Journal of Alloys and Compounds 505 (2010) 348–351

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

Microstructure and mechanical properties of PbSn alloys deposited on carbon fiber reinforced epoxy composites

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ARTICLE INFO

Article history:
Received 3 December 2009
Received in revised form 29 May 2010
Accepted 1 June 2010
Available online 18 June 2010

Keywords:
Plasma spraying
PbSn alloys
Microstructure
Carbon fiber reinforced epoxy composites

ABSTRACT

In this paper, PbSn coatings were deposited on carbon fiber reinforced epoxy composites (CFRE composites) by means of plasma spraying with four groups of processing parameters (g1, g2, g3 and g4). The microstructure and phase composition of the as-sprayed coatings were examined by scanning electron microscopy (SEM) and X-ray diffraction (XRD), respectively. The bonding strength of the coatings to the substrates was detected on a RGD-5 tensile testing machine. It was indicated that high melting extent of the coating, strong bonding between the coatings and the substrate, as well as low porosity could be achieved with the g3 parameters. However, the phase composition and the content of crystalline phases of PbSn coatings varied slightly with different processing parameters.

1. Introduction

Lightweight characteristics in spacecraft operations are highly desirable for cost and emission reductions. Since the late 1980s, there has been substantial interest in developing and testing metal lined composite-overwrapped tanks for launch vehicle and spacecraft applications. Now, more and more aircraft, launch vehicle, and spacecraft systems are requiring composite tanks for chemical or fuel storage [1–3]. Current state of the practice for such tanks includes metal liner and composite-overwrapped layer structures. The thickness of metal liner lies in the range of 0.5–1 mm. In order for the next generation of space vehicles to be affordable, it is critically important to achieve a significant reduction in their structural weight thus reducing the cost of launching payloads into space. Composite tanks with metal coating as liner are being considered for these applications because of their potential to increase mission capabilities and lower production costs. The thickness of metal coating can reach 0.1–0.2 mm. These tanks are projected to offer up to 25% weight reduction compared to current conventional metal lined tanks, allowing increased chemical storage volume and/or reduced total system mass [4–7].

Plasma spraying is a flexible and cost-effective technique to manufacture metal coatings. The feedstock particles are melted and accelerated towards a substrate by a plasma gas stream. The molten droplets are flattened upon impacting on the substrate and quenched, thus forming thin layers or splats. The interbonding between overlapping splats, deposited during repeated torch passes, generates a dense, adherent and homogeneous coatings. In the plasma spraying, the temperature in the middle area of plasma flame can reach 5000 K. The particle velocity lies in the range of 100–300 m/s. The process is simple and can be operated either manually or in an automated manner. It is possible to spray coatings on a wide range of materials including metals, alloys and ceramics [8–10].

In this work PbSn coatings were deposited on carbon fiber reinforced epoxy composites (CFRE composites) by the plasma spraying technique. During the process of plasma spraying, source powders are melted in a plasma flame formed through the ionization of an inert gas such as Ar. The main motivation of this work lies to the fact that although CFRE composites are the most thermal resistant polymer matrix composites, 645.15 K is the upper limit of their working temperature for long time service. The sprayed particles with high melting point can cause degradation of the mechanical properties of CFRE composites, or even damage. Due to the low melting point of PbSn alloy (505 K) [11], the alloy was chosen to fabricate coatings on CFRE composites as metal liner and efforts were made to determine the plasma spraying parameters to prevent any pyrolysis and thermal damage in the substrates.

In recent years, in terms of coatings fabricated by plasma spraying, effects of processing parameters on coatings properties were extensively reported [12–14]. However, most of these papers concentrated on alloy coatings sprayed on metallic or ceramic substrates [15–17]. There was also investigation about HA coating deposited on carbon/carbon composites [18–20]. In many cases, researchers select one parameter and test its influence onto the coating properties. Plasma spraying, however, involves many

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doi:10.1016/j.jallcom.2010.06.067

Table 1
Plasma spraying parameters for PbSn coating.

Group	Current (A)	Spraying power (kW)	Spray distance (mm)	Plasma gas flow rate (l/min) Ar, H_2	Carrier gas flow rate (l/min)	Pressure of compressed air (MPa)
1	300	35	120	35, 9	2.9	0.8
2	300	40	120	35, 9	2.9	0.8
3	400	40	120	35, 9	2.9	0.8
4	400	45	120	35, 9	2.9	0.8

parameters and complex interactions between them. In a more careful approach, one should take into account a few, most significant, process parameters. This way was chosen in a study by Heimann et al. in which a statistical plan of experiments was considered including many process parameters such as current, electric power, plasma forming gas composition and flow rate, carrier gas flow rate, and spraying distance. These authors tested, resulting from the spray experiments, phase composition, adhesion, and porosity of their deposits [21,22].

In order to optimize the current and the spraying power, PbSn coatings plasma sprayed on CFRE composites using four groups of processing parameters (g1, g2, g3 and g4) were investigated in this work. The aim is to investigate the effect of the plasma spray process parameters on the microstructure, phase composition and bonging strength of the coatings to the substrates.

2. Experimental

Experiments were performed with Pb30 wt.%Sn alloys prepared from 99.8%Pb and 99.7%Sn. CFRE composite laminates having dimensions 5 mm × 10 mm × 10 mm were used as substrates and were prepared prior to PbSn deposition according to the following steps: (1) chemical cleaning with acetone to remove grease or moisture, and (2) grit blasting with 46-mesh corundum powders in order to create an anchor tooth profile at the surface of the substrate [23].

R-750C plasma spray system made by Plasma Tech. Co. Ltd. from Switzerland was used to spray the powder to produce the coating. In order to optimize the plasma spray processing parameters, four groups of parameters (g1, g2, g3 and g4) were designed to prepare the coatings at a spraying distance of 120 mm. The four groups of parameters of the plasma spray processes were shown in details in Table 1.

The as-sprayed coating was characterized with bonding strength measurements, scanning electron microscopy (SEM), and X-ray diffraction (XRD). Bonding strength of the coating to the substrate was determined using an INSTRON-5566 tester, according to National standard GB8642-88. A coated sample was glued to a steel plate, with the coating facing the steel plate. The sample was clamped onto an INSTRON-5566 tester and was pulled with the speed of 0.5 mm/s till it failed. The bonding strength was calculated according to $\sigma_p = P/S$, where σ_p was the bonding strength, P was the maximum load applied before the sample failed, and S was the area of the coating glued to the steel plate. When the sample failed at the substrate/coating interface or within the coating, the data were valid. If the sample failed at the coating/glue or glue/steel plate interface, glue with higher strength must be used. Three samples were tested for each data point [24].

SEM was performed using a 20 kV JEOL 840A microscope equipped with an OXFORD ISIS 300EDS analyzer and the necessary software, both the surface and the cross-section of the coating were examined. For the examination of the cross-section, the samples were cut and mounted in bakelite and then polished up to 4 μm alumina emulsion.

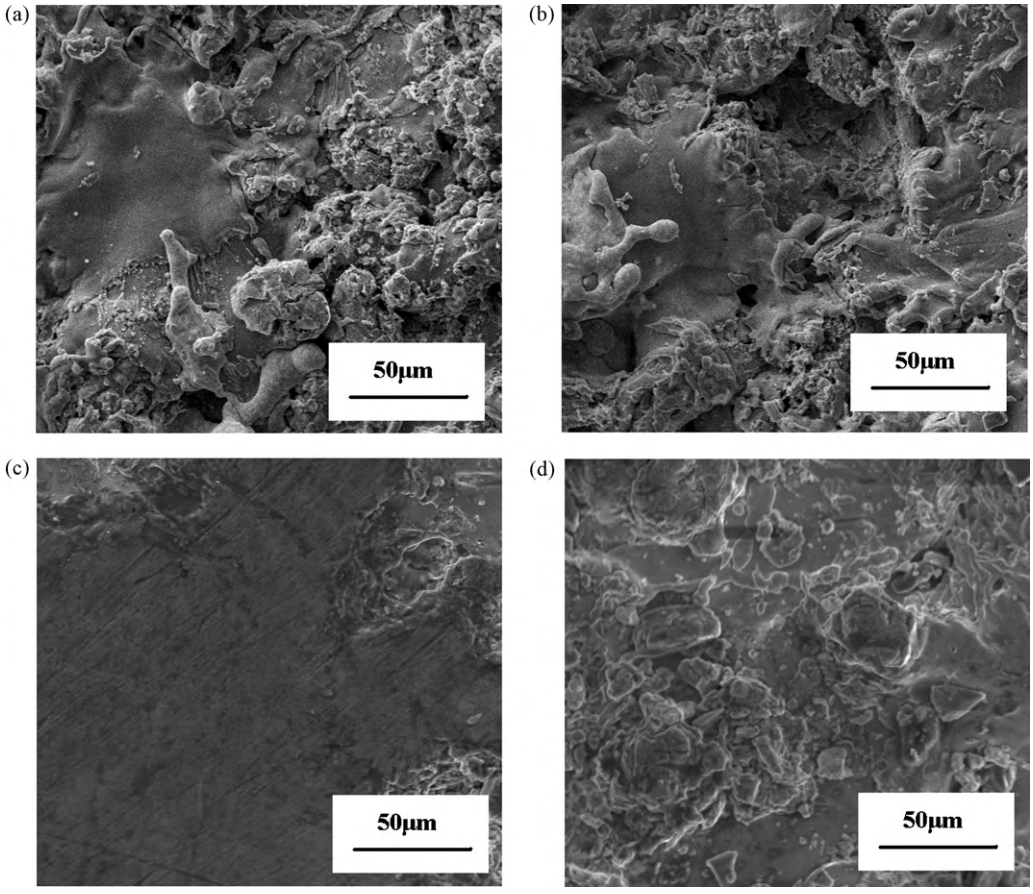

Fig. 1. SEM micrograph of the surface of the coating with four different spraying parameters (a: g1, b: g2, c: g3, d: g4).

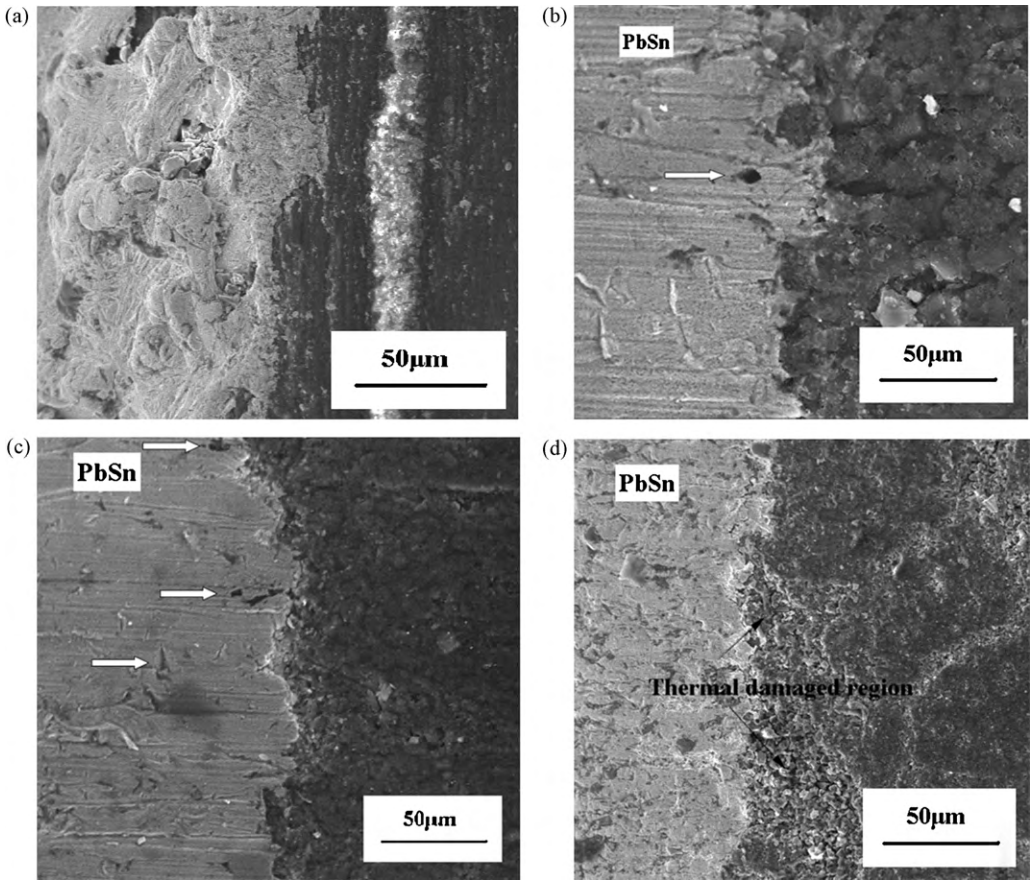

Fig. 2. SEM micrograph of the coating with four different spraying parameters (a: g1, b: g2, c:g3, d:g4). The arrows indicate pores in the coating.

X-ray diffractometry was used to assess the phase composition of the plasma-deposited coatings. A Philips X'pert MRD Diffractometer was used. The sample was scanned by a nickel filtered CuKα ($\lambda = 1.54186$ Å) radiation at a scan rate of 4°/min.

3. Results and discussion

The appearances of the surface of the coatings with different parameters are presented in Fig. 1. On the surface of coating sprayed with g1 parameters, some particles remain unmelted or partially melted as seen in Fig. 1a. The coatings are composed of tightly adhered irregular splats with pores and microcracks. There are some particles that were solid prior to contact with substrate and were disintegrated on impact, indicating that the parameters of g1 could not provide sufficient energy to melt all particles, which results in a non-uniform microstructure in the coatings. The porosity is relatively high and the surface is obviously characterized by high roughness. When sprayed with g2 parameters (Fig. 1b), more particles were completely melted, and well-flattened splats were produced. Almost all of the PbSn powder were totally melted or super-melted and joined each other with g3 and g4 parameters (Fig. 1c and d), and the flattened splats and non-uniform particles (which suggests that some melted particles could have sparked after they reached substrates due to the excessive plasma energy input) can be clearly seen. This is not peculiar, if we take into account the growth method used. Since the coating is formed through the impact of high velocity droplets of molten metal powders on the surface of the substrate, it is impossible to obtain an even surface. This phenomenon becomes more intense because part of the powder is totally or partially solidified before reaching the substrate or the already formed coating. Furthermore, since the freezing time of the deposited particles is very short, the liquid droplets are not likely to encounter a liquid surface. As a result, the surface is obviously characterized by low roughness.

The microstructures of the cross-section of the PbSn coating on CFRE composite substrate at different parameters are showed in Fig. 2. The cross-sectional view of the coating offers more information. It can be seen that the joint of the coating and substrate is not good enough. Moreover, microcracks and delamination are seen in the interface between the coating and substrate (Fig. 2a). When sprayed with g2 parameters (Fig. 2b), the coating is characterized by low integrity but there are less microcracks and delamination in the interface. It is obvious that the coating sprayed with g3 parameters is characterized by high integrity. Especially along the interface of the coating and the substrate neither cracks nor delamination is observed. Therefore good coating adhesion is expected. Nevertheless, far from the surface of the coating, certain pores are detected, which are noted with arrows in Fig. 2. In any case the pores seem to be unavoidable because during thermal spraying the droplets that deposit at the substrate could be semi-molten or even solid and they collide with a solid surface. As a result it is likely that the droplets do not exactly fit the already formed pattern and hence pores are formed around their contact point.

It is clearly from Fig. 2a–c that the quality of the coating increased as the power and current increasing. It is understandable that at high power and current level particles are in good molten state, impinging with higher velocities on the substrate. Hence, uniform and dense microstructures are obtained at such conditions. At lower power and current, not only do the particles get cooled but also their velocities are reduced. This will lead to the greater number of unmelted particles resulting in high porosity and microcracks.

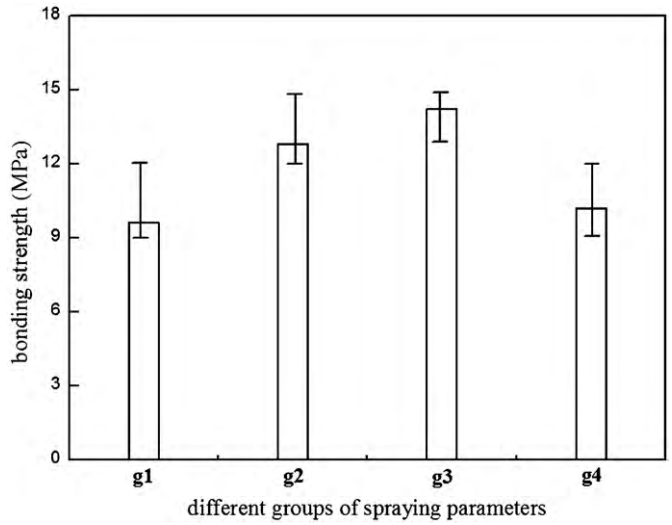

Fig. 3. Bonding strength of Pb–Sn coatings-CFRE substrates with four different spraying parameters.

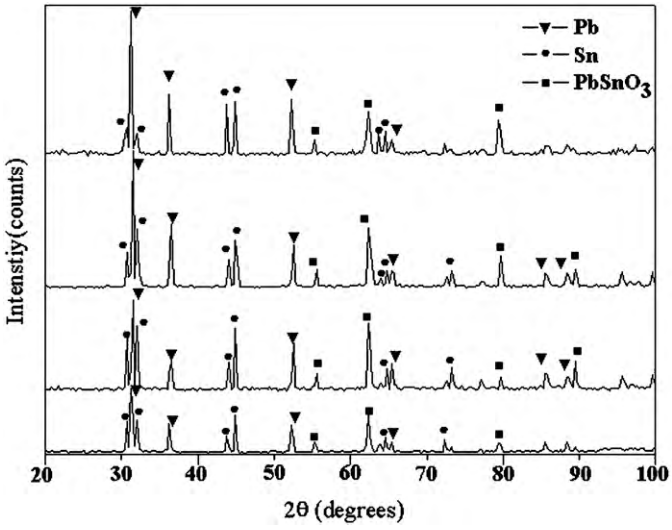

Fig. 4. X-ray diffraction spectra of PbSn coatings with four different spraying parameters (▼: Pb; ●: Sn; ■: $PbSnO_3$).

Fig. 2d showed the cross-section of the PbSn coating on CFRE composite substrate with g4 parameters. No cracks and delamination is found but a thermal-damaged region with broken fiber is found between the substrate and PbSn coating, indicating that the parameters of g4 provide excessive plasma energy to spark some melted particles so as to ablate the carbon fiber. The thermal-damaged region will influence the bonding strength of PbSn coating.

Fig. 3 shows the bonding strength with different groups of spraying parameters. In the figure, as expected, the coating sprayed with g3 parameters has the highest bonding strength of 14.2 MPa. The coating sprayed with g4 parameters only has the third higher bonding strength of 10.2 MPa, which was lower than that of the coating sprayed with g2 parameters, 12.8 MPa. While, the coating sprayed with g1 parameters has very low bonding strength of 9.6 MPa.

XRD patterns for as-sprayed coatings corresponding to g1, g2, g3, g4 parameters are given in Fig. 4. It can be seen that the coatings contain many kinds of crystalline phases. The crystalline phases are primarily a mixture of cubic Pb, tetragonal Sn and a little monoclinic $PbSnO_3$. The intensity of the Pb and Sn peaks corresponding to g1 and g2 parameters are lower than that of peaks corresponding to g3 and g4 parameters. However, the content of crystalline Pb and Sn change slightly with the processing parameters. It can be explained as follows: the crystalline PbSn coatings is arisen from partially melted starting particles determined by the plasma energy input and recrystallized phase determined by the temperature of coatings and substrates during spraying. With a spraying power of 35 kW, most of the particles are partially melted, and the lower temperature of the plasma flame causes the coatings and substrates cooler, which reduces the content of recrystallized Pb and Sn. On the contrary, with the increase of spraying power from 35 to 45 kW, the higher temperature improves both the recrystallization of Pb and Sn and the decomposition of melted particles.

4. Conclusions

The examination of the PbSn coatings sprayed onto CFRE composite laminates with four groups of the plasma spray processing parameters revealed that, PbSn alloy coatings could be fabricated on CFRE composites successfully with g3 parameters. The XRD patterns of coatings indicated that the coatings were composed of crystalline Pb, Sn and a little $PbSnO_3$. The spraying parameters had little effect on the content of crystalline Pb and Sn phase, whereas the melting extent of coating particles and the amount of decomposed phases were severely influenced by the spraying parameters. Moreover, the mean bonding strength of the PbSn coatings-CFRE composite substrates increased from 9.6 to 14.2 MPa as the parameters varied from A to B. Results indicated that the best group of spraying parameters among the tested conditions was g3, in which carbon fiber bundles bonded well with PbSn coatings and the bonding strength was accordingly enhanced.

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