

15 Second XAFS Scans during In Situ Oxidation/Reduction of Copper Doped Ceria WGS Catalyst

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Introduction

In situ XAFS and XRD has been shown to be useful in the characterization of a Cu doped Ceria under WGS reaction conditions[1]. The active form of the catalysts is shown to be metallic copper on the surface of the ceria. The Cu has been shown to go reversibly in and out of the cerium positions in the ceria fluorite lattice. This process is very rapid and therefore we have used a new quick XAFS technique in to study the kinetics of this process.

Materials and Methods

The XAFS data were collect at beam line X18B at the NSLS. The rapid oscillation of the energy for the XAFS scans is obtained with a cam drive on the channel cut monochromator. The intensity of the ion-chambers and other current-integrating detectors (Lytle-detector, PIPS-detector) is determined with current-amplifiers, whose output is directly measured with a fast, multiplexing ADC. The energy of the x-rays is calculated from the angle of the monochromator, which is measured with a rotary Heidenhain encoder. The acquisition system reads the intensity of all detectors simultaneously, and with a small time-delay the position of the monochromator. The cam and additional hard- and software are designed to collect data of a complete XAFS-scan in about 0.5s with about 500 points. Velocities of 20scans/s in the XANES region have been obtained in some tests with reference-foils. Isothermal reductions and reoxidations data were collected in fluorescence mode with a PIPS detector in an in situ plug flow cell at 275, 300 and 325°[2]. The Ce_{0.8}Cu_{0.2}O₂ was prepared in the method of Marcos[1].

Results and Discussion

We have observed clean XANES spectrum during the transformation of the ionic copper at the cerium position in the fluorite lattice to metallic copper at a rate of 15 seconds per 500 eV scan. (Figure 1). Analysis of the kinetics from the data at the three temperatures is in progress.

Significance

The time resolution of in situ XANES has been reduced to 15 seconds per scan.

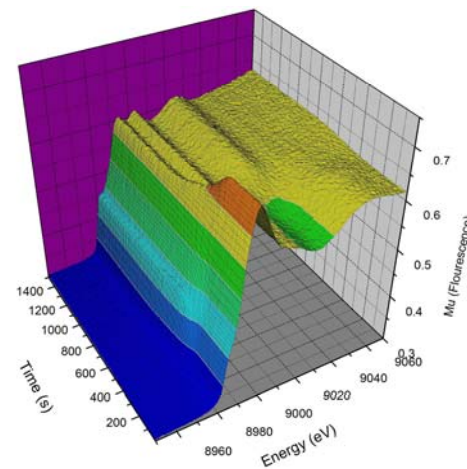


Figure 1. Series of XANES spectra obtained during the reduction of Ce_{0.8}Cu_{0.2}O₂.

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References

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