solids using deep neural networks.

H. Sutterud[*], G. Cassella, W. T. Lou, D. Pfau, J. Spencer, W. M. C. Foulkes,

[*]*Physics, Imperial College London* *e05-bulk-**fou***

The use of deep neural networks (DNNs) for representing fermionic wavefunctions is gaining popularity. DNNs have demonstrated impressive results in systems such as molecules and nuclear systems as well as periodic systems such as the homogeneous electron gas and solids.

One favourable trait is the inherent flexibility of neural networks. Standard ab initio methods for solid systems often rely on uncontrollable approximations, such as the choice of a functional in density-functional theory or the form of the Slater-Jastrow trial wavefunction in quantum Monte Carlo methods. DNN ansätze, on the other hand, are built from multilayer perceptrons, which are universal approximators. The network weights and biases that define a DNN Ansatz are efficiently optimized using variational Monte Carlo methods and beyond-first-order optimizers. This produces results of consistent quality across highly diverse systems, and makes it possible to discover quantum phase transitions without prior knowledge of their existence.

In this talk, we investigate the interaction-driven Mott transition between metallic and insulating states of solids. Although Mott's first paper on the subject was published in 1949, some aspects of the transition are still not fully understood. Using the fermionic DNN architecture FermiNet to represent a supercell of bulk lithium, a metal-insulator transition with the characteristics of a Mott transition occurs spontaneously as the density of the system is adjusted. We will present our findings and discuss the nature of the transition in light of these results.

The effect of ceria surface composition on scavenging reactive oxygen species

Khoa Minh Ta*, David J. Cooke, Lisa J. Gillie, Roger M. Phillips, Marco Molinari

*Department of Chemistry, University of Huddersfield

*e05-surfin-**mol***

Cellular activities produce reactive oxygen species (ROS), such as hydrogen peroxide, superoxide anions, and hydroxyl radicals, which can accumulate and produce oxidative stress. This is linked to many degenerative diseases and cancer. Ceria nanoparticles (CeNPs) have been reported to perform enzyme mimetic activities to scavenge ROS. Here, we study the effect of surface composition on the energetic of the adsorption of such ROS. We use the most stable surfaces of ceria, i.e. {100}, {110}, and {111} surfaces, which are present in many active CeNPs, with surface compositions that include bare stoichiometric, hydroxylated, oxygen deficient and hydroxylated / oxygen deficient surfaces. We found that the three low Miller index ceria surfaces can perform both catalytic (where surface composition remains unchanged) and chemical (where surface composition changed) pathways for the scavenging. Our results suggest that hydroxylated surfaces would have limited scavenging activity compared to oxygen deficient surfaces. However, oxygen deficient surfaces may deteriorate over time due to the irreversible healing of surface oxygen vacancies.

Modelling the effect of H in the formation, stabilisation and evolution of voids in polycrystalline Cu

Vasileios Fotopoulos *, Alexander L. Shluger*

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

Cu is used in a wide variety of applications ranging from power generation to microelectronics. Stress-induced voiding is amongst the most commonly reported defects in Cu.[1] Hydrogen plays a crucial role in Cu embrittlement and can be detrimental to the performance of electronic devices.[2,3] However, the correlation between hydrogen embrittlement and vacancy formation and aggregation leading to voiding in polycrystalline Cu is not yet fully understood. Furthermore, since measuring experimentally the amount of incorporated H in metals remains challenging, a theoretical approach is needed.

In this work, to efficiently describe the effects of H on the mechanical properties of Cu, we combine density functional theory (DFT) with bond order molecular dynamics (BO-MD). Our MD simulations Cu vacancies and H complexes in grain boundaries (GBs) demonstrate that they can withstand temperatures up to 700 K without dissociating.[4] Thus, H increases the stability of vacancy clusters in GBs during annealing. Under uniaxial strain, H at concentrations higher than 10 mass ppm facilitates the formation of partial dislocations close to the GBs, leading to a reduction of the yield strength of the crystal. Using statistical mechanics and inputs from DFT simulations, we determine equilibrium concentrations of vacancy clusters in Cu at different temperatures and study synergy effects between H and Cu vacancies. The H concentration for up to 100 bar partial pressure values and close to the melting point of Cu was orders of magnitude lower than the estimated concentration of Cu vacancies.[5] At this low H concentration, the interaction between H and Cu vacancies is negligible and becomes significant only for H partial pressures of 1 GPa or above. Therefore, the amount of H in Cu is sufficient to play a role in the initiation of voids only under extreme H pressure conditions. These results put thermodynamic limits on the hydrogen-induced vacancy clustering and void formation in bulk Cu.

[1] Wu, Z. et al. 2008. *Microelectronics Reliability*, 48(4), pp.578-583.
[2] Zarnas, P.D. et al. 2021. *International Journal of Solids and Structures*, 213, pp.103-110.
[3] Djukic, M.B. et al. 2019. *Engineering Fracture Mechanics*, 216, p.106528.
[4] Fotopoulos, V. et al. 2023. *TMS 2023 152nd Annual Meeting & Exhibition Supplemental Proceedings*, pp. 1001-1010. Cham: Springer Nature Switzerland.
[5] Fotopoulos V. et al. 2023. *Physical Chemistry Chemical Physics*. 2023. 25, pp.9168–9175.

Simulating Hydrogenation in Magnesium Through the Application of Machine Learning Models

Oliver M. Morrison, Sanliang Ling, David Grant and Gavin Walker

[*] *Advanced Materials Research Group, University of Nottingham* *e05-surfin-lin*

In this contribution, we simulate magnesium hydride systems through the application of machine learning (ML) models, with the goal of identifying the rate limiting steps of the hydrogenation process. If metal hydride systems are to reach their full potential, it is vital that we develop a detailed fundamental understanding of how these systems evolve. Experimentally, it is difficult to pinpoint the factors that contribute to the differences between metal hydride systems, e.g differences in the kinetics of the hydrogenation and dehydrogenation reactions. Accurate molecular dynamics (MD) simulations of metal hydride systems could shed light on the rate limiting steps of the hydrogenation and dehydrogenation reactions, but traditional approaches to approximating the potential energy surface, such as classical force fields and density functional theory (DFT), are either lacking in accuracy or computational efficiency. Accurate large size, long-timescale MD simulations are therefore infeasible via these methods. ML has been applied to an abundance of problems, to assign simple functions to complicated patterns. A generic many-parameter model is designed, and parameter values are tuned to fit the model to specific examples. We apply the same concepts to produce a model that approximates the potential energy surface, mimicking the predictions of DFT with a less complicated, less computationally intensive model. This enables the simulation of large-scale atomic systems for longer time scales. The nature of ML incurs the risk of transferability issues; the model may extrapolate from our examples incorrectly when it encounters a particularly unfamiliar atomic structure. In such an event, we perform DFT on these structures and add them to set of examples used to train the model. Through repeated improvement of the ML model, we hope to identify the rate limiting steps of the hydrogenation process in magnesium, and consequently, potential improvements to the development of new catalysts to accelerate the reaction kinetics of metal hydride systems.

Study of Hydroxyl Nests in Zeolites

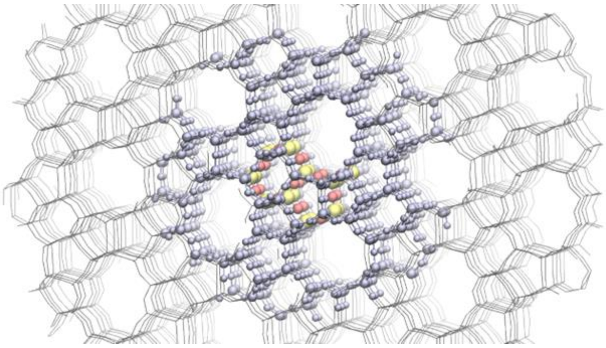
Alec Desmoutier

* *University College London* *surfin-cat*

For a long time, zeolites have been utilised as a material for various experiments in the petrochemical industry as a catalyst, however, the mechanism in which these zeolites interact has remained a mystery. The current theory is that hydroxyl nests are generated as an intermediate step in reaction within zeolites but haven't been tested computationally.1

A hybrid QM/MM approach using the ChemShell software was employed to study CHA/ZSM-5/Alpha Quartz defects. The model comprises a quantum mechanical description of a cluster of ~200 atoms around the reaction site near the porous channels, embedded in a much larger cluster modeled using a classical molecular mechanic's forcefield.2, 3

In this study, we have shown the results for the creation of hydroxyl nests as well as the aggregation of these defects to generate larger pores. Our results show the comparison of generating these defects from a point of origin outwards vs the removal of the key ring from the zeolitic framework. The formation of various mesopores that is found in the literature is a key aspect that is being aimed to emulate and recreate. Study into the introduction of metals (in this case Platinum) into the framework of the hydroxyl nest as an anchoring site for larger metal clusters to form around.



Cluster of ZSM-5 generated, the QM region (yellow and red atoms), and the active region (grey atoms) are highlighted.

Bulk and Surface Contributions to Ionisation Potentials of Metal Oxides

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Obtaining absolute band edge positions in solid materials remains an inherently complex task from both experiment and theory, and indeed there is considerable confusion in the literature. Our work shows clearly that the ionisation potential (IP) and electron affinity (EA) of metal oxides emerge as surface-related parameters, exhibiting high sensitivity to various factors, such as material morphologies and processing histories. A particularly intriguing example that exemplifies this complexity is provided by ceria (CeO_2), where experimental measurements have yielded several electron volts of discrepancies in its IP. Therefore, developing a comprehensive understanding of how the bulk and surface contribute to the IP of metal oxides would shed light on the experimental discrepancies and our understanding of the band alignment between different materials, thereby greatly benefitting the development of novel advanced photocatalysts and photovoltaic cells.

We have employed a combination of theoretical approaches, from classical electrostatics to quantum mechanics, to elucidate the bulk and surface contributions to the IP of metal oxides. By carrying out electrostatic analyses supported by interatomic potential techniques, we discern and quantify the various sources of intrinsic surface effects contributing to energy-level shifting in metal oxides. By employing a specially designed hybrid QM/MM embedded-cluster model that excludes all surface effects, we have determined a theoretical bulk IP for CeO_2 of only 5.38 eV. Changing surface orientation results in intrinsic IP variations from 4.2 eV to 8.2 eV, as revealed by plane-wave DFT calculations with slab models. Such low bulk contributions to IPs (5.4-6.1 eV) and high degree of tunability from surfaces were also found in ZrO_2 and HfO_2, in which surface polarisation plays a crucial role. Our analysis, in addition to rationalising the observed broad range of experimental results, provides a firm basis for future interpretations of experimental and computational studies of oxide band structures. Rational exploitation of these relationships could benefit the design of novel photovoltaic and electronic devices towards higher efficiencies.

Bismuth oxychalcogenide materials for thermoelectric applications

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In order to effectively combat climate change a wide range of approaches are required. One route is to increase the efficiency of energy-intensive process to reduce the associated CO_2 emissions. A 2014 study showed that in the UK alone 48 TWh yr^{-1} of energy is wasted as heat,[1] meaning that recovering 1 % would equate to a reduction of >250,000 Tonnes yr^{-1} of CO_2 emissions.[2]

Oxychalcogenides are a class of prospective high-performance thermoelectric materials that combine the favorable properties of oxides, *viz.* earth abundance and thermal stability with the desirable electrical properties and low thermal conductivity of chalcogenide materials, the latter of which have some of the largest reported ZT values e.g. SnSe ($ZT = 2.9$).

We have applied a fully *ab inito* modelling protocol to predict the ZT of the bismuth oxychalcogenides Bi_2OCh_2 (Ch = S, Se or Te) as prospective p- and n-type thermoelectrics. Lattice-dynamics calculations were performed to model the phonon spectra and lattice thermal conductivity, and electronic structures calculated using hybrid density functional theory were to model the Seebeck coefficient, electrical conductivity and electronic thermal conductivity.

All three oxychalcogenides are predicted to show low lattice thermal conductivities of < 3 W m^{-1} K^{-1} at 300 K. However, in most cases the high electrical thermal conductivity hampers the overall ZT except for p type Bi_2OS_2 and Bi_2OSe_2, for which we predict maximum ZT values of 2.82 and 2.99, respectively.

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Relating the formation energies for oxygen vacancies defects to the structural properties of tungsten oxides

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e05-power-mur

To address the growing demand for low carbon energy, the UK government has recently committed to building the world’s first nuclear fusion power plant by 2040, the Spherical Tokamak for Energy Production (STEP). One of the challenges faced by the STEP project concerns the oxidation of the reactor’s tungsten-based first wall, which may occur during a loss of coolant accident or during remote handling during maintenance.

The oxidation of tungsten is a complex process, starting with the formation of a protective WO2.72 layer. This layer then cracks leading to rapid oxygen transport to the oxide/metal interface and the formation of columnar WO2.9. The abundance of vacancies in WO2.9 leads to rapid growth of the oxide layer and progressive transformation to WO3. WO3 is permeable to oxygen, and its formation rate depends on oxygen ion transport to the WO3/WO2.72 interface. Therefore, in this work we use density functional theory to study the physical properties of all oxide phases involved in the oxidation process. From the DFT simulations we show how key features of the electronic structure of the WOx materials change as the metal-oxygen ratio evolves. Then we calculate the formation energies for oxygen vacancies and activation energies for their diffusion allowing an assessment of their mobility in the different tungsten oxide phases. Our results provide a new level of understanding of the sub-stoichiometric Magnéli phases that are observed during the oxidation of tungsten.

Controlling the heat transport in thermoelectric materials

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Around 60 % of the energy used globally is wasted as heat, with clear implications for climate change. Thermoelectric (TE) power can address this by recycling waste heat to electricity, harnessing the Seebeck effect in a thermoelectric material to extract energy from a temperature gradient. Thermoelectric generators (TEGs) are established in the aerospace industry and have potential applications from IoT devices, to automobile engines, to repurposing decommissioned oil rigs as geothermal power plants.

An ideal thermoelectric material requires a high Seebeck coefficient and electrical conductivity and a low thermal conductivity. However, compared to the electrical properties the heat transport through the lattice vibrations (phonons) is less well understood, and less is known about how to control it.

The lattice thermal conductivity can be modelled using techniques such as the single-mode relaxation-time approximation, and theoretical calculations have proven valuable for understanding the low thermal conductivity in flagship TEs such as PbTe and SnSe. This talk will build on these success to discuss how insight from modelling studies can be used to suggest structural modifications to control the heat transport in thermoelectric materials.

The constant relaxation-time approximation (CRTA) analysis allows differences in thermal conductivity to be attributed quantitatively to the phonon group velocities, which depend on the atomic masses and the chemical bond strength, and the phonon lifetimes, which depend on anharmonic phonon-phonon interactions. In materials with large group velocities the thermal transport can be suppressed by alloying, as in e.g. $Sn(S_{1-x}Se_x)$, or by chemical substitution with "discordant" dopants. In materials with long lifetimes, introducing loosely-bound "rattler" ions, such as in $CoSb_3$, can reduce the thermal conductivity through "resonant scattering" of the heat-carrying modes. While this has yet to be explored in depth, calculations on the hybrid perovskite $(CH_3NH_3)PbI_3$ suggest that small-molecule rattlers may be particularly effective.

The Baskerville Tier-2 Service

Jon Wakelin

University of Birmingham

TBA

The GW4 Tier-2 Service

Steven Chapman

University of Bristol

TBA

Oxygen-functionalised graphene: structure, properties and potential for phosphate sensing

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[*]*Department of Chemistry, University of Sheffield* *e05-enviro-**nat***

Graphene is an outstanding material thanks to its excellent electronic and optical properties. Graphene is also a promising sensor material, which has been used in sensors for volatile gas molecules and as an electrode in electrochemical measurements. Thus, graphene already enables high-sensitivity measurements; however, selectivity is a problem, which may be addressed by functionalizing graphene with various functional groups. In this project we collaborated with experimental partners to develop graphene-based electrodes to detect phosphate, an essential plant nutrient, in water and in soil. In particular, we used theoretical modelling to investigate how the nature of oxygen functionalization affects electronic properties of graphene and its interaction with adsorbed phosphate.

Density-functional theory calculations were used to model flat and curved graphene sheets containing epoxide and hydroxyl groups and substitutional oxygen at different concentrations (1-12% oxygen). Curvature was found to have little effect on the electronic properties of graphene: curved graphenes remained semimetallic, and their optical absorption spectra were similar to graphene. However, oxygenation had significant effect on the properties, with new states at the Fermi level for graphenes containing substitutional oxygen, and gap opening in functionalised graphenes with oxygen content above 6%. Moreover, additional peaks appeared in the simulated optical absorption spectra of oxygen-containing graphenes. These results show that oxygenation significantly changes these materials' optical and electronic properties, potentially leading to new applications in electronics and sensors. Following this, we investigated how different phosphate species (PO_4^{3-}, HPO_4^{2-}, $H_2PO_4^-$, H_3PO_4) can adsorb on pure and oxygenated graphene. All the considered phosphates were physisorbed, with hydroxyl-containing graphene offering the strongest adsorption. Significant changes in electrical conductivities before and after adsorbing phosphate species on these pristine or functionalized graphene sheets were found, suggesting that oxygenated graphene is a good candidate for soil phosphorus sensor material.

An Efficient Model for sp-Lone Pair Cations

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A large class of material both of fundamental and applied interest is made by compounds of lone pair cations, notably, Sn(II), Pb(II) and Bi(III), with localised off-centre lone-pair lobes. In materials, the lone pairs remain chemically inert but are highly polarisable, which results in the stabilisation of distorted crystal structures and a wide range of complex physicochemical phenomena.

We propose a novel type of the force field, based on embedded atomic orbitals centred on lone pair cations to provide an efficient method for the study of this class of material.

To account for the formation of the lone pair lobes on cations, we focus on the hybridisation of s and p type cation valence orbitals and represent them as localised atomic orbitals. In this presentation, we will provide the theoretical basis of the lone pair model, including the full details of energy and derivatives of lone pairs interacting with atoms described in turn with traditional interatomic potentials. We will demonstrate the success of the model on application to nanoclusters of lead monoxide and difluoride. The results show that the lone pair model is efficient and accurate in predicting and reproducing the nanocluster atomic configurations obtained using density functional theory.

Fundamental Principles of a Virtual Tokamak Fusion Reactor

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The ambitious timescale of the international fusion reactor development programme implies the increasing emphasis on virtual engineering, in turn creating a strong drive towards the exploration of digital twins and advanced computer models for materials compatible with virtual engineering, to enable the assessment of a variety of expected operating conditions. This presentation examines the fundamental principles and applications of self-consistent multiscale materials modelling for predicting the gradual evolution of deformations and stresses at the reactor component scale, resulting from the continuous exposure of a mechanically loaded reactor structure to neutron irradiation – a fundamentally atomic scale microscopic phenomenon. The topics addressed in the presentation include the eigenstrain method for evaluating macroscopic deformations from the density of microscopic defect volume tensors, which can now be derived from *ab initio* density functional theory and larger scale atomistic simulations. There are now new dynamic atomic-scale simulation algorithms for treating non-linear high dose effects, predicting the power law statistics of defect structures, also discovered in the recent dedicated experimental observations of complex evolving defect microstructures.

Optimisation of decision sequences from selection of elements to chemical formulae of functional materials

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[*] *Department of Chemistry, University of Liverpool* *e05-discov-**dar***

The workflow for materials discovery – from selection of chemical elements to combine (Fig 1a) to identification of functional high-performing candidates (Fig 1b) to establishing experimentally accessible formulae (Fig 1d) – presents a complex hierarchy of consequential decisions. Development of quantitative guides is thus imperative to aid decision-making, increase the success rate and accelerate the discovery workflow. Recent advances in learning the patterns of properties-materials relationships from historical data have enabled a range of powerful techniques for prediction of functional performance for materials. Applied at the level of the periodic table, these techniques have enabled ranking of elemental combinations regarding the likelihood of forming new materials[1].

In this talk, I will present new capabilities for learning from materials data at two stages of the discovery workflow: phase field selection (PhaseSelect[5]) and stoichiometry optimisation (PhaseBO[6]). PhaseSelect learns from data about chemical elements themselves and discovers their contributions to functional properties of materials, e.g., such as superconductivity or magnetism; then it identifies the promising elemental combinations for new materials in terms of their functional performance. PhaseBO accelerates the exploration of all possible combinations of selected elements and increases the probability of the discovery of practically accessible new materials.

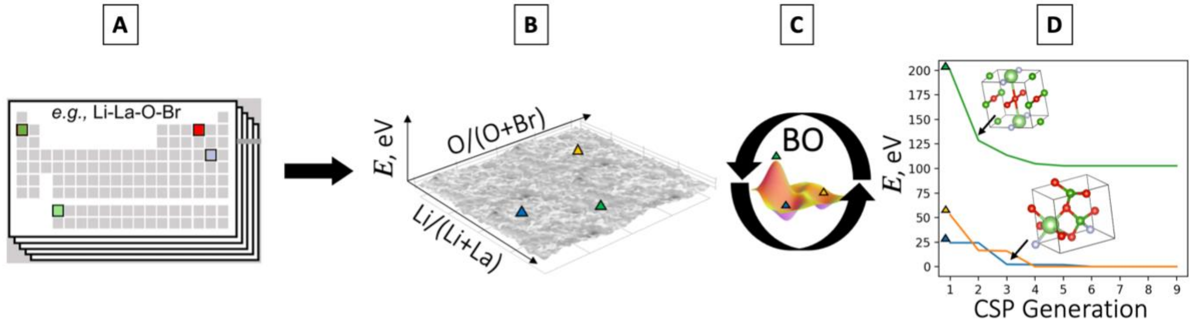

Figure 1. Workflow of materials discovery: from selection of elements for function to identification of practically accessible new materials

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Computational Modelling of Wide Band Gap Transparent Conducting Oxide Sb_2O_5

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Transparent conducting oxides (TCOs) combine optical transparency and electrical conductivity, making them indispensable in the fields of optoelectronics. However, the limited number of post-transition metal TCOs restricts the range of devices they can support. Recently, Ga-doped $ZnSb_2O_6$, an Sb(V)-based oxide with unique band alignment, has been computationally and experimentally examined, offering much-needed diversity to the field.[1] Inspired by this success, an underexplored post-transition binary Sb(V)-based system, Sb_2O_5 is considered a promising candidate due to its wide band gap and comparable electronic structures to the other successful TCOs.

In this work, we use hybrid density functional theory to investigate the electronic structure of Sb_2O_5, where wide fundamental and optical band gaps are found, enabling transparency. Furthermore, we apply the Phonopy to prove the dynamic stability of Sb_2O_5 by analyzing its phonon dispersion.[2] We simulate charge transport properties using the AMSET code.[3] We demonstrate the band alignment using surfaxe,[4] in which Sb_2O_5 has the largest ionization potential and electron affinity among the existing TCOs. A large thermodynamic stability region can be seen by comparing Sb_2O_5 with its competing phases. Lastly, we conduct an in-depth investigation of the intrinsic defect chemistry of Sb_2O_5 where a large dopability window is present. To facilitate this study, we use ShakeNBreak to generate the defective supercells, with subsequent defect analysis carried out using the DOPED code.[5]

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A computational study characterising the intrinsic and extrinsic doping profile of ZnS

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[*]*Department of Chemistry, University College London*

e05-discov-dos

Zinc Sulfide (ZnS) is a widely synthesised material, largely due to its potential in luminescence devices and optoelectronics (sensors, LEDs and photovoltaics, among others) stemming from a wide and direct bandgap of 3.6 eV, alongside an inexpensive manufacturing cost and an environmentally benign nature. Despite this, there have been few studies investigating the electronic and optical properties of the material, and even fewer attempts to form a connection between the two. A noteworthy experimental paper by Hoshina et al. [1] suggested interesting excitonic behaviour dependent on n/p-type co-dopant proximity, and recent advances in computational modelling tools provide an opportunity to investigate this and other metastable states with more certainty.

The aim of this study is to construct a first-principles assessment of intrinsic and extrinsic defects in zinc-blende ZnS, using the VASP [2] density functional theory code to identify the relaxed electronic structures and corresponding optical properties. The hybrid functional HSE06 was used to accurately identify the energies of ZnS and its defect structures, along with competing phases.

This study also uses the *ShakeNBreak* [3] structure searching algorithm to scope the energy landscape and identify the global minimum structure, separating it from local minima that might lead to inaccurate excitation mechanisms. This is performed for intrinsic (antisites, vacancies and interstitials) and extrinsic (n/p- doped) point defects, as well as donor-acceptor defect complexes within ZnS, with appropriate defects generated using the *DOPED* [4] software tool. Following an analysis of the formation energies and spin-dependent charge localisation in each system, we relate each structure to a configuration coordinate diagram, identifying the excitation mechanism and optical properties associated with different dopants. This study will undoubtedly prove useful when assessing the capability of similar structures in optoelectronic applications.

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Organosulfide-Halide Perovskites incorporating Zwitterions

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*e05-discov-**fil***

Halide perovskites have been the leading contender for low-cost and efficient solar cell materials, stimulating efforts to design of novel semiconductors with tunable bandgap in the visible range. Cation and/or halide mixing is a popular strategy to achieve this. However, these give rise to other undesirable effects such as halide segragation. Additionally, chalcogenide perovskites such as $BaZrS_3$ and $SrZrS_3$ also have bandgaps in the visible range with robust stability under ambient conditions. These materials, however, require high temperature for synthesis (800-1000 ◦C). A common characteristic of these materials is that the band edges primarily consist of B-site cations and anions contributions.

The new class of chalco(S)-halide(Cl,Br) perovskites have been synthesised at low temperature (150 ◦C) [1]. In these compounds, the chalcogen S was introduced via the zwitterion cysteamine ($^{+}H_3NCH_2CH_2S^{-}$), where the S occupies 1/3 of the anionic sites and the long organic chain acts as the A-site cation, stabilising these compounds. In this study, we used first principles DFT calculations to compute the electronic properties of this new materials class. We found that these new perovskites are also direct band gap semiconductors, with band gaps in the visible range, tunable by the halogen composition. In this talk, I will present our first principles study of the electronic properties of this new material family in comparison with lead halide perovskites.

We acknowledge funding support from EPSRC (EP/V010840/1), ARCHER2 Supercomputing Service and Brown Science Foundation.

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Prediction of critical micelle concentrations in surfactant mixtures by DPD simulations

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Invited Talk

The critical micelle concentration (CMC) has become a key design target in detergent formulation since the CMC correlates with relevant physicochemical properties. Most of applications feature surfactant mixtures, due to design or to the polydisperse nature of certain surfactants. Hence the motivation on modelling and predicting the behavior of surfactant mixtures and blends.

In this presentation, we follow the methodology presented by del Regno et al. [A. del Regno, J. Phys. Chem. B 125, 5983-5990 (2021).], where the pseudo-phase separation model (PSM) is applied to predict CMC of binary mixtures. The methodology consists on running dissipative particle dynamics (DPD) simulations of high-concentration surfactant mixtures. The molar fractions of every compound in both pseudo-phases is obtained and related to the non-ideal mixing parameters via the theory. Finally, in the limit of low concentrations, the mixture CMC is predicted as a function of the composition. The approach is applied to sodium laureth sulfate (SLES) surfactants with polydispersity in the ethoxylate spacer.

Structural and dynamic properties of poly(styrene-co-maleic anhydride) at the interface of conjugated polymers

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Conjugated polymers are employed in a variety of application areas due to their bright fluorescence and strong biocompatability. For these applications, readily fabricated conjugated polymer nanoparticles (CPNs) are formed using capping agents to provide stable and soluble conjugated polymer formulations. Post-assembly functional modification and chemical doping can be used to further tune CPN properties for target applications.

The nanoscale structure of conjugated polymer materials emerges from interactions between different molecules, and holds significant control over their much-prized optical properties. Reliably controlling the optical properties of conjugated polymers upon aggregation and assembly remains a challenge. The structural properties of the interface between the capping agents and the conjugated polymer play a key role in the ability to then functionalise the surface of the conjugated polymer formulation with any specific molecules required for tuning the delivery of a CPN to a desired target. In this presentation, I will present the results of a series of all-atom molecular dynamics simulations that we have conducted in order to provide a molecular scale understanding of the structural and dynamic properties of poly(styrene-co-maleic anhydride) (PSMA), which is a common capping agents for CPNs, on the interface of the conjugated polymer, poly(9,9-di-n-octylfluorene-alt-benzothiadiazole) (F8BT). I will compare the properties of the PSMA capped interface with those which are found when using another commonly used capping agent, Brij O10. Finally I will discuss the effect of the difference in the interfaces formed on the functionalisation of these polymer interfaces.

Iron-sulfur peptides for an efficient electron transfer to the hydrogenase enzyme

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e05-react-**ter**

In the green alga *Chlamydomonas reinhardtii*, the hydrogenase HydA1 catalyses at an iron-sulfur site the conversion of protons and photosynthetic electrons to H_2. The electron transfer to HydA1 is mediated by the iron-sulfur unit of ferredoxin PetF. However, PetF does not efficiently donate its electrons to HydA1, thus limiting the yield of H_2 [1]. In this presentation, I will investigate the replacement of PetF with two small iron-sulfur peptides (PM1 and FM1 hereafter) that hold potential for an improved electron transfer to HydA1 [2]. Using an energy-based scoring function, I will show that FM1 and PM1 bind to two different surface regions of HydA1. The distance between the two iron-sulfur clusters in FM1/HydA1 is shorter compared to PM1/HydA1, ensuring a more efficient electron transfer. This finding is in line with the reported higher H_2 production rates for FM1/HydA1 in vitro. I will also show that the redox potential of the peptides, and therefore their electron transfer properties, can be changed by single-residue mutations in the secondary coordination sphere of the iron-sulfur cluster. In particular, taking advantage of molecular dynamics, I have designed a PM1 variant with a bulkier side chain that is able to disrupt the hydrogen-bonding network with water. The resulting redox potential, calculated within the linear response approximation [3], is shifted by about –60 mV compared to the original peptide. These results will guide experiments aimed at replacing PetF with novel peptides that enhance the photosynthetic H_2 production in the alga.

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How bulk and surface properties of Ti_4SiC_3, V_4SiC_3, Nb_4SiC_3 and Zr_4SiC_3 tune reactivity: A computational study

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*e05-react-**cat***

The "MAX"-phase of over 100 ternary carbides/nitrides was first identified in the 1960s.[1] They display a very novel combination of properties including high conductivity and extreme resistance to oxidation or heating, which has led to these materials being labeled as "metallic ceramics".[2] Structurally, the surfaces of these materials strongly resemble the pristine (111) facets of early transition metal carbides (TMCs), which have themselves been shown to be efficient catalysts for the hydrogenation of CO_2. Our group has previously undertaken a detailed in silico study into the catalytic activity of the equivalent TMCs to the MAX-phase material under investigation in this work and found the metal terminated (111) facets to be extremely active for CO_2 reduction.[3] The choice of the transition metal components was partly informed by this study and partly by a systematic screening of the bulk and surface properties of a diverse array of carbides.[4] This talk will focus on results published as part of a recent Faraday Discussion will present the results from a computational study into the bulk and catalytic properties of Ti_4SiC_3, V_4SiC_3, Nb_4SiC_3 & Zr_4SiC_3 and compare these properties to those already obtained for TiC, VC, NbC & ZrC.[5] We will show that the addition of an intestinal silicon layer into the bulk of these material increases the lattice paraments beyond those observe for the corresponding carbides. We also show that the pristine surfaces of the MAX-phase materials are much more active towards CO_2, H_2, H_2O and OH. However, our results demonstrate that pristine surfaces are not likely to be present in an oxygenating environment. Instead, the modelling predicts all surfaces will oxidise and barriers for CO_2 reduction will increase in the presence of H_2O, O_2 or OH. Current, work supported by the MCC is looking into the activity of these oxidized surfaces.

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Posters

Non-linear moments modelling of metal, oxygen and hydrogen binding to metallic surfaces

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e05-surfin-shu

The adsorption energy E_{ads} of metal, oxygen and hydrogen adsorbed on a series of strained FCC and HCP transition metal surfaces has been modelled using the first four moments $\mu_{(1-4)}$ of the clean surface d-states [1-2]. For compressive and tensile surface strains of between -5% and +10% the adsorption energy E_{ads} has been shown to be effectively modelled by a degree 6 polynomial expansion of $\mu_{(1-4)}$.

The physical justification for these models is discussed in terms of the distortion of the surface d-states during bonding, and extends the effectiveness of the conventional d-band model the which only uses first adsorption moment $\mu_{(1)}$ i.e. the centre of the surface d-band, to cases where the surface is controllably distorted. The current work discusses the changes to the width, skewness, and bimodality of the surface electronic states of the Pt-block metals under strain, and also shows the non-linear character of adsorption under strain.

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Advances in sustainable catalysis: A computational perspective

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The urgent need for society to move towards a greener and more sustainable future presents a very exciting opportunity for catalytic chemists. Many of the necessary changes in resource management and increased energy efficiency will be propelled by the directed design of new catalysts, for which a detailed theoretical understanding of the activity of current catalysts is a crucial part. Many very different computational techniques are being applied across Hub project's to the characterization of novel catalysts as a preliminary step to the engineering of new and much greener chemical route to important products. This poster will depict the implementation of a QM/MM protocol to explore the bioengineering potential of OleTJE, in order to increase its selectivity toward olefin production. There will also be a demonstration of the use of restricted cluster model calculations to investigate the ability of HOD to catalyze spin-forbidden oxygen activation.

An example of sustainable homogenous catalysis will be a study explored the reactivity of a novel μ-nitrido-bridged diiron-oxo porphyrin that was able to catalyze the methane to methanol reaction under very mild conditions. This study used unrestricted DFT methods to determine that the acidity of the FeO anion was mostly responsible for its increased activity over the related mono-oxygen porphyrin catalysts. The final example explored in this poster will concern several periodic DFT studies into the electronic properties and catalytic abilities of the low-index facets of early transition metal carbides. These studies point to the possibility of green catalytic routes towards the production of fuels and useful chemicals from the utilization of the green-house gas carbon dioxide; as well as the potential for these materials to be used as catalysts in hydrogen fuel cells. Taken together these example will demonstrate some of the powerful computational studies preformed as part of the UK Catalysis Hub's remit to model the next generation of powerful catalytic systems for a future circular economy.

Capturing the ground state electronic and magnetic properties of Cobalt containing metal and mixed metal oxides using density functional theory (DFT).

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e05-surfin-**log**

The Fischer-Tropsch (FT) reaction is important to mitigate chemical waste and produce sustainable hydrocarbon fuels. Hydrogenation of CO is an endothermic reaction, and so requires the use of catalysts to make the process economically viable. Transition metals like Ru, Fe, Co, and Ni have been demonstrated, yet further work is needed to understand the catalyst efficacy.[1] Cobalt-containing metal oxides (e.g. Co_3O_4) have been used as a catalytic precursor and reduced to obtain the active catalysts for Fischer-Tropsch (FT) synthesis, along with transition metal ions such as Mn^{2+} that act as promoters.[2] The active catalyst species is the Co metal atom obtained by reducing the metal oxides, which are dispersed in form of clusters across a support such as TiO_2, SiO_2, or Al_2O_3.[3] In the Mn-promoted Co/TiO_2 FT reaction, an increase in Mn loading enhances the selectivity towards alcohols (oxygenates) and olefinic products. Advanced characterization techniques reveal the presence of spinel-like structures, with the formula $Co_xMn_{3-x}O_4$, in the catalyst precursor, indicating transition metal interactions.[4] However, the nature of the interactions between Co, Mn and the support, contributing to enhanced Co metal dispersion and oxygenate selectivity is still evasive.

As a result, understanding the effect of Mn promoters in the formation of active catalyst would require an accurate description of the properties of metal oxide species involved in catalyst synthesis, from Co_3O_4 to $Co_xMn_{3-x}O_4$ and finally to Co metal. The magnetic properties of Co^{2+}, Mn^{2+} and Mn^{3+} ions in these metal oxides add an additional dimension to the problem. Our simulations of antiferromagnetic (AFM) CoO and MnO, having a rocksalt structure show distortions during geometry optimizations, reducing the symmetry from cubic to rhombohedral. Additionally, equation of state (EOS) calculations show that the antiferromagnetic (AFM) state is energetically preferable. The ordering of the magnetic moments within the crystal systems, like CoO and MnO, significantly distorts the geometry (lattice parameters) below their respective Neel (T_N) or Curie (T_c) temperatures as observed experimentally.[5] Our findings lay the groundwork for the development of models of precursor nanoparticles, with future work focusing on the impact of spin on surface structures.

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Modelling hole trapping in hydrogen-related defects at the c-Si/a-SiO_2 and c-Si/a-SiO_2/a-HfO_2 interface

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The performance metal-oxide-semiconductor field-effect transistors (MOSFETs) is affected by random telegraph noise and bias temperature instability. These effects are caused by the trapping and release of holes and electrons in the dielectric oxide layer, but the atomistic nature of these processes is unclear. This research is focused on the DFT modelling of the effect of Negative Bias Temperature Instability (NBTI) in c-Si/a-SiO_2/a-HfO_2 devices. We aim to evaluate the accuracy of the Reaction Diffusion Drift (RDD) model, which suggests that the hole-induced dissociation of hydrogen-passivated defects can explain the trap generation kinetics in many types of devices.

We investigate how H-related defects in amorphous SiO_2 can be involved in hole trapping and hydrogen release, making them possible candidates for NBTI. Using non-local density functional we investigate how H-passivated oxygen vacancies and 3-coordinated silicon centres trap holes and release protons in bulk a-SiO_2 by reporting barriers for proton dissociation and defect charge transition levels with respect to the Si/SiO_2 interface using the rigid band approximation. We also probe the effect of trap levels of pre-existing defects with respect to the distance to the interface using models of c-Si/a-SiO_2/a-HfO_2 stacks to simulate both thick and thin film devices. Distributions of one-electron levels are calculated for oxygen vacancy centres and H-induced defects as a function of the distance to the interface. The thermodynamic nature of interstitial hydrogen is studied with respect to its position in the Si/SiO_2/HfO_2 stacks to assess its behaviour according to the RDD model. The charge state of interstitial H depends on the material in which it is present, and it can drift toward HfO_2 with a low energy barrier, as proposed by the RDD model.

Computational Infrared and Raman Spectroscopy in ChemShell

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To aid the interpretation of experimental vibrational spectra, we developed and implemented computational infrared and Raman facilities in the ChemShell computational chemistry environment using hybrid quantum mechanical and molecular mechanical approach. Density functional theory for the electronic structure calculations and classical forcefields for the environment are employed. Computational vibrational spectra for chemical active sites are reported using electrostatic and fully polarizable embedding to achieve more realistic vibrational signatures of material systems, including solvated molecules, proteins, zeolites and metal oxide surfaces, providing useful insight into the effect of the chemical environment on the signatures obtained from experiment. This work has been facilitated by the efficient task-farming parallelism implemented in ChemShell for high-performance computing platforms.

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

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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.

Potential thermoelectric materials: zinc-based oxychalcogenides $SrZn_2Ch_2O$ (Ch = S, Se)

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In the process of primary energy consumption, a large amount of waste heat is generated. Thermoelectric materials can convert waste heat energy into electric energy, which is crucial to solving the current energy crisis and global warming. In general, the performance of thermoelectric materials can be determined by the dimensionless figure of merit, ZT, that is evaluated by electronic and thermal properties. However, it is difficult to maximize ZT due to the interaction between different parameters. Although some high ZT thermoelectric materials such as Bi_2Te_3 and PbTe have been found, they have not been widely used because they contain rare or toxic elements.[1,2]

Recently, $SrZn_2S_2O$ was proposed as a novel photocatalyst for water splitting applications.[3] Because of its complex structure, relatively high thermal stability, and the low toxicity and easy synthesis of oxides, we think that $SrZn_2S_2O$ may be a potential thermoelectric material. Using density functional theory (DFT), we first studied the phononic stability of $SrZn_2S_2O$ and its variant - $SrZn_2Se_2O$. Then based on the $SrZn_2Ch_2O$ (Ch = S, Se) electronic structure calculated with hybrid-DFT, electronic transport properties were calculated using AMSET[4]. Our calculation results indicate that both oxychalcogenides may become thermoelectric materials for high-temperature applications.

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Intrinsic defects in amorphous Ga_2O_3

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Ga_2O_3 is a promising material for power electronics devices and solar blind UV photodetectors, owing to its high breakdown electric field and wide bandgap (~5 eV). However, defects that occur during the growth, fabrication, or operation of Ga_2O_3-based devices can affect their performance and electronic properties.

In this study, we used classical dynamics (MD) and density functional theory (DFT) with the Gaussian plane-waves method to model amorphous phase of Ga_2O_3 and investigate the impact of point defects, such as oxygen vacancies and hole trapping on the electronic properties of Ga_2O_3. We employed the PBE0-TC-LCR functional, a non-local functional, to obtain more accurate and reliable predictions for the geometry relaxation and charge transition levels of the defects.

Our findings revealed that oxygen vacancy defects in amorphous Ga_2O_3 exhibit additional charge transition levels (CTLs) compared to those observed in the crystalline phase. This behavior indicates that additional electrons tend to become trapped around oxygen vacancies. Furthermore, we discovered that hole trapping in amorphous Ga_2O_3 is highly localized and occurs at deeper trapping energy compared to the crystalline phases. Our study could provide insights for designing Ga_2O_3-based electronics by understanding the impact of defects on the material's electronic properties.

Developing Standardised Modelling Workflows for QM/MM Simulations of Metal Oxides

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Modelling plays a key role in advancing our understanding of materials, assisting in both the discovery of novel condensed phase systems and development of their applications. Thus, models of solid state materials need to be both accurate and efficient, yet there is often a compromise between accuracy and computational cost. QM/MM (Quantum Mechanical/Molecular Mechanical) simulations achieve high accuracy with good computational efficiency by limiting the full quantum mechanical calculation to a small region of interest where preserving physicality is critical; the interactions with the quantum region are then modelled though coupling to an environment of classical charges.[1] QM/MM has advantages over periodic density functional theory (DFT) simulations; the latter method can be resource inefficient, scale poorly with increasing sizes of cells and require corrections for nonphysical interactions, such as defect interactions, due to limitations in cell size.[2]

We have performed calculations on neutral and charged defects in bulk MgO, employing QM/MM simulations (validating these against periodic DFT calculations) to develop a full workflow for producing accurate and realistic simulations. Our initial methodology is based on the simple system of bulk magnesium oxide (MgO) to facilitate accurate model development, allowing us to design heuristics for easily setting up embedding simulations.[3-6] Currently, we are exploring the optimal size/shape of the QM and MM regions for accurate and efficient simulation, building on previous work in this area that suggests using a unit cell based partitioning approach over the simpler radial partitioning method.[7, 8] A study by Shushko, Shluger and Catlow explored MgO surfaces with a similar but distinct embedding method, and we hope to be able to replicate their results using our newer models.[9] The outcomes of this will show if the best method of partitioning can be rigorously identified from chemical observables of the bulk periodic system. In the future, we hope to use this work to investigate species that are more challenging to model for their applications to photocatalytic hydrogen production.

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Study of three-phase contact angles via DPD simulations

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e05-surfin

The removal of oil droplets from solid substrates is the core problem in numerous applications of home and personal care products. When surfactants adsorb at an oil-water interface, they lower the interfacial tension, which is a key driver for oily soil detergency [eg E. Kissa, Textile Res. J. 45, 736-41 (1975)]. Thus, understanding and predicting the effect of surfactants on the three-phase contact angles of solid—oil—water systems is paramount in wash formulation design.

In this work, we use dissipative particle dynamics (DPD) to study solid—oil—water systems, where the substrate is represented as a continuous wall. A collection of systems with different wall interactions is prepared to portray hydrophobic and hydrophilic cases. Our aim is to predict the equilibrium contact angle of the oil droplets using Young's equation. The required interfacial tensions are obtained from oil—water and solid—liquid systems. The approach is validated by the slower simulation of explicit cylindrical droplets, which involves multiple boxes of different size to extrapolate the contact angle at infinite droplet radius. We report a good agreement between both methodologies for different surface interactions. We will then work towards more real world situations where surfactants and other surface properties need to be considered.

Influence of Solvent on Selective Catalytic Reduction of Nitrogen Oxides with Ammonia over Cu-CHA Zeolite

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e05-react-sok

In this new study, we made a comparison between water- and ammonia-solvated and bare Cu species [1]. Our results show the promoting effect of solvent on the oxidation component of the NH_3-SCR cycle since the formation of important nitrate species is found to be energetically more favourable on the solvated Cu. Conversely, both solvent molecules are predicted to inhibit the reduction component of the NH_3-SCR cycle. Diffuse reflectance infrared fourier-transform spectroscopy (DRIFTS) experiments exploiting (concentration) modulation excitation spectroscopy (MES) and phase-sensitive detection (PSD) identified spectroscopic signatures of Cu-nitrate and Cu-nitrosamine (H_2NNO), important species which had not been previously observed experimentally. This is further supported by the QM/MM-calculated harmonic vibrational analysis. Additional insights are provided into the reactivity of solvated active sites and the formation of key intermediates including their formation energies and vibrational spectroscopic signatures, allowing the development of a detailed understanding of the reaction mechanism. Further, we made a significant addition to the just-published work and now extended the study to Fe Beta-zeolite. The new results are significantly promising since we found the formation of key intermediates in a system containing Fe-framework is highly exothermic as compared to Al-Framework zeolite.

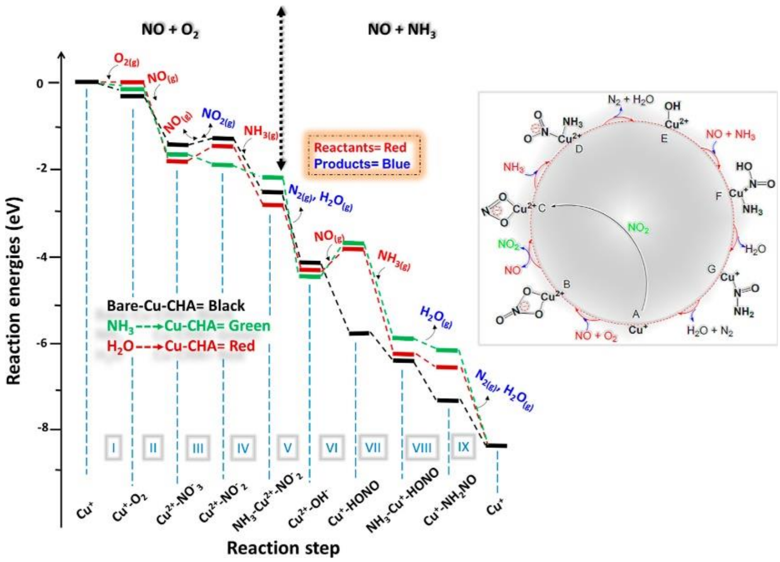

Figure 1. Potential reaction-energy landscape for NH_3-SCR on the activated Cu-CHA site and (black) on the bare site, (green) with physisorbed ammonia and (red) with physisorbed water. Inset right: NO-activated NH_3-SCR of the NOx catalytic cycle.

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Removal of VOCs in cooking fumes via adsorption method on zeolites

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e05-enviro-cat

The growth of public awareness of hazard from gaseous pollutants emitted by cooking activities spawned the implement of new laws and regulations as well as new technologies for cooking fumes emission control. Considering the emission characteristics of large air volume and intermittence of cooking fumes, MFI zeolites were applied as an efficient adsorbent for enriching the low-concentrated complex humid volatile organic compounds (VOCs) from cooking fumes. In order to illustrate the superiority of Ti-O-Si framework as a cooking VOCs adsorbent, zeolites with the same MFI topology to TS-1 but different framework compositions of Si-O-Al (ZSM-5) and Si-O-Si (silicalite-1, hereinafter referred as S-1) were also investigated for comparison. Related characterizations were performed to reveal the difference among three zeolites. It was found that textural properties, surface silanol density and defect content significantly affect the adsorption site accessibility, water resistance as well as organophilicity of zeolites adsorbents, all of which play a decisive role in the adsorption process of cooking fumes. Furthermore, adsorption energy between water/VOCs molecular and zeolites with Si/Al/Ti center was calculated through hybrid quantum mechanical/molecular mechanical (QM/MM) technique, revealing the adsorption interaction between different framework and VOCs as well as the water resistance.

Thermoelectric properties of the *Pnma* and *R3m* phases of GeS and GeSe

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As the impact of climate change becomes ever more apparent, coupled with dwindling fossil fuel reserves and an ever-growing demand for more energy, there is an intense focus on finding technologies that can provide clean and sustainable energy. As large amounts of current energy usage is wasted as heat (60 %), using thermoelectric generators to capture waste heat and recover it as useful electrical energy could significantly enhance efficiency and sustainability. Furthermore, thermoelectric generators are easily scalable to a wide range of applications including in the automotive, manufacturing and power industries.

Tin monoselenide SnSe is one of the most promising TE materials and was reported to have a very high *ZT* of 2.6 at 923 K. The germanium analogues GeS and GeSe have also been predicted to have good TE performance, and, recently, the rhombohedral (*R3m*) phase of GeSe, was reported to have a *ZT* value > 1. However, the TE properties of *R3m* GeS, a structural analogue of *R3m* GeSe, have not yet been investigated.

We have used first-principles modelling to perform a comprehensive evaluation of the structural and electronic properties of the orthorhombic (*Pnma*) and *R3m* phases of GeS and GeSe. Our results show that n-type *Pnma* GeSe can support a large *ZT* of 2.23 at 940 K, due to a combination of its low lattice thermal conductivity and comparatively high electrical conductivity. This suggests that, if n-type doping of *Pnma* GeSe is possible, it could be a good match for p-type semiconducting material (*e.g.,* SnSe) in a thermoelectric couple device.

Stability of Ru@Sn_9 Zintl cluster on a CeO_2 (111) surface and its catalytic activity in Water-Gas-Shift (WGS) reaction.

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Cerium Oxide (CeO_2) is one of the most efficient compounds, supporting surface, because of the facile changes in oxidation state between Ce^{4+} to Ce^{3+}, and is widely used in high-performance oxygen storage applications and catalytic redox reactions, solid oxide fuel cells, water-gas shift reactions, etc. The use of Zintl clusters or metalloids rather than isolated noble metal atoms as the catalytic centre offers the potential to exploit the interactions between transition and main-group metal to achieve low-barrier reactions, and recent work from our collaborators in Tianjin suggests that Zintl clusters are effective catalysts for the water gas shift (WGS) reaction. Recently, Sun et al. have reported the selective reduction of CO_2 over highly dispersed $RuSnO_x$ sites, derived from a $[Ru@Sn_9]^{6-}$ Zintl cluster but an atomic-level understanding of mechanism, and how it relates to the electronic properties of the cluster remain unclear.[1]

In this work, we firstly investigate the stability of the Ru@Sn_9 Zintl cluster on the CeO_2 surface by comparing the optimised energy of the cluster on the surface with a dispersed structure where the Ru and Sn atoms are dispersed on the surface, using periodic Density Functional Theory (DFT). Interestingly, in many cases clusters prove not to be stable on surface, but rather are disrupted by strong metal-support interactions (SMSI).[2] An important first objective therefore is to establish the stability of the Ru@Sn_9 cluster on the surface under normal reaction conditions. It has been found that Ru@Sn_9 cluster will disperse over the surface and accumulate in a monolayer where Ru would prefer to stay at the edge. Later, we report some initial studies on the $RuSn_9/CeO_2$ (111) catalyst for WGS reaction where possible reaction mechanisms have been explored and activation energy barriers calculated. This catalyst shows a significantly lower energy barrier using different reaction path, compared to other reported heterogeneous catalysts such as Au, Pt, Cu nanoparticles on supported surfaces.

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Utilisation of Machine Learning for Lithium Titanium Sulphate (LTS) Solid Solutions Energy Prediction

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Battery development is critical for the transition from fossil fuels to renewable and more sustainable energy as rate of climate change is rapidly increasing. However, traditional material discovery methods are time-consuming and resource intensive. An ideal approach would be to study the desired properties to identify a potentially suitable material. While computational methods have been successful in assisting material discovery using this ‘inverse design’ method, ab-initio methods, such as DFT and Hartree Fock, requires significant computational cost and time.

To address these challenges, Machine Learning is used to facilitate the material discovery of Lithium Titanium Sulphide for batteries. DFT were used to generate the energies of the structures which are used as the labels for training the machine learning model, while descriptors were used to represent the structures generating features.

The data obtained showed Many Body Tensor Representation (MBTR) descriptor to have the most consistent performance with all machine learning model whilst Smooth Overlap of Atomic Orbital (SOAP) have the lowest mean absolute error (MAE) overall when paired with support vector regression (SVR). This project showcases the potential of Artificial Intelligence in accelerating material discovery and advancing the development of sustainable battery technology.

Effects of Electronic Polarization on the Convergence of Mott-Littleton Calculations of $BaSnO_3$

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Barium Stannate ($BaSnO_3$) is a perovskite oxide material with a wide range of applications in various electronic devices thanks to its attractive electronic and thermal properties. Point defects in such materials can introduce additional charge carriers, modify band structures, and affect carrier mobility, resulting in changes in conductivities as well as catalytic activities. Understanding and controlling point defects in perovskite materials is crucial for tailoring their properties to specific applications and advancing their technological potential. The Mott-Littleton method evaluates the defect energy accurately by calculating the energy difference between the perfect and the defective crystals using interatomic potentials, which require significantly less computational resources, compared to *ab initio* methods. Partitioning the regions around the defect centre also provide straightforward and intuitive interpretations on defect energies based on factors that affect the formation and stability of the defect.

We show that converging the defect energy in Mott-Littleton calculations can be challenging due to effects of electronic polarization due to the charged point defect, even for simple crystal structures in bulk phase, and analysing charge distributions in localised cluster can provide further insights on how the model can be improved and adapted to surfaces and interfaces, where addressing defect-induced dipole and quadrupole effects remains problematic.

Ketonisation of Carboxylic Acids over ZrO_2-based catalysts for Biomass Valorisation: The Role of Surface Acid-Base Sites

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Production of liquid fuels from biomass have gained increasing industrial and academic attention as alternative energy source to fossil-based fuels. Biofuels are usually obtained as a mixture of organic species with high content of oxygenated derivatives, which required chemical upgrading for improvement of its physicochemical properties prior to their use as fuels.[1] Acid–base bifunctional catalysts are particularly desirable in this context, which can be tuned by different approaches in order to optimise their activity and selectivity.[2] Here, we investigate how the reaction mechanism and energy profile of the ketonisation of carboxylic acids are affected by modification of the acid-base properties of zirconia-based catalysts.

The ketonisation reaction has been proposed to occur via several putative mechanisms. Currently, the β-ketoacid mechanism is the most plausible one based on previous experimental and computational results.[1] The two most important steps of the β-ketoacid mechanism are investigated here: enolisation of the adsorbed carboxylic acid and C-C coupling step to afford α-hydroxy γ-carboxy alkoxide intermediate. These reaction steps were studied over pristine c-ZrO_2 (111) and t-ZrO_2 (101) surfaces, as well as over three modified surfaces: Ca- and Y-doped c-ZrO_2 (111), and, sulfated t-ZrO_2 (101). We show that distinct strengths of the acid-base sites on the surface of these catalysts can influence the adsorption modes and protonation state of the reactants and intermediates, resulting in significant changes in the computed energy barriers and the identity of the rate-determining step in the ketonisation of organic acids.

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Computational investigation of CO_2 hydrogenation over Rh-based catalysts

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The alleviation of CO_2 emissions has become an urgent issue, requiring the development of technology for carbon dioxide capture, storage and utilization. Energy-efficient catalytic CO_2 conversion using renewable energy, has attracted considerable attention worldwide as a potentially feasible means to mitigate CO_2 emissions, and sustainably produce commodity fuels and chemicals. It is thermodynamically feasible to convert CO_2 through hydrogenation processes to produce and hydrocarbons. Among them, methane (CH_4) is an attractive target molecule as it can be directly injected into the existing natural gas infrastructure for distribution and storage. In addition, CO_2 hydrogenation to CH_4 is regarded as a feasible process for the long term chemical storage of electricity produced from renewable sources. Among the metal catalysts, Rh has exhibited high activity for CO_2 hydrogenation and selectivity towards methane. However, the various mechanistic routes for CO_2 hydrogenation to CH_4 are multiple and complex. A powerful tool for exploring the reaction mechanisms under realistic conditions is provided by the combination of Density Functional Theory (DFT) and kinetic Monte Carlo (kMC) simulations which can complement operando experimental techniques and identify descriptors of activity and selectivity for CO_2 hydrogenation.

In this context, we aim to investigate all the mechanistic pathways for CO_2 hydrogenation over Rh surfaces, providing a valuable benchmark study to inform the rational design of high-performance catalysts for CO_2 hydrogenation. Here, DFT simulations have been used to explore the reaction mechanisms for CO_2 hydrogenation over the Rh(111) surface, including 53 reversible reaction steps, 6 desorption steps, CO_2 adsorption, and all the relevant intermediates. It has been found that the Rh catalyst can promote the CO_2 activation and H_2 dissociation based on the geometric and electronic analysis of the adsorption structures for CO_2 and H_2 and lower activation energies for H_2 dissociation. Based on the present work, we plan to carry out kMC simulations to explore the product distributions and the dominant reaction pathways involved in CO_2 hydrogenation over the Rh surface, which will provide an in-depth insight into the reaction mechanisms for the CO_2 hydrogenation over the Rh surface under experimental conditions.

High-dimensional neural networks for interatomic potentials: Applications to metal organic frameworks

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Machine learning interatomic potentials (MLIP) are revolutionising our way of describing and understanding how atoms and molecules interact. Precise simulations of atomic and molecular chemical systems is hampered by the poor scalability of first principles methods such as Density Functional Theory (DFT), or by the limitations of classical force fields due to their empirical nature. A major challenge in the development of MLIPs is creating a training set that allows the system to learn all of the relevant physics and chemistry of a given system. Active learning is a technique that can solve this problem by building a training set that systematically explores phase space. In this work we use neural network potentials and active learning to study UiO-66 metal organic framework (MOF), a system of interest due to its stability, pores and filtration abilities. We compare the radial distributions functions of configurations from simulations using our system with simulations using DFT methods and explore the scalability and transferability of our potential.

USING POTENTIALS FOR MODELLING COMPOUNDS THAT UNDERGO THE METAL – INSULATOR PHASE TRANSITION

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The metal-insulator phase transition is the process by which a compound that conducts electricity becomes an insulator (or vice versa) by means of a change in its atomic and electronic structure. One of the most studied compounds is vanadium dioxide (VO_2). The computational method most widely used for its study is DFT since the process has its origin in a Peierls distortion caused by the presence of a d^1 electron in vanadium. Moreover, there are other dioxides that undergo the same process by means of a Peierls distortion: NbO_2 and TaO_2, being both niobium and tantalum in the same group as vanadium (Group V).

Interatomic potentials are a convenient computational method since they allow to study large systems and/or a high number of configurations without being too computationally expensive. Considering that DFT is a computationally expensive one, interatomic potentials are a good alternative for studying and optimising the structure of these compounds. All in all, this project aims to adapt the potentials used in VO_2 for using them also in NbO_2 and TaO_2 with the finality of analysing the effect of their differences in the parameters of the potentials and developing an easy-going approach to study the elements in the same group. The potentials that are being used are Buckingham Potential (for optimising the general structure), Morse Potential and Double-Well Morse Potential (for reproducing the Peierls distortion) using the Package GULP.

Electronic and Structural Properties of the 2D Photocatalyst *g*-C_3N_4

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Increased efficiency in the performance of photocatalytic materials can be effected by the formation of nanocomposites, often with carbon-based 2D systems. Graphitic carbon nitride (*g*-C_3N_4) has shown great promise for photocatalytic water splitting when combined with semiconductor materials such as TiO_2. It is highly stable, semiconducting and has favourable electronic properties. However, much is not understood about the properties of the material and the mechanisms by which it facilitates photocatalytic processes. We survey different density functional theory approaches to model *g*-C_3N_4 in order to understand fundamentally its structural and electronic properties. We also study the effects of applied strain on its properties, to simulate realistic structures when the 2D system is interfaced on a semiconductor surface. Our results help us understand catalytic processes that are observed by our experimental collaborators who grow samples and measure their activity.

Modeling the Effect of OSDA on Nucleation Process of TS-1 Zeolite

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Zeolites are porous materials that come in many topologies and have multiple catalytic functions. The majority of zeolite synthesis relies on organic structure-directing agents (OSDAs). However, most OSDAs designs are based on trial and error, chemical intuition, or large-scale screenings rather than rational design. Thus, this empirical labor-intensive task makes the design of OSDA a significant technical threshold. In this study, we analyzed the effects of over 80 different OSDAs on the nucleation process using TS-1 zeolite as an example. Through high-throughput computational simulations, literature mining, synthesis, and characterization, we revealed the energetic, geometric, and electrostatic descriptor effects of OSDAs on the entire nucleation process.

High-throughput computational design and discovery of conductive materials in the CSD MOF subset

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e05-power

MOFs are innovative porous materials, and have been widely studied for the past two decades for applications in different areas including gas storage, gas separation and catalysis. Here, we aim to study electrical conductivity in MOFs, a less explored but interesting property that can bring promising opportunities in energy storage and sensing application. Due to their high porosity and surface area, MOFs are poor electrical conductors. Here, to identify promising conductive structures, we performed high-throughput screening of the existing ca. 90,000 structures in the CSD MOF subset[1]— characterising the band gap and examining the presence or absence of metallic behavior. The first set of selection criteria was developed based on the nature of MOFs' surface chemistry with a focus on the type of the secondary building unit and the ligand. We focused on MOFs containing open shell metals, metal clusters and highly conjugated linkers that can facilitate through-linker charge transfer between metals. The second set of criteria involved the presence of linkers containing metal-S, or -N coordination, redox-active linkers, π-π stacking, and mixed valence metals. For the ca. 1000 structures shortlisted, we then performed DFT calculations to derive useful insights into structure-conductivity relationships in MOFs, identify top-performing conductive MOFs, and to delineate key chemical and physical features in MOFs that influence their conductive properties for the first time. The results guide MOF researchers to assess and design conductive structures for electronics, energy storage and sensing applications.

Reference:

[1] Peyman Z. Moghadam *et al*. (2017) Chemistry of Materials, 29, 2618–2625

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