Synthesis of uranium nitride powders using metal uranium powders

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1. Introduction

Uranium nitride (UN) is a potential fuel material for advanced nuclear reactors [1-3] because of their high fuel density, high thermal conductivity, high melting temperature, and considerable breeding capability in LWRs. Uranium nitride powders can be fabricated by a carbothermic reduction [4] of the oxide powders, or the nitriding of metal uranium [5].

The carbothermic reduction has an advantage in the production of fine powders. However it has many drawbacks such as an inevitable engagement of impurities, process burden, and difficulties in reusing of expensive N^{15} gas. Manufacturing concerns issued in the carbothermic reduction process can be solved by changing the starting materials from oxide powder to metals. However, in nitriding process of metal, it is difficult to obtain fine nitride powders because metal uranium is usually fabricated in the form of bulk ingots.

In this study, a simple reaction method was tested to fabricate uranium nitride powders directly from uranium metal powders. We fabricated uranium metal spherical powder and flake using a centrifugal atomization method. The nitride powders were obtained by thermal treating those metal particles under nitrogen containing gas. We investigated the phase and morphology evolutions of powders during the nitriding process. A phase analysis of nitride powders was also a part of the present work.

2. Experimental

A centrifugal atomization process was applied to fabricate U powders and flakes. By adjusting the processing parameters, we obtained spherical, and flake shape depleted U metal powders. Nitriding of fabricated metal powders was conducted using an electrical heating furnace. Uranium metal particles in an alumina crucible were heated up to 1000℃ at a heating rate of 5℃/min in a N_2+50% H_2 atmosphere, and then annealed for 5 hours to fully form a nitride phase. The obtained nitride sample was annealed again at 1500℃ for 2 hours under a 10vol% H_2-Ar mixed gas atmosphere to obtain a single uranium mononitride phase.

The morphology and microstructure of the particles were observed using optical and scanning electron microscopy (SEM). The phase evolution of nitrides powders were investigated using an X-ray diffractometer.

3. Results

Fig. 1 shows the sphere- and flake- shaped metal U powders fabricated trough a centrifugal atomizing method adjusting the different process parameters. The thickness of a metal flake was about several tens of micrometers. The maximum diameter of a spherical uranium particle was about 45μm. The X-ray diffraction patterns for the both uranium particles revealed that both are single-phase γ-U.

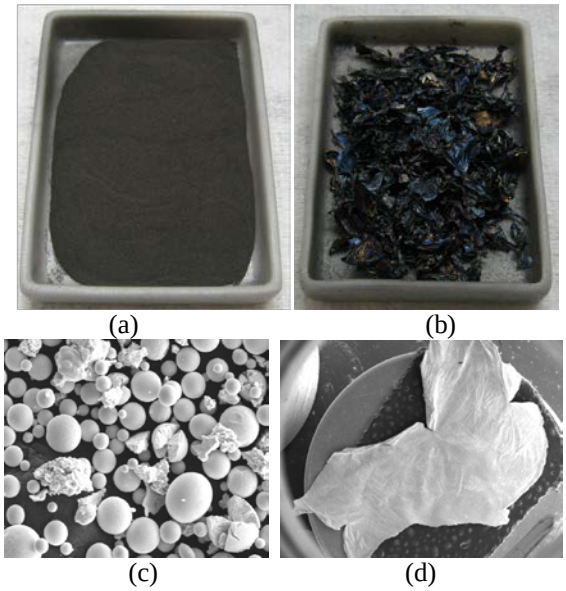
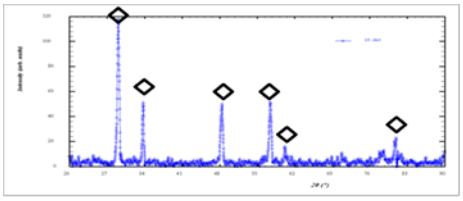


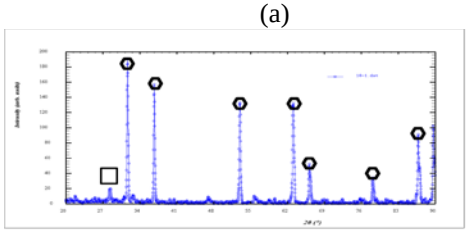
Fig. 1. Optical and SEM images of (a), (c) spherical powder and (b), (d) metal flakes

Fig. 2(a) shows an X-ray diffraction pattern for flakes after the annealing at 1000℃ in a N_2+50% H_2 atmosphere. The diffraction peaks in the pattern indicate the formation of a single phase of UN_2. Those particles of UN_2 phase have converted to UN particles by the further annealing it at 1500℃ under a 10vol% H_2-Ar mixed gas, as shown in Fig. 2(b). Traces of a UO_2 impurity phase were also observed in the pattern. The oxide phase may be caused by oxygen impurities in mixed gas or infiltrated air through an unexpected leakage during the annealing.



◇: Uranium di-nitride (UN_2)

(a)



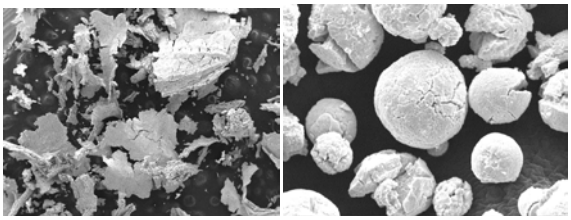
⬡: Uranium mono-nitride (UN)

□ : Oxide impurity (UO_2)

(b)

Fig. 2. X-ray diffraction patterns for the nitrides powders obtained by annealing the metal powders (a) at 1000℃ in a N_2+50% H_2 atmosphere and then further annealing (b) at 1500℃ in a 10vol% H_2-Ar mixed gas.

(a) (b)



(c) (d)

Fig. 3. Morphology changes of nitrides powders after the thermal annealing. Optical photos for (a) UN_2 flakes and (b) UN flakes. SEM images for (c) UN flakes and (d) UN spherical particles.

Fig. 3 shows the optical and SEM morphology evolutions during the sequential annealing to fabrication nitride powders. Optical photographs for flake particle revealed that flakes were increasingly fragmented into smaller particles during the nitriding process. A detailed examination on nitride particles using SEM also showed that numerous cracks were developed on the particles, as shown in Figs. 3(c) and (d). The theoretical density of UN_2 and UN are much smaller than that of γ-U and UN has higher density than UN_2. Therefore, observed fragmentation and cracking of particles are caused by sequential volume changes of expansion and contraction.

4. Summary

A simple reaction method was tested to fabricate nitride fuel powders directly from uranium metal particles. Direct nitriding the U metal particle obtained through centrifugal atomization is a simple and effective method to fabricate the uranium nitride powders.

ACKNOWLEDGEMENT

The authors acknowledge that this work has been performed under the Nuclear R&D Projects supported by the Ministry of Education, Science and Technology in Korea.

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