Investigation of Deposited Materials on Carbon Electrodes in Molten Salts by *in-situ* X-ray Diffraction Measurements

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Introduction

The use of molten salt media for pyrochemical separation process in nuclear fuel cycle has emerged as an important field for engineering and scientific interest for recent years. For reprocessing of oxide nuclear fuels, the spent nuclear fuels are dissolved into molten NaCl·2CsCl eutectic salt, and then the mixed oxide fuels (MOX) are co-electrodeposited onto carbon electrodes [1)].

However, the detailed reaction mechanism is not clear on the electrochemical process. It is necessary to perform the *in-situ* X-ray diffraction (XRD) measurements of the electrodeposition process to elucidate the detailed reaction mechanism. However, the *in-situ* XRD measurements are difficult due to the high absorption of X-ray by the molten salt.

For the *in-situ* measurements, the XRD measurements were performed by reflection method to the electrodeposited material in the carbon container as an electrode to avoid the effect of the absorption. For the preliminary experiment, the XRD pattern of electrodeposited nickel film inside a carbon container was measured by reflection method using a multi-axis diffractometer at BL19B2 beamline of SPring-8. The time dependence of the XRD pattern was investigated and the variation of the nickel film with the molten salt was discussed.

Experimental

The nickel metal was electroplated in a carbon container from nickel electroplating bath with current density of 10mA/cm^2. The electroplating bath consists of nickel sulfate ($NiSO_4.6H_2O$) 150 g/*l* , ammonium chloride (NH_4Cl) 15 g/*l* , and boric acid (H_3BO_3) 15 g/*l*. The carbon container with nickel thickness of 100μm was filled with anhydrous NaCl·2CsCl salt. The container was set in high temperature furnace shown in Fig. 1, and heated up to 923K. The XRD patterns were measured by the reflection method using the multi-axis diffractometer at BL19B2 beamline of SPring-8 with 30keV photon energy (λ=0.413 Å) at 923K. The alignment of incident beam was adjusted to the inside of the carbon container. The XRD measurements were performed by 2θ scan from 5 to 30 degree of 2θ with incident angle θ of 5°.

Results and Discussion

Figure 2 shows the variation of XRD pattern of the carbon container with heating time at 923K. The XRD patterns were shown as a function of momentum transfer, Q ($Q = 4\pi\sin\theta/\lambda$). All X-ray patterns at 923K showed graphite peaks which are observed for the carbon container at 298K. Sharp peaks assigned to (111), (200), (220) and (311) of nickel metal were observed at 0min at 923K. It is noted that the peaks of nickel decreased with time. After 160min, the peaks of nickel had disappeared. It was found that the color of the molten salt changed to green after 160min at 923K. It shows that the deposited nickel inside the carbon container dissolved into the molten salt as nickel ion.

In this study, it is found that this method is very useful for the *in-situ* XRD measurement of the electrodeposition process in molten salts.

References

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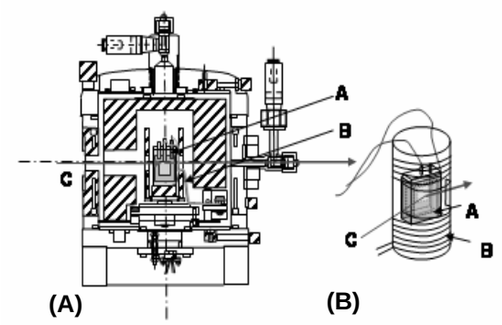

Figure 1 Scheme of high temperature furnace (A) and sample cell (B) for *in-situ* XRD measurement. A: carbon container (cathode), B: heater, C: X-ray beam.

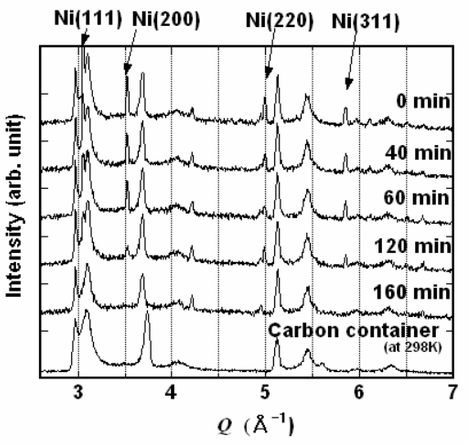

Figure 2 Time dependence of XRD pattern for deposited nickel in a carbon container at 923K