Advances in Materials Research, *Vol. 6 No. 4 (2017) 343-348*
DOI: https://doi.org/10.12989/amr.2017.6.4.343

Preparation of gas-atomized Fe-based alloy powders and HVOF sprayed coatings

Joseph Lik Hang Chau [*1], Alfred I-Tsung Pan [2] and Chih-Chao Yang [1]

[1] *Materials and Chemical Research Laboratories, Industrial Technology Research Institute, Tainan City, Taiwan*
[2] *Strategy and R&D Plan Office, Industrial Technology Research Institute, Hsinchu, Taiwan*

(*Received October 27, 2017, Revised March 13, 2018, Accepted April 2, 2018*)

Abstract. High-pressure gas atomization was employed to prepare the Fe-based $Fe_{50}Cr_{24}Mo_{21}Si_2B_3$ alloy powder. The effect of flow rate of atomizing gas on the median powder diameter was studied. The results show that the powder size decreased with increasing the flow rate of atomizing gas. Fe-based alloy coatings with amorphous phase fraction was then prepared by high velocity oxygen fuel spraying (HVOF) of gas atomized $Fe_{50}Cr_{24}Mo_{21}Si_2B_3$ powder. Microstructural studies show that the coatings present dense layered structure and low porosity of 0.17% in about 200 μm thickness. The Fe-based alloy coating exhibits an average hardness of about 1230 HV. Our results show that the HVOF process results in dense and well-bonded coatings, making it attractive for protective coatings applications.

Keywords: alloy; coatings; HVOF; gas atomization; powder

1. Introduction

Metallic glasses have been of interest because of their good physical and chemical properties (Suryanarayana and Inoue 2013). The random arrangement of atoms in these materials lead to the lack of microstructural features such as grain boundaries and dislocations, resulting in extraordinary high strength, hardness and excellent wear resistance (Inoue and Takeuchi 2011). Combinations of several engineering properties such as high strength, high hardness and good corrosion resistance rendered them as potential applications in engineering materials (Nishiyama *et al*. 2004). However, metallic glasses suffer a severe drawback of inherent brittleness that limit their applications as structural materials. Samples of amorphous metallic alloys are usually prepared only as thin ribbons and rods of several millimeters, because the critical cooling rate required to form the glassy phases in these alloy compositions was of the order of 10^5-10^6 K/s (Chen 2011). Accordingly, much attention has been paid to the coating applications with a view to widen their uses in industrial fields.

Among the various metallic glass systems, Fe-based bulk glassy alloys are very attractive because of their excellent soft magnetic properties, high electrical resistivity, high mechanical strength and relatively low materials cost. Nowadays, many coating methods can be used to prepare Fe-based amorphous metallic coatings, for example, plasma spraying, high velocity

∗Corresponding author, Ph.D., E-mail: jchau@itri.org.tw

http://www.techno-press.org/?journal=amr&subpage=5 ISSN: 2234-0912 (Print), 2234-179X (Online)

oxygen fuel spraying and magnetron sputtering (Kobayashi *et al*. 2008, Guo *et al*. 2011, Pei *et al*. 2011). High velocity oxygen fuel (HVOF) sprayed coatings have been widely used for industrial applications due to the relatively low operation temperature and a superhigh velocity (~1000 m/s) creating an extremely dense and well-bonded coatings (Fauchais *et al*. 2014). Because of the high particle kinetic energy available in the HVOF thermal spray process partially molten or "soft" particles are capable of forming dense coatings without the requirement of complete particle melting (Ni *et al*. 2009). This condition can facilitates the retainment of amorphous phase. Previous work reported that high hardness Fe-based alloy coating with low porosity can be fabricated by plasma spraying (Chau *et al*. 2017). In this work, HVOF technique was employed to prepare Fe-based metallic alloy coatings on stainless steel SS304 substrate with the self-prepared gas atomized Fe-based alloy powders as the feedstock powders.

2. Experimental

Gas atomization of molten alloys has benefits for production of large scale of metallic materials. This technique is particularly suitable for producing metallic glasses because of the rapid solidification occurs during the process. In this study, high-pressure gas atomization with a typical cooling rate of 10^3-10^4 K/s was employed to prepare the Fe-based alloy powders. A mixture of pure elements of Fe, Cr, Mo, Si and B with 99.9wt% purity was induction melted in a copper crucible under nitrogen atmosphere to form $Fe_{50}Cr_{24}Mo_{21}Si_2B_3$ master ingot. It is generally accepted that the addition of small amounts of specific elements like Mo, Si and B greatly enhance the glass forming ability of alloy, while Cr can enhance the corrosion resistance by providing a high passivation ability of the metallic glasses. Powders of the same composition were then prepared using high pressure nitrogen gas atomization method. Under vacuum condition (1×10^{-2} torr), the master ingot was heated up to the operating temperature (Tm = 1700 K), subsequently the molten alloy was ejected through a nozzle with a N2 pressure of 2.2 MPa. The gas-atomized powders were then sieved and divided into different size ranges from 40 (350 mesh) to 149 (100 mesh) μm. HVOF thermal spraying (JP-5000HP/HVOF system) was applied because of its ability to produce high density coatings with lower residual stress and improved metallurgical bonding to the substrate. The detailed spraying parameters are presented in Table 1. All these parameters were selected from numerous trial runs. The as-atomized alloy powders with particle sizes in the range of 40 to 74 μm were used for thermal spraying onto the 304 stainless-steel flat substrates. All the substrates were degreased by acetone, dried in air, and grit-blasted prior to deposition.

Scanning electron microscopy (JSM- 6330TF, JEOL) equipped with an energy-dispersive X-

Table 1 Detailed parameters of deposition process

Conditions	Value
Oxygen flow rate (m^3/s)	1.45×10^{-2}
Kerosene flow rate (m^3/s)	5.25×10^{-6}
Nitrogen flow rate (m^3/s)	6×10^{-3}
Powders feed rate (g/s)	0.42
Traverse velocity (mm/s)	800
Spraying distance (mm)	350
Spraying time (s)	7s/Pass

ray (EDS) spectroscope and X-ray diffraction (XRD, Philips Xpert) using Cu Kα radiation (λ = 1.5406Å) at 2 theta-step of 0.02° and scanning duration of 1 s from 10° to 90° were used for the microstructure and phase analysis of the gas-atomized powders and as-sprayed Fe-based alloy coatings. Particle size distributions were analyzed by laser particle size analyzer (Malvern-Mastersizer 2000). Image analysis software (Optimas) was used for measurement of coating porosity. Five SEM micrographs of the cross-sections of coatings with magnification of 1000 × were selected for the measurement of porosity. The Vickers microhardness of coatings was measured by a microhardness instrument (Matsuzawa MMT-X3) under a load of 25 g with a dwell time of 10 s. An average of 10 measurements for each sample was recorded as a microhardness value.

3. Results and discussion

Fig. 1 displays the X-ray diffraction pattern of the $Fe_{50}Cr_{24}Mo_{21}Si_2B_3$ master ingot. Because of the low cooling rate of the copper molding process (typically 10^2~10^3 K/s) (Srivastava *et al*. 2002), high fraction of crystalline phases are present in the ingot sample. After gas atomization of the

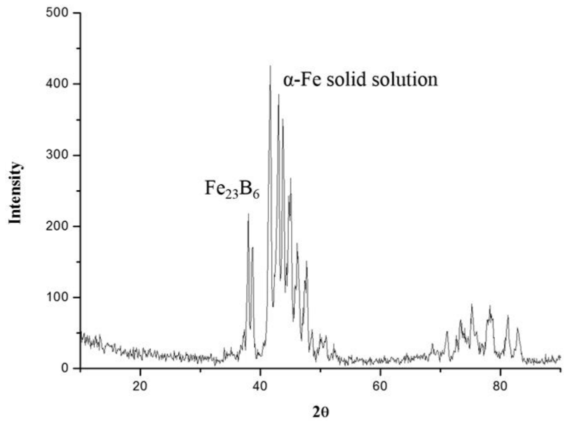

Fig. 1 X-ray diffraction pattern of the $Fe_{50}Cr_{24}Mo_{21}Si_2B_3$ master ingot

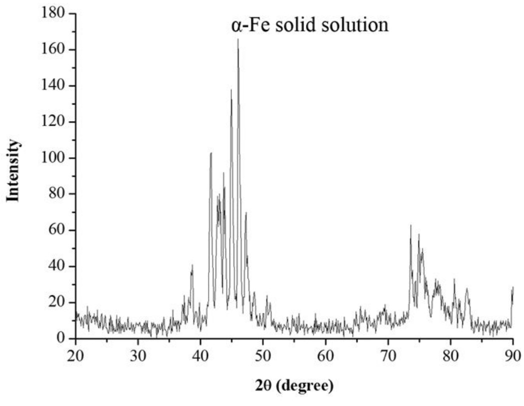

Fig. 2 X-ray diffraction pattern of the gas atomized powder particles

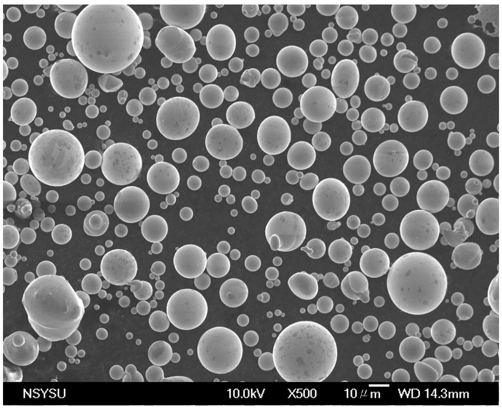

Fig. 3 SEM images of the $Fe_{50}Cr_{24}Mo_{21}Si_2B_3$ powder particles

Table 2 Effect of flow rate of atomizing gas on the median powder diameter

Flow rate of atomizing gas (Nm^3/min)	Powder median diameter D_{50} (μm)
10.66	26.25
13.74	25.55
15.52	23.76

alloy ingot, highly uniform and spherical powder particles were formed. Fig. 2 shows the X-ray diffraction pattern of the gas atomized powder particles. It can be seen that although crystallinity was reduced due to relatively faster cooling rate (typically 10^3~10^4 K/s) during the gas atomization process, some phases still present in the powder sample.

Fig. 3 shows the SEM image of the $Fe_{50}Cr_{24}Mo_{21}Si_2B_3$ powder particles before sieving process. It can be seen that the majority of the particles were spherical or near-spherical in shape, consistent with the typical morphology of gas atomized powder particles. During the gas atomization, the molten liquid breaks into small droplets that solidify in the presence of nitrogen gas during their flight inside the chamber. Owing to the rapid solidification conditions, the majority of the particles produced by gas atomization in nitrogen atmosphere are spherical or near-spherical in shape.

Table 2 summarizes the effect of flow rate of atomizing gas on the median powder diameter. It can be seen that the powder size decrease with increasing the flow rate of atomizing gas. Specifically, with each additional 3 Nm^3/min increase in atomizing gas flow rate, the median diameter D_{50} is decreased by approximately 2 μm. The cooling rate of droplet with given diameter is governed by the values of combined heat transfer coefficient depending mainly on the relative velocity of the droplet and atomizing gas (Grgač *et al*. 2012). Increase in the relative velocity of the atomizing gas and droplet increase the efficiency of gas atomization, thus the deeper initial undercooling of a droplet below the equilibrium liquidus temperature can be achieved. According to the X-ray diffraction pattern of the Fe-based alloy particles (Fig. 2), it can be observed that α-Fe solid solution and several unknown phases presents in the powders.

Fig. 4 displays the X-ray diffraction pattern of the Fe-based alloy coating. It can be seen that the coating possess of amorphous phase. In a typical HVOF spraying process, fuel and oxygen are generally pressed into a combustion chamber in a continuous flow, producing a jet of combustion products at supersonic speed. The gas and particle velocities are commonly much higher than those achieved using plasma spraying. Powders injected into this gas steam are accelerated to a very high velocity. The gas-atomized alloy particles may be partially or fully melted to form splats that conform and adhere to the substrate surface. The molten droplets cool at very high rate (10^5~10^6 K/s) and rapid solidification occurs when hitting the substrate because of the extremely fast velocity (Liu *et al*. 2009). Thus, the cooling rate of HVOF process is much higher than in the gas atomization process (10^3~10^4 K/s) resulting to a higher fraction of amorphous phase in the coatings.

Fig. 5 shows the SEM image of Fe-based alloy coating with a thickness of about 200 μm. It can be seen that no significant un-melted particles and large size pores in the coating. In addition, macro or micro cracks are not found in the coatings. It shows that the coating was compact because of the strong splat-to-splat cohesion in the HVOF process. From the cross-sectional SEM image, the Fe-based alloy coating has dense and uniform structure. The porosity of the coating analyzed by SEM imaging is 0.17%. The Fe-based alloy coating exhibits an average hardness about 1230 HV, which exhibited much higher microhardness than the substrate of 300 HV.

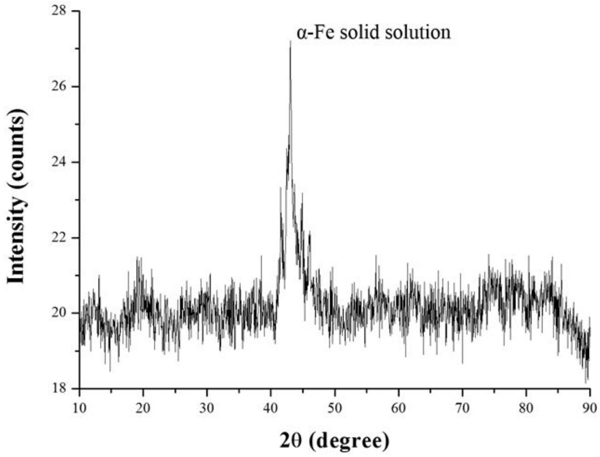

Fig. 4 X-ray diffraction pattern of Fe-based alloy coating

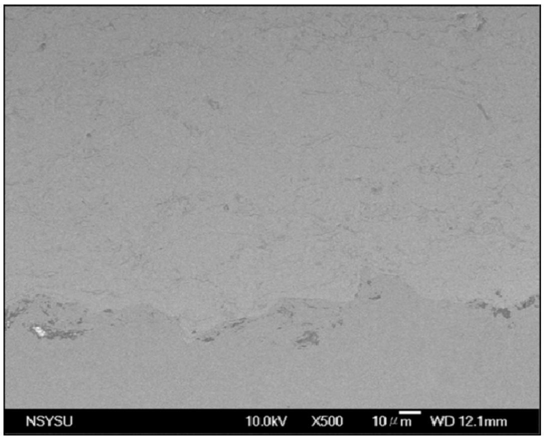

Fig. 5 SEM cross-sectional images of the Fe-based alloy coating

4. Conclusions

Fe-based $Fe_{50}Cr_{24}Mo_{21}Si_2B_3$ alloy powders can be prepared by high-pressure gas atomization process. The powder size decreases with increasing the mass flow rate of atomizing gas. Fe-based alloy coatings with amorphous phase was prepared by HVOF thermal spraying of gas atomized $Fe_{50}Cr_{24}Mo_{21}Si_2B_3$ powder. Microstructural studies show that the coatings present dense layered structure and extremely low porosity of 0.17% in about 200 μm thickness. The Fe-based alloy coating exhibits an average hardness about 1230 HV. Our results show that the HVOF process results in dense and well-bonded coatings, making it attractive for protective coatings applications.

Acknowledgments

Financial support from the Ministry of Economic Affairs of Taiwan is appreciated.

References

Chau, J.L.H., Hou, Y.Y., Pan, A.I.T. and Yang, C.C. (2017), "Fabrication of high hardness and corrosion resistance coatings from Fe-based alloy powders", *Particul. Sci. Technol.*: *Int. J.*, **35**(2), 209-213.

Chen, M. (2011), "A brief overview of bulk metallic glasses", *NPG Asia Materials*, **3**(9), 82-90.

Fauchais, P.L., Heberlein, J.V.R. and Boulos, M. (2014), *Thermal Spray Fundamentals*: *From Powder to Part*, Springer, New York, NY, USA.

Grgač, P., Behúlová, M., Moravčík, R. and Mesárošová, J. (2012), "Semi-quantitative model of the microstructure development in the high-alloyed iron based alloy during atomization", *Mater. Res.*, **15**(5), 705-712.

Guo, R.Q., Zhang, C., Chen, Q., Yang, Y., Li, N. and Liu, L. (2011), "Study of structure and corrosion resistance of Fe-based amorphous coatings prepared by HVAF and HVOF", *Corrosion Science*, **53**(7), 2351-2356.

Inoue, A. and Takeuchi, A. (2011), "Recent development and application products of bulk glassy alloys", *Acta Materialia*, **59**(6), 2243-2267.

Kobayashi, A., Yano, S., Kimura, H. and Inoue, A. (2008), "Mechanical property of Fe-base metallic glass coating formed by gas tunnel type plasma spraying", *Surf. Coat. Technol.*, **202**(12), 2513-2518.

Liu, Z.Q., Zheng, Y.G., Chang, X.C., Hou, W.L., Wang, J.Q., Tang, Z. and Burgess, A. (2009), "Microstructure and properties of Fe-based amorphous metallic coating produced by high velocity axial plasma spraying", *J. Alloys Compounds*, **484**(1-2), 300-307.

Ni, H.S., Liu, X.H., Chang, X.C., Hou, W.L., Liu, W. and Wang, J.Q. (2009), "High performance amorphous steel coating prepared by HVOF thermal spraying", *J. Alloys Compounds*, **467**, 163-167.

Nishiyama, N., Amiya, K. and Inoue, A. (2004), "Bulk metallic glasses for industrial products", *Mater. Transact.*, **45**(4), 1245-1250.

Pei, H.J., Lee, C.J., Du, X.H., Chang, Y.C. and Huang, J.C. (2011), "Tension behavior of metallic glass coating on Cu foil", *Mater. Sci. Eng. A*, **528**(24), 7317-7322.

Srivastava, R.M., Eckert, J., Löser, W., Dhindaw, B.K. and Schultz, L. (2002), "Cooling rate evaluation for bulk amorphous alloys from eutectic microstructures in casting processes", *Mater. Transact.*, **43**(7), 1670-1675.

Suryanarayana, C. and Inoue, A. (2013), "Iron-based bulk metallic glasses", *Int. Mater. Rev.*, **58**(3), 131-166.

CC