

In the Lab

UK Research on Materials for Electrochemical Devices

Johnson Matthey Technology Review features new laboratory research

Introduction

A select group of researchers are profiled here, all of whom are involved in the design and characterisation of materials for electrochemical energy storage and conversion devices. These include a broad range of battery types, fuel cells, supercapacitors, photovoltaics and devices for the production, storage and utilisation of hydrogen.

Many are pioneering the use of advanced techniques for characterising energy materials, enhancing our understanding of the fundamental kinetic, structural, electronic and magnetic properties which distinguish materials as being well suited to a particular application. Some are also developing novel techniques for accurately assessing properties which are currently not easy to measure, for example: Sam Cooper and Ainara Aguadero's work on isotopic labelling for the quantification of surface exchange and solid-state diffusivity of battery and fuel cell materials.

The performance and function of an energy material is often strongly linked to its microstructure, both in terms of its homogenised bulk properties and certain forms of heterogeneity. Understanding this link is key to enhancing manufacturing methods, through the processing of materials to component and device construction, by tailoring materials for optimum performance in the target device. Experimental techniques are complemented by computational models, providing important insights into physical and chemical processes happening at the nanoscale.

Once reliable assessment techniques are established, it will be possible to screen materials rapidly and build up a database of material properties. This high-throughput screening and a variety of machine

learning tools will accelerate the identification of novel functional materials, composites and synthesis techniques for a specific purpose. A comprehensive materials library with powerful data mining capabilities can also provide diagnostics for materials from a degraded device, aiding our understanding of the mechanisms behind device ageing and failure.

Some of the research groups covered here have developed expertise in synthesising new energy materials, with provable success in controlling the resultant materials' properties. They make use of composites, incorporating nanostructures and other exotic ingredients to introduce specific properties to an already stable and reliable base material, as well as a range of innovative techniques, such as electrospinning, to control microstructure.

The researchers presented here engage with energy research across a range of scales, from the development of atomistic mechanisms all the way up to techno-economics and policy. Beyond this, they are also all active in areas beyond energy, including sensors, catalysts and memristors, as well as the development of new experimental techniques and synthesis routes.

JACQUELINE EDGE*

Department of Mechanical Engineering, Imperial College London, South Kensington Campus, London, SW7 2AZ, UK

*Email: j.edge@imperial.ac.uk

About the Research

1. Electrochemical Energy Storage

The most exciting aspects of Sam's current research focus around two main topics within the realm of materials for electrochemical energy

Samuel J. Cooper



- **Department:** Faculty of Engineering, Dyson School of Design Engineering
- **University:** Imperial College London
- **Address:** South Kensington Campus, London
- **Post Code:** SW7 2AZ
- **Country:** UK
- **Email:** samuel.cooper@imperial.ac.uk
- **Website:** <https://www.imperial.ac.uk/people/samuel.cooper>

storage and many of these projects are undertaken in collaboration with various members of Imperial College London's Electrochemical Science and Engineering group.

Firstly, he is using isotopic methods to characterise the surface exchange and bulk diffusivity of electrode active materials, in collaboration with Ainara Agudero. Similar methods were deployed with great success to understand oxygen ion transport and surface exchange for fuel cell systems (1, 2). However, battery materials present specific challenges, in particular room temperature operation and moisture sensitivity, which require these methods to be redesigned. Sam's group is currently trialling four distinct approaches to this problem, which is a major undertaking, but the potential rewards, in terms of high throughput screening of cathodes and electrolytes, are significant.

Secondly, he is looking at the analysis and design of electrode microstructures, in collaboration principally with Nigel Brandon, also at Imperial College London. X-ray and ion beam three-dimensional (3D) imaging techniques are pushed to their limits by multiphase, nanoscale battery materials, but the last few years have seen significant progress in their application (3, 4), in particular for investigating unusual microstructures (5, 6). Sam has previously focused on developing open-source software to allow the community to

standardise their analysis approach (5, 6), but more recently he is working on machine learning techniques and multiphysics parametric studies to generate design rules.

In addition to his material research, Sam collaborates with Billy Wu at Imperial College London on device level characterisation to understand the state of health and optimum designs for battery cells and packs. Simplified cell models typically do not incorporate mechanisms to capture cell-to-cell variation, and yet this is known to be a key feature limiting the performance of battery packs, especially in the context of potential second-life applications. By implementing novel thermal voltammetry methods (7), combined with multidimensional cell grouping, they are looking to overcome this complexity with a data-driven approach. Finally, Sam is using recurrent neural networks to predict trends in the grid-scale market to accelerate the implementation of next generation electrochemical energy storage.

Much of this work is currently being funded by the Faraday Institution's Multiscale Modelling project, as well as a variety of Faraday-associated Innovate UK projects, including: Advance Battery Life Extension (ABLE), IMproving Power bAttery Cooling Technologies (IMPACT) and A holistic battery design tool: From materials to packs (Mat2Bat).

2. Optimisation of Ion-Dynamics in Electrochemical Systems

Ainara Agudero



- **Department:** Faculty of Engineering, Department of Materials
- **University:** Imperial College London
- **Address:** South Kensington Campus, London
- **Post Code:** SW7 2AZ
- **Country:** UK
- **Email:** a.agudero@imperial.ac.uk
- **Website:** <https://www.imperial.ac.uk/people/a.agudero>

Ainara's current research focuses on the study and optimisation of ion-dynamics taking place in electrochemical systems, with a special focus on solid state devices, including secondary batteries, fuel cells, electrolyzers and memristors. The common aim is the analysis of how the different ion dynamics affect the performance and degradation of these systems. In order to reveal this, her group uses a combination of structural, electrochemical and chemical characterisation techniques. More specifically, they use surface-sensitive analysis and isotopic labelling to reveal and differentiate different ion kinetics taking place at the bulk as well as at the surfaces and interfaces of materials.

One of the biggest topics of research focuses on development of solid state batteries, in which Ainara studies the effect of processing on lithium dynamics (8, 9) and seeks to understand the origin of dendrite formation (10, 11). Her group is also developing new isotopic labelling methods to evaluate the bulk diffusivity and surface exchange kinetics of Li in different battery materials. This work takes place in collaboration with Sam Cooper from the Dyson School of Engineering at Imperial College and will be used to correlate battery performances with variations in the Li kinetics, for instance in systems with dynamic interfaces and cation inter-diffusion processes (12).

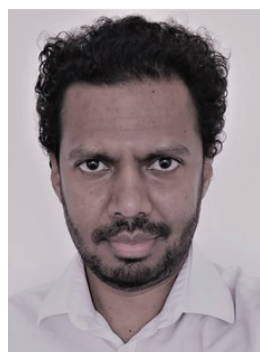
Another important area of research is the development of fast oxygen conductors (13) and the study of the potential topotactic redox capabilities of oxides (14) and their applications for fuel cells, electrolyzers, hydrogen production, memristive switching or catalysts (15). This work takes place in collaboration with John Kilner and Stephen Skinner at Imperial College London and with universities in the UK, Europe and elsewhere.

Finally, in the area of surface analysis techniques, the group has a strong background in the study of energy materials using secondary ion spectroscopy and low energy ion scattering (16). At the moment, the group is also developing a unique, worldwide facility called Hi5 (strategic equipment grant EP/P029914/1) with a plasma ion source and dual positive and negative ion detection capabilities for *in situ* characterisation (T, bias) of electrochemical devices, from the nm to the mm scale. Hi5 will be housed in the Department of Materials at Imperial College London.

Ainara has received funding from a number of Engineering and Physical Sciences Research Council (EPSRC) grants, the Science and Technology Facilities Council (STFC) Futures Early Career Award, Energy Cooperative Research Centre (CIC energiGUNE), the Bosch Energy Research Network and The Faraday Institution, among others.

3. Chemistry and Physics of Materials

Chandramohan George



- **Department:** Faculty of Engineering, Dyson School of Design Engineering
- **University:** Imperial College London
- **Address:** South Kensington Campus, London
- **Post Code:** SW7 2AZ
- **Country:** UK
- **Email:** chandramohan.george@imperial.ac.uk
- **Website:** <http://www.imperial.ac.uk/people/chandramohan.george>

Chandra's research activities in the broad areas of chemistry and physics of materials seek to understand charge-carrier dynamics, ion-diffusion, charge-transport and light-matter interactions in solids and metal-organic frameworks for renewable energy. Against this backdrop, shape-controlled synthesis was successfully extended to battery materials *via* a colloidal route, producing phospho-olivines in the form of thin platelet crystals, which in the case of lithium iron manganese phosphate has led to a fine-tuning of metal redox energies due to cation intermixing (17) and in the case of lithium iron phosphate with an etched surface, enabled ultrafast battery charging (17). Using hierarchical carbon pre-patterned structures, ultra-flexible Li-ion battery design capable of offering fold radii down to 0.5 mm was proposed (18). By integrating solar cell materials such as organic dyes (19) and organo-halide perovskites (20) in Li-ion cell configuration, new design principles of photo-rechargeable batteries are being advanced. Lastly, by exploiting epitaxial growth relationships, bi-functional oxygen cathodes made of iron oxide nanoparticles and carbon nanotubes are shown to regulate the morphology of discharge products, enabling a fully reversible Li-air battery (21).

Current research into the development of next generation Li-ion batteries with value added features (mechanical pliability and shape-conformity) are supported by The Royal Society.

4. Sustainable Materials

Magdalena Titirici



- **University:** Imperial College London and Queen Mary, University of London
- **Address:** South Kensington Campus, London and Mile End Road, London
- **Post Code:** SW7 2AZ and E1 4NS
- **Country:** UK
- **Email:** m.titirici@imperial.ac.uk
- **Website:** <https://titiricigroup.org>

The research interests in Magda's group are in sustainable materials, in particular porous carbon and hybrids produced from available resources such as bio- and plastic waste and abundant metals (i.e. iron, manganese and nickel). Her group produces carbon and carbon hybrids using hydrothermal processes which allow scale-up and continuous processes. They can produce up to 1 kg carbon per day and can control exactly the morphology, pore structure, pore size and shape required for each application. They have a great degree of control over the degree of graphitisation, ranging from hard carbons to soft graphitic carbons.

The group applies designer carbon materials to energy storage and conversion technologies, for example as anodes for sodium-ion batteries, electrodes in supercapacitors, cathodes in lithium-sulfur batteries and as electrocatalysts in fuel cells, electrolyzers and metal-air batteries. They pay a great deal of attention to understanding the fundamentals involved in structure-function relations using advanced characterisation tools applied *ex situ* and *operando* such as: small angle X-ray spectroscopy (SAXS), small-angle neutron scattering (SANS), X-ray absorption near edge structure (XANES), transmission electron microscopy (TEM), nuclear magnetic resonance (NMR) and magnetic resonance imaging (MRI), working collaboratively with experts in these areas.

The group is well-funded and formed of around twenty researchers, with funds from EPSRC, the European Union, Innovate UK, the Royal Society, the British Council, the Royal Society of Chemistry (RSC) and industry. Their publications are highly cited and recognised internationally with 18,000 citations from 160 publications, five patents, ten book chapters and one edited book. The Principal Investigator, Professor Titirici, has been recognised internationally with the RSC Corday Morgen Prize, the IOM³ Rosenheim Medal, the Chinese Academy of Science President Award and an Honorary PhD from Stockholm University, Sweden.

5. Computational Modelling of Fundamental Processes

Pooja Goddard



- **Department:** Advanced Materials Modelling, Department of Chemistry
- **University:** Loughborough University
- **Post Code:** LE11 3TU
- **Country:** UK
- **Email:** p.goddard@lboro.ac.uk
- **Website:** <http://am2-rg.wixsite.com/home>

Pooja's research group, based at Loughborough University's Department of Chemistry, focuses on computational modelling of fundamental processes in complex materials at the atomic or quantum scale. Their multiscale modelling approach combines inter-ionic potential-based methods and density functional theory (DFT) simulations in synergy with experimental groups and industry.

This requires a good understanding of the structural, electronic, magnetic and transport properties which are crucial in identifying novel functional materials for sustainable energy and catalytic applications. The nature of defects in inorganic solids as well as their effect on electronic and transport properties is also important, not

only in understanding the key structure-property relationships, but also in the next phase of materials design with enhanced performance. In addition to this, a sound understanding of nano-ionic properties can yield a wealth of materials with significant technological impact.

The computational methods range from atomistic potentials-based methods, where the forces are dominated by the long-range electrostatic interactions, but also includes short range, van der Waals attractions, electron-electron repulsions and polarisability, to DFT at varying levels of theory. Molecular dynamics is also used to study the transport properties as a function of time and temperature.

Further to this, expansion towards more sophisticated time dependent density functional theory and embedded cluster methods is being pursued.

The areas of research within the group are wide-ranging with a focus on the next generation energy storage systems, thin film photovoltaics, fuel cell materials and, more uniquely, fingerprint detection materials and biomarker detection.

Pooja has received funding from several EPSRC grants and her current collaborations include: Professor Laurence Hardwick (University of Liverpool, UK); Professor David Scanlon (University College London, UK); James Cookson (Johnson Matthey Plc, UK); Professor Olle Eriksson and Biplab Sanyal (Uppsala University, Sweden); Professor Frank Tietz (Forschungszentrum Jülich, Germany) and Professor Michael Walls (Centre for Renewable Energy Systems Technology (CREST), Loughborough University, UK).

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