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Research article

Sytentesis of Al-B_4C composite coating on low carbon steel by mechanical alloying method

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Abstract

In this study, the Al/B_4C composite coatings were fabricated on the steel substrate by mechanical alloying method. A novel coating method, mechanical alloying, was used to layer a Fe-Al/ B_4C composite coating on the surface of low-carbon steel. The coating process was performed in a planetary ball-mill using a milling time at 10 hrs, a ball-to-powder ratio of 10:1, and a ball-mill velocity of 200 rpm. During the mechanical alloying process, moving balls and 5 wt% B_4C particles and 95 wt% Al powder were impacted the substrate surface The coating thickness, morphology and cross-section microstructure of the composite coatings were investigated by Scanning Electron Microscopy (SEM). Experimental results showed that the coatings were formed on the surface of the low-carbon steel substrate by the formation of Al/B_4C composite.

Keywords: *Composites, coatings, mechanical alloying*

1. Introduction

Metal matrix composites (MMCs) have attractive mechanical/physical properties and enhanced elevated temperature capabilities. Likewise, aluminum matrix composites reinforced with ceramic particles have received considerable attention as they can be formed by standard metalworking practices [1]. Numerous aluminum matrix composite systems with discontinuous reinforcing phases such as B_4C, SiC, Al_2O_3, BN and AlN have been developed using different production techniques [2]. B_4C is the third hardest technical material after diamond and cubic boron nitride at room temperature [3]. Aluminum has a low melting point and is commonly used to increase the fracture toughness of the composite [4]. Al/B_4C composites have a potential to be used as armor materials in body protection, helicopters, military aircrafts and vehicles. For such kind of applications, mechanical properties such as hardness, bending strength, compressive strength, elastic modulus and fracture toughness need to be balanced carefully along with low density and a homogeneous microstructure. Moreover, aluminum and its alloys have a high corrosion resistance in atmospheric conditions in that thin and strongly adhering

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aluminum oxide layer provides a protection and slows down the electrochemical corrosion processes [5].

Many techniques have been developed to produce composite coatings, such as chemical vapor deposition (CVD), physical vapor deposition (PVD), electrophoresis, electro-deposition, and sol gel technique that combine chemical and physical (high energy ball-milling) methods [6]. The chemical composition of additives hardly remains unchanged due to the dissolution into the metal liquid or metallurgical reactions with the environment resulting in the deterioration of the toughness and crack resistance of MMCs [7]. In order to overcome these limits, in-situ synthesis of composite coatings by high energy ball-milling methods could be investigated as a new research area for potential industrial applications. In this method, mechanical alloying (MA), as a high energy ball-milling technique is a practical processing route for synthesis of metal powder and ceramics mixtures. When a mixture of powder and ceramic is processed by MA, part of the milled powders forms a coating film over the milling balls and on the inside wall of the container. It was recognized recently that this phenomenon could be utilized as a flexible method to composite coat on the surface of an object e.g. a plate attached to the wall of the milling container. The impacts of the milling balls activate and harden the surface. They also deliver and attach the powder particles and sometimes also initiate chemical interaction between target material and coating powder [8-12].

The purpose of the present work was to: (a) investigate and identify a new composite coating technique; (b) study microstructure, distribution of B_4C particles within the coatings as well as their surface roughness and micro hardness.

2. Experimental

A steel substrate (12x12x3 mm^3), Al2024 powders (d50: 75 μm) and B_4C (d50: 47 μm) were charged into the vial by maintaining a ball-to-powder ratio of 10:1. Al and B_4C powders were blended in ratios of 95:5. Milling was performed in a Fritsch Pulverisette 6 planetary ball mill in an argon atmosphere. The milling processes were performed at a speed of 200 rpm and milling times of 6, 12, 18, 24 and 30 hrs. The specimens were cut from steel substrate using a cutting machine before coating process. The heat-treatment was carried out in an argon environment to prevent oxidation of the coating. The samples were placed in the furnace, which was evacuated of air, filled with argon and heated gradually (5°C min^{-1}). The samples were held for 120 min at 600°C and then they were cooled down in the furnace to minimize the internal stresses. The coatings thicknesses, surface morphologies and the microstructure features of cross section of coated specimens were measured from all samples using a Zeiss EVO LS1O Scanning Electron Microscope (SEM). Vickers hardness was determined using a micro hardness tester (Innovatest 422D) with a load of 0.05 kg and an indentation time of 10s. The average of 3 indents was used to ensure acquisition of a reasonably representative value. All indents were kept away from porous locations. Surface roughness of the coated samples was determined by roughness tester MarSurf 1 MAHR at a high sensitivity setting. Five measurements were carried out from each group to obtain an average roughness value (Ra). The phase analysis of composite layer was evaluated by X-ray diffractometry (XRD) (Rigaku Corporation, Japan) using CuK_α radiation.

3. Results and Discussion

3.1. Morphology

During the milling, centrifugal forces are produced by the vials rotating around their own axes and by the support disk orbiting around the central axis. The forces are then exerted on the balls, powders and substrate to be ground. Since the vials and the supporting disk rotate in opposite directions, the centrifugal forces alternately change their directions. Under this condition, the balls are primarily carried up the inner surface of the vials and then run down the inside wall, producing a so-called "friction effect" on the inner wall. Subsequently, the balls, powders and substrate material being ground are propelled off the inner wall and travel freely across the inner chamber of the vial, colliding against the opposite inside wall. In this situation, another new mechanism, a so-called "impact effect", is developed during the interaction. The energy developed through the impacts is several times higher than that of conventional ball mills [12].

Two different kinds of coating mechanisms, i.e. friction effect and impact effect, act alternately on the vial and its contents. In particular, the impact effect during milling causes the powder materials and grinding balls to lift off and then travel close to the inner wall of the vial. It is assumed that at high content of Al and B_4C powders, coating can be mechanically alloyed on carbon steel substrate by means of the following three steps:

(i) Because of repeated collisions between the milling balls and the inner wall, the steel substrate surface suffered a large degree of plastic deformation and, thus, became activated. A fraction of the Al, consequently, easily transferred and adhered to the activated substrate surface. Subsequently the B_4C powders are embedded into the coated Al layers.

(ii) Further ball-substrate-ball collisions elevated the impact effect significantly. More and more adhered Al and B_4C powders were hammered together on the highly activated low-carbon steel surface, owing to the repeated action of cold welding. The present coating layer, meanwhile, became denser and smoother due to the incident fraction effect. Additionally, the impact effect initiated the diffusion of Fe atoms from the substrate to the interior of the coating.

(iii) At prolonged milling time, sufficient inter-diffusion and alloying between individual atoms. [13,14].

The initial morphology of the B_4C particles is angular or polygonal in shape while the Al powders has a distribution of ligaments or irregular shapes. The size of the B_4C particles was smaller than Al powders. Mean average particle size of Al and B_4C powders were measured as 75 μm and 47 μm, respectively. Fig. 1 showed that the microstructural characteristics of coating layer (packing, embedded and B_4C distribution) were significantly influenced by the applied milling times and Fig. 2 shows the development of the Al/B_4C composite coating structure as a function of the milling time. The Al/B_4C composite coating formed very rapidly when the milling time reached 6 hrs (Fig. 1a). Al particles were soft and their tendency to weld on the steel substrate was high in this stage.

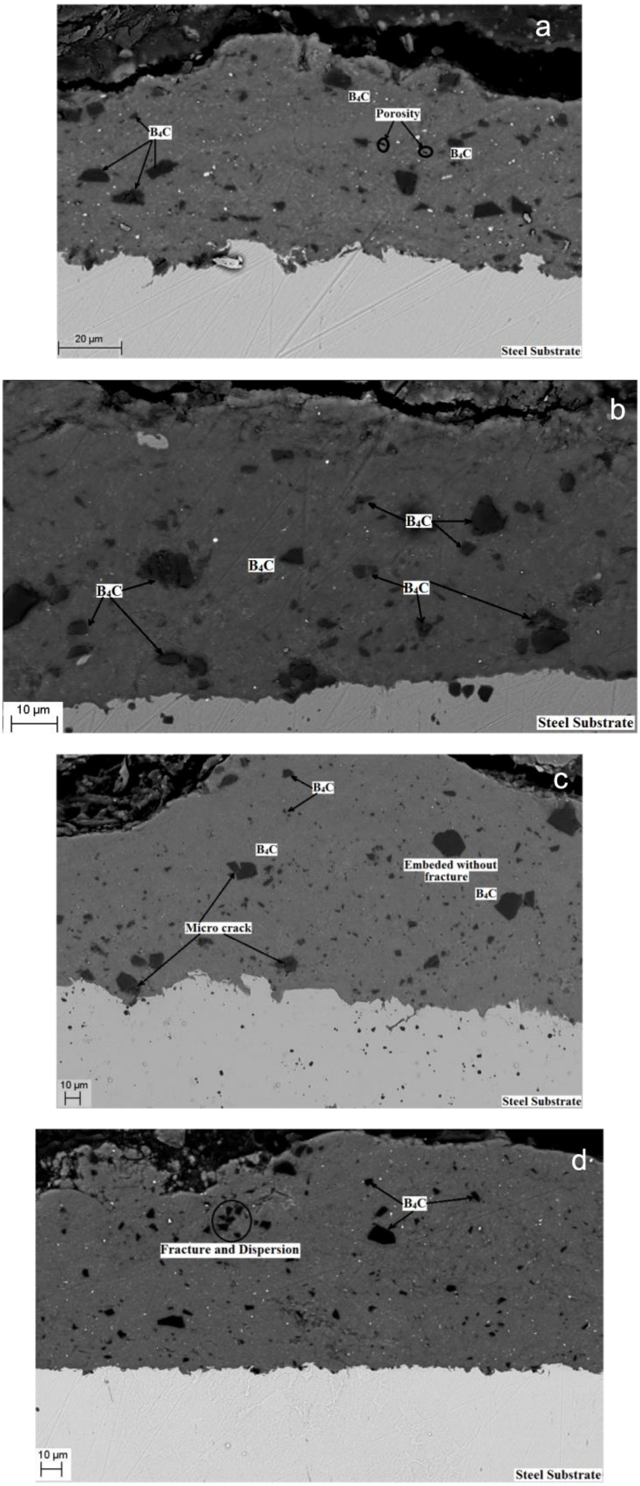

Fig. 1 SEM images of the composite coating layer; (a) 6 hrs (b) 12 hrs (c) 18 hrs and (d) 30 hrs

It should be noted that distribution of B_4C particles was not homogeneous and coating layer included embedded B_4C particles which were fractured and unfracture in the initial stages of coating process. Major composite coating characteristics in this stage were high porosity, the very roughness surface between coating and substrate interface and coarse B_4C particles. As shown in Fig. 1b, an increase in the number of fractured B_4C particles increased the embedded B_4C particles. Moreover, continuous impact effect occurred by ball-substrate-ball was an important factor in reducing the porosities in the coating layer. In this stage, milled powders still showed ductile behavior and because of the heavy plastic deformation experienced by powder particles during milling, particles tend to get cold-welded to each other, especially for the ductile Al powders having a ductile nature. As shown in Fig. 1c, the amount of B_4C in composite coating layer continued to rise with the increasing milling time from 6 to 12 hrs. The formation of micro cracks on the B_4C particles began in this stage due to the increase in ball-substrate-ball collisions. This increase can be attributed to the decrease in the number of coated ductile Al powders. With increasing the milling time to 18 hrs, the mechanically alloyed coating had a significantly denser structure. As shown in Fig. 1d, smooth substrate surface and homogeneously distributed B_4C powders were the major characteristics of these coatings in the final stage.

The weight of coating layer increased from 9 to 55 mg with increasing the milling time from 6 to 30 hrs (Fig. 2) with a homogeneous distribution of B_4C. Further, the amount of B_4C particles embedded without fracture was low in the Al/B_4C composite coating layer in this stage. Also, it was observed that the amount of coarse B_4C particles decreased gradually from the coating to the substrate with increasing milling time and reached to minimum in the final stage. The porosities were filled with fractured B_4C particles. Very rough substrate surface were flattened by hard B_4C particles and upgrade ball impacts. It is known that the longer the milling times, the higher would be the energy input into the powder and milling balls. As a consequence, higher centrifugal forces are generated within the milling system, thereby intensifying the strength of impact effect. The porosity and surface roughness of Al/B_4C composite coating layer decreased with the increasing impact effect.

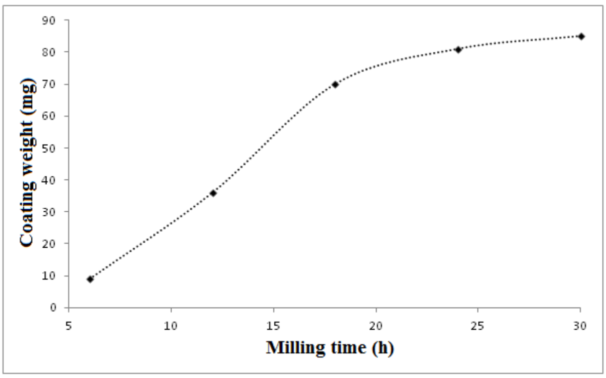

Fig. 2 The change of coating weight of as a function of milling time

The reaction between the low-carbon steel substrate and the Al_20_3 powder was examined by XRD. Fig. 3 shows the XRD pattern of the coating surface for 10 hrs of milling after heat treatment at 600°C. The XRD patterns confirmed the presence of both elemental Fe and Al and compounds such as Fe_3Al, Fe_2Al_5 and FeAl.

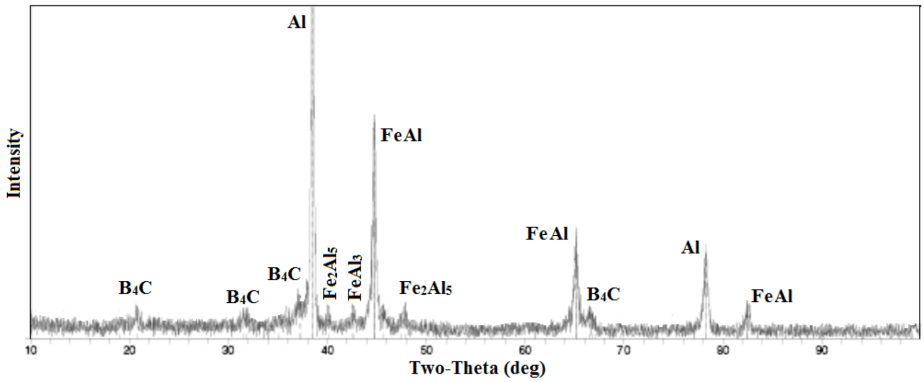

Fig. 3 XRD pattern after mechanical alloying (30 hrs) and subsequent heat treatment (600°C)

3.2. Surface roughness and hardness

Surface roughness (Ra) values of composite coatings at different milling times were shown in Fig. 4. The surface roughness was low at short milling time (6 hrs) and it increased to the optimum coating thickness with the increase in milling time. At a relatively low milling time of 6 hrs, formation of a heterogeneous coating surface consisting of flattened Al/B_4C composite powders which produced by cold welding with less inter-particle contact, was observed. It was found that the steel substrate could be covered with a very thin and non-homogeneous composite coatings layer in the initial stage of coating (6 hrs) with a high surface roughness. The surface roughness of the composite coating at the end of 12 hrs was higher than that for 6 hrs of milling due to the increase in thickness of the coating. After 12 hrs milling the MA processed coating became denser and was free of any apparent pores. By increasing the milling time, the surface roughness was decreased due to ball-substrate-ball collisions. Interestingly, on further milling up to 24 hrs, a smooth, highly consolidated, and fully dense coating surface was obtained by excess ball impacts. Surface roughness decreased from 10.2 to 3 μm when milling time was increased from 12 to 24 hrs for Al/B_4C composite coatings.

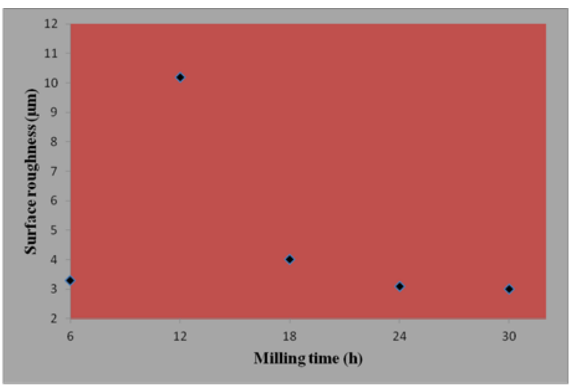

Fig. 4 The surface roughness variation as a function of milling time

Fig. 5 shows the Vickers micro hardness values as a function of milling time for the Al/B_4C composite coatings. The distance was determined from the interface of the coating and substrate to study the effect of milling time on the hardness along the depth in the substrate. The micro hardness of all coating layers was higher than the steel substrate ones because of the existence of B_4C particles and Fe-Al intermetallic. The average hardness of 5, 15, 25 and 40 µm away from the substrate surface in the composite coatings were in the range of 220, 205, 180 and 160 Hv for 6 hrs, respectively.

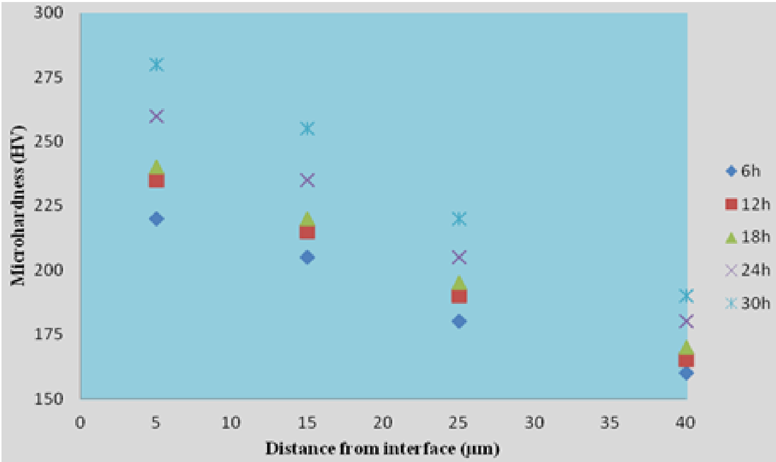

Fig. 5 The micro hardness (HV) variation as a function of milling time

The value of micro hardness was decreased towards coating surface from substrate. The highest hardness (280 Hv) was obtained at 30 hrs. Low porosity, increasing B_4C content in the coatings and homogeneity distribution of fractured B_4C were the major reasons for the increase of hardness at 30 hrs. Other reasons for this increase can be listed as below;

- The refined structure of substrate and coating as a result of the impact of ball-steel substrate-ball collisions (work hardening). This can be attributed to the higher impact force caused by ball-powder-ball and ball-steel substrate-ball collisions.
- The enhanced grains in surface of substrate and coating.
- The existence of Fe-Al intermetallic compounds and B_4C in the composite coatings [15].

4. Conclusions

The some important remarks can be given below:

- Ball-substrate-ball collisions produced a composite coating consisting of Al as matrix and B_4C as reinforced material, then the structure was steadily refined and consequently the inter-layer spacing decreased. This led to a decrease in the porosity in the composite coating layers.
- Heat treatment process provided the formation of Fe-Al intermetallic which increases the hardness of composite coating layer. Heat treatment of the green samples coated Al/B_4C composite led to the ordering of the Fe(Al) solid solution and its transformation to Fe-Al ordered phase.
- With increasing the milling time, the coating weight and hardness of the Al/B_4C composite coating increased due to work hardening of coating layer and homogenous distribution of B_4C particles.

- The hardness of the Al/B_4C composite coating at a distance of about 5 µm from the interface between the coating and substrate is nearly 1.5 times than that of the Al/B_4C composite coating at a distance of about 40 µm. The surface hardness of composite coating layer from 6 to 12 hrs of milling then decreased between from 12 to 24 hrs and remained nearly constant to 30 hrs.

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