

Mixed valence states of amorphous Ce-Ru alloy system

Ikuo NAKAI*¹, Yingjie LI¹, Yusuke AMAKAI², Shigeyuki MURAYAMA²

¹Tottori Univ., Tottori 680-8552, Japan

²Muroran Institute of Technology, Muroran, Hokkaido, 050-8585, Japan

Introduction

Structure disordered Ce-Ru alloy system shows a variety of electronic properties such as a heavy fermion like behavior in a Ce-rich concentration and superconductivity in a Ru-rich region [1-3]. It is well known that the Ce atom has both of trivalent ($4f^1$) and tetravalent ($4f^0$) configurations. Therefore it is considered that the valence state of Ce atoms is a key to understand them. In the work we have measured XANES spectra at a Ce L_3 edge of the amorphous $\text{Ce}_x\text{Ru}_{1-x}$.

The amorphous phase of Ce-Ru alloys ($\text{a-Ce}_x\text{Ru}_{100-x}$) with Ce content x of 9, 43 and 80 at% was prepared by DC sputtering. The cubic Laves phase intermetallic compound CeRu_2 was also prepared as a reference

XANES spectra were measured at 295 K in the transmission mode at the 9A beam line of PF.

Results and discussion

Figure 1 shows the normalized Ce L_3 edge XANES spectra for $\text{a-Ce}_x\text{Ru}_{100-x}$, which consist of three terms such as transitions from $2p_{3/2}$ to the continuum and to a couple of unoccupied $5d$ states corresponding to the $4f^1$ (peak G) and the $4f^0$ (peak H) configuration. The solid line is the best fit to the observed data denoted by the open circle. The broken line represents each term. It is clear that the tetravalent component decreases with increasing Ce concentration.

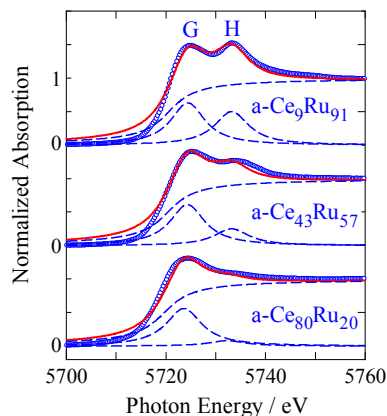


Fig. 1. Normalized Ce L_3 absorption of $\text{a-Ce}_x\text{Ru}_{1-x}$ alloys plotted as a function of photon energy.

The energy E_0 at onset of the continuum and the valence of $\text{a-Ce}_x\text{Ru}_{100-x}$ estimated from the area of peaks G and H are plotted as a function of Ce content in Fig. 2. It is also shown for the CeRu_2 compound as comparison. These parameters for $\text{a-Ce}_x\text{Ru}_{100-x}$ as well as the CeRu_2 compound depend linearly on Ce content as shown by the

solid line. Although the valence tends toward trivalence with increasing the Ce content, the extrapolation predicts that an amorphous Ce pure metal, if it exists, will be still in the mixed valence state. The heavy fermion like behavior in $\text{a-Ce}_x\text{Ru}_{100-x}$ with the high Ce content comes from strongly correlated $4f$ electrons [1-3].

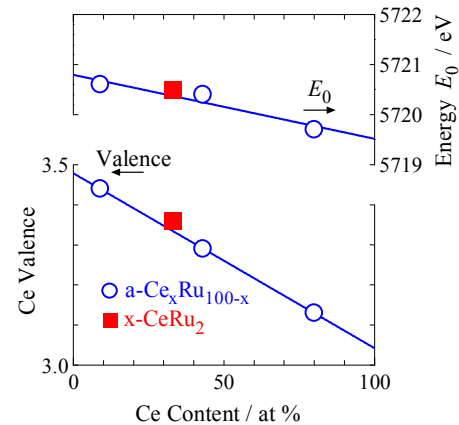


Fig. 2. The Ce valence and the energy E_0 plotted against Ce content.

It is easy to understand from Fig. 2 that E_0 increases with increasing Ce valence. This is a well-known phenomenon as the chemical shift that the absorption edge shifts to higher energy with increasing valence. In the case of the Ce based materials, the energy shift arises from Coulomb attraction between the $2p_{3/2}$ core hole and the $4f$ and/or $5d$ electron and from Coulomb repulsion between the $4f$ electron and the excited $5d$ electron. It is considered that the energy shift of E_0 with valence is mainly attributed to the Coulomb attraction, while the Coulomb repulsion has only a small effect. The Coulomb attraction at the $4f^1$ final state is larger than that of the $4f^0$ final state. In other words, the Ce $2p_{3/2}$ level moves toward lower energy with increasing valence. The valence state of Ce depends on the structure around Ce atoms. Investigation on the local structure of $\text{a-Ce}_x\text{Ru}_{100-x}$ is now in progress.

References

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* nakai@ele.tottori-u.ac.jp