2001A0465-NX-np BL01B1

EXAFS Study of Local Structural Disorder in Heavy-fermions and Non-Fermi Liquid Materials

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Introduction

Several intermetallic compounds of Ce exhibit non-Fermi liquid scaling behaviors such as linear temperature dependence of resistivity and logarithmic scaling of specific heat at low-temperatures. Recent experimental and theoretical studies strongly suggest that structural and chemical disorder might play a crucial role in the non-Fermi liquid behaviors in these materials. In this experiment, we have applied the x-ray absorption fine structure (XAFS) to investigate the disorder in heavy-fermion and non-Fermi liquid compounds with $ThCr_2Si_2$ type and $BaNiSn_3$ type tetragonal crystal structure.

Experimental

Polycrystalline samples of $CeCu_2Si_2$, $CeNi_2Ge_2$, $CeCoSi_3$ and $CeCoGe_{1.5}Si_{1.5}$ were prepared by arc metling and were homogenized by annealing under vacuum at the prescribed temperatures for several hours. The XAFS spectra were measured in transmission mode at Ce K edge in the range of 8–295 K (also at Co K and Cu K edges in few samples at RT) in $CeCu_2Si_2$, $CeNi_2Ge_2$, $CeCoSi_3$, $CeCoGe_{1.5}Si_{1.5}$ using a Si(311) double crystal monochromator at BL01B1.

Results and Discussion

Fugure 1 displays the Fourier transforms of Ce K edge XAFS spectra in $CeNi_2Ge_2$, $CeCu_2Si_2$, $CeCoSi_3$ and $CeCoGe_{1.5}Si_{1.5}$. The first peak in the |F(r)| at 2.4 ~ 2.8 Å corresponds to the first nearest neighbor Ge/Si atom. The amplitude of this peak is strongly reduced in the later samples, particularly severe attenuation is observed in $CeCoGe_{1.5}Si_{1.5}$. These features indicate the degree of static disorder in each sample. Further the double peak structure (2.44 Å, 2.96 Å) found in $CeCoGe_{1.5}Si_{1.5}$ may be signaling a two phase behavior. These results contribute to the understanding of the relation between type of disorder and the heavy-fermion or non-Fermi liquid behavior.

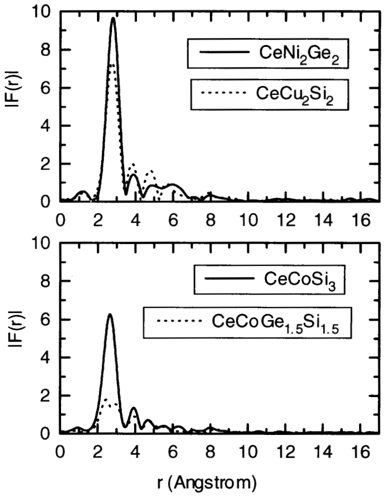

Fig. 1: Fourier Transforms of Ce K edge XAFS spectra in Ce based heavy-fermion or non-Fermi liquid materials.

2001A0473-NX –np BL01B1

Study of specific structure of micro zirconium oxides dispersed on the porous materials which surface area exceeds 1,000 m^2/g

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Supported Zr oxide catalysts are active for the selective reduction of unsaturated carbonyl compounds to corresponding unsaturated alcohols. The activity and selectivity significantly depend on kind of support and loading of Zr. In this study, effect of physico-chemical property of support was investigated on the structure of Zr oxide supprted on porous material with highly surface area (>1000 m^2/g).

XANES spectra of the supported Zr catalysts was indicated in Fig. 1. The spectra of the reference samples, ZrO_2 and $ZrSiO_4$, are also indicated in Fig. 1. The XANES spectra of the catalysts were similar to that of ZrO_2, whereas a significant shoulder peak was observed between 18038 and 18040 eV on Zr/SiO_2, Zr/MCM41, and Zr/TiO_2 also as observed in the spectra of $ZrSiO_4$. These results suggest that different coordination of zirconium oxide will be expected on Zr/SiO_2, Zr/MCM41, and Zr/TiO_2. The Zr K-edge EXAFS are shown in Fig. 2. On Zr/SiO_2 and Zr/MCM41, two different oxygen coordination were observed at ca. 2.0 and 2.3~2.4 Å, in addition to Zr-Zr coordination (ca. 3.4 Å). The active carbon-supported Zr indicated a different EXAFS from Zr/MCM41. MCM41 and active carbon have markedly high surface area, more than 1000 m^2/g. These results indicate that the state of zirconium was strongly affected by surface nature of supports. Zirconium oxide was not simply dispersed on the high surface area supports. It should be noted that the markedly low coordination numbers of oxygen were obtained on Zr/MCM41.

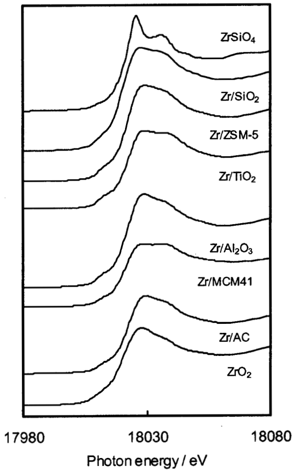

Fig. 1 Zr K-edge XANES spectra of the supported Zr catalysts

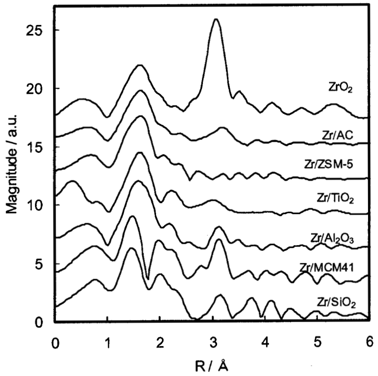

Fig. 2 Zr K-edge EXAFS of the supported Zr catalysts (Zr: 5 wt%)