

Industrial Research on Catalysis using XAS at Diamond Light Source

Anna B. Kroner¹, Sin-Yuen Chang¹, Miren Agote-Aran^{1,2,3}, Ellie K. Dann^{2,3}, Ines Lezcano-Gonzalez^{2,3}, Peter P. Wells^{1,3,8}, Emma Gibson^{3,4}, Alexandre Goguet⁷, Tugce Eralp Erden⁵, Agnes Raj⁵, Christopher Hardacre^{3,6}, Paul Collier⁵, Andrew M. Beale^{2,3}, Elizabeth J. Shotton¹

¹*Diamond Light Source, Harwell Science and Innovation Campus, Didcot, OX11 0DE, UK*

²*Department of Chemistry, UCL, London, WC1H 0AJ*

³*UK Catalysis Hub, Research Complex at Harwell, OX11 0FA*

⁴*School of Chemistry, University of Glasgow, G12 8QQ*

⁵*Johnson Matthey Technology Centre, Sonning Common, RG4 9NH*

⁶*School of Chemistry, The University of Manchester, M13 9PL*

⁷*QUB, School of Chemistry and Chemical Engineering, Belfast, BT9 5AG*

⁸*University of Southampton, School of Chemistry, SO17 1BJ*

anna.kroner@diamond.ac.uk

The requirements for high selectivity and activity of catalysts are among the most crucial demands for a successful commercial application. Therefore, catalyst characterisation provides a unique opportunity for industry to develop new challenging materials for energy, chemistry and environmental technologies. Over the past decades, great efforts have been devoted to developing methods for catalyst characterisation under real operating conditions.

In order to facilitate *operando* experiments, in our Industrial Liaison team at Diamond, we developed a suite of sample environments which allows us to mimic operational conditions and, at the same time, it gives flexibility to study structural properties using a wide range of techniques. The sample environments include capillary-based reaction cell [1] and gas systems operational at ambient and high pressure (up to 20 bar). We also collaborate in a research project dedicated to the development of a combined EXAFS/DRIFS/MS set up which was then applied to study Pd/Al₂O₃ catalysts. In this work the formation of PdO nanoparticles supported on γ -Al₂O₃ has been followed throughout impregnation and subsequent calcination from two alternative molecular precursors; Pd(NO₃)₂ and Pd(NH₃)₄(OH)₂ [2]. Synthesised Pd/ γ -Al₂O₃ catalysts were additionally studied in the process of CO and CH₄ oxidation by the newly developed EXAFS/DRIFS/MS set up.

In our collaborative work with Johnson Matthey, UCL and Research Complex at Harwell Campus we study Mo containing zeolites that can be efficiently applied in the process of methane dehydroaromatisation (MDA) which converts methane directly into light olefins and aromatics [3]. In spite of the promising reactivity of Mo/zeolites, the carbon deposition during reaction and rapid catalyst deactivation is a limiting factor for the commercialisation of this process. Debate regarding nature and location of active sites is still ongoing and catalyst deactivation mechanism is yet not well understood [4]. Performed combined XAS/XRD/MS experiment on Mo/zeolites under *operando* MDA conditions has allowed us to resolve the structures of evolving Mo species (i.e. Mo-oxo, MoO_xC_y and MoC_x) and to draw conclusions regarding their structure-activity relationship [5]. Furthermore, detachment of MoC_x from the zeolite and the early sintering of active Mo species was identified as the key step in catalyst deactivation.

These examples will be used to highlight how collaborative projects between industrial partners, university researchers and Diamond scientists have contributed significantly to the development of next generation materials and industrial processes in the field of catalysis.

References

1. A. B. Kroner, K. M. H. Mohammed, M. Gilbert, G., Duller, L. Cahill, P. Leicester, R. Woolliscroft, E. J. Shotton, *Proceedings of the 12th International Conference on Synchrotron Radiation Instrumentation – SRI2015*, 2016, **1741**, 030014-1-4
2. E. K. Dann, E. K. Gibson, R. A. Catlow, Paul Collier, Tugce Eralp Erden, D. Gianolio, C. Hardacre, A. Kroner, A. Raj, A. Goguet, and P. P. Wells *Chem. Mat.*, 2017, **29**, 7515-7524.
3. Z. R. Ismagilov, E. V. Matus, L. T. Tsikoza, *Energy Environ. Sci.* 2008, **1**, 526-541
4. U. Olsbye, S. Svelle, M. Bjørgen, P. Beato, T. V. W. Janssens, F. Joensen, S. Bordiga, K. P. Lillerud, *Angew. Chem. Int. Ed.* 2012, **51**, 5810 – 5831
5. M. Agote-Arán, I. Lezcano-González, H. Ubayda-Islam, A. B. Kroner, D. S. Wragg, W. A. Slawinski, A. M. Beale, submitted to *Angew. Chem. Int. Ed.*