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Synthesis and structure of Ce nitrides in high pressure and temperature

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Introduction

The combination of the diamond anvil cell and YAG laser heating (LASER-DAC) is a useful technique for the synthesis of metal nitrides in a supercritical nitrogen fluid. The synthesis and crystal growth of metal nitrides of simple metal, transition metal and rare earth metal using the LASER-DAC system [1-5] have been reported.

Recently, we have also succeeded in synthesizing two new lanthanum nitrides in a supercritical nitrogen fluid at high pressure (about 30 GPa) and high temperature (about 2000 K) using the LASER-DAC heating system. Since La nitrides are usually synthesized as the NaCl-type structure under ambient pressure, they are novel La nitrides which are different from NaCl-type nitrides. Other Lanthanide nitrides are also usually synthesized as the NaCl-type structure under ambient pressure. Accordingly, it is expected that novel Ce ones can be obtained by the LASER-DAC as well as the La nitrides. This article reports the synthesis of other Ce nitrides in a supercritical nitrogen fluid at about 30 GPa by the LASER-DAC in comparison with the novel La nitrides.

Experimental Procedure

The YAG laser (100W CW multimode) was used for sample heating. The sample was observed during heating using a CCD camera. The culet size of the diamond anvil was 450 μm in diameter. A rhenium gasket was used and the gasket hole was about 200 μm in diameter. Synthesis pressure was about 30 GPa, which was measured before and after laser heating using the ruby fluorescence technique at room temperature. Starting materials were solid Ce metal and gaseous nitrogen.

High-resolution angle dispersive X-ray diffraction experiments were carried out using synchrotron radiation at the Photon Factory in Tsukuba. A monochromatized X-ray was collimated to a thin beam and used to irradiate the sample. The diffracted X-ray was detected by an imaging plate (IP). The X-ray intensities recorded on the IP were measured by a laser-scanning reader and converted into two-dimensional digital intensity data. These data were then integrated along the polar axis which coincided with the Debye rings observed on the IP. Typical exposure times were about 20 min.

Results and Discussion

The pattern at about 30 GPa and room temperature before heating is identified by the combination of tetragonal (*tI*2) Ce metal and solid nitrogen since the starting materials, Ce metal and nitrogen, do not react at this stage and the NaCl thermal insulator was not used in this experiment. The X-ray diffraction pattern at about 30 GPa and room temperature after heating at 30 GPa and 2000 K is completely different from the pattern above, indicating that the Ce metal has reacted with the nitrogen. The X-ray diffraction pattern just after the decompression was also measured at 0.1MPa and room temperature in an atmosphere of the nitrogen gas. The Ce nitride(s) pattern remains almost the same as that at 30 GPa although the diffraction intensity was changed because of the sample movement during the decompression. After measurement at 0.1MPa and 300K, the X-ray diffraction pattern was measured again in air. The pattern was a broad halo, indicating that the synthesized nitrides got glassy due to the decompression or oxidation. This is the same result of the new La nitrides reported before [5].

The well-known reported Ce nitride is of the simple NaCl-type structure. No X-ray diffractions of the NaCl-type structure cannot be found in the pattern of the recovered sample. There is another reported Ce nitride which crystallizes the La_2O_3-type structure. This structure is the same as that of one of the new La nitrides recently reported by us. Besides, the results of the TEM-EDS analysis indicated that the Ce metal was nitrided. These results indicate that one (or some) new Ce nitride(s) has (have) been synthesized in this study. Although we have tried to analyze the X-ray diffraction pattern, it cannot be identified so far.

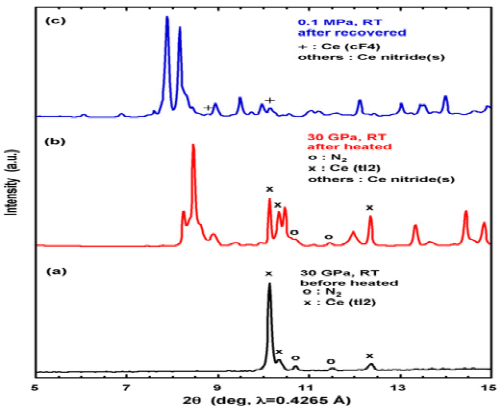

Fig. 1. XRD patterns at room temperature when synthesizing Ce nitride. (a) At about 30 GPa before laser-heating; (b) at about 30 GPa after laserheating at about 30 GPa, 2000 K; (c) after recovered to 0.1MPa.

References

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