

# Formation of Fe-base Metal Glass Coating by Gas Tunnel Type Plasma Spraying<sup>†</sup>

KOBAYASHI Akira\*, YANO Shoji\*\*, KIMURA Hisamichi\*\*\*, and INOUE Akihisa\*\*\*

## Abstract

*Metal glass has excellent functions such as high toughness and corrosion resistance. Therefore it is one of the most attractive materials, and various developmental research works have been conducted by many researchers. However, the metal glass material is expensive and a composite material is preferred for the industrial application. Thermal spraying method is one of potential candidates to produce such metal glass composites. The gas tunnel type plasma system, which has high energy density and efficiency, is useful for smart plasma processing to obtain high quality ceramic coatings such as Al<sub>2</sub>O<sub>3</sub> and ZrO<sub>2</sub> coatings. In this study, the Fe-base metal glass coatings were produced by the gas tunnel type plasma spraying, and their microstructure and mechanical properties were investigated. The Fe-base metal glass coatings of 100 μm in thickness were formed densely with Vickers hardness of Hv=1100 at a higher plasma current.*

**KEY WORDS:** (Metal glass), (Gas tunnel type plasma spraying), (Fe based materials), (microstructure), (Hardness).

## 1. Introduction

Metal glass has excellent physical and chemical functions such as high toughness and corrosion resistance<sup>1-3</sup>). Therefore it is one of the most attractive advanced materials, and many researchers have conducted various developmental research works. Metal glass is expected to be used as a functional material at high temperature for example. However, as the metal glass material is expensive, the application for small size parts has been carried out only in some industrial fields. In order to widen the industrial application fields, a composite material is preferred for the cost performance. In the coating processes of metal glass with the conventional deposition techniques such as plasma sputtering and laser-assisted methods, there is a problem of the difficulty to form thick coatings due to their low deposition rate. Thermal spraying method is one of potential candidates to produce metal glass composites. Because of the cheaper method compared to other conventional ones, metal glass coatings can be applied to the longer parts and therefore widen the application field.

The gas tunnel plasma spraying is one of the most important technologies for high quality ceramic coating

and synthesizing functional materials, because the plasma jet has high speed and high energy density under various operating conditions. Also, it operates at a high voltage and with easy control of power. The performances of gas tunnel type plasma jets were clarified in previous studies<sup>4-6</sup>). For example, a gas tunnel type plasma jet, of the 200 kW class, has a high temperature of more than 20,000K and high energy density. Also, the thermal efficiency of gas tunnel plasma jet is about 80 %, which is much higher than that of a conventional type of plasma jet (≈ 50 %).

It is superior to the properties of other conventional type plasma jets<sup>7</sup>). Therefore this plasma has great possibilities for various applications in thermal processing<sup>8</sup>). As to the formation of high performance materials, high quality ceramic coatings were obtained by the gas tunnel type plasma spraying method<sup>9, 10</sup>); for example, typical alumina coatings produced had a high Vickers hardness of Hv =1200-1600<sup>11</sup>). Also, it is possible to produce sprayed coatings of refractory materials<sup>12</sup>) such as W. As another application, the gas tunnel type plasma jet was applied to the surface nitridation of titanium. This experiment also investigated the possibility of the speedy formation of a high

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functionally thick TiN coating<sup>13, 14</sup>.

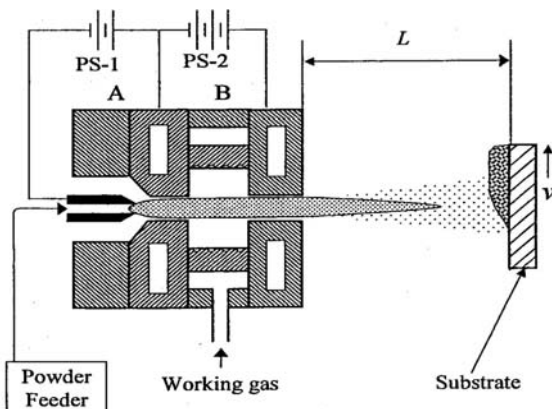
In this study, the gas tunnel type plasma spraying was used as the smart plasma spraying method, and the metal glass coating was formed on the stainless-steel substrate by gas tunnel plasma spraying. The formation process and the properties of the metal glass coating were investigated. The microstructure and surface morphology of the metal glass coatings were examined using Fe based metal glass powder as starting material. For the mechanical properties the Vickers hardness was measured on the cross section of the coating.

## 2. Experimental

### 2.1. Preparation of metal glass coatings

Fe-based metallic glass powder (Fe-B-Si-Mo) was used in this study. This powder was atmospherically plasma sprayed (APS) on flat 304 stainless-steel substrate by using a gas tunnel type plasma spraying torch which is shown in **Fig. 1**. Fe-based metallic glass powder was internally fed inside the plasma flame stream to obtain a maximum temperature because metal glass has high melting point  $\approx 2000$  °C.

Experiments were carried out under the spraying conditions as shown in **Table 1**. The plasma torch was operated at power levels up to 20kW and the arc current was chosen  $I=160, 300, 400$  A, respectively. The plasma jet was generated with the aid of argon gas was supplied at 180 l/m. The torch was maintained at a spray distance of 50 mm from the substrate surface. The powder injection was internal to the torch and directed parallel to the plasma flow and parallel to the torch trajectory. The powder feed rate was about 12 g/min. The spray configuration was a combination of a rotating sample holder and fixed torch uniformly. The defined substrate was traversed 4 times, and the spraying time was 30s. To avoid adhesion problems due to different thermal expansion coefficients between the coating and the substrate and limit the stress level, nitrogen cooling gas was applied to the spraying configuration to lower the coating temperature during deposition.



**Fig. 1** Systematic diagram of gas tunnel type plasma spraying torch.

**Table 1** Spraying condition.

Arc current	160, 300, 400 A
Voltage	40-50V
Spraying distance	50 mm
Working gas Ar flow rate	130 l/min
Feed gas flow rate	10 l/min
Powder feed rate	12 g/min
Traverse number	4 times
Spraying time	30 s

The components of this Fe-based metallic glass powder used in the experiment are shown in **Table 2**. The Fe content was 60% or more and there was 30% Mo, and some B content. The micrograph of this Fe-based metallic glass powder is shown in **Fig. 2**. The particle size of the powder was 30-60  $\mu\text{m}$ .

The grit blasted substrates were used for plasma spraying of metal glass. The substrates of dimension 50 x50 x2 mm<sup>3</sup> were grit-blasted with alumina grit on one side to clean and roughen the surface (5-7  $\mu\text{m}$ ) and followed by ultrasonic cleaning using acetone to remove any grease and other contaminations.

### 2.2. Characterization techniques

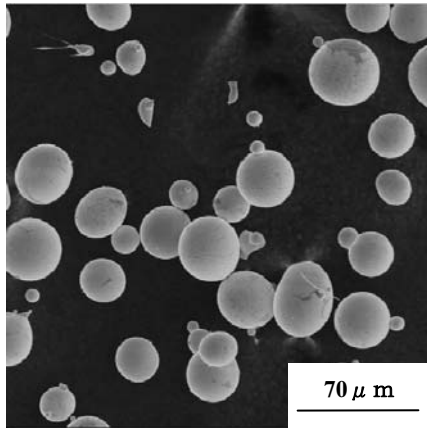
Microscopic observation of the coatings was performed using an optical microscope. Each specimen was mounted in conductive resin, ground with SiC paper and finally polished with 0.05 micron alumina slurry. The average thickness of the sprayed coatings was observed by optical microscope.

The surface morphology of the feedstock powder and the metal glass coating cross-section was examined by an ERA8800FE scanning electron microscope. The examined cross-section samples were mounted in epoxy resin using (HMP-Molding hand press, Wingo Seiki Co., Ltd. Osaka, Japan), polished by using (Buehler Metaser V Grinder-polisher) and buffed with alumina paste (1.0, 0.3, and 0.05  $\mu\text{m}$ , respectively) to obtain a mirror finished surface. All examined samples were coated with a thin film of gold using gold ion sputtering system (Thermo VG Scientific Polaron Sc7620 Sputter Coater) to make them electrically conductive before SEM observation.

Phase constituents of feedstock powder and metal glass coating were identified by using a JEOL JDX-

**Table 2** Fe-based metallic glass –composition.

Fe-Si-B-Mo		
component	Content (wt %)	Size ( $\mu\text{m}$ )
Fe	62.46	30-60
Si	0.66	
B	5.24	
Mo	31.64	



**Fig. 2** SEM micrographs of metal glass feedstock powder. Spherical shape “30-50 μm”

3530M X-ray diffractometer system with  $\text{CuK}\alpha$  radiation source at voltage of 40 kV and current of 40 mA.

Vickers microhardness measurement was made on polished sample surfaces using a load of 50g on each material. Indentation parameters were set as 20s loading time and average thickness was derived from five measurements.

### 3. Results

#### 3.1. Microstructure of feedstock powder

**Figure 2** shows the SEM graphs of Metal Glass feedstock powder. It is clear from SEM micrographs that the particle is a spherical type and the size of the powder ranged from 30 μm to 60 μm. **Table 3** shows some properties of Fe-based metallic glass. The density is similar to Fe. Yield strength is 700MPa. Stable crystallinity is at 560°C. Elastic modulus was 6000MPa. Hardness was  $\text{Hv}_{50}=990$ .

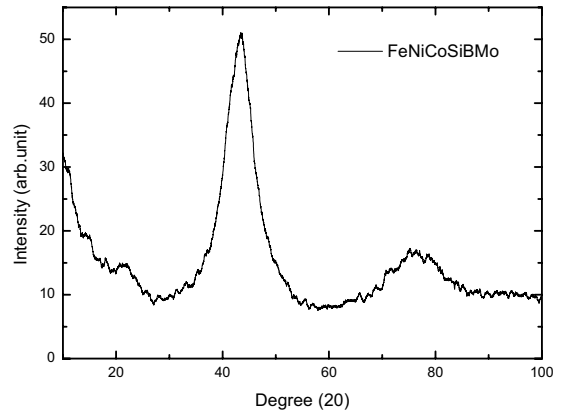
The XRD pattern of typical feedstock powder is depicted in **Fig. 3**. There are no crystalline peaks corresponding to Fe, Ni, Mo, etc. And the broad amorphous phase (Phase center is about 43 degree) was observed clearly in the pattern.

#### 3.2. Microstructure of plasma-sprayed metal glass coatings at different arc current

**Figure 4** shows the optical micrographs of the cross-section of plasma-sprayed Fe-Si-B-Mo metal glass

**Table 3** Fe-based metallic glass (Fe-B-Si-Mo) – properties.

Melting point	unknown
Hardness	990 (Hv 50g load)
Density	7.50 (g/cm <sup>3</sup> )
Yield strength	7x10 <sup>8</sup> N/m <sup>2</sup>
Stable crystalline	560 °C
Molecular weight	4911 X10 <sup>-3</sup> Kg/mole
Elastic modulus	>60x10 <sup>9</sup> N/M <sup>2</sup>



**Fig. 3** XRD pattern of the metal glass powder.

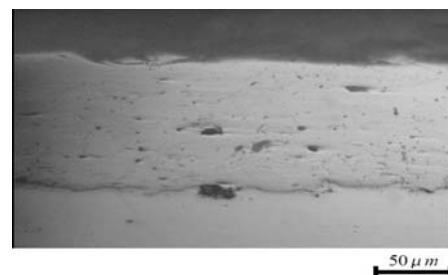
coatings sprayed at different plasma current of (a) 160 A, (b) 300 A and (c) 400 A at a spraying distance of  $L=50$  mm. In this case, the primary plasma gas flow rate was 130 Ar l/min, and the powder feed rate was 12 g/min (feed gas flow rate 10 l/min) during the spraying time of 30 sec.



(a) 160A



(b) 300A



(c) 400A

**Fig. 4** Optical micrographs of the cross-section of the metal glass sprayed at: plasma current: (a) 160 A, (b) 300 A and (c) 400 A. Carrier gas of Ar flow rate: 4 l/min, Ar plasma gas flow rate: 120 l/min, spray distance: 50 mm, and spray time: 30 sec.

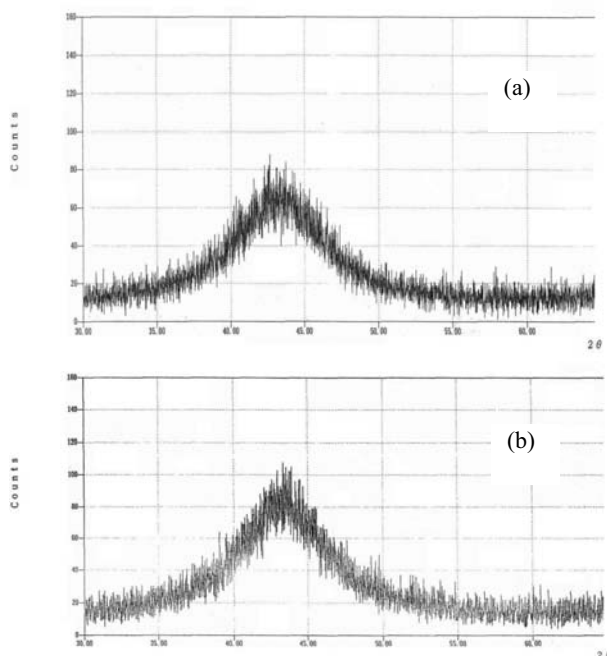
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Fig. 4 (a) shows the cross-section of the Fe-Si-B-Mo coating at: plasma current: 160 A. The cross-section at 300 A, Ar flow rate: 4 l/min, Ar plasma gas flow rate: 130 l/min, spray distance: 50 mm, and spray time: 30 sec are shown in Fig. 4 (b). The coating thickness is 100  $\mu\text{m}$  and very dense. At 400 A after increasing the plasma power, porosity was decreased and a thick coating was obtained.

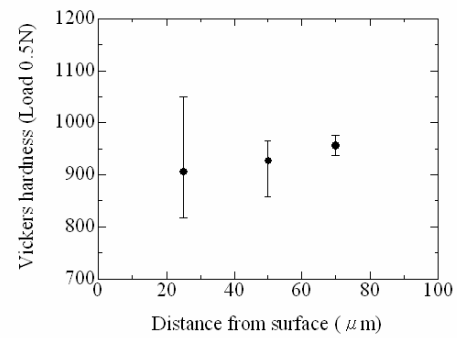
It was revealed that the Fe-Si-B-Mo coating was compact, smooth, dense, free from pores, adhering well, and with no cracking. The absence of pores from Fe-Si-B-Mo coatings indicated that the Fe-Si-B-Mo particles did not undergo decomposition during plasma spraying.

The XRD pattern from the surface of the coating is shown in Fig. 5 (a), also, the result of that for metal glass powder is shown in Fig. 5 (b), which revealed that the broad peaks corresponding to amorphous phase sprayed films. There were no crystalline peaks in the XRD pattern of the coating. This pattern of Fig. 5 (a) is similar to the of the metal glass powder shown in Fig. 5 (b), and in Fig. 3. Also, other peaks corresponding to the stainless-steel substrate, and any other oxidized materials were not detected.

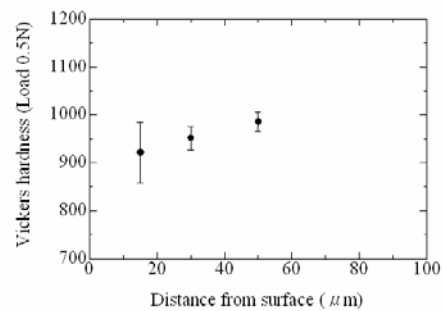
When the feedstock metal glass powder is injected into the plasma jet, particles in the plasma high temperature region are heated and may simultaneously decompose. So, there is the possibility of Fe, Si, Mo, etc. crystalline peaks, but there were no peaks from Fe or Si, Mo detected in the XRD spectra.



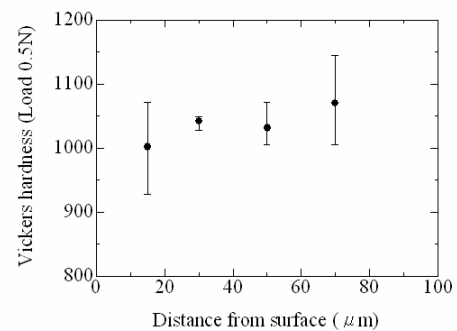
**Fig. 5** XRD patterns of the metal glass (a) on stainless-steel substrate sprayed at plasma current 160 A, and (b) powder used on this study.



(a) 160A



(b) 300A



(c) 400A

**Fig. 6** Distributions of Vickers hardness of Fe-based metal glass coating, deposited at different plasma current of (a) 160 A, (b) 300 A and (c) 400 A.

### 3.3. Vickers hardness of plasma-sprayed metal glass coatings

**Figure 6** shows the distribution of Vickers hardness on the cross section of metal glass coatings formed by gas tunnel type plasma spraying. In this case, the plasma current was respectively, (a) 160 A, (b) 300 A and (c) 400 A. The spraying distance was  $L=50\text{mm}$ , and the traverse number was 4. The coating thickness approximately varied from 70 to 100 $\mu\text{m}$ .

The Vickers hardness on the cross section of metal glass coating increased with an increase in the distance from the surface. This means at the coating near the substrate, the sprayed particle was rapidly cooled by the

substrate when deposited. So, more amorphous phases would be realized under those conditions. The Vickers hardness of metal glass coatings is around  $Hv_{50}=900-1070$ , wherever measured in the cross section of the coating. This hardness was similar to that of the original powder.

At higher currents, a high hardness coating was obtained because the coating would be thick and of high density due to a lower percentage of porosity, as shown in Fig. 4(c). Also, the plasma current affects the cooling rate when the particle is deposited on the substrate. In the case of higher current, large power gives higher cooling rate of the deposit metal glass powder. The highest value of  $Hv_{50}=1070$  was obtained near the substrate due to higher cooling rate. In this case, however, the hardness was uniform at the middle region of the coating.

Thus, Fe based metal glass coating was formed effectively by the gas tunnel type plasma spraying. This allows the development of thick and high functional metal glass coatings, which will be useful for various industrial applications.

#### 4. Conclusions

The Fe-based metal glass coatings sprayed by gas tunnel type plasma spraying produced the following results.

- (1) X-ray study confirms the presence of amorphous phases in the sprayed coatings and Fe based metal glass particles did not undergo decomposition of oxidation during deposition.
- (2) The sprayed metal glass formed on the stainless-steel substrate has a compact, smooth, and dense morphology with low porosity, especially at high plasma current.
- (3) The thickness of the sprayed metal glass coating was about 100  $\mu\text{m}$  at high current, depending on the spraying parameters.
- (4) The Vickers hardness of metal glass coating was around  $Hv_{50}=1000$ , in the cross section of all coating regions at different plasma currents. Vickers hardness near the substrate became a little higher due to higher cooling rate. Vickers hardness was increased up to about  $Hv=1100$  at a higher plasma current.

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