

Johnson Matthey Highlights

A selection of recent publications by Johnson Matthey R&D staff and collaborators

Pore-Scale Filtration Model for Coated Catalytic Filters in Automotive Exhaust Gas Aftertreatment

M. Plachá, P. Kočí, M. Isoz, M. Svoboda, E. Price, D. Thompsett, K. Kallis and A. Tsolakis, *Chem. Eng. Sci.*, 2020, **226**, 115854

This paper reviews the filtration efficiency of automotive exhaust gas filters with the impact of catalyst distribution. Using 3D XRT the structure of filter wall is reconstructed. This includes catalytic material subject to spatial distribution. A custom solver developed and implemented in OpenFOAM with the Lagrangian method for soot particles simulates the filtration process. Gas velocity and particle size are used to predict clean filtration efficiencies. The filtration of particles smaller than 50 nm is strongly improved with the Brownian motion. The clean filtration efficiency was significantly increased with on-wall catalyst layer. Experimental data from engine test bench were used to compare the obtained results.

A Sustainable Chemicals Manufacturing Paradigm Using CO₂ and Renewable H₂

R. R. Bommareddy, Y. Wang, N. Pearcy, M. Hayes, E. Lester, N. P. Minton and A. V. Conradie, *iScience*, 2020, **23**, (6), 101218

Continuous gas fermentation of CO₂ and hydrogen by the cell factory is demonstrated in this study, with *Cupriavidus necator*, to (*R,R*)-2,3-butanediol and isopropanol. While a high carbon efficiency of 0.75 C-mol_{product} C-mol_{CO₂}⁻¹ is demonstrated, it is technoeconomically infeasible for producing commodity chemicals. The energy inefficiency is overcome by heat integration between exothermic gas fermentation and endothermic supercritical water gasification. The potential for sustainable manufacturing using renewable feedstocks by combining the carbon efficiency of biocatalysis with energy efficiency enforced through process engineering is shown in this study.

Synergistic Electrolyte Additives for Enhancing the Performance of High-Voltage Lithium-Ion Cathodes in Half-Cells and Full-Cells

A. Kazzazi, D. Bresser, M. Kuenzel, M. Hekmatfar, J. Schnaidt, Z. Jusys, T. Diemant, R. J. Behm, M. Copley, K. Maranski, J. Cookson, I. de Meatza, P. Axmann, M. Wohlfahrt-Mehrens and S. Passerini, *J. Power Sources*, 2021, **482**, 228975

A new combination of electrolyte additives is introduced to address insufficient stability of the electrolyte towards oxidation in high-voltage lithium-ion cathode materials. It targets the stabilised electrode/electrolyte interfaces and interphases for both the anode and cathode. The performance of the cathode half-cells and also of lithium-ion full-cells with a cell voltage higher than 4.5 V can be significantly improved due to the synergistic effect of tris(trimethylsilyl) phosphite (TTSPi) and bis(2,2,2-trifluoroethyl) carbonate (TFEC). It can be shown that there is enhanced cycling stability, increased capacity and improved coulombic efficiency with cells using both TTSPi and TFEC as additives.

Electrocatalyst Performance at the Gas/Electrolyte Interface under High-Mass-Transport Conditions: Optimization of the “Floating Electrode” Method

X. Lin, C. M. Zalitis, J. Sharman and A. Kucernak, *ACS Appl. Mater. Interfaces*, 2020, **12**, (42), 47467

A floating electrode technique was used to facilitate, at the gas/electrolyte interface, high-mass transport to a catalyst layer comprised of an ultralow loading of catalyst (1–15 μg_{Pt} cm_{geo}⁻²). System cleanliness, break-in procedure, hydrophobic agent, ionomer type and measurements of catalyst surface area and loading need to be taken into account to acquire reproducible results with high performance. The

gas permeability of the hydrophobic agent was an important factor for improving the HOR and ORR performance. Evidence of more than a local mass transport barrier is provided showing suppression of the HOR and ORR introduced by the Nafion[®] ionomers.

Controlling the Drying-Induced Peeling of Colloidal Films

A. Osman, L. Goehring, H. Stitt and N. Shokri, *Soft Matter*, 2020, **16**, (36), 8345

A series of experiments were carried out using polyethylene oxide (PEO) additives and drops of aqueous mixtures of colloidal silica dispersions. It is shown that film peeling can be discouraged and finally eliminated with an increase in PEO concentration and molecular weight. Time-lapse digital microscope images of the evaporating droplets were used. The drying front length across the evaporating surface is modified as a result of the additives modifying the suspension properties. A greater understanding of the physics of film failure was the result and is pertinent information for a variety of industrial processes including inkjet printing and coating applications.

Exhaust Energy Recovery via Catalytic Ammonia Decomposition to Hydrogen for Low Carbon Clean Vehicles

S. Sittichompoo, H. Nozari, J. M. Herreros, N. Serhan, J. A. M. da Silva, A. P. E. York, P. Millington and A. Tsolakis, *Fuel*, 2021, **285**, 119111

On-board exhaust assisted catalytic ammonia conversion to hydrogen-nitrogen by direct reaction with part of the exhaust gas (reforming) or by using only exhaust heat (decomposition) was demonstrated. Up to 15% more energy than the reactant ammonia is contained in the resulting hydrogen-nitrogen gas mixture from the exhaust heat driven thermochemical energy recovery processes. It was revealed that complete ammonia conversion occurs at typical GDI engine exhaust gas temperatures (450–550°C). Up to 30% reduction in CO₂ and fuels consumption can be achieved by partially replacing gasoline in a GDI engine with the resultant hydrogen-nitrogen products.

Understanding the Deactivation Phenomena of Small-Pore Mo/H-SSZ-13 During Methane Dehydroaromatisation

M. Agote-Arán, A. B. Kroner, D. S. Wragg, W. A. Sławiński, M. Briceno, H. U. Islam, I. V. Sazanovich, M. E. Rivas, A. W. J. Smith, P. Collier, I. Lezcano-González and A. M. Beale, *Molecules*, 2020, **25**, (21), 5048

Operando XAS, *in situ* synchrotron powder diffraction and electron microscopy were used to study the structure of the small pore Mo/H-SSZ-13 during

catalyst preparation and reaction. When compared to the medium pore Mo/H-ZSM-5, the small pore catalyst demonstrated less effective molybdenum dispersion and ion exchange. Al₂(MoO₄)₃ particles and mesopores formed as a result. During methane dehydroaromatisation, part of the molybdenum species in Mo/H-SSZ-13 underwent a full reduction to Mo⁰, something not observed in Mo/H-ZSM-5. Varying molybdenum speciation and low performance on small pore zeolites was ascribed to ineffective ion exchange into well dispersed Mo-oxo sites and mesopore formation during calcination.

Environment-Dependent Catalytic Performance and Phase Stability of Co₃O₄ in the Preferential Oxidation of Carbon Monoxide Studied *In Situ*

T. M. Nyathi, N. Fischer, A. P. E. York and M. Claeys, *ACS Catal.*, 2020, **10**, (20), 11892

Unsupported Co₃O₄ nanoparticles were studied under different CO-PrOx environments using *in situ* PXRD and magnetometry, and for the first time, the individual and combined effect of hydrogen, water and CO₂ on the progress of the targeted carbon monoxide oxidation process was shown. The presence of hydrogen alone led to the reduction of Co₃O₄ to Co⁰. Co-feeding water decreased Co₃O₄ reducibility and carbon monoxide oxidation activity, while co-feeding CO₂ had no effect on Co₃O₄ reducibility.

Structural Stability, Surface and Photocatalytic Properties of CdGeP₂: A DFT Study

E. Rugut, D. Joubert and G. Jones, *Comput. Mater. Sci.*, 2021, **187**, 110099

The stability, surface and photocatalytic properties of ternary cadmium germanium diphosphide were investigated using the first principle technique. The compound displayed mechanical, vibrational and structural stability, which was established by its elastic constants, phonon dispersion and cohesive energy. Computed results demonstrated that the ternary cadmium germanium diphosphide was able to straddle the water redox potential and therefore had the viability to split water into oxygen and hydrogen. This study, therefore, offers a step toward solving the imminent energy crisis by identifying, characterising and optimising materials that are exceptional for artificial photocatalytic applications.

Mechanistic Insight in NO Trapping on Pd/Chabazite Systems for the Low-Temperature NO_x Removal from Diesel Exhausts

R. Villamaina, U. Iacobone, I. Nova, E. Tronconi, M. P. Ruggeri, L. Mantarosie, J. Collier and D. Thompsett, *Appl. Catal. B: Environ.*, 2021, **284**, 119724

Pd-CHA systems were investigated as passive NO_x

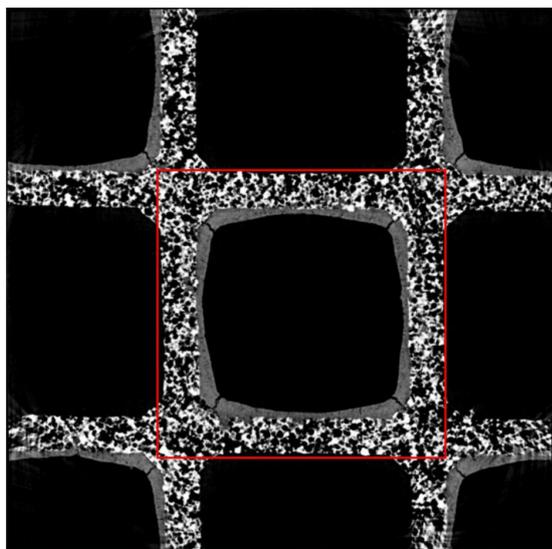


Fig. 1. A horizontal slice from an XRT scan with the marked representative channel. Reprinted under Creative Commons Attribution 4.0 (CC BY)

adsorbers for low-temperature NO_x removal from diesel exhausts. Gaseous mixtures of the main exhaust components (carbon monoxide, oxygen and water) were studied through TPD and nitric oxide adsorption experiments at various temperatures. The key parameter controlling nitric oxide storage and release was the palladium redox state. Nitric oxide and carbon monoxide were shown to reduce Pd(II)OH to Pd(I), the primary nitric oxide storage site. The strong reducing power of carbon monoxide led to enhanced nitric oxide storage. Water also enhanced carbon monoxide storage. Gaseous mixtures with greater oxidising strength favoured nitric oxide desorption at lower temperatures.

Washcoating of Catalytic Particulate Filters Studied by Time-Resolved X-Ray Tomography

M. Blažek, M. Žalud, P. Kočí, A. York, C. M. Schlepütz, M. Stampanoni and V. Novák, *Chem. Eng. J.*, 2021, **409**, 128057

The washcoating process in catalytic monolith filters was imaged with time-resolved XRT (**Figure 1**). Several alumina washcoats with different particle size distributions were studied. The investigation gave new understanding of the drying dynamics of the porous filter wall, as well as the on-wall catalyst layer. For instance, catalyst layer shrinking and cracking during drying was found to be dependent on particle size distribution. Mercury intrusion porosimetry and SEM images of the final structures were used to supplement the XRT results. Further experiments were undertaken to assess the impact of washcoat structure on filter pressure loss.

4D *In-Situ* Microscopy of Aerosol Filtration in a Wall Flow Filter

M. P. Jones, M. Storm, A. P. E. York, T. I. Hyde, G. D. Hatton, A. G. Greenaway, S. J. Haigh and D. S. Eastwood, *Materials*, 2020, **13**, (24), 5676

Time-resolved *in situ* synchrotron micro X-ray CT was demonstrated for the first time experimentally to investigate aerosol filtration. Pore scale deposits of titania nanoparticles were directly imaged in 4D, which is 3D plus time. 3D tomograms were obtained at a rate of one per minute. The penetration depth of particulate deposits, filtration rate and reduction of filter porosity over time were quantified. Porosity changed from 0.60 to 0.56 after 20 min. The data set from this study can be used to validate simulations of automotive wall flow filters.