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Characteristics of Coating Slurry Solution for Inner Surface Coating Layer of Quartz Mold Using Metal Fuel Rod Fabrication Process

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1. Introduction

In the case of metallic fuel for next generation, it consists of 3 or 4 elements-alloy containing RE(Rare-Earth)s in existing U-Zr alloy fuel [1]. This requires a more precise control in the fuel fabricating process, since it shows differences in fabrication characteristics and process, which are quite different from binary alloys of conventional U-Zr alloys [2, 3]. Therefore, it is important to develop the characteristics of the alloy and the customized manufacturing process according to the addition of each constituent element.

Generally, metal-fuel production is performed by selecting metal components having proper composition ratios according to the characteristics of the reactor core, melting them in a high-temperature melting furnace, containing and cooling them into a proper mold material and fabricating them into fuel rods [4]. This study relates to a mold coating solution used for manufacturing process of a metal fuel rod based on a conventional U-Zr alloy fuel fabrication. The Y_2O_3 (or ZrO_2) slurry solution, coating material of the inner surface of the quartz tube which is a mold material, was selected and used as coating slurry solution.

In this study, the properties such as viscosity, particle size, and dispersion stability of coating slurry solution that was used to a coating at the inner surface of the quartz tube were measured. Also, to obtain the basic data to improve the characteristics of the inner surface of quarts tube, surface modification experiments used a variety of the corrosion solutions were carried out.

2. Technical Data

To understand the basic properties of the slurry solutions used in this study, we obtained technical data through literature survey and internet information. Yttria slurry solution was composed of Y_2O_3 particles mixed with magnesium silicate and a small amount of organic compounds. And the viscosity of the raw solution was appeared about 300 to 700 cP, acetone or ethyl alcohol is used to as dilution medium [5].

When the slurry solution is coated on the inner surface of the quartz tube by brushing method, it becomes white when dried, and does not dissolve in water after dried. The coating layer was known to be usable up to 1900℃ under a vacuum atmosphere. The related data are briefly shown in the table 1.

Table 1. Physical Properties of Coating Slurry Solutions

	Y2O3 slurry (Y aerosol brushable)	ZrO2 slurry (Z aerosol brushable)
Active ingredient	Y2O3	ZrO2
Binder phase	Magnesium silicate + small amount of organic	←
Viscosity	Brookfield , 300~700 cP (P;cm/g.sec)	Brookfield , 425 cP
Specific gravity	1.3	1.5
Using method	Brushing, traditional air-spraying, dipping	←
Temperature	Air/inert/Vac. without Carbon , room~1900℃	←
Dilute solution	Faster drying : Acetone (CH3COCH3)	←
	Slower drying : Ethyl alcohol (C2H5OH)	←
Caution	Avoid breathing of vapors	←
Product	White, will grey out on first heat up to 700℃ by small organic binder	Cream, will grey out on first heat up to 700℃ by small organic binder
Other	Nearly water insoluble after drying	←

3. Experiments and Results

3.1 Viscosity measurements

Generally, yttria(or zirconia) slurry solution is used as a coating material on the inner surface of the mold during metal fuel production because it is known that the coating layer obtained from these solutions can withstand up to 1500℃.

Rheosence viscometer was used to measure the viscosity of yttria slurry solution at room temperature. The average viscosity of the yttria slurry which was diluted medium was measured to be about 35 cP which was measured three times in these tests. Considering that the viscosity of the un-diluted solution is about 700 cP, it may be appropriate to dilute the un-diluted solution to acetone (or ethyl alcohol) 10 to 15 times in order to apply it to the actual process later.

Also, the availability of zirconia slurry solution as a coating material in mold to withstand at the reaction with high temperature melt containing uranium has been reviewed and viscosity also was measured. The viscosity of the zirconia slurry solution was measured in the same manner as described above, the measurement results of the two materials are also shown in Fig.1.

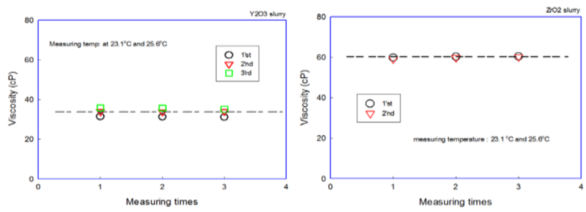

Fig. 1. Viscosities of slurry solutions (left :Y_2O_3, right:ZrO_2).

3.2 Particle size analysis

The yttria slurry solution is in a state in which the Y_2O_3 particles are dispersed in a mixed solution of a magnesium silicate binder and a small amount of organic matter as shown in the above table 1. The particle size and shape of the yttria particles (or zirconia particles in case of zirconia slurry) formed on the coating layer were observed when the slurry solution was used to fabricate the coating layer.

Experiments were performed by hand-brushing on a flat glass plate using the yttria slurry solution and zirconia slurry solution, and then dried at room temperature for 2 days. Particle size and shape by SEM photographs of the dried coating layer are shown in Fig. 2. The average size of the particles was observed to be 2-3 ㎛ for the yttria and 1.5-2 ㎛ for the zirconia.

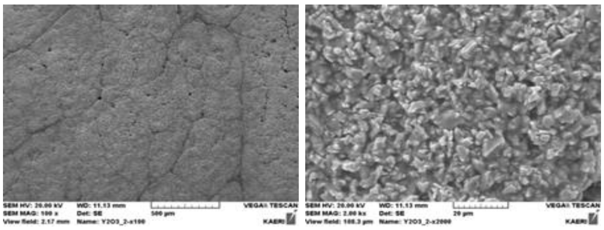

Fig. 2. SEM photographs of coating layers (left : Y_2O_3, right : ZrO_2).

3.3 Elemental analysis of coating layer

In the course of observing the SEM of the coating layer, the components of the coating layer were analyzed by an EDS analyzer attached to the SEM and shown in Fig. 3. As a result of the EDS analysis, as expected in advance, the yttria slurry coating layer was mainly analyzed for Y, and some Si and Mg element peaks were observed together with the O peak. But relative intensities of peaks were slightly different.

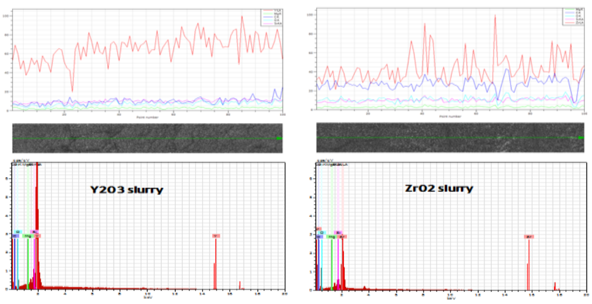

Fig. 3. EDS profiles of slurry solutions (left : Y_2O_3, right : ZrO_2).

3.4 Particle size and dispersion stability

The particle size distributions and dispersion stability of the two slurry solutions were analyzed using a Zetasizer Nano-ZS90 and LUMisizer, respectively. As a result of the particle size analysis, the two sample solutions were diluted sufficiently with ethyl alcohol, and its' result was shown in Fig. 4. In both samples, particles with a size of 1 to 3μm were found to be dispersed in the form of mono-dispersion, which are the results obtained after repeated analysis.

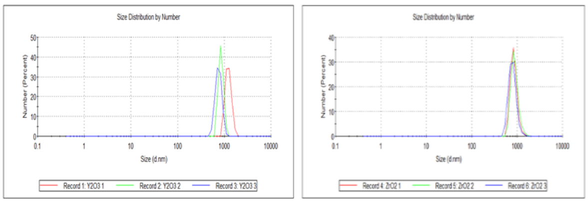

Fig. 4. Particle size distribution of slurry solutions (left : Y_2O_3, right : ZrO_2).

In the process of measuring the particle size distribution, the dispersion stability of the two slurry solutions was simultaneously measured. As can be seen from the Fig. 5, the stability of the particles dispersed in the solution was found to be relatively stable in the case of the zirconia slurry than that of yttria slurry.

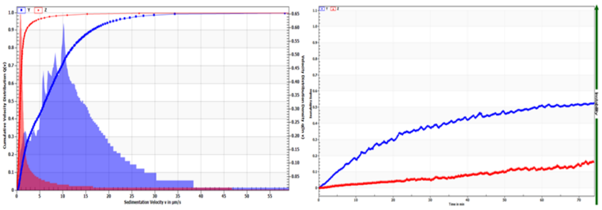

Fig. 5.Dispersion stability of slurry solutions (left :Y_2O_3, right : ZrO_2).

3.5 Quartz tube surface modification

When the yttria(or zirconia) slurry is coated on the rounded-quartz tube surface, the molten metal often enters the inner surface of the quartz tube due to the coating layer troubles during the production of the metal fuel rod and damages the coating layer. Therefore, as a solution for preventing such a problem, the improving research is needed to increase the adhesion strength of the coating layer. In this study, preliminary experiments for the adhesion improving of the coating layer were carried out. This is the surface-modification experiments for the surface of the quartz tube by corrosion medium. In here, the corroded-surface serves as an anchor for adhesion improving when meeting to the yttria slurry solution.

As a part of this study, we first observed the surface roughness, one of the surface characteristics, using by corroded-quartz tube surface made by the various corrosion solutions. The specimens obtained after the corrosion test were observed with a general Zoom photographs and a Streoscopic images, and the surface roughness were measured. The results of the modified-surface shape and the surface roughness measurement values according to the corrosion solution are summarized in table 2 and Fig. 6.

4. Conclusions

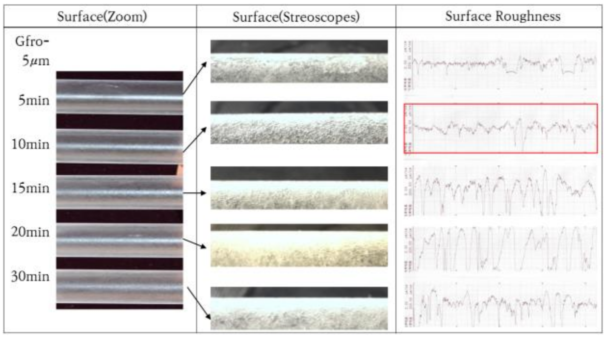

Fig. 6. Surface roughness measurements after corrosion treatment (corrosion media: Gfro-5μm solution).

Table 2. Summary of Quartz Surface Roughness After Corrosion Tests.

Solutions	Time	Ra	Rz	Remarks
Original quartz	0	0.017μm	0.128μm	
GT-2000(NH_4HF_2) (1st test)	1Hr	0.034	0.809	
	2	0.080	1.654	
	3	0.026	0.488	
	4	0.047	1.046	
	5	0.114	1.932	
HF-15% (2nd test)	10min	0.018	0.210	
	90	0.044	1.310	
GT-250MG4	1Hr	0.028	0.650	
HF-25%	0.5Hr	0.280	5.482	
	1.0	0.180	2.549	
	1.5	0.401	4.479	*2
	2.0	0.191	2.391	
Gfro-5μm (3nd test)	5min	0.646	3.222	
	10	0.556	4.099	*1
	15	2.772	11.540	
	20	1.432	9.284	
	30	1.303	8.480	

In this study, the process of coating the inner surface of the quartz tube mold with yttria slurry solution in the initial fuel rod manufacturing process was studied. From these studies, we have obtained the physical properties of yttria slurry and zirconia slurry solutions which are an inner coating slurry solution. As a basic study for applying these slurry solutions to the present fabrication process, we analyzed the viscosity, particle size, size distribution, and the dispersion stability of slurry solutions. Also, in order to increase the adhesion of the coating layer to the inner surface of the quartz tube used as a mold material, a basic corrosion experiments were conducted.

Also, preliminary experiments were carried out to increase the adhesion of the slurry solution by increasing the surface roughness of the mold surface through corrosion tests of quartz tube surfaces using various kinds of corrosion solutions. As a method of improving the adhesion of the coating layer, it could be pre-confirmed that the inner surface of the quartz tube could be modified by a chemical method. Later, this procedure using the surface modified-tubes can be applied in metal fuel rod production.

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