An Evaluation of the Glass-Ceramic Solidification Characteristics of Uranium Catalyst Waste by Green Body Pressure

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1. Introduction

We have developed a process to treat a large volume of problematic radioactive waste, namely, a spent uranium catalyst which was used for production of acrylonitrile by a private company in South Korea.[1] The Korea Atomic Energy Research Institute (KAERI) is currently developing a volume reduction method for the uranium catalyst presently stored [2], for which the final solids generated in the process should be made into stable solids and satisfy the disposal conditions of solid radioactive waste in Korea.

Common immobilization methods for radioactive waste include, cement, glass, asphalt and polymers. Although using cement has advantages, such as inexpensive material cost and accumulated technology, it has a disadvantage that its chemical durability is relatively weak and its volume is larger compared with other solidifying media for the same initial waste volume. In the case immobilization of uranium waste by cement, CaO which is the main component of the cement solidification medium changes to $Ca(OH)_2$ when it reacts with water, and under this condition, uranium oxide is converted to uranium hydroxide ion ($(UO_2(OH)_X{}^{Y-})$), thus potentially becoming mobile and leaching out.[3] Therefore, it is necessary to develop technology for manufacturing solidified bodies with higher volume reduction factors and better physiochemical properties.

In this study, we investigated a method of sintering and immobilizing uranium-containing solids (Si-U-Sb-Fe-Al) Ox which are generated from treatment of the uranium catalyst waste. A glass solvent such as SiO_2 and Sb_2O_3 was used to form a U_3O_8 incorporated glass-ceramic structure possessing high mechanical strength and chemical stability. The solidification method and final sintered solid waste forms were characterized. It was found that the volume of the waste is reduced during the sintering process and is made into a stable solid.

In this paper, we studied the change in physicochemical properties of sintered bodies due to changes in molding pressure during the manufacturing of Green Bodies (GB) for the production of uranium-containing glass-ceramic sintered bodies.

2. Experimental

2.1 Experimental method

Components of uranium catalyst, analysis by EDS(Energy Dispersive X-ray Spectroscopy), shows that they are glass former or flux components for soda-lime or borosilicate glasses.(Table 1)

Table 1. EDS result of Uranium waste

Element	Si	Sb	U	Fe	Al	O
Atomic wt (%)	8.77	16.9	4.40	12.2	0.37	45.0

A mass of uranium waste catalyst (3 g, 1.06 g / ml) and 0.6 ml of distilled water, as a binder, were added to a mold. The molding pressure was set at 15, 30, 60, 120, and 240 MPa pressure using a pellet press (diameter 13 mm).

The resultant GB was heated to 1100°C (heating rate 10K / min) by using a furnace, and then subjected to isothermal heat treatment for 4 hours (Fig. 1).

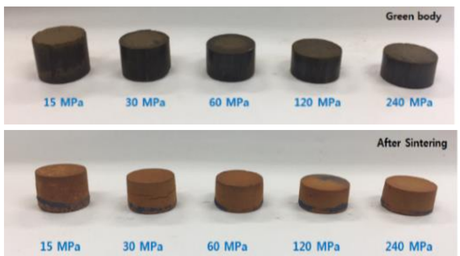

Fig. 1. Pictures of Green bodies and Sintered solids as a function of production pressure.

Several properties of density, porosity and compressive strength testing were measured.

2.2 Evaluation of sintered body characteristics

The volume and density of the sintered bodies were measured by using a hydrostatic weighing method using Archimedes principle and a gas volume meter based on Boyle's law. Bulk density and true density were obtained by two methods and porosity was measured by the difference of the values.

Final volume reduction of uranium catalyst waste by the glass-ceramic was evaluated by the difference between volumes of sintered body and initial uranium catalyst solid.

The compressive strength obtained at minimum molding pressure of 15MPa used in this work was over 3.45MPa which is the requirement for disposal of immobilized waste in Korea.(ASTM C39 Compressive Strength Standard Test)

3. Results and discussion

The volume reduction after sintering was 71.99 ~ 74.21% depending on the molding pressure used, as shown in Table 2. The difference in volume reduction due to molding pressure change was negligible when considering the high molding pressures used.

Table 2. Compression Test by Molding & Volume Reduction from original Uranium waste

Molding pressure (MPa)	After Sintering	
	Compressive strength (MPa)	Volume Reduction (%)
15	5.01	71.99
30	11.40	72.67
60	19.07	73.24
120	34.00	74.21
240	31.61	74.21

The minimum pressure of 15MPa was found that for forming a GB satisfies the compressive strength criteria of 3.45MPa, with the actual strength being 1.4 times more than indicated in the ASTM C39 standard. Table 3 shown in the porosity decreased with molding pressure, but the difference was not significant when the large difference in molding pressures used is taken into consideration. Therefore, there was little difference in durability of glass-ceramic sintered body according to molding pressure.

Table 3. After Sintering Density and Porosity Result by molding pressure

Molding pressure (MPa)	Density (g/cm^3)		Porosity (%)
	Archimedes	Pyconometer	
15	3.78	4.61	17.84
30	3.88	4.63	16.26
60	3.96	4.59	13.71
120	4.11	4.66	11.83
240	4.11	4.61	10.88

Although using the minimum pressure produces satisfactory compressive strength results, a higher molding pressure is felt necessary owing to safety concerns during handling, transport and disposal of the waste form. However, because the cost of molding increases as the pressure of the compact increases, a molding pressure of 30 MPa was evaluated to sufficient. This pressure alone is more than four times the compressive strength criterion.

The chemical stability test of sintered body prepared at 30MPa of molding pressure was carried out, which is not present in the paper, the leaching standard for uranium test (ASTM C1285-02: PCT). It was observed to satisfy

In this works the durability and leaching characteristics of the glass-ceramic sintered body will be farther studied.

REFERENCES

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