Ce Mn mixed oxides for low temperature catalytic after treatment applications

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Introduction

Manganese oxides molecular sieves (OMS) materials have 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. Cryptomelane is an OMS-2 mateiral which has a microporous, nano-tunnel structure composed of edge shared MnO_6 octahedra that form a 2 x 2 arrangement, with a K^+ cation positioned within the framework [1]. The structural incorporation of various dopants and tunnel cations can further enhance the functionality of OMS-2 [2]. Other synthesis routes can be used for the 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 and tested for applications in automotive emission abatement. Due to the high emissions of lean burn diesel engines operating at low temperatures and during cold start, and issues with catalyst deactivation, the requirement for a catalyst which is active at low temperature is one of the main challenges in automotive emission control [4]. In particular, we have investigated the use of manganese oxide hybrid catalyst supports for their applicable use in automotive after treatment. We have studied 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 $Ce(NO_3)_3$ and deionised water, using a theoretical Ce/Mn ratio of 0.5, and stirred for 48 h at room temperature and the resulting sample was dentoed as Ce[0.5]-OMS-2. Amorphous Ce/Mn mixed oxides were synthesised by adding $Ce(NO_3)_3 \cdot 6H_2O$ during the synthesis of OMS-2, prior to the addition of maleic acid. A Ce/Mn molar ratio of 0.5 was used in the precursor solution and the sample was denoted as Ce[0.5]-Mn.. 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 using the same equipment for 24 h at 873 K with a feed of 5% H_2O and 10% O_2, with Ar used as the balance gas. Following the ageing process, the activity of the sample was tested 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[0.5]-OMS-2 than for OMS-2:40. This suggests that K^+ tunnel cations are exchanged for Ce^{3+} or Ce^{4+} inside the tunnel framework. XRD analysis also showed that Ce[0.5]-Mn was amorphous and there was no presence of the distinct OMS-2 tunnel structure. Furthermore, ICP-OES analysis also confirmed that Ce[0.5]OMS-2 contained a Ce concentration of around 5 wt.%, while Ce[0.5]-Mn had a Ce content of around 40 wt.%, despite using the same Ce/Mn mole ratio in the precursor solution.

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-2:40 by ion extraction in Ce[0.5]-OMS-2, resulted in the CO T_{50} to increase by around 20 K compared to the un-doped sample. However, Ce[0.75]-Mn which was synthesized using the one-pot sol-gel method, showed a reduction of the CO T_{50} value of around 30 K. After ageing, CO light-off was over OMS-2 was reduced significantly. The CO T_{50} value was increased by 136 K over OMS-2, however, the Ce/Mn catalysts showed remarkable resistance to ageing, with CO T_{50} values increasing by only 145 K and 25 K over Ce[0.5]-OMS-2:40 and Ce[0.75]-Mn respectively. A similar effect was observed for C_3H_6 light-off.

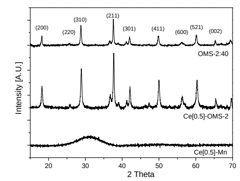

Figure 1 – XRD patterns of pure OMS-2:40, Ce[0.5]-OMS-2 and Ce[0.5]-Mn

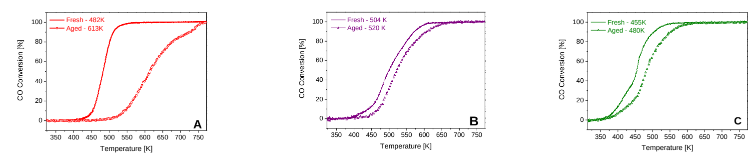

Figure **Errore. Nel documento non esiste testo dello stile specificato.** – CO conversion curves over fresh and aged samples of OMS-2:40 (A), Ce[0.5]-OMS-2 (B) and Ce[0.5]-Mn (C)

Significance

This work has showed that the improvement of the catalytic activity for low temperature oxidation of automotive emissions through the incorporation of ceria and manganese oxides, particularly as a mixed oxide material. The addition of ceria has also shown to significantly improve resistance to ageing compared to conventional OMS-2, highlighting their potential for automotive emission control applications.

References

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[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.

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Plenary Speakers

Tim Bugg

'Bacterial Enzymes for Lignin Degradation'

Tim Bugg is Professor of Biological Chemistry at the University of Warwick. His academic career started at the University of Southampton in 1991, where his group studied enzymes involved in the bacterial degradation of aromatic compounds and enzymes involved in bacterial peptidoglycan assembly. Since moving to Warwick in 1999, his group has more recently studied enzymes involved in bacterial degradation of lignin, and the application of biocatalysis to convert lignin into renewable aromatic chemicals. He is the author of the undergraduate textbook "Introduction to Enzyme and Coenzye Chemistry".

Alessandra Quadrelli

'Surface Organometallic Chemistry on MOFs, POPs and Inorganic Oxides for CO2 and N2 Reduction: En route to Renewable Energies Storage'

Alessandra is director of research of the French National Centre for Scientific Research, CNRS, at the nanochemistry platform of the C2P2 labs. She also chairs the CPE Lyon Engineering School Sustainable Development Chair and is Associate Editor of the RSC journal "Green chemistry".

Her research focuses on developing molecular understanding of the interaction between organometallic precursors and solid surfaces of SiO2, MOFs and 2D wafers (among other solids). She applies this understanding to the synthesis of heterogeneous catalysts and thin films aimed at renewable energy utilization. She considers her Top-3 professional achievements: A new mechanism for N2 cleavage (SCIENCE, 2007), the creation of the "CO2 forum" conferences (http://co2forum.cpe.fr) and the synthesis of a MoS2 monolayer by Atomic Layer Deposition, ALD (NANOSCALE, 2017).

Enrico Tronconi

'The NH3-SCR Redox Cycle over Cu-CHA: Insights from Transient Response Methods'

Enrico Tronconi is a Professor of Chemical Engineering at the Department of Energy of Politecnico di Milano, Italy. His research interests concern the applications of Catalytic Reaction Engineering to environmental protection and energy conversion. Enrico has investigated DeNOx aftertreatment technologies during the last twenty years. He is also active in the study of novel structured catalysts and reactors for process intensification.

Programme

Monday, 7 September 2020

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13:10		**Plenary Speaker - Enrico Tronconi - The NH3-SCR redox cycle over Cu-CHA: Insights from transient response methods** Chair Nancy Artioli Zoom link Passcode: 564253			
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Paraskevi Panagiotopoulou https://zoom.us/j/97399415584
Effect of operating conditions on the catalytic performance of supported Rh catalysts for the reaction of LPG steam reforming

Sebastián Gámez https://zoom.us/j/92504964904
Carbon Black-Polydopamine-Ruthenium composite as an efficient and recyclable boomerang catalyst for the oxidative cleavage of oleic acid

Olívia Salomé Soares https://zoom.us/j/91947639213
Tuning the surface properties of carbon supporting materials to achieve efficient Ni based catalysts for CO2 methanation

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Madan Behera https://zoom.us/j/95673586731
NO reduction using Pt-zeolite catalysts in O2 and CO2 rich gas atmosphere

Alain Li https://zoom.us/j/99638543980
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Luke Roebuck https://zoom.us/j/93843924326
Rare-Earth Doped Ceria-Zirconia Nanodispersions: Oxygen Storage Materials for Gasoline Particulate Filters

Theodora Ramantani https://zoom.us/j/95216791843
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Aidan Doyle https://zoom.us/j/94588987681
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Daniela Pietrogiacomi https://zoom.us/j/93672747929
Oxidative dry reforming of methane for syngas production: a promising activity of Ni/ZrO2 catalysts

Lioudmila Nossova https://zoom.us/j/95479118807
Co- and Zr-doped barium cerate perovskite catalyst for simultaneous NOx storage and soot oxidation

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Effect of ball-milling and plasma treatment on microcrystalline cellulose on the H2 production via cellulose photoreforming

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Aleksandra Borcuch https://zoom.us/j/91333867881
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Aneta Święs https://zoom.us/j/99716227131
Catalytic performance of modified ferrierites as effective catalysts for catalytic reduction of NO with ammonia

Nicolaas van Strien https://zoom.us/j/91282813973
Unique pathway to platform chemicals - 2,5-furandicarboxylic acid and muconic acid from sugar acids

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Effect of zironium on catalytic combustion of vinyl chloride over Co3O4-based catalysts

Tamara Kharlamova https://zoom.us/j/93846412069
Ceria-supported Pt–Ag bimetallic catalysts for CO oxidation and hydrogenation of nitrophenol

Małgorzata Sieradzka https://zoom.us/j/92460388107
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Transient Response Method to delve into NOx removal process using a hybrid NSR-SCR system

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Cyril Thomas https://zoom.us/j/98488294172
Exceeding the 2 wt% Ag Loading Frontier on Al2O3 for C3H6-SCR: Insights into the identification of the Al2O3 sites of importance

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Adrian Mizera https://zoom.us/j/97269257932
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