

# Impact of low-temperature sintering on the Fe-based Amorphous coatings

Amjad Iqbal Falak<sup>\*1</sup>, Ayesha Iqbal<sup>2</sup>, Ahmed Zubair Jan<sup>3</sup>, Muhammad Bilal Hafeez<sup>4</sup>

<sup>1</sup>Department of Advance Materials and Technologies, Faculty of Material Engineering, Silesian University of technology Gliwice Poland, e-mail:Amjad.iqbal@polsl.pl

<sup>2</sup>Center of Leadership and Decision Making, Faculty of Behavioral and Social Sciences, University of the Punjab Lahore, Pakistan, e-mail:ayeshafalak565@gmail.com

<sup>3</sup>Department of Mechanical Engineering, Wroclaw University of Science and Technology, Wroclaw, Poland e-mail: Ahmed.jan@pwr.edu.pl

<sup>4</sup>Department of Mechanical Engineering, Faculty of Mechanical Engineering and Ship Technology, Gdansk University of Technology Gdansk, Poland, e-mail: muhammad.bilal.hafeez@pg.edu.pl

## Abstract

Bulk metallic glasses (BMGs) have gained a lot of attention in recent years due to their outstanding properties such as high hardness and excellent wear and corrosion resistance. However, they are restricted in industrial applications due to their extreme brittleness. Iron based amorphous coatings from BMGs are the best solution to use them by overcoming the problem of extreme brittleness. The coatings can be sprayed by various thermal techniques such as flame spraying (FS), high velocity air fuel (HVOF), High velocity oxy fuel (HVOF) spraying process and plasma spray (PS) process. In this study, we have developed iron-based amorphous coating via HVOF and found interesting effect of structural relaxation at low temperature. Iron-based amorphous alloy powder with Fe<sub>48</sub>Mo<sub>14</sub>Cr<sub>15</sub>Y<sub>2</sub>C<sub>15</sub>B<sub>6</sub> composition was used as the feedstock powder. The coatings sprayed by HVOF the techniques showed compact coatings as observed by the Scanning electron microscopy (SEM) results and amorphous phase was confirmed by the X-ray diffraction analysis at various temperature. Further investigations on the mechanical properties and chemical properties show that low temperature sintering improve the property of coatings.

**Keywords:** Bulk Metallic Glasses, Iron-based Amorphous coating, Thermal spray Process

## 1. Introduction

Bulk metallic glasses (BMGs) have been widely taking the attention, especially, when Fe base amorphous coatings (an alternative form of BMGs materials), due to their ultrahigh strength, high hardness, elasticity, hydrophobicity, magnetic properties and exceptionally good wear and corrosion resistance. From materials and industrial point of view, iron-base amorphous systems are trending due to unique alloying compositions. Due to low cost and good glass forming ability (GFA), their demand is increased [1-2]. Huang et al. found that Fe base amorphous BMGs can work at high Temperature ranging from (300-571K) and shows excellent wear resistance behaviour than other traditional alloys, and BMG systems [3]. R.Q. et al. experimented with Mo<sub>7.4</sub>W<sub>1.6</sub>B<sub>15.2</sub>C<sub>3.8</sub>Si<sub>2.4</sub> (material composition chosen because of its high neutron absorbing ability from Boron) and concluded that excellent wear and corrosion resistance in underground environment is due to “blocking effect by passive layers” [4]. With their prominent use there is also a hindrance while choosing these materials, they show brittleness at room temperature. To overcome these challenges, research areas like BMG composites to enhance plasticity and Fe-base amorphous thermal spraying techniques are efficient ways [5]. Coating technologies are just trying to replace the old tradition of alloying. Some Reports of novel alloys also suggest that they have issues related to health and recyclability. Thermal coatings seeking consideration due to their low cost and availability of various choices regarding substrate and materials. Recently X. Shan et al. synthesize a protective, thermal shock and oxidation resistance coating by slurry-spraying method on Cr Mn alloy substrate [6].

The Defense Advanced Research Projects Agency (DAPRA) of United States found Fe<sub>48</sub>Mo<sub>14</sub>Cr<sub>15</sub>Y<sub>2</sub>C<sub>15</sub>B<sub>6</sub> (SAM1651) as high-performance corrosion resistant alloys due to its better performance and can be use in defense applications [7].

G.L. Hou et al. work with HVOF for its low flame temperature and high flame velocity in his research of heat treatment on wear behavior of WC-(WCr)<sub>2</sub>C-Ni coating [8]. Thermal spraying techniques are promising choice for amorphous coatings due to their high cooling rate. Different thermal spray techniques are available e.g., flame spraying (FS), high velocity oxy-fuel (HVOF) spraying, plasma spraying (PS), and electric arc spraying (EAS). Careful optimization and selection of a technique is required, because preparation is the basis of a good coating

(i.e., low porosity and cracks, homogeneous deposit). High velocity oxygen fluid (HVOF) is the most widely used coating technique, due to its improved metallurgical strength with substrate and high dense coatings. Although a lot of work has been done on amorphous coatings, but still research is continuing to improve quality of these systems. Author and his group also try to coat SAM series alloy with HVOF spraying technique for this involving industrial benefit at low cost. We found an interesting impact of low sintering temperature effect on the coatings.

## 2. Material and Methods

**Substrate and coating material:** Low carbon steel (AISI 1018) was used as a substrate, due to its high demand in improve drilling, machining, threading, and punching processes. Low carbon steel was transformed into  $5 \times 5 \times 1 \text{ cm}^3$  through cutting edge technology. One clear piece of Low carbon steel was degreased in polar solvent before spraying. To increase the roughness of coating, we have done grit blasting at 0.35 MPa. Profilometer was used to measure desired roughness. Roughness of substrate helps us in better adhesion with coated material. Composition Fe48Cr15Mo14C15B6Y2 were used as a powder. This commercially available (SAM1651) has better properties in recent available amorphous systems [9-10].

**Spraying Process:** Thermal spraying process involves heating the alloy composition until it becomes molten, then by high pressure it is sent on roughly surface of substrate where it is solidifying. HVOF was used and coating thickness was achieved in the range of 300-350  $\mu\text{m}$ . Spraying parameters for HVOF we have taken are given in **Table 1**. Before operating HVOF machine, a leakage test is performed on all gas cylinders including propane, oxygen, and nitrogen.

Nitrogen was used as powder feeder gas while propane and oxygen involve in combustion process. A required pressure was set by opening gauges of cylinder. Powder is kept in an oven for 2h, temperature range (25-150 °C) to remove moisture. HVOF can work at very low temperature (1000-3000 °C) and super high velocity which 3 to 4 times higher than speed of sound. High velocity also reduces the oxygen content in coatings. This process is typically ideal for metals depositing, ceramic and dense coatings [11-13]. The principle working of HVOF is that it uses hydrocarbon as fuel (propane was used in this Experiment), oxygen and pressure for combustion at high speed, while nitrogen work as carrier gas which import powder from powder feeder to spray gun.

The powder feeding rate is also one of parameter that build thickness of coatings. The cooling pipes are attached to spray gun; continuously cold water is flowing through pipes to cool down the gun. The cooling rate of HVOF is 107–101  $\text{K s}^{-1}$ . These all steps are involved in generating a high speed “supersonic flame” [14-15].

## 3. Characterizations

Mono-layer amorphous coated samples of HVOF were transformed into square shape, as per requirement of SEM, XRD, and epoxy moulding process requirements. Their grinding and polishing were performed to get mirror image. Cross section of coatings has been scanned by SEM MIRA3 TESCAN, amorphous phase was confirmed by XRD (GNR analytical instrument group Explorer), and hardness was measured by Micro VICKER hardness NBK NABEYA VLS 3858 apparatus. Other tests like corrosion testing were performed through Gamry apparatus. Finally, the comparison has been done on the investigated properties.

### A. SEM Morphology

A cross section has been seen through scanning electron microscopy (SEM) Mira3 Tescan. It is confirmed that a coating has been prepared. Amorphous powder properly adheres with substrate. The thickness of coating via HVOF is 315.98  $\mu\text{m}$ . **Figure 1** show cross section image of coatings. Scanning has been done by moulding the sample. It shows stresses between the layers.

### B. XRD Analysis

During this study XRD was done with Cu  $K\alpha$  rays, a crystalline graphite analyzer and Step scanning was performed from 20° to 80° (2 theta).

XRD results of sprayed coatings and SAM1651 powder shows no significant Bragg diffraction peak which confirms that both coating and powder have amorphous structure. It can be seen in **Figure 2**. Furthermore, we performed the sintering at low temperature and performed XRD to check crystalline phases in the coatings. It can be viewed in the Figure that interestingly we observed no phase transformation in the coatings at low temperature. It shows amorphous phase is highly stable in the iron-based amorphous coatings.

### C. VICKER HARDNESS

We performed the hardness test to analyze the coating property. It was observed that coatings have very good hardness level. Through this test we observed that low level of hardness can be increase by employed iron-based amorphous layer. It was also observed that low temperature sintering effect the hardness property. It can be seen in **Figure 2** The sintering improves the hardness level due to structure relaxation in the system and creating more strengthen in the metallurgical bonding.

#### D. Corrosion analysis

Corrosion behavior of fabricated layer was analyzed by Gamry instrument The Reference 3000™. 35 grams of solid NaCl and 1000 mL distilled water were mix together in a 1200ml beaker to make 3.5 wt% an electrolyte solution for the experimentations. We are using 3.5 wt% NaCl due to the concentration of 3.5% of NaCl (by weight) in aqueous solution is often used as, probably similar to the concentration of this salt present in sea water. First sample was dipped in 3.5% NaCl for approximately 1h to stabilize the surface. To calculate the corrosion current ( $I_{corr}$ ) and corrosion potential ( $E_{corr}$ ) by potentiostat apparatus, we use voltage from -1.2 to 1.2V. The result obtained by this experiment is mentioned in **Table 2**. It can be concluded that the corrosion rate is almost 40 times higher through HVOF spraying. It can be observed that sintering improved the corrosion resistance of amorphous layer. We conclude this investigation, as coating layer has a lot of porosity and un-melted particles with in it. These all are collective generating stresses and decreasing corrosion current density. But as we perform the process of sintering the amorphous layer at two different temperatures, this sintering at low temperature produce the structure relaxation in the system. Heating decreases the porosity level, microstructure have re-organized them in the layer and stresses are removed in the form of hot corrosion products. Sintering also make HVOF coatings layer into more compactness between the layer and metallurgical bonding become stiffer. Hence it can be summarized, that the iron-base amorphous coatings can be superior coated with HVOF, but the effect of sintering is highly affecting them. This effect not only improves the structure property of the coatings but also help them to resist as an outstanding way under saline environment.

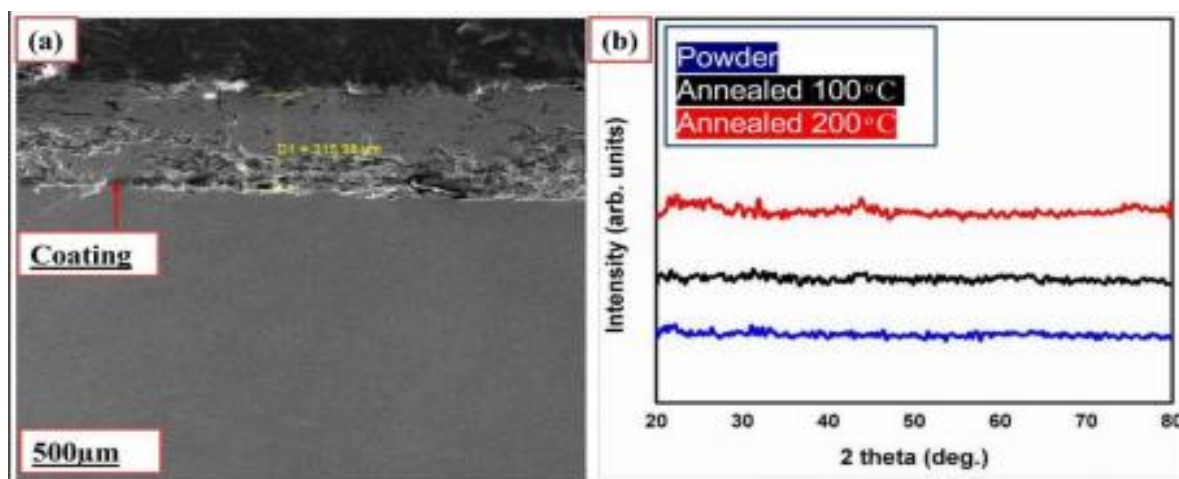


Figure 1. (a) SEM image of cross-sectional analysis at 20.0kv (b) XRD patterns

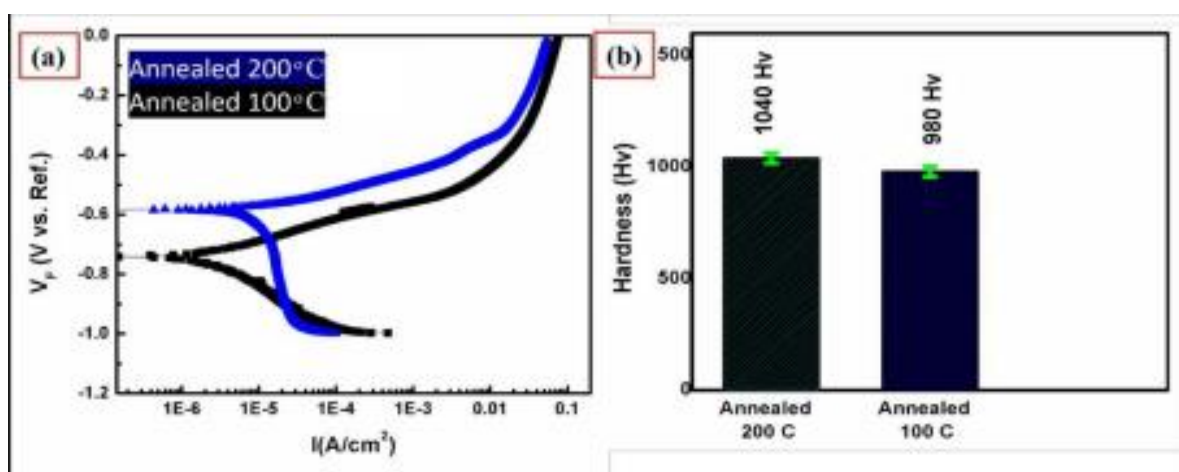


Figure 2. Electrochemical analysis of Fe- based amorphous coating: (a) Tafel fitted Potentiodynamic (b) Micro Vickers Hardness under 8N load for 20 second

Table 1. Parameter for High Velocity oxyfuel

S. No	Parameters	Values
1.	Fuel flow (L/h)	25 Lh <sup>-1</sup>

2.	Oxygen flow(m <sup>3</sup> /h)	35 m <sup>3</sup> h <sup>-1</sup>
3.	Spraying distance (mm)	300 mm
4.	Powder feeding rate (g /min)	24 gmin <sup>-1</sup>

**Table 2. Potentiodynamic parameter and Micro-Vickers Hardness**

S. No	Samples	Corrosion Rate	E <sub>corr</sub> (V)	I <sub>corr</sub> (μA/cm <sup>2</sup> )	Hardness
1.	Annealed 100°C	9.67 (mpy)	-0.586	11.4	980 HV
2.	Annealed 200°C	1.775(mpy)	- 0.724	2.09	1040 HV

#### 4. Conclusions

Through this research it was revealed that HVOF thermal spray can be used for fabrication of iron-based amorphous layer. The quality of obtained coating is good. The impact of sintering make them more efficient in the form of hardness, and their anti-corrosion property was increase in much greater time.

#### Acknowledgement

The authors are grateful to the surface engineering laboratory, Institute of space technology, Islamabad for providing the required facilities, and Silesian University of technology, for providing the research atmosphere.

#### Author Contributions

Conceptualization, A.I.F.; Formal analysis, A.I.F, A.Z.J and M.B.H; Investigation, A.I.F, A.I, A.Z.J.; Methodology, A.I.F, A.Z.J, M.B.H; Project administration, A.I.F, M.B.H; Visualization, A.I.F. and A.I; Writing—original draft, A.I.F.; Writing—review & editing, A.I.F, A.I. All authors have read and agreed to the published version of the manuscript.

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