Ce Mn mixed oxides for low temperature emission control catalysts

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Introduction

Manganese oxides molecular sieves (OMS-2), have a microporous, nano-tunnel structure composed of edge shared MnO_6 octahedra that form a 2 x 2 arrangement [1]. OMS-2 has a mixed valency of Mn^{3+} and Mn^{4+} cations that contributes to its highly active redox properties, which make it particularly interesting for emission control applications. Functionality of OMS-2 can be further extended by structural incorporation of various dopants and tunnel cations[2]. Other synthesis routes can for incorporation of a high concentration of dopants can also lead to the inhibition of the crystalline[3]. In this work we have synthesized a range of manganese oxide based supports doped with Ce for emission control applications. Currently, some of the main challenges is to provide a catalyst which is active at low temperature, due to the high emissions of combustion engines during cold start and low operating temperatures of diesel combustion engine, along with the issue of catalyst deactivation[4]. In particular, we have investigated the use of manganese oxide hybrid catalyst supports with 1wt% Pt for their activity in the oxidation reactions of CO and C_3H_6, before comparing their activity with a commercial diesel oxidation catalyst.

Materials and Methods

OMS-2 molecular sieves were prepared by a sol-gel method. $KmnO_4$ was dissolved in deionized water before slowly adding Maleic acid. The solution and stirred for 40 minutes. The subsequent gel was then washed with deionized water and dried overnight at 90 °C. The gel was then crushed and calcined in air at 450 °C for 4 h. Ce doped OMS-2 was prepared by ion exchange. A sample of OMS-2 was suspended in a solution of $Ce(NH_4)_2(NO_3)_6$ (denoted Ce/OMS:AN) or $Ce(NO_3)_3 \cdot 6H_2O$ (denoted Ce/OMS:NH) and deionised water, using a theoretical Ce/Mn ratio of 0.75, and stirred for 48 h at room temperature. Amorphus Ce/Mn mixed oxides were synthesised by adding $Ce(NO_3)_3 \cdot 6H_2O$ during the synthesis of OMS-2, before the addition of maleic acid. The synthesised samples are expressed as Ce[***X***]-Mn, where ***X*** is the molar ratio of Ce/Mn used in the precursor. To investigate the effect of the dopants on the catalytic activity of the OMS-2 supports, light-off tests were carried out from 303 K to 773 K at a rate of 5 K/min in the presence of 10% O_2, 4.5% H_2O, 2000ppm CO, CH_4, C_3H_6, each and 200ppm NO (flow of 100ml/min). The samples were aged for 24 h at 873 K with a feed of 5% H_2O and 10% O_2, with Ar used as the balance gas. The catalytic activity of the aged catalyst was tested immediately after ageing, by carrying out two cycles of the light-off test described above.The exit stream was analysed using an online Pfeiffer Vacuum quadrupole mass spectrometer. Further characterisation on the materials has been carried out including XRD, ICP BET,SEM.

Results and Discussion

XRD patterns of the catalyst samples are shown in figure 1 and show that Ce doping by ion exchange maintains the structural integrity of the OMS-2. Peaks positions are at the same 2θ values for Ce/OMS-2:AN and Ce/OMS-2:NH than for OMS-2. This shows that K^+ tunnel cations were successfully exchanged for Ce^{3+} or Ce^{4+} .XRD analysis also showed that Ce[0.75]-Mn was amorphous. Furthermore, ICP-OES analysis also confirmed that Ce/OMS-2:AN and Ce/OMS-2:NH contained low concentrations of Ce (approx. 5 wt.%) while Ce[0.75]-Mn had a Ce content of around 25 wt.%.

Figure 2 shows CO conversion, before and after catalyst aging, as a function of temperature for the different catalyst samples. It shows that the addition of Ceria has an impact on the activity of the OMS-2. The addition of Ce to OMS-2 samples Ce/OMS-2:AN and Ce/OMS-2:NH has provided an increase of the T_{50} value for CO oxidation of 8K and 21K respectively, compared to the un-doped OMS-2. However, Ce[0.75]-Mn showed a reduction of 14 K for the T_{50} value for CO oxidation. After ageing, CO light-off was over OMS-2 and CeO_2 was signifcanlty impacted. The CO T_{50} value was increased by 136 K over OMS-2 and 50% conversion wasn't achieved at 773 K over CeO_2. However, the Ce/Mn catalysts showed remarkable resistance to ageing, with CO T_{50} values increasing by 23 K, 4 K and 25 K over Ce[0.75]-Mn, Ce/OMS-2:NH and Ce/OMS-2:AN. Similar results were realised for C_3H_6 light-off.

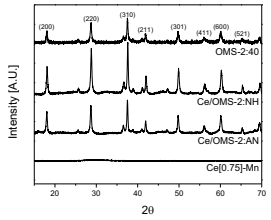

Figure 1 – XRD patterns of pure OMS-2:40, Ce doped OMS-2:40 (CeOMS-2:AN and Ce/OMS-2:NH) and a mixed Ce/Mn oxide (Ce[0.75]-Mn)

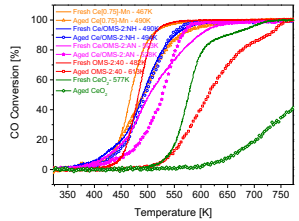

Figure 2 – CO light-off curves of OMS-2:40, CeOMS-2:AN, Ce/OMS-2:NH and Ce[0.75]-Mn compared to conventional CeO_2, before and after hydrothermal ageing.

Significance

This work has showed that the addition of Ceria to OMS-2 has provided catalysts which provide high activity for the oxidation of the exhaust gases in low temperature conditions and also have superior resistance to ageing compared to conventional catalysts, making them very suitable materials for automotive emission control.

References

[1] L. Pahalagedara *et al.*, "Applied Catalysis B : Environmental Benchmarking of manganese oxide materials with CO oxidation as catalysts for low temperature selective oxidation," *"Applied Catal. B, Environ.*, vol. 204, pp. 411–420, 2017.

[2] C. Chen *et al.*, "Structural Distortion of Molybdenum-Doped Manganese Oxide Octahedral Molecular Sieves for Enhanced Catalytic Performance," 2015.

[3] Y. Liu and J. Hou, "Ce ion substitution position effect on catalytic activity of OMS-2 for benzene oxidation," *Mater. Res. Bull.*, vol. 118, no. May, p. 110497, 2019.

[4] T. Montini, M. Melchionna, M. Monai, and P. Fornasiero, "Fundamentals and Catalytic Applications of CeO2-Based Materials," *Chem. Rev.*, vol. 116, no. 10, pp. 5987–6041, 2016.

UKCC 2020

6th UK Catalysis Conference, 7-9 January 2020
Loughborough, UK

Tuesday, 7th January			
11:00	Registration desk opens at Burleigh Court Hotel		
12:30	Lunch at Holywell Park		
13.50	Welcome – Conference commences at Holywell Park		
	Chair - Catlow		
14.00	Duncan Wass *(Turing Lecture Theatre)*		
14.45	Coffee		
	Session A *(Turing Lecture Theatre)*	*Session B* *(Brunel/Murdoch Lecture Theatre)* CatalysisHub session	*Session C* *(Stephenson Lecture Theatre)*
Chair/IT	Garforth/Deshmukh	Lennon/Shiels	Diez-Gonzalez/Keogh
15.15	K1	O22	O47
15.35		O23	O48
15.55	O1	O24	O49
16.15	O2	O25	K9
16.35	O3	O26	
16.55	Coffee		
Chair/IT	Taylor/Keogh	Kondrat/McDermott	Marr/Isah
17.25	O4	K6	O50
17.45	O5		O51
	Chair - Hardacre		
18.10	Johannes Lercher *(Turing Lecture Theatre)*		
20.00	Dinner		

Wednesday, 8th January			
	Chair - Hutchings		
9.00	Angelika Brückner *(Turing Lecture Theatre)*		
	Session A (Turing Lecture Theatre)	*Session B (Brunel/Murdoch Lecture Theatre)*	*Session C (Stephenson Lecture Theatre)*
Chair/IT	McGregor/Sun RSC INTEREST GROUP SURFACE REACTIVITY & CATALYSIS session	Fan/McDermott	Wood/Tanvir
9.50	K2	O27	O52
10.10		O28	O53
10.30	O6	O29	O54
10.50	Coffee		
Chair/IT	Thompson/Akor	Wu/Keogh	Reina/Hao
11.20	K3	O30	O55
11.40		O31	O56
12.00	O7	O32	O57
12.20	O8	K7	O58
12.40	O9		O59
13.00	Lunch		
	Chair - Manyar		
14.00	José Odriozola *(Turing Lecture Theatre)*		
14.45	Coffee		
	Session A (Turing Lecture Theatre)	*Session B (Brunel/Murdoch Lecture Theatre)*	*Session C (Stephenson Lecture Theatre)*
Chair/IT	Paterson/Yue RSC INTEREST GROUP APPLIED CATALYSIS session	Moody/McDermott	Whiston/Deshmukh
15.15	K4	K8	O60
15.35			O61
15.55	O10	O33	O62
16.15	O11	O34	K10
16.35	O12	O35	
16.55	Coffee		

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Chair/IT	Kroner/Shiels	Berlier/Sun	Raveendran/Akor
17.25	O13	O36	O63
17.45	O14	O37	O64
18.05	O15	O38	O65
18.30	**BP Poster session**		
20.00	Conference Dinner		
Thursday, 9th January			
	Session A (Turing Lecture Theatre)	*Session B (Brunel/Murdoch Lecture Theatre)*	*Session C (Stephenson Lecture Theatre)*
Chair/IT	Beale/Keogh	Artioli/Deshmukh	Upadhyayula/Tanvir
9.00	O16	O39	O66
9.20	O17	O40	K11
9.40	O18	O41	
10.00	K5	O42	O67
10.20		O43	O68
10.40	Coffee		
Chair/IT	Minova/Shiels	Mitchell/Deshmukh	Hintermair/McDermott
11.10	O19	O44	K12
11.30	O20	O45	
11.50	O21	O46	O69
	Chair - Davidson		
12.20	Stewart Parker *(Turing Lecture Theatre)*		
13.05	Closing remarks		

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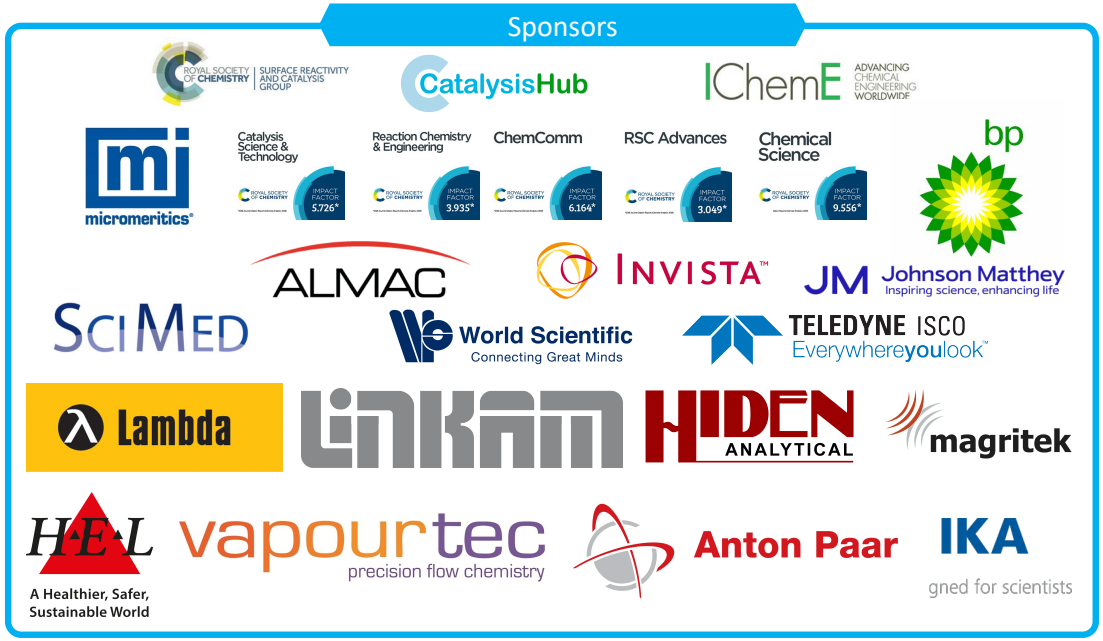

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List of Talks UKCC 2020

#	Title	Authors
Pl 01	Catalytic Conversion of Renewable Feedstocks to Advanced Synthetic Fuels	Duncan Wass
Pl 02	New strategies for enhancing catalytic rates	Johannes Lercher
Pl 03	Identifying active sites and mechanisms: Opportunities and limitations of in situ and operando spectroscopy in catalysis	Angelika Brückner
Pl 04	From electrons to reactors: The WGS revisited	José Odriozola
Pl 05	What's on your catalyst? Characterization of surface species on Pd and Pt catalysts	Stewart Parker
K 01	Elementary Steps in the Formation of Olefins from Surface Methoxy Groups in ZSM-5 and SAPO-34 Seen by Operando Infrared Microspectroscopy (OIMS)	Ivalina Minova, Santhosh Matam, Alex Greenaway, Richard Catlow, Mark Frogley, Gianfelice Cinque, Paul Wright and Russell Howe
K 02	Simultaneous removal of NO_x and soot particulate from diesel exhaust by in-situ catalytic generation and utilisation of N_2O	Anna Cooper, Catherine Davies, Kate Thompson, Stuart Taylor, Stan Golunski, Maria Bogarra Macias, Omid Doustdar and Athanasios Tsolakis
K 03	Cu-CHA for NO_x Selective Catalytic Reduction: insights into Cu speciation and reaction mechanisms by in situ spectroscopic techniques	Gloria Berlier
K 04	Insights into the CO_2 formation pathways over bimetallic Fischer-Tropsch catalyst for selective production of synthetic diesel: A theoretical & experimental study	Shashank Bahri and Sreedevi Upadhyayula
K 05	Catalytic scissoring of lignin C–C and C–O bonds	Wang, Luo, Liu and Li
K 06	Complex Transition Metal Oxides in Zeolitic Crystalline Forms As Selective Oxidation Catalyst	Wataru Ueda
K 07	Recent Strategies for the Application of Molecular Catalysts to Aqueous Substrates	Andrew Marr
K 08	(Trans)Forming C-N and C-O Bonds with Copper Catalysis	Silvia Díez-González
K 09	Renewable Furan building Block for Biorefinery Applications	Chandrashekhar Rode

K 10	Insight into the mechanism of hybrid non-thermal plasma catalysis system	Xiaolei Fan, Huanhao Chen, Yibing Mu and Chris Hardacre
K 11	MAX Phases and MXenes as Efficient Heterogeneous Catalysts	Shiju Raveendran
K 12	The Effect of a bimetallic Pd/Pt species on Catalyst Activity for Methane Oxidation	Jillian Thompson, Tang Son Nguyen and Andrew Beale
O 01	Investigating Mass Transport in Hollow Mesoporous Zeolites Used for Fluid Catalytic Cracking (FCC)	Luke Forster, Carmine D'Agostino, Xiaolei Fan and Yilai Jiao
O 02	Unravelling mass transport in hierarchically porous catalysts	Carmine D'Agostino, Neil Robinson, Mark Isaacs, Chris Parlett, Karen Wilson and Adam Lee
O 03	Effect of Flue Gas Impurities on the Capture and Utilisation of CO_2 in Superbase Ionic Liquids	Rebecca Taylor, Adam Greer, Helen Daly, Chris Hardacre, Matthew Quesne, Richard Catlow and Johan Jacquemin
O 04	Elucidating the Significance of Nitrate Speciation in Small-pore Cu-containing Zeolitic Materials for the NH_3-SCR reaction	Leila Negahdar, Naomi Omori, Mark Frogley, Fernando Cacho-Nerin, Wilm Jones, Stephen Price and Andrew Beale
O 05	Understanding fluorescence emission dynamics from zeolite crystals to yield insight into framework-adsorbate interactions	Naomi Omori, Alex Greenaway, Paul Collier and Andrew Beale
O 06	Using high throughput experimentation technology to understand effects in large scale reactors	Chris Mitchell and Xander Nijhuis
O 07	Understanding the Mechanochemical Synthesis of Perovskite $LaMnO_3$ and its Catalytic Behavior	Blackmore Rachel, Maria Elena Rivas-Velazco and Peter Wells
O 08	Perovskites decorated with exsolved Ni nanoparticles; operando monitoring of phase and structural changes that dictate redox methane conversion to syngas	Leonidas Bekris, Kalliopi Kousi, Dragos Neagu, Evangelos I. Papaioannou and Ian S. Metcalfe
O 09	Introduction to the High-Resolution Benchtop NMR	Anna Gerdova
O 10	Hydrogen partitioning as a function of time-on-stream for an un-promoted iron-based Fischer-Tropsch synthesis catalyst applied to CO hydrogenation	Alisha Davidson, Paul Webb, Stewart Parker and David Lennon
O 11	Simultaneous In Situ Study of Fischer-Tropsch Catalyst Series by XRD-CT	Jay Pritchard, Andrew Beale and James Paterson
O 12	Observing the Effects of Mn-promotion in Co-based Fischer-Tropsch Catalysts using In-situ Gas Cell Scanning Transmission Electron Microscopy	Matt Lindley, Sarah Haigh and James Paterson
O 13	Tuneable transesterification of glycerol with dimethyl carbonate for synthesis of	Gunjan Deshmukh and Ganapati Yadav

	glycerol carbonate and glycidol on MnO_2 nanorods and efficacy of different polymorphs	
O 14	Modified Red Mud as an Efficient Catalyst for the Synthesis of Glycerol Carbonate by the Transesterification of Glycerol	Bikashbindu Das and Kaustubha Mohanty
O 15	Solar water remediation: efficient removal of ciprofloxacin from aqueous solution using WO_3/TiO_2 photoanodes	Natalia S. Sabatin, Jonas H. Costa, Caio R. Silva, Taicia F. Pacheco, Jose R Guimarães and Claudia Longo
O 16	Alkyl lactate formation from the depolymerization of polylactic acid by metal complex catalysts	Luis Antonio Roman Ramirez, Paul McKeown, Fabio Lamberti, Matthew D. Jones and Joseph Wood
O 17	Hydrocracking of Post-Consumer Polyolefins	Abdulrahman Bin Jumah and Arthur Garforth
O 18	Catalytic Cracking of Polymers over Zeolites in a Twin Screw Compounder	Isaac Campbell, Aleksander Tedstone and Arthur Garforth
O 19	Tetralin and Naphthalene as Exemplar of Poly-aromatic in Heavy Oil Upgrading using $NiMo/Al_2O_3$ Catalyst Heated with Steel Balls via Induction	Abarasi Hart, Mohamed Adam, John Robinson, Sean Rigby and Joseph Wood
O 20	Catalytic upgradation of bio-oil derived phenolic compounds to fuel precursors	Gul Afreen and Sreedevi Upadhyayula
O 21	One-pot transformation of glucose to HMF using a dual acidic catalyst	Firdaus Parveen, Shashank Bahri and Sreedevi Upadhyayula
O 22	Hierarchical Porosity in Zeolite Catalysts for Plastic Hydrocracking	Aleksander Tedstone
O 23	Treatment of high ionic strength wastewater	Xiaoxia Ou, Chris Hardacre, Simon Beaumont, Arthur Garforth, Xiaolei Fan and Helen Daly
O 24	Advances in sustainable catalysis: A computational perspective	Matthew Quesne, Fabrizio Silveri, Nora de Leeuw and Richard Catlow
O 25	Improvement of biocatalyst performance using continuous flow	Sebastian C. Cosgrove, Itziar Peñafiel, Ashley P. Mattey, Nigel S. Scrutton, Nicholas J. Turner
O 26	Management of data objects derived from computational chemistry research for catalysis	Abraham Nieva de la Hidalga, Nitya Ramanan, Brian Matthews
O 27	$CeFeO_x$ catalysts for the total oxidation of propane and naphthalene VOCs: Influence of cerium precursor and molar ratios	Kieran Aggett and Stuart Taylor
O 28	Synthesis and catalytic application of Titanium Silicoaluminophosphate Molecular Sieves	Rekha Yadav, Shashank Bahri, Kanthi Pusapati and Sreedevi Upadhyayula
O 29	Extracting structural information of Au colloids at ultra-dilute concentrations: identification of growth during nanoparticle immobilization	George Tierney, Paul Collier, Nikolaos Dimitratos and Peter Wells

O 30	Metagenomic enzyme discovery to commercial bioprocessing	Thomas Moody, Megan Smyth and Scott Wharry
O 31	Application of Ru-tethered catalyst to generate optically active value added chiral alcohols	Vijyesh Vyas, Richard Knighton, Bhalchandra Bhanage and Martin Wills
O 32	A heterogeneous platform for biocatalytic asymmetric deuteration	Jack Rowbotham, Miguel Ramirez Hernandez, Oliver Lenz, Holly Reeve and Kylie Vincent
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O 34	Highly selective reduction of α,β-unsaturated aldehydes, ketones and carboxylic acids under ambient conditions using tetraalkylphosphonium ionic liquids	Stephen Mc Dermott, Kathryn Ralphs, Eadaoin McCourt, Christopher Ormandy, Thiago A. Carneiro de souza, Peter Nockemann, Johan Jacquemin and Haresh Manyar
O 35	Catalytic Hydrogenolysis of 5-hydroxymethylfurfural via Polyphenylene Supported Ruthenium Catalyst	Xuze Guan, Ryan Wang and Qiming Wang
O 36	Electrochemical oxidation of dibenzothiophene and 4,6-dimethyldibenzothiophene on a silver/polyaniline modified electrode	Adeniyi Ogunlaja
O 37	Isolated Pd sites as selective catalysts for electrochemical and direct hydrogen peroxide synthesis	Simon Freakley
O 38	Pt-Pd Single Atom Alloys supported on carbon for the Oxygen Reduction Reaction in combination with XAFS	Sushila Marlow, Ruoyu Xu and Feng Ryan Wang
O 39	A DFT Study of SO_x (x = 0 – 3) and H_2O Reactivity on Pt (111) surface	Nora H. de Leeuw, Cornelia G.C.E. van Sittert, Marietjie J. Ungerer and David Santos-Carballal
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O 41	Computational studies on poisoning of Ni catalyst in Methane Steam Reforming	Sai Sharath Yadavalli, Glenn Jones and Michail Stamatakis
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O69	Effect of steam de-alumination on the interactions of propene with H ZSM-5 zeolites	Alex Hawkins, Andrea Zachariou, Stewart F. Parker, Paul Collier, Iain Hitchcock, Ian P. Silverwood, Russell Howe and David Lennon