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# Microstructure and wear resistance of steel matrix composite coating reinforced by multiple ceramic particulates using SHS reaction of Al–TiO<sub>2</sub>–B<sub>2</sub>O<sub>3</sub> system during plasma transferred arc overlay welding

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#### ABSTRACT

The steel matrix (SHS-free) coating and its composite (SHS-produced) coating reinforced by multiple ceramic particulates were developed by plasma transferred arc (PTA) overlay welding. 5% and 10% weight percentages of mixtures of aluminum (Al), titanium dioxide (TiO<sub>2</sub>), and boron oxide (B<sub>2</sub>O<sub>3</sub>) powders by sequence weight ratio of 9:8:7 were used as precursors. Aluminothermic reduction of these oxides, being highly exothermic in nature, essentially leads to a self-propagating high-temperature synthesis (SHS) of multiple ceramic particulate reinforced steel matrix composite coatings. Composite coatings have been subsequently characterized by X-ray diffraction (XRD), optical microscopy, scanning electron microscopy (SEM) with energy dispersive spectroscopy (EDS) attachment, transmission electron microscope (TEM), nanoindentation, and sliding wear measurement. The results show that the hypoeutectic microstructure exists in the steel matrix coating where it consists of  $\gamma$  (Fe, Ni),  $M_7C_3$ , and (Fe, Cr)<sub>2</sub>B phases. Adding mixtures of Al–TiO<sub>2</sub>–B<sub>2</sub>O<sub>3</sub> by sequence weight ratio of 9:8:7 changes its microstructure into pseudoeutectic characteristic whose crystal growth is cell dendrite in 5% SHS-produced coating but dendrite in 10% SHS-produced coating. Not only the metastable Al<sub>2</sub>O<sub>3</sub> with nanometer and TiB<sub>2</sub>, but the TiB and TiC can be formed in the SHS-produced coating, except for those phases that existed in the steel matrix coating. Hardness and sliding wear resistance of the SHS-produced coatings increase in comparison with that of the steel matrix coating. The best sliding wear resistance can be obtained in the 5% SHS-produced coating for its high ratio of hardness to elastic modulus. © 2013 Elsevier B.V. All rights reserved.

#### 1. Introduction

Metal Matrix Composites (MMCs) commonly have a good combination of hard ceramic reinforcements and ductile metallic matrix, which make them a promising candidate for being used in wear resistance applications [1]. Among various techniques, *in situ* has been expected to be one of the best techniques to produce these high-performance ceramic reinforced MMCs in view of its outstanding interfacial compatibility between matrices and reinforcements, thermodynamically stable and distribution uniformity of the reinforcements in the matrix [2]. During the past few decades, self-propagating high temperature synthesis (SHS) technology, as one of the fabrication techniques to synthesize *in situ* ceramic particulate reinforced MMCs, has attached much attention because of its low energy consumption and highly product purity [2–4].

However, one of the drawbacks of conventional SHS reactions for industrial application is that the final products might be highly porous [5–7]. Therefore, this process must be combined with a densification step, such as hot-pressing, extrusion, quasi-isostatic pressing, shockwave compaction [8], or casting routes [7]. Unfortunately, such combing procedures are expensive and time consuming to produce a bulk

composite, especially due to the fact that wear mainly occurs on the surface of components. Thus, surface modifying technology appears as an alternative way to extend the application of MMCs by forming a protective composite coating on the surface of the substrate material. In the previous work, microstructures, hardness and wear resistance of ceramic particulate reinforced MMC coatings by means of SHS reaction during overlay welding were investigated [9-11]. However, most of the work thought the thermodynamic behavior in synthesizing ceramic particulates by SHS reaction during nonequilibrium overlay welding followed the same rule with that of the equilibrium solidification procedure [9–11]. Few efforts have been focused on the effects of the matrix materials, such as steel matrix alloy in which Fe, C, Cr, Ni, and B elements might be included, on the characteristics of ceramic particulates synthesized by SHS reaction during the nonequilibrium solidification process. Besides, little work has been paid to the influence of the addition of the precursor particulates on the matrix microstructure of the coating reinforced by ceramic particulates produced using SHS reaction during overlay welding. It is obvious that the characteristics of the phase and microstructure have an important effect on the wear resistance of the deposited coatings.

Therefore, this paper focused on the influences of the matrix material and the contents of the precursor particulates on the microstructure, phase characteristic, and properties of the multiple ceramic particulate

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reinforced steel matrix alloy coating produced using SHS of Al– $TiO_2$ – $B_2O_3$  system during one of the overlay welding, plasma transferred arc, which is more commonly used on account of its extremely high temperature, excellent arc stability, high surfacing speed, high energy exchanging efficiency, low cost in equipment and operation [12–15].

#### 2. Experimental details

Plates of Q235A steel with dimensions of  $200 \times 30 \times 20 \text{ mm}^3$  whose composition is listed in Table 1 were used as substrates during PTA overlay welding. They were polished using emery paper and then washed with acetone to provide a clean surface. No heat treatment was taken before PTA overlay welding. Powder mixtures of Al (99.9% purity and ~10  $\mu$ m in diameter), TiO<sub>2</sub> (99.5% purity and ~2  $\mu$ m) and B<sub>2</sub>O<sub>3</sub> (99.5% purity and ~3  $\mu$ m in diameter) were used as the precursor materials. The weight ratio of Al:TiO<sub>2</sub>:B<sub>2</sub>O<sub>3</sub> was chosen as 9:8:7 based on the following reaction [5,16,17]:

$$10Al + 3TiO_2 + 3B_2O_3 = 5Al_2O_3 + 3TiB_2$$
 (1)

The standard Gibbs free energy change,  $\Delta G_T^0$  has been calculated using data shown in reference [18]. The thermodynamic calculation indicates that  $\Delta G_T^0$  value can be expressed as Eq. (2) in which T is the Kelvin [17]:

$$\Delta G_T^0 = -2761726 - 833T + 160T \ln T - 87T^2 + 27 \times 10^5 T^{-1} \text{ J/mol } (2)$$

It can be calculated approximately that  $\Delta G_{T}^{0}$  is negative within the temperature range of PTA overlay welding. Therefore, the reaction can occur with the evolution of heat. The first and foremost requirement for a reaction to become self-sustaining is that the reaction must be highly exothermic. It can be obtained from Eq. (2) that the reaction is highly exothermic within the temperature range of PTA overlay welding, indicating that the reactions would be self-sustaining.

In order to achieve a uniform distribution, precursor powders were mixed in a blender for 1 h within an argon atmosphere. The precursor mixtures then were added into a steel matrix alloy (referred to as SHS-free) powder by weight percentages of 5% and 10% (referred to as 5SHS and 10SHS, respectively), and then mixed in a blender for 1 h within an argon atmosphere to obtain the composite powder. The used steel matrix alloy powder whose nominal compositions are given in Table 1 was spherical in shape with 75–185 µm in diameter. The composite powder was further mixed with a proprietary organic binder to form a slurry and then was prepared on the Q235A steel substrates. Prefabricated alloy layers then were heated in an electric heat drier for 4 h at 100 °C to remove the moisture within an argon atmosphere. Therefore, uniform prefabricated alloy layers with an average thickness of 2.5 mm were obtained.

Deposited coatings were carried out using an L4-400PC overlay welding machine. Detailed PTA overlay welding conditions, as listed in Table 2, were optimized by dilution, formability, surface hardness and deposition thickness after several experiments. The PTA torch was held by oscillation of 25 times per minute above the specimens with a shielding gas of pure argon supplied.

The microstructure and compositions of the coatings on the crosssection were observed by optical microscopy and JSM-6490LV scanning electron microscopy (SEM) with energy dispersive spectroscopy (EDS)

**Table 1**Chemical composition of the substrate and steel matrix alloy powder (wt.%).

| Alloy                                  | С | Si           | Mn | В         | Cr | Ni         | Fe           |
|--|---|--------------|----|-----------|----|------------|--------------|
| Substrate<br>Steel matrix alloy powder |   | 0.30<br>4.42 |    | -<br>3.51 |    | -<br>10.31 | Bal.<br>Bal. |

attachment. A Rigaku D/MAX-3C X-ray diffractometer with Cu  $K\alpha$  radiation was used to analyze the phase compositions of the coatings using plate specimens. Details about phase characteristics in the coatings were investigated by H-800 transmission electron microscope (TEM) using thin-film samples. Techniques for preparing TEM samples are similar to those described in our previous works [19].

Hardness (H) and elastic modulus (E) of the coatings on the polished cross-section were evaluated using an MTS nanoindenter NanoXP<sup>TM</sup> instrument with a Berkovich diamond indenter by continuous stiffness measurement method with the capacity of controlling the loading about 100 micro-Newtons ( $\mu$ N). Forty indentations were made across the cross-section from near the substrate to near the surface of each coating. All experiments were performed at about 25 °C. E value was then calculated from the Oliver–Pharr model [20,21] considering the pile-up [22,23] and the results were averaged. The Poisson's ratio of the different deposited coatings was assumed equal to 0.30.

Sliding wear test was conducted on the deposited coatings using a ring-block wear testing machine (MM200, PR China). Specimens with  $30 \times 6.5 \times 6.5 \text{ mm}^3$  dimensions were ground using mesh 230–800 emery papers before the wear test. Then the obtained specimens were fixed on one metal fixture as the upper sample (block specimens). One 40 wt.% WC-strengthen nickel-based alloy coating (~1050 HV<sub>0.5</sub>) obtained by laser cladding was used as a ring (counterpart material, inner bore is 16 mm and external diameter 40 mm). The coated block specimens were worn by the ring as the rotation axis running. The test was performed under 49 N loads at room temperature for 1200 s without a lubricant. Weight losses of the deposited coatings were measured to be within 0.01 mg and the average value for each specimen was calculated using three data.

#### 3. Results and discussion

#### 3.1. Phase compositions and structural characteristics

Fig. 1a–c shows the XRD patterns obtained from the surface of the coatings with varying precursor powder mixtures of Al–TiO<sub>2</sub>–B<sub>2</sub>O<sub>3</sub> addition by sequence weight ratio of 9:8:7 in the steel matrix alloy. It can be found from Fig. 1a that the SHS-free coating is mainly composed of  $\gamma$ (Fe, Ni), M<sub>7</sub>C<sub>3</sub> (M = Fe, Cr), and (Fe, Cr)<sub>2</sub>B phases.

Comparing with the SHS-free coating, adding mixtures of Al–TiO<sub>2</sub>–B<sub>2</sub>O<sub>3</sub> (SHS-produced) by sequence weight ratio of 9:8:7 leads to the change of the XRD patterns greatly. Far more diffraction peaks for TiB<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> phases can be indexed apart from those phases existing in the SHS-free coating, confirming that TiB<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> could be synthesized in Al–TiO<sub>2</sub>–B<sub>2</sub>O<sub>3</sub> system by SHS reaction during PTA overlay welding, as shown in Fig. 1b and c. As reported by references [24–26], following aluminothermal reaction might take place during SHS of Al–TiO<sub>2</sub>–B<sub>2</sub>O<sub>3</sub> system:

$$Al + B_2O_3 \rightarrow Al_2O_3 + [B] \tag{3}$$

$$Al + TiO_2 \rightarrow Al_2O_3 + [Ti] \tag{4}$$

$$[Ti] + [B] \rightarrow TiB_2 \tag{5}$$

**Table 2**Parameters of the PTA overlay welding.

| Parameters              | Units     | Value   |  |
|-------------------------|-----------|---------|--|
| Transferred arc current | Α         | 210-225 |  |
| Transferred arc voltage | V         | 38-45   |  |
| Main gas pressure       | MPa       | 0.3     |  |
| Power supply            | g/min     | 58      |  |
| Oscillation rate        | times/min | 27      |  |
| Travel speed            | mm/min    | 110     |  |

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