Materials Science

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Nanocrystalline Lu_2O_3:Tb studied by X-ray absorption spectroscopy

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Introduction

Recently, lutetium-based compounds such as Lu_2O_3 attract increasing interests due to their good physical and chemical properties as scintillant for the detection in the field of high-energy and particle physics, nuclear science and medical imaging.[1,2] However, the single crystal preparation of the Lu_2O_3 using Ceochralski technology is an expensive, time-consuming and size-limiting process because of its high melting point of 2450ºC. One method is to use nanocrystalline, which can be synthesized at low temperature. The luminescence property of the nanocrystalline is influenced by the local structure around luminescence center. In this work, we use X-ray absorption spectroscopy to study the valence and local structure of Tb ions for different particle sizes Lu_2O_3:Tb.

Experimental

Different particle sizes (29 nm, 20 nm, 15 nm and 5 nm) nanoscale Lu_2O_3:Tb(1%) was prepared by combustion method using glycine as a fuel. Tb L_3 edge X-ray absorption spectroscopy were made at beamline BL-12c at PF-KEK in fluorescence mode with a 13 elements Ge solid state detctor. All spectroscopy were measured at room temperature. EXAFS Data analysis was performed using Athena and Artemis package.

Result and Discussion

Figure 1 shows the Tb L_3 edge XANES spectra of different particle size Lu_2O_3:Tb samples and Tb_4O_7. The Tb_4O_7 spectrum has two much broader peaks centered at 7518 eV and 7525.4 eV, which result from $2p^65d^04f^8 \rightarrow 2p^55d^14f^8$ (Tb^{3+}) and $2p^65d^04f^8 \rightarrow 2p^55d^14f^7$ (Tb^{4+}) transition, respectively. The XANES spectra of Lu_2O_3:Tb with particle size larger 15 nm show only one strong white line at 7518 eV, which indicates the main valence of Tb ions is +3. But when the particle size decreases to 5 nm, an obvious peak around 7525 eV appears which suggests that considerable Tb^{4+} exist in the sample.

Fig.2 shows the Fourier transformation of EXAFS at Tb L_3 edge. When the particle sizes decrease to 5 nm, the first peaks exhibit a decreasing of the intensities and become broad while the second and the third peaks are strongly attenuated and nearly disappear. The phenomena indicate that there are dramatic changes of local structure in the small particle size samples.

The fitting result shows that, when the particle size reduce to 5 nm, Tb-O bond lengths increase from 2.27 Å to 2.37 Å and the coordination numbers slightly increase from 6 to 6.6. Meanwhile, the Debye-waller factors increase obviously indicating large disorder in the small size sample. The obtained local structure information will be used to explain the size dependence of luminescence property of Lu_2O_3:Tb.

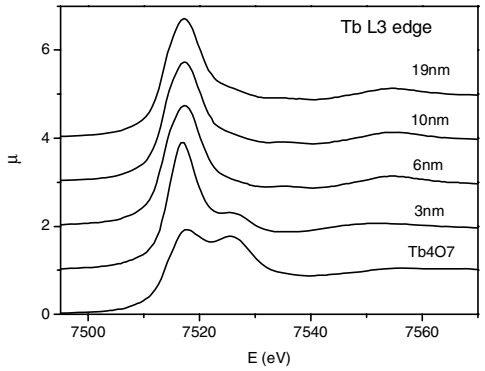

Fig.1 Tb L3 XANES of different particle size Lu_2O_3:Tb and Tb_4O_7

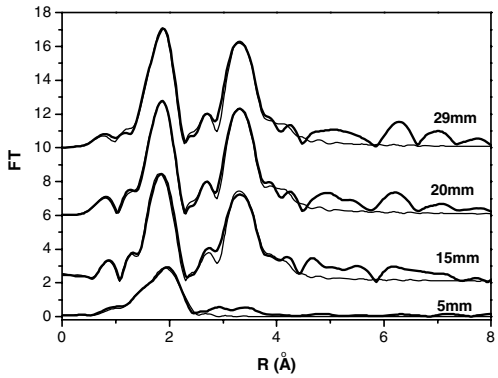

Fig.2. Fourier transformation of EXAFS at Tb L_3 edge for different particle size Lu_2O_3:Tb. thick line: experimental; thin line: fitting

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