Preparation of Uranium Powder having Reactive Shape using Uranium Hydridation

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1. Introduction

The primary incentive for nuclear fuel with higher burnup and longer cycle is the economic benefits. The fuel cycle costs could be reduced by extending fuel discharge burnup and fuel cycle length. The longer fuel cycle can increase reactor operational margin and reduce the refueling time and the spent fuel management burden.

However, increment of fuel burnup and cycle length might result in the acceleration of material aging for nuclear fuel components. Then, the safety and integrity of nuclear fuel will be degraded. Therefore, an enhancement of the safety and economics of the LWR fuel through the fuel burnup and cycle extension must be simultaneously considered. Moreover, after the Fukushima daii-chi nuclear accident, the accident tolerance of the LWR fuel has become a primary matter of concern. So, it is indispensable to develop the innovative nuclear fuel material concepts and technologies which can overcome degradation of fuel safety and integrity.

Uranium nitride fuel has been proposed as a potential fuel material for advanced nuclear reactors because nitride fuel has the advantages of both metallic and oxide fuels. That is, the high melting point, high uranium density, and high thermal conductivity are the representative merits of nitride fuel [1, 2]. Nitride fuel is also considered as a fuel material [3] for the accident tolerant fuel of current LWRs to compensate for the decrease in fissile fuel material caused by adopting a thickened cladding such as SiC composites.

However, nitride fuel has a critical disadvantage of a serious reaction with water at a typical LWR condition. Bulk uranium nitride is known to be dissolved in water at a temperature above 230 °C. Uranium nitride powder is more unstable and reacts with water at about 150 °C. Therefore, the water-proof nitride fuel must be developed to apply to current LWRs. Several strategies to prevent or reduce the reaction of nitride fuel with water have been suggested [4].

KAERI is developing uranium nitride-oxide composite fuel pellet that is expected to have higher fuel performance and lower water reactivity. In the development of the fabrication technologies of uranium based composite fuel pellet, uranium nitride powder should be prepared, first.

We have considered a simple reaction method to fabricate uranium nitride powders directly from metal uranium powders. Also, to create reactive uranium powder with nitrogen, it is applied that the uranium powder is pretreated in the hydrogen atmosphere [5]. In this study, to investigate the behavior of the uranium powder hydriding process, thermal analysis tests were performed. And the morphology evolution of powder during the hydriding process was observed.

2. Experimental and Results

The spherical uranium powder that was fabricated using a centrifugal atomization method was used in the test. The thermo-gravimetric and thermal analysis tests of uranium powder hydridation were performed using TG/DTA (SETARAM, Setsys Evolution). The device allows measuring continuously weight change of a sample and heat generated while following a thermal cycle in a hydrogen atmosphere. Small quantities of powder of about some tens of milligrams are used. Crucible and components in device were made out of tungsten to minimize a reaction with hydrogen gas. The morphology and microstructures of the particles were observed using scanning electron microscopy (SEM).

Figure 1 shows differential thermal analysis (DTA) results about uranium hydridation as a function of temperature. The uranium powder was heated up to 600 °C in the hydrogen atmosphere. In the results, the uranium hydride (UH_3) began to form in the temperature range of 290 to 330 °C. And the peak of the decomposition [6] of uranium hydride was detected in the range of 440 to 480 °C. Based on the results, the optimized pretreating method to make more reactive uranium powder can be designed. That is to say, in the first pretreating step, the uranium powder reacts with

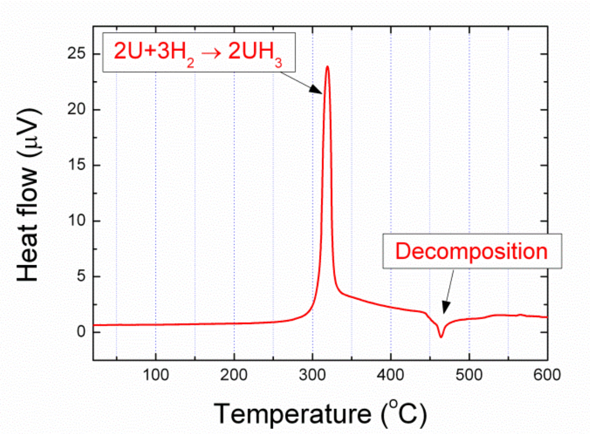

Fig. 1. DTA results about uranium powder hydridation

hydrogen gas in the temperature range of 350 to 420 °C. In the second step, the uranium hydride powder is decomposed by heat treatment above 480 °C. Then, it is expected that the uranium powder having more reactive shape with nitrogen gas in the uranium nitriding process will be obtained.

Figure 2 shows the morphology evolution in the uranium powder hydriding and decomposition process. Hydrogen diffuses through uranium metal powder and forms a network of brittle hydride over the grain boundaries. The uranium hydride powder will be shaped porous and fine due to a volume expansion. And the hydrogen will be reversibly removed by further heating.

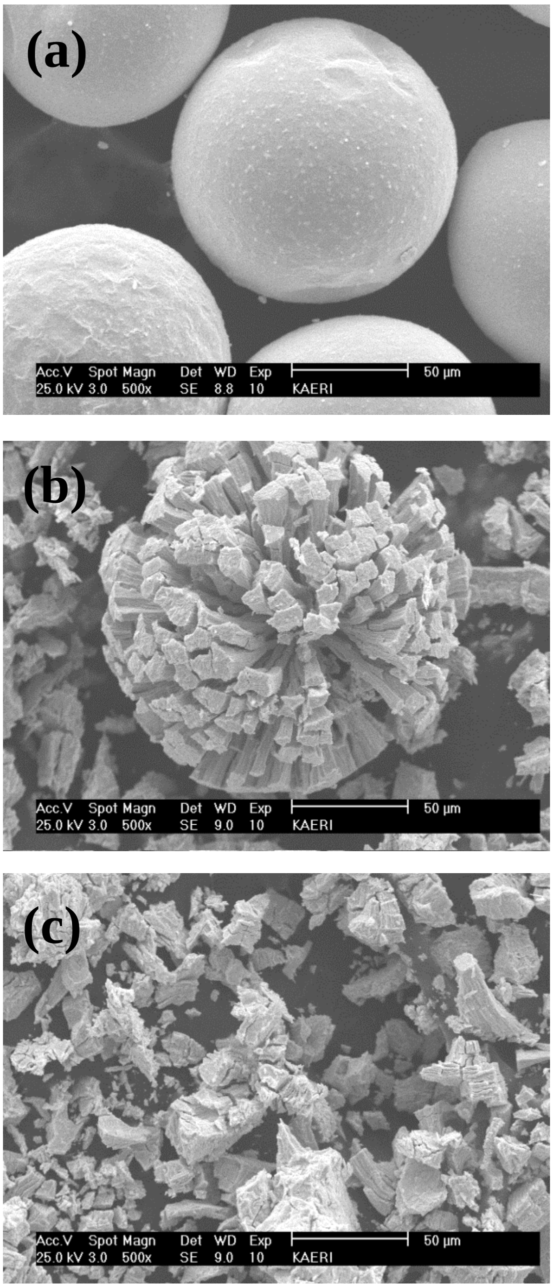

Fig. 2. SEM images of (a) initial, (b) intermediate, (c) final reactive uranium powder morphology

3. Summary

To create reactive uranium powder with nitrogen, the pretreatment of uranium powder in the hydrogen atmosphere was considered. To investigate the thermal behavior of the uranium hydriding process, thermal analysis (TG/DTA) tests were performed. Based on the results, the optimized pretreating method to make more reactive shape of uranium powder could be designed.

ACKNOWLEDGEMENT

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MEST) (No. 2013000720).

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