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Developments in the fabrication technology of low density MOX pellets for fast breeder reactor fuel

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Abstract

The fabrication technology of low density MOX pellets was studied using some organic compounds as pore formers. The pore former, K3 showed the best performance among five organic compounds in the 7.5 kg-UOX runs, but K3 lost this in the 36 kg-MOX run, possibly because plutonium decay heat affected its performance. Comparison of thermal stability between K3 and a newly introduced pore former, Avicel, of high softening temperature, showed that K3 lost its spherical particle shape and consequently, its pore forming ability at 70 °C which was below its reported melting point (84–88 °C) while Avicel could maintain its spherical particle shape up to 150 °C. The pore forming performance of Avicel was also confirmed in the 36 kg-MOX run which is the same scale as for mass production of MOX fuel pellets.

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1. Introduction

A significant extension of mixed oxide (MOX) fuel pin life is critically important for fast breeder reactors (FBRs) to economically compete against light water reactors. When the MOX fuel is irradiated up to a high burn up beyond 100 GWd/t, the fission gas release fraction and swelling rate become large, affecting the integrity and consequently, the fuel pin life. The use of low density or annular pellets is a powerful solution to these problems. Both of these pellet types can increase the free volume and improve the storage capacity of fission gas and mitigation ability towards swelling in the fuel matrix. Annular pellets moderate the temperature in the central region, but their fabrication technique is complicated, because the diameter of FBR fuel pellets is so small. Thus, the fabrication technology of low density pellets was studied here.

A number of papers have described developments in fabrication of low density pellets [1–5]. One of the most important properties in low density pellets is their dimensional stability during the initial stage of irradiation. Low density pellets containing large diameter pores have to be fabricated to prevent irradiation-induced densification. Up to now, two methods have been tried to fabricate these pellets. The first one uses less active raw powder

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[1–3] and the second one uses mixed organic additives as pore formers with the MOX powder [4,5].

In this study, the fabrication process of low density pellets was investigated, based on the addition of pore former to raw MOX powder. Twenty organic compounds were surveyed, and from their decomposition temperature, ease of granulation and green pellets stability, six pore formers were selected as candidates for process development study. UO_2 (UOX) and MOX runs were carried out using six pore formers and the fabrication process of low density MOX fuel pellets was established.

2. Experimental

2.1. Pore former and pellet fabrication test

Table 1 shows the properties of the six candidate pore formers. Nos. 1–5 had melting points below 150 °C. Nos. 4 and 5 were the same material, but differently processed. No. 4 was processed as hollow spherical particles and No. 5 was processed as powder.

Their performance as pore formers was examined in two pellet fabrication tests, UOX runs on a 7.5 kg batch scale (7.5 kg-UOX runs) and MOX runs on a 36 kg batch scale (36 kg-MOX runs). The 36 kg-MOX runs correspond to conditions used for the practical mass production of MOX fuel pellets. Fig. 1 shows the process flow sheet applied to fabrication of low density pellets in this study.

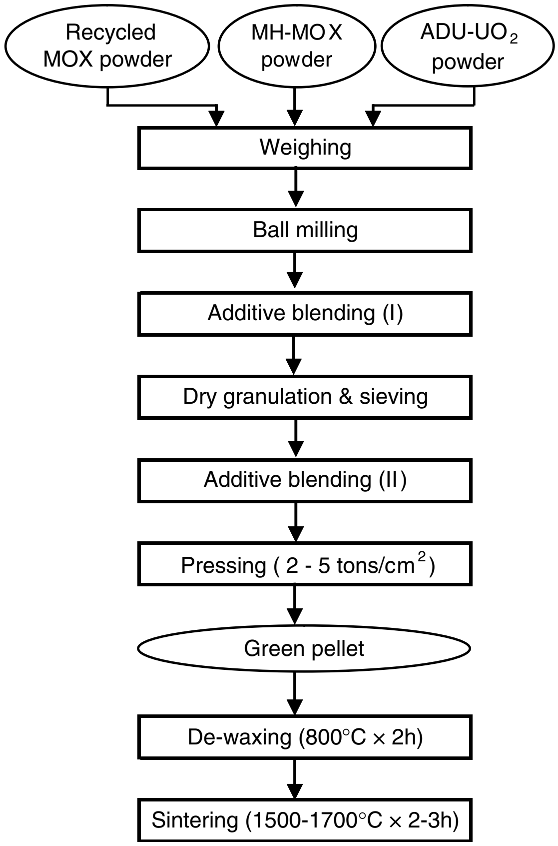

Fig. 1. Process flow used in UOX and MOX pellet fabrication runs.

2.2. Preparation of feed powders

In the 7.5 kg-UOX runs, natural UOX powder prepared by the ADU (ammonium diuranate) process was used as a feed powder after ball milling for 6 h. In the 36 kg-MOX runs, MOX powder, ADU UOX powder and recycled MOX powder were weighed and mixed in the ball mill for 6 h, and this mixed powder was utilized as a feed MOX powder. The properties of these feed powders are shown in Table 2.

2.3. 7.5 kg-UOX runs

The stability of dry granulation was checked by a fabrication test of low density pellets using natural

Table 1
Properties of candidate pore formers

No.	Poreformer		Molecular formula	Molecular mass	Melting point (°C)	Bulk density (g/cm^3)
	Product name	Chemical name				
1	Benzoic acid	Benzenecarboxylic acid	C_6H_5COOH	122.1	122–126	0.48
2	Stearic acid	Octadecanoic acid	$CH_3(CH_2)_{16}COOH$	284.5	69–72	0.48
3	Alflow-H-50S	1,2-Bis(octadecanamido)ethane	$C_{17}H_{35}CONHCH_2CH_2NHOCOC_{17}H_{35}$	593	135–146	0.61
4	Alflow-H-50T					0.37
5	K3	Glycerin trihydroxystearate	$CH_2COOR–CHCOOR–CH_2COOR$, $R = (CH_2)_{10}–CH(OH)–(CH_2)_5–CH_3$	938	84–88	0.93
6	Avicel	Crystal cellulose	$C_6H_{10}O_5$	162	350–400[a]	0.33

[a] Decomposition temperature.