



# Laminar Fe-based amorphous composite coatings with enhanced bonding strength and impact resistance

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## Abstract

Laminar coatings comprising a multi-amorphous  $\text{Fe}_{48}\text{Mo}_{14}\text{C}_{15}\text{Y}_2\text{C}_{15}\text{B}_6$  layer and a crystalline NiCrAl layer were prepared by the high-velocity oxygen fuel technique. The structures of the coatings were studied with X-ray diffraction, scanning electron microscopy and cross-sectional transmission electron microscopy, while the various mechanical properties, such as hardness, strength, interfacial toughness, bonding strength and impact resistance, were investigated by micro/nanoindentation, three-point bending and adhesion testing as well as by tup-drop impacting coupled with finite element method simulations. The results show that composite coatings exhibit an almost fully compact structure at interfaces between the amorphous layer and the NiCrAl layer with an attractive combination of high hardness and bonding strength, good interfacial toughness and high impact resistance with respect to the monolithic amorphous coating. The maximum bonding strength and impact resistance obtained in the laminar coatings reached 40 MPa and 21.2 J, respectively, which are respectively a  $\sim 54\%$  and a 10-fold increase over the monolithic coating. The enhancement of the bonding strength results from the formation of the localized metallurgical bonding as well as complete wettability between the NiCrAl layer and the amorphous layer, which is observed by high-temperature contact angle tests. The improvement in the impact resistance of the composite coatings results from hindering crack propagation and mitigation of stress concentration due to the addition of the soft NiCrAl multilayers. The present work may pave a way to the development of durable, tough and highly impact resistant amorphous–matrix composite coatings for applications in severe loading conditions.

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**Keywords:** Thermal spraying; Fe-based amorphous coating; Multilayers; Mechanical properties; Finite element simulation

## 1. Introduction

Bulk metallic glasses (BMGs) are amorphous in nature, with high strength and large elastic strain [1,2]. However, their poor ductility and limited product sizes restrict their widespread applications [3–5]. Amorphous coatings, an alternative form of BMGs, have recently attracted increasing interest in many industrial applications, because of the possibilities of synthesizing a large-scale surface protection layer with an excellent combination of properties, such as

high hardness and wear resistance, superior corrosion resistance and neutron-absorption ability [6–20].

While a great deal of research on amorphous coatings has been focused on corrosion and tribological properties, a limited amount of research has been directed towards the evaluation of bonding strength, which is actually of primary importance for the coating lifetime in service. It has been reported recently that amorphous coatings have insufficient bonding strength with the steel substrates. For example, Komaki et al. [9] reported that the thermally sprayed  $\text{Fe}_{70}\text{Cr}_{10}\text{P}_{13}\text{C}_7$  amorphous coatings have a bonding strength of  $<15$  MPa, which cannot meet the requirements for most industrial applications. Our previous

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experimental results also verified the low bonding strength (<30 MPa) for relatively thick (typically >400  $\mu\text{m}$ ) amorphous coatings [21]. The failure mechanism of the amorphous coating was found to be mainly adhesive fracture at the interface between the amorphous layer and the substrate, where cracks usually initiated from the boundaries of non-melted particles and then propagated along the coating/substrate interface. It has been suggested that a decrease in interfacial porosity, an increase in the wettability of the sprayed particle on the substrate and the introduction of an adhesive interlayer are the main approaches to improving the bonding strength of the amorphous coatings [9,21].

Apart from the bonding strength, the impact resistance is another important property that would strongly affect the lifetime and other properties of the amorphous coatings. Recently, Farmer et al. [10] investigated the impact resistance of a Fe-based amorphous coating and demonstrated that the amorphous coatings exhibited poor impact tolerance, as revealed by severe interfacial delamination failure and extensive surface cracking when they underwent a dynamic impact. This is probably due to the insufficient adhesion between the coating and the substrate and is also due to the brittle nature of monolithic amorphous materials. It has been predicted that the composite coatings with a mixing structure of amorphous matrix and ductile (or hard) crystalline reinforcements can enhance the fracture toughness and the impact resistance, in which the crystalline phases and their interfaces can act as barriers to crack propagation [22–27]. A few amorphous–matrix composite coatings reinforced by ceramic particles (e.g. WC–Co,  $\text{B}_4\text{C}$  and TiN) have been exploited in recent years [28–30]. Thanks to the satisfactory adhesion between the reinforcements and the amorphous matrix as well as the inhibiting effect of the additions to crack propagation, the composite coatings show high hardness, good bonding strength and significantly improved tribological properties. Terajima et al. demonstrated that the composite coating with the maximum 8% volume fraction addition of WC–Co phase exhibited a 15% increase of the bonding strength as compared with the monolithic Fe-based amorphous coating [28]. However, the bonding strength of the composite coating is closely related to the volume fraction, size and spacing of the reinforcements; an optimal combination of them is required, otherwise it would deteriorate the properties of the composite coatings [31]. How to precisely control the structures of the particle-reinforced composite coatings was a big challenge due to the difficulties in achieving homogeneous dispersion of reinforcements in the parent matrix accompanied by avoiding chemical reactions of pre-mixed composite powders during the spraying process [32,33].

The concept of laminar materials (which consists of two alternate layers with different physical and mechanical properties) can be applied to the amorphous coatings to improve the mechanical properties with well-controlled microstructures of the coatings. In fact, numerous

amorphous/crystalline thin films with laminar structures have been developed recently, and they exhibited high strength, high ductility and superior damage resistance over their monolithic counterparts [34–41]. For instance, an amorphous ZrCu/crystalline–Cu nanolaminate prepared by magnetron sputtering presented a high flow stress of  $\sim 1.1$  GPa and large tensile ductility (with  $13.8 \pm 1.7\%$  strain) [35], while the metallic glass/Zr laminated composite showed significant enhancement of the toughness due to crack redirections at the crystalline layers upon deformation [34]. More recently, such geometric configuration was applied to thermally sprayed coatings with enhanced properties. One example came from the work by Watanabe et al., who reported that the optimal WC–Co/Cu multi-layer composite coatings achieved 2-fold higher work of fracture and a comparable flexural strength over the monolithic WC–Co coating [42]. However, the studies on Fe-based amorphous–matrix coating with laminar structures have yet to be reported.

In this paper, using a similar structural design concept, we have developed a few Fe-based amorphous composite coatings with a laminar structure via the high-velocity oxygen fuel (HVOF) thermal spraying technique. The resulting coatings comprise thick amorphous layers with alternate thin, adhesive and oxidation-resistant NiCrAl crystalline layers [21,43]. It will be demonstrated that the laminar coatings display pronounced improvement in mechanical properties, including interfacial toughness, bonding strength and impact resistance over the monolithic amorphous coating. The underlying mechanisms of the enhancement of the properties in the composite coatings are discussed on the basis of structural observation, high-temperature wetting measurements and finite element simulation.

## 2. Experimental procedure

### 2.1. Preparation and structure characterization of coatings

The  $\text{Fe}_{48}\text{Mo}_{14}\text{Cr}_{15}\text{Y}_2\text{C}_{15}\text{B}_6$  amorphous powder produced by high-pressure inert gas atomization with a size range of 33–55  $\mu\text{m}$  [44], and a commercial Ni–(16–22 wt.%) Cr–(3–7 wt.%) Al powder (hereinafter NiCrAl) with an average size of <45  $\mu\text{m}$  was used as the feedstock powder. All coatings were fabricated by means of HVOF thermal spraying in open air, using the same spraying parameters as described in our previous work [44]. AISI 1045 mild steel plate with dimensions of 100 mm  $\times$  100 mm  $\times$  6 mm was used as the substrate, which was mechanically polished, degreased by acetone and subsequently grit-blasted prior to thermal spraying. Two types of laminar coatings with the same ratio of NiCrAl layers (18 vol.%) were synthesized, i.e., four-layered coating (two NiCrAl layers + two amorphous layers) and six-layered coating (three NiCrAl layers + three amorphous layers). In all coatings, the NiCrAl layer was first sprayed on the steel substrate to get a good bonding strength and the outermost layer was an

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