

Preliminary experimental study of high temperature interaction between uranium nitride pellet and eutectic lead-bismuth coolant

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1. Introduction

A small modular liquid-metal fast reactor (SMLFR) cooled by lead-bismuth eutectic (LBE) [1] is under development in the Department of Nuclear Engineering at Ulsan National Institute of Science and Technology (UNIST). For this LBE-cooled SMLFR design, uranium mononitride (UN) fuel is adopted owing to its high thermal conductivity, high melting point, and large fissile density; however, the interaction between UN fuel with LBE coolant has not been reported in the open literature. As a preliminary study of UN fuel safety under LBE coolant, perhaps during overpower transient with breached cladding, we report an interaction test between UN pellet and liquid LBE at 800 °C.

2. Experimental Methodology

2.1. Sample preparation

UN powder was synthesized from depleted uranium (DU) metal using hydride-nitride method. A piece of DU metal was heated to 235 °C for 4 h in an air-tight furnace refilled with pure hydrogen (99.999 %) of 50 mL/min flow rate. The hydride powder obtained from the process was ground using a zirconia mortar and pestle. The ground UH_3 powder is heated in the furnace up to 500 °C at a nitrogen flow of 200 mL/min, which yields uranium sesquinitride (U_2N_3) powder. The U_2N_3 powder was decomposed to UN at 1200 °C for 8 h under an argon atmosphere at a flow rate of 200 mL/min. The UN powder was spark plasma sintered (SPS) at 1800 °C for 10 min under 70 MPa uniaxial pressure; heating and cooling rates were both 100 K/min.

2.2. Description of interaction test

The interaction test between the LBE coolant and UN fuel pellet was performed in an alumina crucible (Fig. 1a). A piece of uranium nitride pellet (~0.5 g) was placed inside the crucible with LBE alloy pieces (~3 g). The crucible was heated in a furnace to 800 °C for 50 h with an argon flow rate of 200 ml/min; this much higher temperature (800 °C) than the anticipated fuel periphery temperature during normal operation (< 550 °C), was purposely selected to accelerate the reaction between UN pellet and LBE alloy. After the interaction test, the sample was polished to reveal the cross-section of the UN/LBE interface (Fig. 1d).

2.3. Sample characterization

The crystal structure of the sample cross-section was analyzed using X-ray diffraction (XRD, Rigaku, SmartLab SE) to confirm whether any secondary phase formed in the UN/LBE interface. The XRD characterization was performed under $\text{Cu K}\alpha$ radiation ($\lambda = 1.54056 \text{ \AA}$) at 40 kV and 25 mA. The measurement range (2θ) was from 20 to 75 ° at 0.04 ° scanning steps. The sample morphology was also observed using SEM/EDS (COXEM, EM-30ax plus).

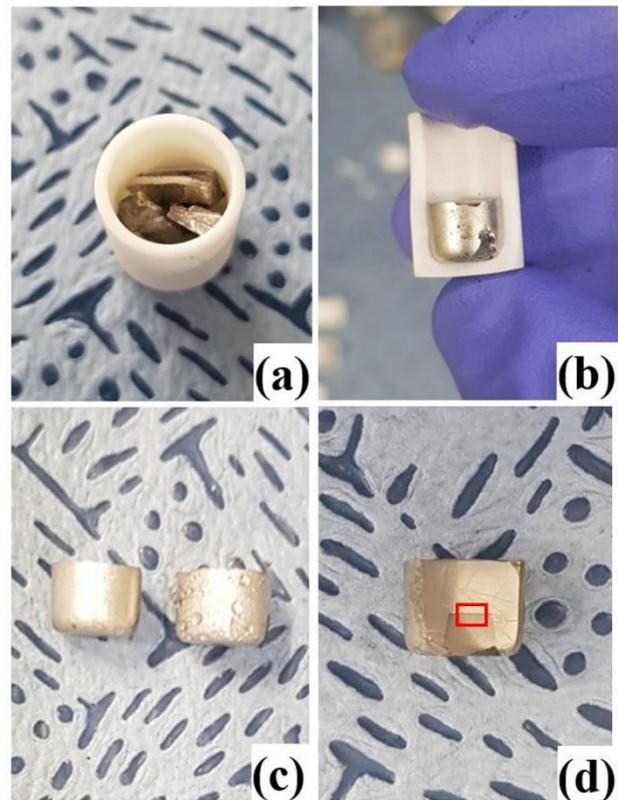


Fig. 1. Visual images of UN/LBE interaction test sample: (a) before; (b) after interaction test; (c) as-tested samples with alumina crucible removed; (d) cross-section of the sample with LBE (top, silvery) and UN (bottom, dark)

3. Results & Discussion

Visual images of the UN/LBE interaction test sample are displayed in Fig.1. The surface of LBE (Fig. 1c) after the test does not show any sign of oxidation. This implies

that oxygen partial pressure of the LBE during the test was maintained below its equilibrium value at 800 °C (P_{O_2} : $\sim 6.5 \times 10^{-5}$ atm) [2]. Also, the measured oxygen partial pressure (P_{O_2} : $\sim 1.0 \times 10^{-4}$ atm) of refilling argon gas was slightly higher than the equilibrium partial pressure of LBE, which may suggest potential presence of oxygen sinks inside the LBE matrix.

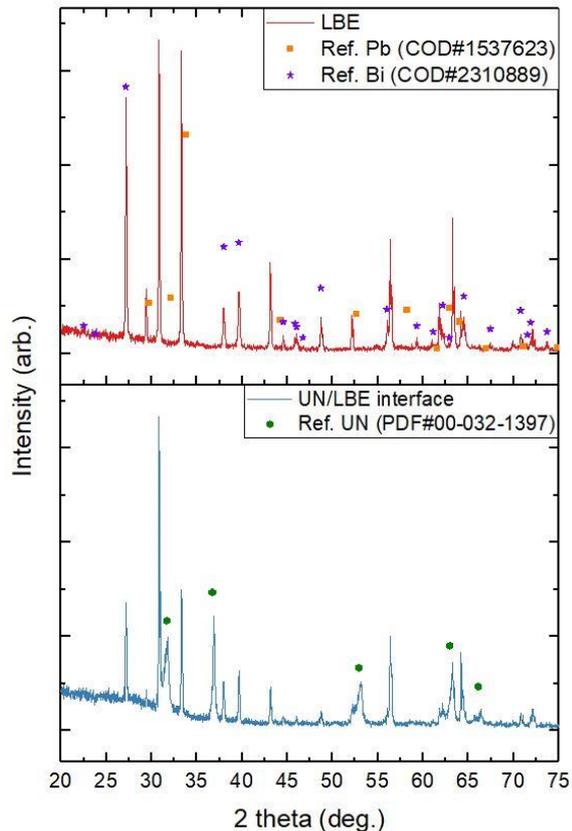


Fig. 2. XRD patterns of LBE (red) and UN/LBE interface (blue).

The XRD patterns of LBE and UN/LBE interface are shown in Fig. 2. According to the Pb-Bi phase diagram [3], at room temperature, solid LBE consists of two phases, intermetallic ϵ -phase and solid-solution of Pb in Bi. The XRD patterns of LBE (Fig. 2, red line) clearly show bismuth peaks (purple dots), although the peak intensities are slightly mismatched. The other peaks are assumed to represent ϵ -phase because at this moment the crystal structure information of the ϵ -phase is not available in the open literature.

The XRD patterns of UN/LBE (Fig. 2, blue line) show pure UN peaks in addition to the above-mentioned LBE peaks. The UN patterns are well-matched with the ICDD reference (PDF#00-032-1397). Other than UN and LBE, no secondary phase was observed.

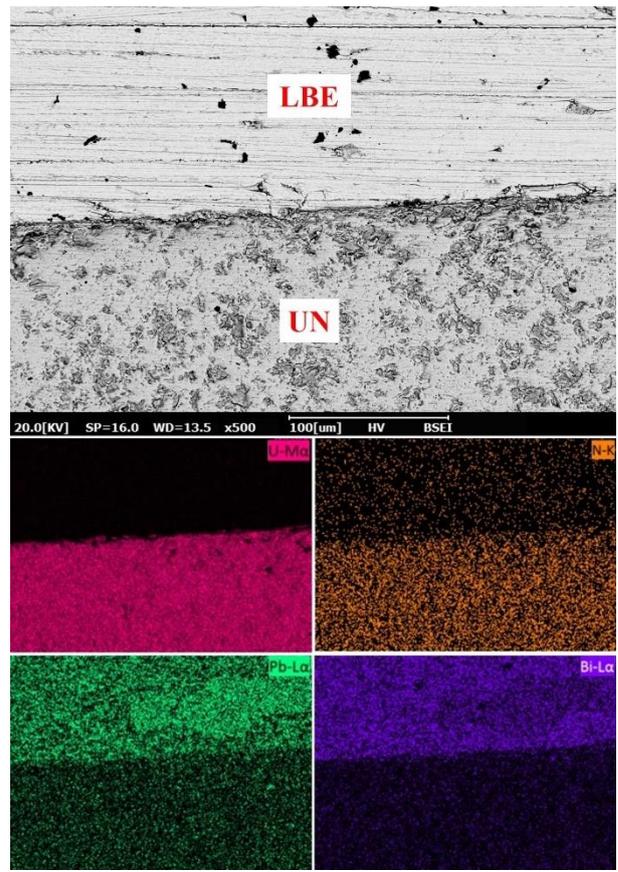


Fig. 3. SEM image of the selected area (red zone in Fig. 1d) with EDS mapping. LBE (top) and UN (bottom)

Fig. 3 shows the SEM image and EDS mapping obtained from of the selected interface (red zone in Fig. 1d) of UN and LBE. Dark strains in Fig. 3 indicates the rough surface of the UN pellet, which was practically unavoidable from the polishing of two disparate materials, ceramic UN and metallic LBE. On the interface between UN and LBE, no sign of interaction layer or compound containing uranium, lead, or bismuth was observed under the instrument resolution limits ($\sim 1.6 \mu\text{m}$).

4. Conclusion

The compatibility test between UN pellet and LBE coolant was conducted for the safety analysis of small modular liquid-metal fast reactor. The test condition of 800 °C was selected to accelerate the reaction between the UN and LBE. SEM/EDS image analysis shows no sign of interaction layer at the interface of UN/LBE. The XRD patterns of the UN-LBE specimen show no secondary phase either. Further study is planned to examine chemical reactions at higher temperatures up to 1400 °C.

ACKNOWLEDGEMENT

This work was supported by the National Research Foundation of Korea(NRF) grant funded by the Korea government(MSIT) (No. NRF-2017M2A8A2018595)

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