Using Ion-beam Sputtering to modify Heterogeneous Nanodispersed Catalysts

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Introduction

Interface science is at the center of the development of new materials for interesting technically materials. Ion beam engineering is one technique which is used for the development of such materials[1]. The work carried out in this project shows that ion beam sputtering can be used as a technique for the controlled surface modification of solid heterogeneous catalysts. The surface nanostructure of these materials can be strategically manipulated to enhance their catalytic performance and resistance to aging. Through doing so this can also reduce the noble metal substitution on the catalysts oxide support[2]. The technique has been investigated using a Pt/$Ce_{0.7}Zr_{0.3}O_2$ catalyst which is a conventional emission control catalyst used mobile combustion engines[3][4]. The technique has been studied by changing different parameters of the ion beam to understand how they independently effect the catalyst. Modeling of the treatment has also been introduced to the describe erosion of the surface through ion sputtering, and how this is influenced by the type of ion used, the number of doses and the current of the ion beam.

Materials and Methods

A sample of Pt (1% w/w) $Ce_{0.5}Zr_{0.5}O_2$ was commercially sourced. Three samples were then bombarded with N^+ ion beams with an energy of 1.5KeV, a current of 20mA and an incident angle of 28˚ to the catalyst surface. Each sample received a different number of doses, with Pt1.5(32) treated 32 times, Pt1.5(16) treated 16 times and Pt1.5(8) treated 8 times. The samples were mechanically agitated between each dose to expose untreated surfaces. The treated samples were compared with an untreated sample, Pt fresh. Catalytic testing was carried out on all samples by using a reaction mixture composed of 10% O_2, 4.5% H_2O, 2000ppm CO, 2000ppm CH_4, 2000ppm C_3H_6, and 200ppm NO with a total flow of 100ml/min. The temperature was increased from 303K to 773K at a rate of 5K/min and the cycle was repeated three times to test the stability of the catalyst. The outlet from the reactor was analysed using a Pfeiffer Vacuum quadrupole mass spectrometer. Extended X-ray Absorption Fine Structure (EXAFS) were carried out at Diamond Light Source Ltd. UK, using fluorescence mode on beamline B18. Spectra were collect at the Pt L_3-edge on each of the samples to investigate the size and distribution of the Pt nanoparticles on the catalyst surface.

Results and Discussion

The catalytic activity was assessed and the results are reported in terms of CO and C_3H_6 conversion in fig. 1. The results show that the catalyst showed improved catalytic activity for oxidation of both CO and C_3H_6 after ion bombardment. The temperature at which 50% conversion was achieved (T_{50} values) for CO oxidation were 503K, 503K and 511K for Pt1.5(32), Pt1.5(16) and Pt1.5(8). This shows improved catalytic activity compared to the untreated sample Pt fresh which had a T_{50} values of 513K. The T_{50} values for C_3H_6 oxidation over Pt1.5(32), Pt1.5(16) and Pt1.5(8) were 528K, 533K and 538K respectively, which also shows the enhanced activity at low temperatures compared with Pt fresh which had a T_{50} values of 546K.

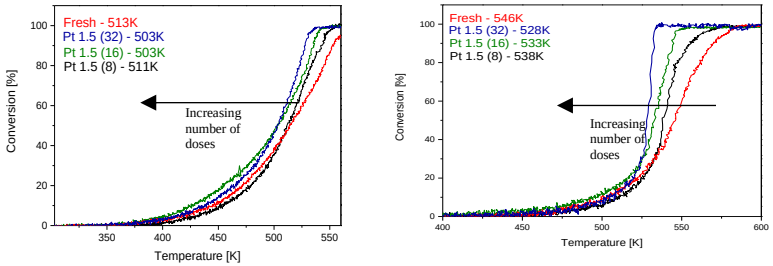

Figure 1. CO conversion (a) and C_3H_6 conversion (b) as a function of temperature for the Pt (1% w/w) $Ce_{0.5}Zr_{0.5}O_2$ fresh and traeted with different ion doses (8-16-32).

The results are in line with EXAFS Fourier transform analysis (Fig. 2) which revealed that the sample, after ion bombardment, is characterized by a distribution of Pt nanoparticles which is decreasing with the intensity of the bombardment, as well as by the formation of atom vacancies and incomplete terraces (HRTEM and H_2 reduction FTIR studies).

Significance

Ion beam irradiation has been used as a post synthesis technique for surface modification of typical heterogeneous emission control catalysts. The study has been carried out using Pt 1w/w% $Ce_{0.5}Zr_{0.5}O_2$, which is a typical emission control catalyst. The results show that the technique can be used as a method for the manipulation of the surface properties of the material, and thus the enhancement of its catalytic properties, lowering the temperature at which the material is active. This opens up the idea of using ion bombardment as a useful tool for the controlled processing of nanodispersed heterogeneous catalytic materials.

References

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Columbus ABCDEF	Columbus GH	Columbus IJ	Columbus KL
2:10 PM **Wed-CHAB-1410** Thermodynamic Stability of Molybdenum Oxycarbides Formed from Orthorhombic Mo_2c in Oxygen-Rich Environments. **Carrie Farberow**, *National Renewable Energy Laboratory, USA.*	2:10 PM **Wed-CHGH-1410** Effect of Ag Metal Promoter Dispersion on the Single Step Conversion of Ethanol to Butadiene over $Ag/ZrO_2/SiO_2$ Catalysts. **Austin Winkelman**[1,2], Sneha Akhade[3], Libor Kovarik[2], Roger Rousseau[2], Vassiliki-Alexandra Glezakou[2], Yong Wang[1,2], Vanessa Dagle[2] and Robert A. Dagle[2], *(1)Washington State University, USA, (2)Pacific Northwest National Laboratory, USA, (3)Lawrence Livermore National Laboratory, USA.*	2:10 PM **Wed-CHIJ-1410** Converting 'Nearly Dry' Methane to Electricity at a Practical Temperature By Integration of Electro- and Thermal- Catalysis. Yu Tang[1], Yu Chen[2], Ben deGlee[2], Meilin Liu[2], Ziyun Wang[3] and **Franklin Tao**[1], *(1)University of Kansas, USA, (2) Georgia Institute of Technology, USA, (3)The Queen's University, United Kingdom.*	2:10 PM **Wed-CHKL-1410** OMS-2 Molecular Sieves Doped with Ceria for the Development of New Emission Control Catalyst. **Nancy Artioli**, Haresh Manyar, Kathryn Ralphs, Maxime Grolleau, Ruiari O'Donnell and John Duffin, *Queen's University Belfast, United Kingdom.*
2:30 PM **Wed-CHAB-1430** A Topological Model for the Adsorption of Polycyclic Aromatic Hydrocarbons on Late-Transition Metal Surfaces. **Zhao-Bin Ding**, Matteo Tommasini and Matteo Maestri, *Politecnico di Milano, Italy.*	2:30 PM **Wed-CHGH-1430** Isobutene from Ethanol: Describing the Synergy between in_2O_3 and $m\text{-}ZrO_2$. Priscila da Costa Zonetti[1], Vivian Bridi[1], Guilherme G. Gonzalez[1,2], Carla R. Ramos[1], Odivaldo C. Alves[3], Roberto R. Avillez[4] and **Lucia Gorenstin Appel**[1], *(1)Instituto Nacional de Tecnologia, Brazil, (2)Universidade Estadual do Rio de Janeiro, Brazil, (3)Universidade Federal Fluminense, Brazil, (4) Pontifícia Universidade Católica do Rio de Janeiro, Brazil.*	2:30 PM **Wed-CHIJ-1430** Independent Tuning of Active Sites in Bifunctional Methanol Electro-Oxidation Catalysts. **Adam Baz** and Adam Holewinski, *University of Colorado Boulder, USA.*	2:30 PM **Wed-CHKL-1430** Bow-tie-Shaped $NiCo_2O_4$ Catalysts for Low-Temperature Methane Combustion. **Yiling Dai**, PavanKumar Vanama, Chujie Zhu, Haiyan Wang, Kevin Smith, Michael Wolf and Mark MacLachlan, *University of British Columbia, Canada.*
2:50 PM **Wed-CHAB-1450** Degree of Mass Transfer Control and CFD Analysis of Ammonia Oxidation Catalyst: A Tool for Investigating Mass Transfer Limited Catalytic Processes. **Michael Haas**, *Technische Universität Darmstadt, Germany.*	2:50 PM **Wed-CHGH-1450** Mechanism and Kinetics of Ethanol and Acetone Conversion to Isobutene over $Zn_xZr_yO_z$. **Julie Rorrer**[1], Alexis T. Bell[1] and F. Dean Toste[1,2], *(1) University of California, Berkeley, USA, (2) Lawrence Berkeley National Laboratory, USA.*	2:50 PM **Wed-CHIJ-1450** Understanding How Platinum Excels at the Hydrogen Evolution Reaction. **Andrew A. Peterson**, Per Lindgren and Georg Kastlunger, *Brown University, USA.*	2:50 PM **Wed-CHKL-1450** Using Ion-Beam Sputtering to Modify Heterogeneous Nanodispersed Catalysts. **Ruiari O'Donnell**, Salvatore Scaglione, Rosa Chierchia, Veronica Celorrio and Nancy Artioli, *Queen's University Belfast, United Kingdom.*
3:10 PM – 3:40 PM: Coffee Break			