

Densification of magnesia-based inert matrix fuels using asbestos waste-derived materials as a sintering additive

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ABSTRACT

A new concept for densification of minor actinide-containing inert matrix fuels (IMFs) using asbestos waste-derived materials was proposed for the effective utilization of resources and health protection of the general public. In this concept, magnesium silicates, which are mainly generated by the decomposition of asbestos in low temperature heat-treatments, are used as a sintering additive to achieve high density magnesia (MgO)-based IMFs at relatively low sintering temperature. In the present study, preliminary fabrication tests of MgO-based IMFs with magnesium silicates were carried out using cerium oxide (CeO₂) as a representative of minor actinide oxides. The sintered densities of MgO-based IMFs increased with use of the additives. The sintering behavior of MgO-based IMFs with magnesium silicate additives was discussed from the viewpoints of the effects of magnesium silicates on the densification of the MgO and CeO₂.

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1. Introduction

Transmutation of minor actinides (MAs) in a fast reactor (FR) is currently considered one of the promising options for future nuclear cycle technology, which is essential to reduce the environmental burden caused by their disposal and to provide a sustainable energy supply for the future. MA-containing inert matrix fuels (IMFs) are now being developed as a high-performance device for controlling the inventory of MAs (Osaka et al., 2007a,b; Croixmarie et al., 2003; Maschek et al., 2008; Jorison et al., 2007). IMFs are a composite of a high content of MAs host phase and an inert matrix (IM). They can make possible the rapid transmutation of MAs. Such composite-type fuels have a relatively long development history as high-performance devices for the transmutation of MAs (Osaka et al., 2007a,b; Croixmarie et al., 2003; Maschek et al., 2008; Jorison et al., 2007). Although a number of new technologies for such IMFs have been proposed, e.g. fabrication technology based on sol-gel (Croixmarie et al., 2003) and porous-bead infiltration (Richter et al., 1997) techniques which differ from the standard technologies for UO₂ and MOX fuels, the related technologies for IMFs should be adaptable to the currently-used commercial technologies for the rapid deployment of IMFs at the introductory phase of the FRs.

The development of a fabrication technique for IMFs containing MAs based on the traditional powder sintering route is now underway (Osaka et al., 2007a,b; Miwa et al., 2007, 2009b,c, 2010; Osaka et al., 2006; Miwa and Osaka, 2009). Table 1 shows the fundamental specifications of the IMFs investigated (Osaka et al., 2007a; Miwa et al., 2007, 2009b). The IMs, MgO, Mo and Si₃N₄, were selected based on an evaluation of their high manufacturability, high melting temperature, relatively high thermal conductivity, and chemical and physical stabilities (Miwa et al., 2007, 2009b). The MgO-, Mo- and Si₃N₄-based IMFs with good characteristics, i.e. having no defects, a high density and a homogeneous dispersion of host phase, were successfully fabricated by a simple powder sintering technique (Miwa et al., 2007, 2009b; Osaka et al., 2006; Miwa and Osaka, 2009; Miwa et al., 2009c; Osaka and Tanaka, 2008; Yamane et al., 2008; Yano et al., 2010). In the fabrication of MgO-based IMFs, however, the relative high sintering temperatures of 1873 K were required to achieve the high densities above 90% of theoretical (%TD) (Miwa et al., 2007, 2009a,b; Osaka et al., 2006; Miwa and Osaka, 2009). For the practical use of IMFs, since Am has high volatility (Haire, 1994), a lower sintering temperature is required to prevent its loss during sintering. In addition, lowering the temperature can lead to reduce fabrication cost of IMFs.

To satisfy these needs, a new concept was proposed for densification of IMFs by using asbestos waste-derived materials as a sintering additive (Miwa et al., 2009a). Since asbestos materials have resistance to heat, electrical and chemical damage, they were widely used as insulators in buildings. However, the inhalation of asbestos

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Table 1
Fundamental specifications of MgO-, Si₃N₄- and Mo-based inert matrix fuels (Osaka et al., 2007a; Miwa et al., 2007, 2009b).

		MgO-based	Si ₃ N ₄ -based	Mo-based
Fuel shape		Columnar pellet		
Fuel Density		>90% of theoretical		
Inert matrix	Chemical form	MgO	Si ₃ N ₄	Mo
Host phase	Chemical form	(Pu,Am)O _{2-x}	(Pu,Am)O _{2-x}	(Am,U,Th)O _{2-x}
	Weight fraction	0.30–0.50	0.25–0.40	0.50–0.90
	Form	Sphere, >φ100 μm Particle, <φ10 μm	Grain boundary phase	Particle, <φ10 μm

fibers may cause mesothelioma and lung cancer (Hodgson and Darnton, 2000), and disposal of asbestos materials when old buildings are torn down or remodeled has become a serious public health issue. Therefore, this concept can contribute to the protection of public health by disposing of the toxic asbestos waste through its use in the IMFs in addition to the sophistication of the fabrication procedure for MgO-based IMFs. In the concept, magnesium silicates such as enstatite (MgSiO₃) or forsterite (Mg₂SiO₄), which are found to be formed by the decomposition of asbestos in a relatively low temperature heat-treatment (Gualtieri and Tartaglia, 2007), are used as a sintering additive to achieve high-performance IMFs having a high density at relatively low temperature sintering.

In this study, the effects of magnesium silicate additives on densification of MgO-based IMFs were experimentally investigated for the purpose of establishing a sophisticated fabrication procedure for MA-containing MgO-based IMFs based on powder sintering techniques. Cerium oxide (CeO₂) was chosen to represent oxides of MAs for the host phase, and preliminary fabrication tests of MgO-based IMFs with magnesium silicate additives were carried out.

2. Experimental

Fig. 1 shows the fabrication procedure of the micro-dispersed type MgO-based IMF pellets (size of host phase: <φ10 μm). It was based on a powder sintering method, and almost the same procedure as the previous studies (Miwa et al., 2009c) except for the additives addition. Table 2 shows the fundamental specifications of raw powders. Enstatite or forsterite were milled by planetary ball-milling for 20 min at 600 rpm. MgO (Ube Material Co., Ltd.) and CeO₂ (Sigma-Aldrich Corp.) powders and pre-determined amounts of the magnesium silicate additives were mixed in an agate mortar with a pestle in acetone medium for 15 min. The starting amounts of CeO₂ and the enstatite or forsterite were 40 wt.% and 1wt.% of the

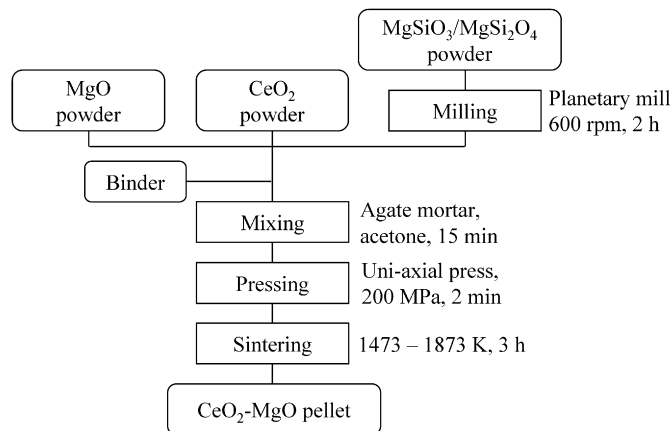


Fig. 1. The fabrication procedure of the micro-dispersed type MgO-based IMF pellets with magnesium silicate additives.

Table 2
Fundamental specifications of raw powders.

	MgO	CeO ₂	Enstatite (MgSiO ₃)	Forsterite (Mg ₂ SiO ₄)
Purity [%]	99.99	99.95	–	–
Specific surface area [m ² /g]	12.6	–	–	–
Mean particle size [μm]	0.1	5	2.5	2.5
Impurity [%]	–	–	Al ₂ O ₃ : 0.07, CaO: 0.06	Al ₂ O ₃ : 2.6, CaO: 0.28

final mixed powder, respectively. Mixed powder was pressed at 200 MPa for 2 min into a compact. Sintering tests were carried out at 1473–1873 K for 3 h in air atmosphere.

The densification behaviors were characterized by the density and microstructure. The sintered density was obtained from metrological results. The theoretical densities of each material were calculated on the assumption that the sintering additives did not react with matrix material by the following equation:

$$\rho_{TH} = (C_M/\rho_M + C_H/\rho_H + C_A/\rho_A)^{-1} \quad (1)$$

where ρ_{TH} is the theoretical density of the composite, and ρ_M , ρ_H and ρ_A are the theoretical densities of the IM (MgO: 3.59 g/cm³), host phase (CeO₂: 7.25 g/cm³) and additives (enstatite/forsterite:

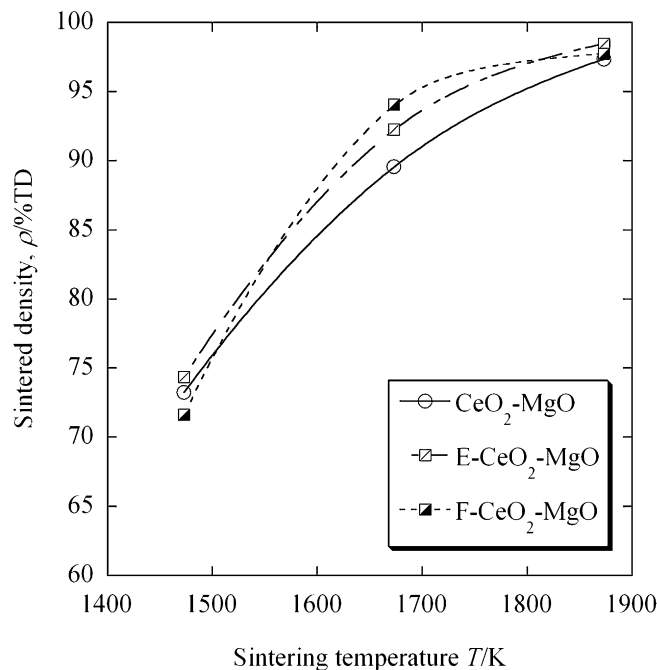


Fig. 2. The sintered densities of 1wt.% E-CeO₂-MgO and 1wt.% F-CeO₂-MgO as a function of sintering temperature.

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