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A practical fabrication method for the advanced heterogeneous fuel with magnesia containing minor actinides

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ABSTRACT

Fabrication tests on advanced heterogeneous fuel with MgO were carried out for the purpose of establishing a practical fabrication method. Advanced heterogeneous fuel consists of spheres (diameter greater than 100 μm) of a minor actinide oxide and MgO inert matrix (macro-dispersed type fuel). Macro-dispersed type fuel pellets with a high density above 90% T.D. were successfully fabricated. In addition, the fabricated pellets showed a homogeneous dispersion of near spherical host phase granules. These were attained by optimization of the fabrication process and conditions; i.e. a preliminary heat treatment of raw powders of host phase, an adjustment of pressure at the granulation process, deployment of a spray-dry process for MgO sphere preparation and sintering in a He atmosphere. From these results, a practical fabrication method for MgO-based macro-dispersed type fuel based on a simple powder metallurgical technique was established.

1. Introduction

Magnesia (MgO)-based heterogeneous fuel with a high content of minor actinides (MAs) is currently considered as one promising option for the rapid incineration of MAs in a future fast reactor cycle system [1–4]. An advanced form of heterogeneous fuel, a so-called macro-dispersed type fuel, is considered to be a high-performance form of heterogeneous fuel [1–6]. It consists of spheres (diameter greater than 100 μm) of an MA oxide host phase dispersed into an inert matrix. The main advantage of this type of fuel is the small degradation of thermal properties made possible through the suppression of irradiation damage to the MgO matrix by fission products [5].

Advanced fabrication technologies such as a sol–gel [6] and porous bead infiltration techniques [7] have generally been required for the macro-dispersed type fuel as it has a completely different form from the standard homogeneous fuel. However, the fabrication process should be adaptable to the currently used commercial fabrication technology for the rapid deployment of macro-dispersed type fuel at the introductory phase of the fast reactors. Furthermore, such a simple process is indispensable for the remote fabrication of fuels containing highly radioactive MAs. To satisfy these needs, a practical fabrication technology for MgO-based macro-dispersed type fuel based on a traditional powder metallurgy is now being developed [1–4].

The purpose of this study is to establish a practical fabrication method for MgO-based macro-dispersed fuel containing minor actinides by using the conventional powder metallurgical route only. As a representative of MA oxide, PuO_2 was chosen and fabrication tests on macro-dispersed type PuO_2 fuel in MgO (PuO_2–MgO) were carried out.

2. Experimental

The fundamental specifications for the MgO-based macro-dispersed type fuel are as shown below. $(Pu,Am)O_{2-x}$ spheres are homogeneously dispersed in the MgO inert matrix [1–4]. The content of $(Pu,Am)O_{2-x}$ is 50 wt%. The size of $(Pu,Am)O_{2-x}$ spheres is 120–350 μm.

Table 1 shows the characteristics of PuO_2 and MgO powders. MgO fine powders of approximately 100 nm in diameter were supplied by Ube material Industry, Co. Ltd.

Fig. 1 shows the fabrication test procedure for the MgO-based macro-dispersed type fuel. PuO_2 spheres with a diameter of up to 350 μm were prepared by a simple powder treatment as mentioned below. Fine PuO_2 powders with 3 wt% of polyvinyl alcohol binder were milled in ethanol with Yttria-Stabilized zirconia balls by oscillating mill for 1 h. The PuO_2 powders were dried under reduced pressure atmosphere of about 10^{-3} MPa for 24 h. The PuO_2 powders were then compacted into columnar pellets by uni-axial press at a pressure of 10–30 MPa. The PuO_2 compact was gently crushed in an agate mortar, and PuO_2 granules were obtained. To obtain the PuO_2 spheres, the granules were rounded by rotation

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Table 1
Characteristics of PuO_2 and MgO powder.

	PuO_2	MgO[a]
Metal impurities (ppm)	<1065	<113
Particle radius (μm)	<0.2	N/A[b]
Specific surface area (m^2/g)	7.27	12.5

[a] Ube material Industry Co. Ltd, 1000A.
[b] N/A: Not analyzed.

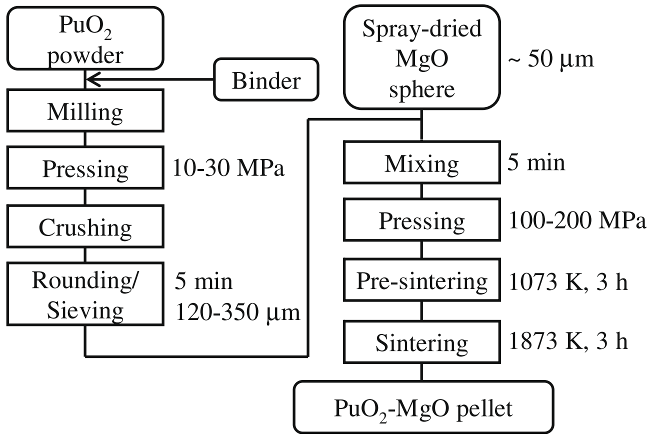

Fig. 1. Fabrication test procedure for macro-dispersed type fuel.

in a mill pot for 5 min. The spheres were then sieved and a specific size fraction, 120–350 μm, was selected.

MgO spheres were prepared by commercial spray-dry apparatus. This procedure is added so as to enhance the homogeneous mixing of PuO_2 and MgO spheres with a short mixing time. This aims to avoid the destruction of PuO_2 spheres. The size of MgO spheres was optimized in a previous study using CeO_2 as a surrogate of minor actinides oxides; MgO spheres with a 50 μm diameter were used [3].

Then each sphere was weighed to assure a composition of 50 wt% PuO_2 and 50 wt% MgO. After uni-axial compaction of the mixed granules of PuO_2 and MgO into a columnar pellet at 100–200 MPa, the pellets were heated at 1073 K in the same atmosphere as sintering for 3 h to remove the organic binder. Sintering was carried out at 1873 K in a He or 0.5%H_2–He atmosphere for 3 h.

The PuO_2–MgO pellets were characterized by density and microstructure; i.e. the shape and dispersibility of the PuO_2 granules were examined.

The sintered density was obtained from metrological results. The theoretical densities of the PuO_2–MgO pellet were calculated by the following equation.

$$\rho_{TH} = \left(\frac{c_M^W}{\rho_M} + \frac{c_H^W}{\rho_H}\right)^{-1}, \tag{1}$$

where ρ_{TH} is the theoretical density of the composite (PuO_2–MgO), ρ_M and ρ_H are the theoretical densities of the inert matrix (MgO; 3.59 g/cm^3) and host phase (PuO_2; 11.67 g/cm^3), respectively. c_M^W and c_H^W are the weight fractions of the matrix (MgO) and host phase (PuO_2), respectively. In this study, the composition of the PuO_2–MgO pellet was determined by the weighing condition of each raw powder. The theoretical density of PuO_2–MgO pellets with a composition of 50 wt% PuO_2 and 50 wt% MgO is 5.49 g/cm^3.

The dispersibility of PuO_2 spheres in MgO was evaluated by 'Homogeneity Distribution' (HD) factor obtained using image processing software for a horizontal cross section of a columnar pellet [6,8].

3. Results and discussion

Table 2 shows the sintered densities of the PuO_2–MgO pellets. A higher density was obtained for pellets compacted at 200 MPa than at 100 MPa. In both cases, sintering in a He atmosphere produced a high density, while sintering in a 0.5%H_2–He atmosphere produced a lower density. Similar sintering behavior was observed for the MgO-based micro-dispersed type fuel [9]. This behavior is attributable to the different sintering behavior of PuO_{2-x} host phase according to the sintering atmosphere. Previous work shows that the rate of PuO_2 densification was greatly enhanced in a reductive atmosphere [10]. Therefore, a mismatch of the densification behavior between PuO_2 and MgO should have occurred in a reductive 0.5%H_2–He atmosphere. This mismatch could cause transient stress in the MgO matrix, which could lead to the formation of large pores in the MgO matrix [11–13]. Thus, the lower density of PuO_2–MgO pellets sintered in the 0.5%H_2–He atmosphere might be caused by the poor sinterablity of the MgO matrix. From these results, it is believed that the higher compact pressure and the higher oxygen potential in the sintering atmosphere are effective for the densification of MgO-based macro-dispersed type fuel.

Fig. 2 reproduces ceramographic images of the microstructure of the PuO_2–MgO pellets sintered in the He atmosphere. The bright gray-colored and the dark gray-colored images show the PuO_2 phase and the MgO phase, respectively. While there were relatively large pores in the matrix, there were no visible cracks. The PuO_2 granules found to be fracture-free up to 200 MPa pressing.

The PuO_2 granules in Fig. 2(a) were prepared by pressing at 30 MPa and crushing. They exhibited a multangular shape, and this was attributed to the strongly-agglutinated fine raw powders (<0.2 μm diameter) resulting from the relatively higher compaction pressure. In addition, gaps between PuO_2 and MgO, which cause poor thermal conductivity in the macro-dispersed type fuel, were formed, as shown in Fig. 2(a). It is believed that the PuO_2 powder was so fine and strongly-agglutinated that the densification of PuO_2 powder was enhanced to a greater degree than MgO powder was. This was confirmed by the pore-less structure of PuO_2 granules as shown in Fig. 2(a). This mismatch of densification behavior led to the break up of interface between the host phase and the matrix, and circumferential cracks formed [11–13].

In order to obtain well-shaped spheres and integrity of the MgO–PuO_2 interface, two approaches were tested: one was lowered compact pressure, intended for the suppression of the agglomeration of PuO_2 powder; the other was use of heat treated raw PuO_2 powders. This process is able to consolidate the raw PuO_2 powders prior to sintering, which could lead to the resistance of excessive PuO_2 sinterability. Fig. 2(b) and (c) show the ceramographic images of PuO_2 granules prepared by pressing at 10 MPa without heat treatment and pressing at 30 MPa with heat treatment of PuO_2 powder at 1073 K for 3 h in a He atmosphere, respectively. The microstructures of both granules were nearly the same. The PuO_2 granules had a nearly spherical shape, and the formation of gaps between PuO_2 and MgO were suppressed, compared to the

Table 2
Densities of the PuO_2–MgO pellets.

Compact pressure (MPa)	Sintering atmosphere	Density (% T.D.)
100	He	86.8
	0.5%H_2–He	81.5
200	He	90.8
	0.5%H_2–He	87.2

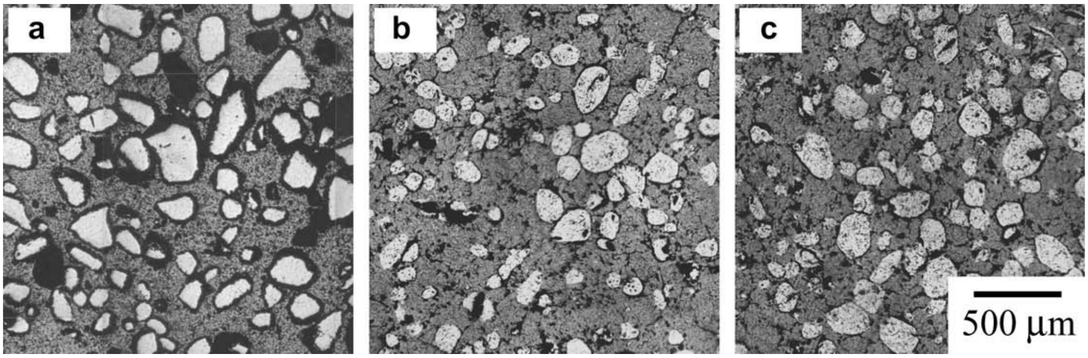

Fig. 2. Ceramographic images of the microstructure of PuO_2–MgO: (a) 30 MPa compaction, (b) 10 MPa compaction and (c) heat treatment at 1073 K/30 MPa compaction.

case of pressing at 30 MPa only. These results show that the PuO_2 spheres and the integrity of the PuO_2–MgO interface could be attained by optimization of the granulation process and conditions according to the characteristics of raw powders in view of suppressing the agglomeration of PuO_2 powder at compaction and matching of the densification behavior between the host phase and the matrix.

Table 3
HD values of the macro-dispersed type fuel pellets.

Composition	Particle size (μm)	HD
PuO_2–MgO	120–350	11.6 ± 1.7 (this study)
$AmO_{1.6}$–MgO (ECRIX)	<50	10–15 [6]
(Am, Y, Zr)O_{2-x}–MgO (CAMIX-2)	30–50	12 [6]
(Am, Y, Zr)O_{2-x}–MgO (COCHIX-3)	90–130	7 [6]

Table 3 compares HD values of the PuO_2–MgO pellets sintered in a He atmosphere with those of the similar MgO-based fuel, i.e. ECRIX (micro-dispersed type fuel) and CAMIX-COCHIX (macro-dispersed type fuel) [6]. The HD value for the present fuel was comparable with the values for both ECRIX and CAMIX-COCHIX, which were fabricated by the advanced fabrication technique, i.e. the sol–gel supported method. The homogeneous dispersibility of host phase was attained after only a short mixing time of 5 min by adopting highly flowable MgO spheres.

4. Concluding summary

Fabrication tests on the MgO-based macro-dispersed type fuel were carried out for the purpose of establishing a practical fabrication method.

A high density above 90% T.D. without any defects was given by compaction of 200 MPa and sintering in a He atmosphere. Near spherical host phase granules, together with the integrity of the interface between the host phase and the MgO matrix, were attained by optimization of the powder treatment condition at the granulation process, e.g. a preliminary heat treatment of raw powders of host phase and optimization of pressure at the granulation process. A homogeneous dispersion of host phase granules into the MgO matrix was attained by adopting highly flowable MgO spheres. Only a short mixing time, 5 min, was deployed for the homogeneous dispersion.

From these result, the MgO-based macro-dispersed type fuel with good characteristics, i.e. having no defects, a high density and a homogeneous dispersion of host phase, was successfully fabricated by a simple powder metallurgical technique with the optimization of the fabrication process and conditions.

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