

# Alumina-based Nano Composite Coating Prepared by Plasma Spray

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In order to obtain nano-structured ceramics composite coating for high temperature application, pre-mixed  $\text{Al}_2\text{O}_3/\text{Y}_2\text{O}_3$  powders were plasma-sprayed in this study. Plasma spraying of spray-dried  $\text{Al}_2\text{O}_3/\text{Y}_2\text{O}_3$  powder resulted in the formation of amorphous coating of metastable  $\text{Al}_2\text{O}_3\text{-Y}_2\text{O}_3$  solid solution. After the heat treatment,  $\text{Al}_2\text{O}_3/\text{YAG}$  nano-structured composite coating was successfully obtained via eutectic reaction between  $\text{Al}_2\text{O}_3$  and  $\text{Y}_2\text{O}_3$ . It was possible to control the sizes of  $\text{Al}_2\text{O}_3$  and YAG particles by heat treatment with proper condition. Hardness of the coatings showed close relationship with their microstructure.

Key words: plasma spray, nano composite, CMC (Ceramics Matrix Composite), eutectic reaction

## 1. INTRODUCTION

Nano-composite material processing is recognized as one of the most important technology, and much effort has been done worldwide. Among many processes for nano-composites, thermal spray is considered as one of the promising process especially for surface modification applications, and a lot of studies have been already reported on this topic (for example [1, 2]). This study tried to prepare nano-structured ceramics matrix composite (CMC) coating by plasma spray for high temperature application, such as gas turbine and/or jet engine and so on. In the plasma spraying of ceramic materials, ceramic particles are expected to be totally molten during their flight and quickly solidified on the substrate. This makes the plasma spray process very difficult to control particle size precisely and obtains nano-composite coatings in conventional way.

In this study, agglomerated  $\text{Al}_2\text{O}_3\text{-Y}_2\text{O}_3$  powder is plasma-sprayed to obtain nano-composite CMC coating. This  $\text{Al}_2\text{O}_3\text{-Y}_2\text{O}_3$  system is known as starting material for uni-directionally solidified eutectic  $\text{Al}_2\text{O}_3/\text{Y}_3\text{Al}_5\text{O}_{12}$  (YAG) composite [3]. This eutectic reaction, which nucleates  $\text{Al}_2\text{O}_3$  and YAG particles from  $\text{Al}_2\text{O}_3\text{-Y}_2\text{O}_3$  liquid phase, would have large possibility to form fine-structured composite. In addition, this eutectic composite is expected to be one of an attractive candidate materials for high temperature structural applications, such as land base gas turbine, jet engine and so on, because of its high bending strength and excellent creep resistance at high temperature.

## 2. EXPERIMENTAL PROCEDURE

### 2.1 Sample preparation

Coating samples were prepared by means of conventional APS process with the parameters shown in Table I. Agglomerated powder was used as a spraying powder, which was prepared by spray dry method with the mixture of sub-micron-sized  $\text{Al}_2\text{O}_3$  and  $\text{Y}_2\text{O}_3$  powder (Table II and Fig. 1). Post heat treatment was carried out for all the coating samples at  $1200^\circ\text{C}$  in air to evaluate the effect of elevated temperature on the structure and properties of the coating samples.

Table I Plasma spray parameter.

Plasma gun	F4 VB (Sulzer Metco)
Powder	agglomerated (+27-63 $\mu\text{m}$ )
Power input	40 kW
Spray distance	150 mm
Plasma gas	Ar: 42 L/min. $\text{H}_2$ : 10 L/min.
Cooling	air, 300 L/min.

Table II Powder preparation.

- Starting powder : 0.42 $\mu\text{m}$  in average  
 $\text{Al}_2\text{O}_3$  (Nippon Light Metal Co., Ltd.)  
 $\text{Y}_2\text{O}_3$  (Nippon Yttrium Co.)
- Method : Agglomeration (Spray Drying)
- Size : +27 -63  $\mu\text{m}$
- Composition :  $\text{Al}_2\text{O}_3 : \text{Y}_2\text{O}_3 = 8 : 2$   
(in mole ratio)

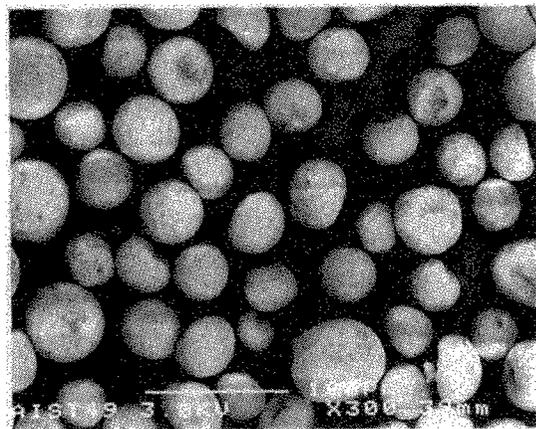


Fig. 1 Agglomerated  $\text{Al}_2\text{O}_3/\text{Y}_2\text{O}_3$  powder.

### 2.2 Evaluation of coating properties

Several examinations were carried out to evaluate structure and properties of the obtained composite coating. Microstructure observation was carried out with using scanning electron microscope (JEOL-6300F) on the polished cross section of the coating samples. X-ray

diffraction measurement was done to identify crystalline phases, and determine mean crystalline particle diameter of  $\text{Al}_2\text{O}_3$  and YAG phases from Scherrer's equation. Vickers microhardness was measured with the load of 0.49N for 30s. Ball on Disk Test was also carried out to evaluate friction coefficient of the coatings at room temperature.

### 3. RESULTS AND DISCUSSION

#### 3.1 Coating structure

XRD patterns and SEM images of the powder, as-sprayed and heat-treated coatings are shown in Fig. 2 and Fig. 3, respectively. As shown in Fig. 2, the as-sprayed coating was metastable amorphous phase of Y-Al-O solid solution. After the heat treatment at  $1200^\circ\text{C}$ ,  $\alpha\text{-Al}_2\text{O}_3$ /YAG nano composite structure was obtained;  $\alpha\text{-Al}_2\text{O}_3$  (seen as dark gray particles in Fig. 3) and  $\text{Y}_2\text{O}_3$  (seen as white ones in the same figures) precipitated and no other crystalline phases were observed (Fig. 2). Thus, it was possible to obtain nano-structured  $\alpha\text{-Al}_2\text{O}_3$ /YAG composite by plasma spray of agglomerated powder and post heat treatment via the eutectic precipitation.

In order to evaluate the stability of these nano-structured coatings at high temperature, the mean crystalline diameter of YAG and  $\text{Al}_2\text{O}_3$  in the heat-treated coating was estimated from the full width of half maximum of the XRD peaks by Scherrer's equation. The results are shown in Fig. 4. The crystalline diameter

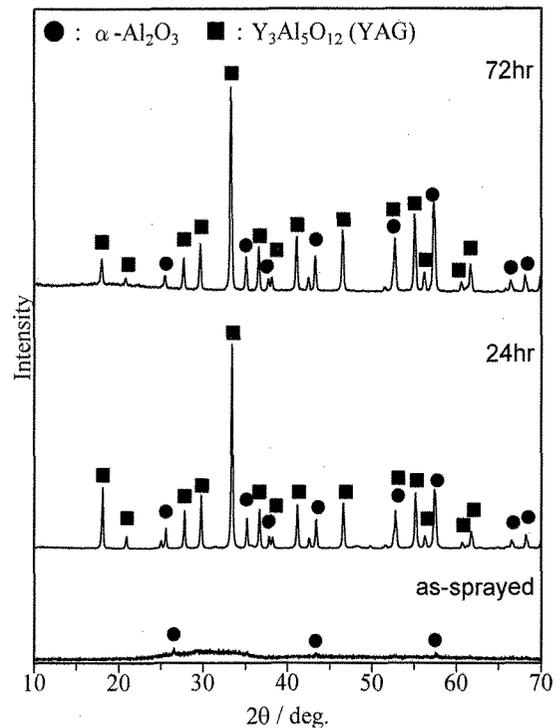


Fig. 2 XRD patterns of the coatings before and after the heat treatment.

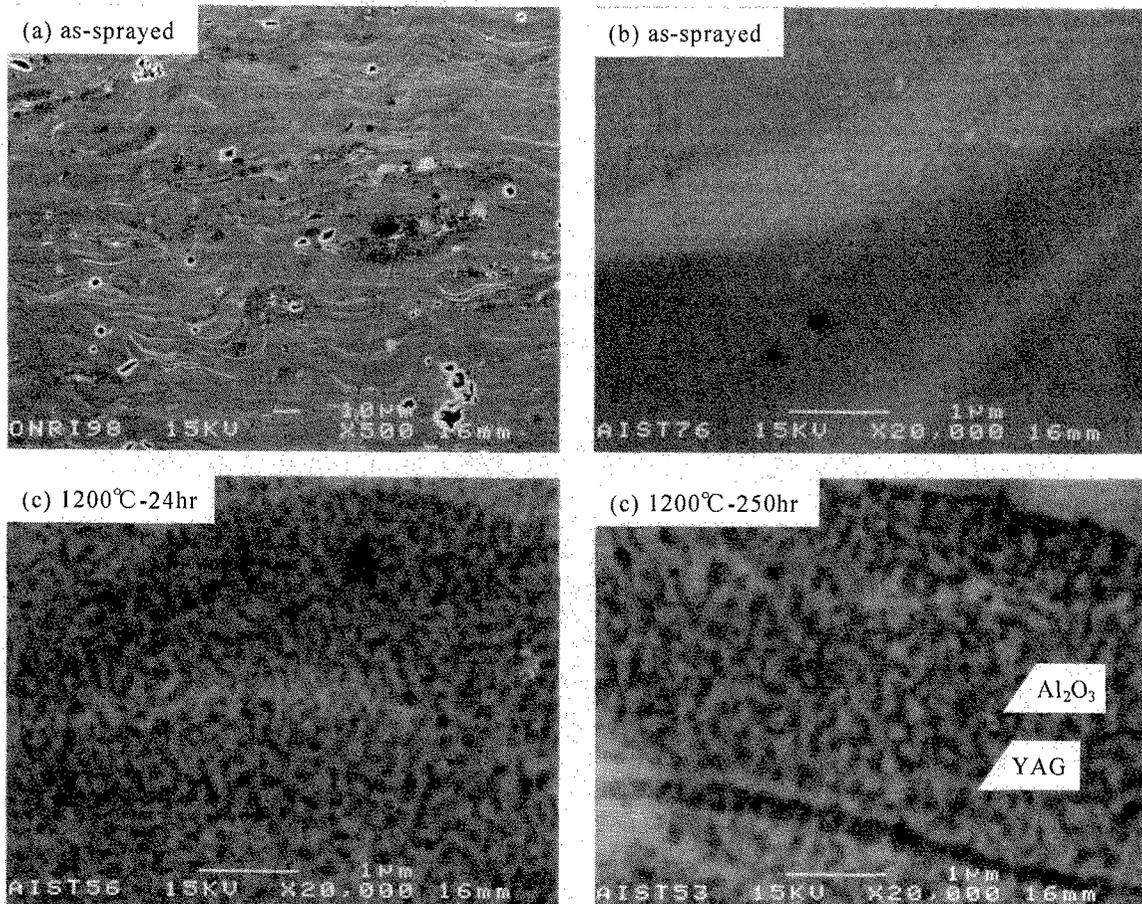


Fig. 3 SEM images of the cross section of the coatings before and after the heat treatment.

was about 40nm after the heat treatment at 1200°C. for 3hr. They showed slight increase with an increase of heat treatment time. However, it remains around 60nm, which is preferably small as a ceramics coating.

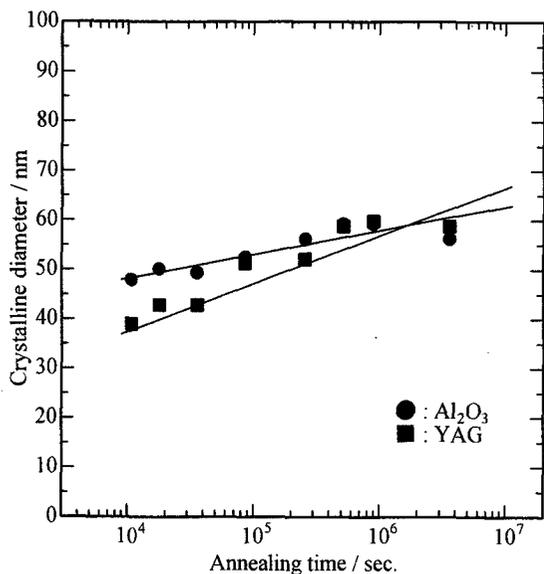


Fig. 4 Crystalline diameter of Al<sub>2</sub>O<sub>3</sub> and YAG in the heat-treated coatings.

### 3. 2 Coating properties

Vickers hardness is shown in Fig. 5. Hardness was kept constant around Hv=1200 after the heat treatment at 1200°C for 1hr, and then began to increase with longer heat treatment. The maximum value is almost equivalent to  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>, Hv=1800. This behavior could be deeply related to the particle size, dispersion and crystallinity of “ $\alpha$ -Al<sub>2</sub>O<sub>3</sub>” particle, since  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> is a dominant phase in the hardness of this  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>/YAG

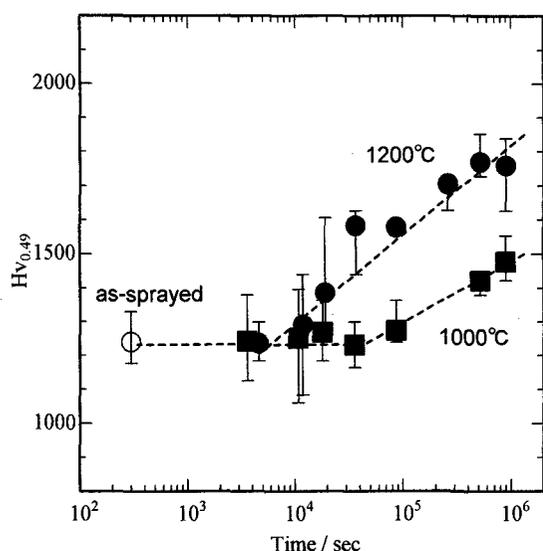


Fig. 5 Vickers microhardness of the heat-treated coatings.

system.

Friction coefficient of the coatings at room temperature is shown in Fig. 6. In this measurement, load, frequency and stroke were 10N, 60Hz and 1mm, respectively. Both of the as-sprayed and heat-treated coatings showed almost constant value through the test.

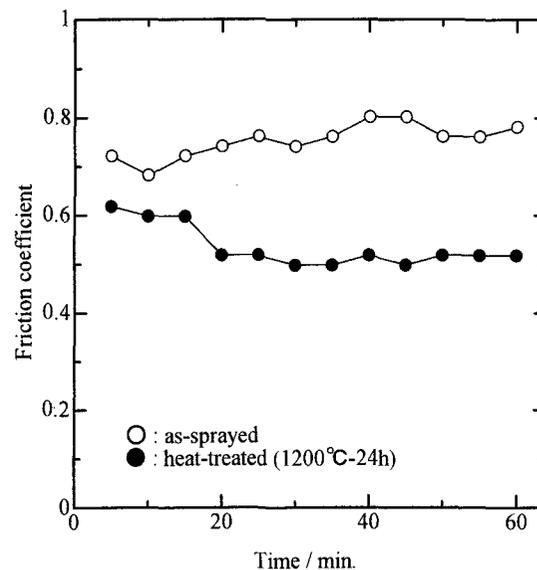


Fig. 6 Friction coefficient of the coatings before and after the heat treatment.

Coefficient of the heat-treated coating is relatively low; this shows that the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>/YAG nano composite coating might have high potential as an anti-wear coating.

### 4. SUMMARY

Effect of the post heat-treatment on the structure and properties of the plasma-sprayed  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>/YAG composite coatings were evaluated. Results are summarized as follows;

- 1) Amorphous phase of Y<sub>2</sub>O<sub>3</sub>-Al<sub>2</sub>O<sub>3</sub> solid solution is obtained in the coatings with the plasma spraying of the agglomerated Al<sub>2</sub>O<sub>3</sub>/Y<sub>2</sub>O<sub>3</sub> powder. After the heat treatment, both of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> and YAG precipitate by eutectic reaction. Their crystalline particle size can be controlled widely with proper heat treatment parameter. Nano-structured  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>/YAG composite coatings are obtained after the heat treatment at 1200°C.
- 2) Particle size increases with longer heat treatment time. However, even after the heat treatment at 1200°C for 250hr, the particle size was around 100nm or even smaller.
- 3) Friction coefficient of the heat-treated coatings showed lower value than that of the as-sprayed one. This shows that nano-structured  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>/YAG composite coating has high potential as anti-wear coating. However, further evaluation should be done for the improvement of the properties at high

temperature.

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#### REFERENCE

- [1] E. Petrovicova, R. Knight, L.S. Schadler and T.E. Twardowski, "Nylon 11/Silica Nanocomposite Coatings Applied by the HVOF Process: Part II. Mechanical and Barrier Properties", *J. Appl. Polymer Sci.*, (1999)
- [2] J. He, M. Ice, J.M. Schoenung, D.H. Shin and E.J. Laverna, "Thermal Stability of Nanostructured Cr<sub>3</sub>C<sub>2</sub>-NiCr Coatings", *J. Thermal Spray Tech.*, vol. 10, No.2 (2001) 293.
- [3] Y. Waku, "Dislocation mechanism of deformation and strength of Al<sub>2</sub>O<sub>3</sub>-YAG single crystal composites at high temperature above 1500°C", *J. Euro. Ceram. Soc.* 20 (2000), 1453.
- [4] M. Suzuki, "Microstructure of Plasma-Sprayed Al<sub>2</sub>O<sub>3</sub>/YAG Coating", *J. High. Temp. Soc. Jap.* Vol.28, Supplement (2002), 258

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