Enhanced Fabrication of UN Pellet from U_2N_3 Powder using Spark Plasma Sintering

Jungsu Ahn, Sangjoon Ahn*
Department of Nuclear Engineering, Ulsan National Institute of Science and Technology
50 UNIST-gil, Ulju, Ulsan, 44919, Republic of Korea
**Corresponding author: sjahn99@unist.ac.kr*

1. Introduction

Uranium mononitride (UN) nuclear fuel has been widely studied recently due to its superior fuel performance such as high thermal conductivity, fissile density, and melting temperature. However, the low fabricability of UN itself makes it difficult for the commercial application of nuclear fuel. For example, at least 2300 ℃ sintering temperature is required to fabricate the 95 %TD UN pellet using conventional sintering, which temperature potentially decompose UN to metallic uranium and nitrogen gas [1].

However, the spark plasma sintering (SPS) method, an advanced sintering technique using DC pulse current, could achieve higher pellet density at even lower sintering temperature and dwell time. Our prior study of UN fabrication using SPS showed ~97 %TD UN pellet was achieved at 1800 ℃ sintering temperature with 5-minute dwell time [2].

At this point, we explored even faster UN pellet fabrication with higher density by adopting a hyper-stoichiometric fabrication method. In this study, spark plasma sintering process was started directly with uranium sesquinitride (U_2N_3) powder to fabricate UN pellet, instead of UN powder.

2. Experimental

2.1. U_2N_3 and UN powder preparation

The feedstock depleted metal uranium was used to synthesis of the U_2N_3 and UN powder. All the synthesis works were conducted in the argon-purged glovebox (P_{O2} < 5 ppm & P_{H2O} < 5 ppm) due to the pyrophoricity of UH_3, U_2N_3, and UN powder. The U_2N_3 and UN were synthesized by continuous hydride, nitride, and denitride process, which includes following reactions:

$$\text{Hydride: } U + \frac{3}{2}H_2 \rightarrow UH_3$$

$$\text{Nitride: } 2UH_3 + \frac{3}{2}N_2 \rightarrow U_2N_3 + 3H_2$$

$$\text{Denitride: } U_2N_3 \rightarrow 2UN + \frac{1}{2}N_2$$

The depleted uranium ingots were soaked in 60% nitric acid for 30 seconds to remove the uranium oxide layer and loaded into Al_2O_3 crucible wrapped by tantalum foil to prevent further oxidation during heating. The metal uranium was heated at 235 ℃ for 4 hours in an hydrogen atmosphere (99.999%) to hydride. As-synthesized UH_3 was ground using mortar and pestle, and sieved less than 20 μm. The obtained UH_3 powder was converted to U_2N_3 at a 500 ℃ for 30 minutes under 99.999% nitrogen gas atmosphere. In order to reduce the U_2N_3 powder to UN powder, temperature was increased up to 1200 ℃ for 2-4 hours with 5 ℃/min of ramping rate under 99.99% argon atmosphere. The resultant U_2N_3 and UN powders were identified by X-ray diffractometry (XRD, Rigaku, SmartLab SE) with the range of 20 to 80° 2θ angle at a 1°/min scan rate. For the XRD measurement of U_2N_3 powder, polyimide tape was attached over the powder to prevent ignition on the air.

2.2. Spark Plasma Sintering

The spark plasma sintering was performed using DrSinter SPS-211LX. Approximately 4 g of powder was loaded into graphite mold wrapped with 0.2 mm-thickness graphite paper to avoid sample-mold reaction. The sintering temperature was in the range of 1500~1800 ℃ and the dwell time was 20 minutes. A 20 MPa uniaxial pressure was loaded to powder during the sintering process.

3. Results

Figure 1 shows the XRD patterns of as-synthesized U_2N_3 and UN powder through continuous hydride-nitride and denitride process, and both patterns were well matched with reference data of U_2N_3 and UN. The high background intensity of U_2N_3 powder is due to attached polyimide tape to prevent powder ignition during measurement.

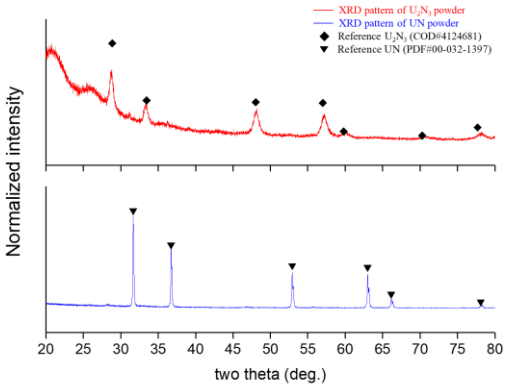

Fig. 1. X-ray diffraction pattern of as-synthesized U_2N_3 powder (top) and UN powder (bottom).

The temperature, vacuum pressure, and z-axis displacement was measured during spark plasma sintering (Fig. 2). The vacuum pressure and the z-axis displacement were rapidly increased above ~900 ℃ for the U_2N_3 powder sintering, while it was maintained for UN sintering. This concurrent increase might indicate the decomposition of low-density U_2N_3 powder (11.3 g/cm^3) to high density UN powder (14.32 g/cm^3).

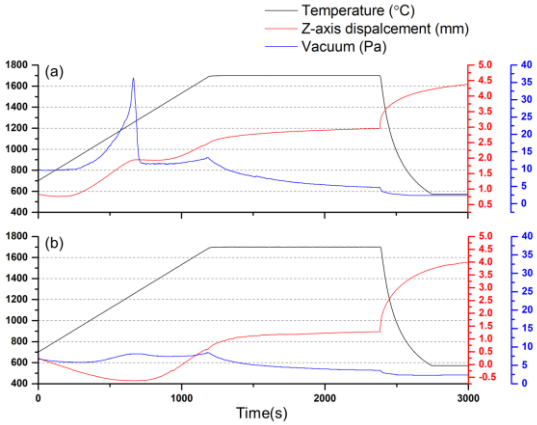

Fig. 2. Measured temperature, z-axis displacement, vacuum pressure during spark plasma sintering (a) U_2N_3 powder, and (b) UN powder at 1700 ℃.

The XRD analysis on the spark plasma sintered pellets shown in Fig. 3 affirmed all major peaks and relative intensities were well matched with the UN reference data regardless of the sintering temperature range of 1500 ℃ to 1800 ℃.

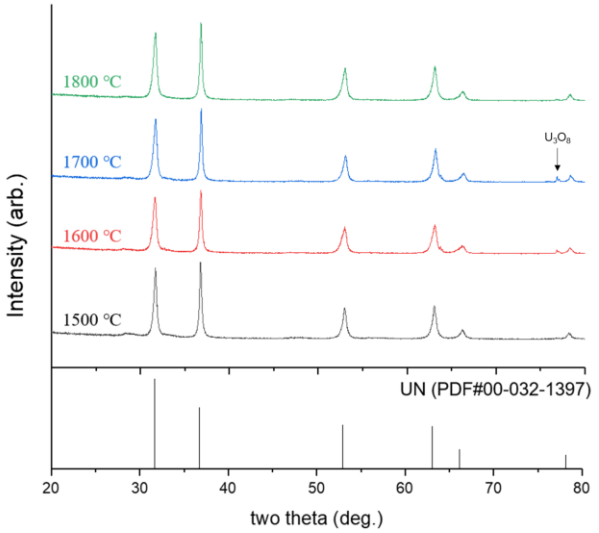

Fig. 3. X-ray diffraction pattern of spark plasma sintered pellets using U_2N_3 powder with the temperature range of 1500 ℃ to 1800 ℃.

Figure 4 shows the relative density of UN pellets spark plasma sintered using U_2N_3 powder and UN powder each. The relative density of both U_2N_3-/and UN-sintered pellet was increased with sintering temperature, and the U_2N_3-pellet showed relatively higher density (85.3 %TD) than that of UN-pellet (79.6 %TD) at low sintering temperature. The density gap between U_2N_3- and UN-pellet was closed with increasing sintering temperature.

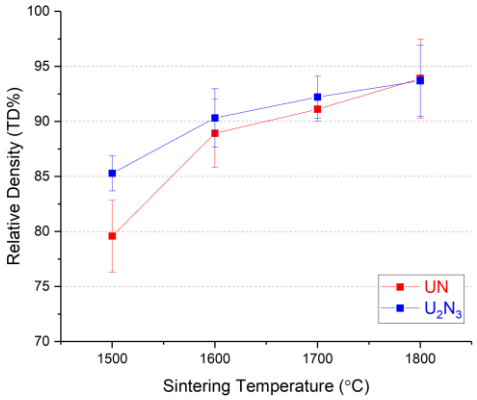

Fig. 4. Relative density of UN pellets spark plasma sintered using U_2N_3 powder (blue dots) and UN powder (red dots).

4. Conclusions

A hyper-stoichiometric UN fuel fabrication via spark plasma sintering method was explored. The UN pellets were spark plasma sintered using U_2N_3 and UN powder at various sintering temperature (1500 ℃ to 1800 ℃). During the SPS of U_2N_3 powder, the decomposition of low-density U_2N_3 powder to high-density UN powder was observed above ~900 ℃. The XRD analysis on U_2N_3-sintered pellets showed well matches with UN reference data for all tested sintering temperatures. The relative density of U_2N_3-sintered pellets was higher than that of UN-sintered pellets at low sintering temperature (1500 ℃), but the density gap was closed with increasing sintering temperature.

ACKNOWLEDGEMENT

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