

Structure evolution of Fe-50%Al coating prepared by mechanical alloying

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Abstract. The deposition of Fe-50%Al coating (in at%) on low carbon steel was successfully prepared by using mechanical alloying (MA). The coating process was performed in a shaker mill with variation of milling times from 30 minute to 180 minutes. The deposited coating was then heat treated at 600°C for 2 hour in a vacuum furnace of 5.6 Pa. The structure evolution of mechanical alloyed samples before and after heat treatment was investigated by scanning electron microscopy (SEM) equipped with energy dispersive X-ray spectrometer (EDX) and X-ray diffractometer (XRD). The results revealed that before heat treatment, the deposited coating on low carbon steel is composed of Fe and Al. The Fe(Al) solid solution was mostly formed after 180 minutes of milling time. Metallographic observation indicated that the surface of Fe-Al coating was rough and the coating thickness was increased with increasing milling time. Meanwhile, the heat treatment process led to structural evolution by forming FeAl intermetallic phase on the surface of low carbon steel.

1. Introduction

Recently, FeAl intermetallic compounds have become promising material for structural and coating applications. This is due to the fact that they have a good strength at intermediate temperatures, excellent resistance to oxidation and corrosion at high temperatures. In addition, the FeAl intermetallic compound is lighter than steels, so they are promising to substitute the applications of stainless steels at room temperature [1]. However, one of the main drawbacks of intermetallic compounds is very low ductility at room temperature and low mechanical strength above 600°C [1-3]. Key features of low ductility are restricted to cross-slip and intense planar slip. Reducing the crystallite size to the nanometers range or introducing fine hard incoherent particles to induce slip homogenization can improve their ductility [4]. Accordingly, modified coating materials as crystallite size reduction are required.

Mechanical alloying is one of the top-down techniques used for synthesis of nanomaterials. This technique is an effective way for preparing the nanocrystalline alloys. High energy ball milling becomes based-principle of the mechanical alloying methods to produce ultra fine and uniform powders. The aforementioned method involves deformation, fragmentation, cold welding and diffusion process, which forms refined powders with a homogeneous chemical composition [5].

Since firstly developed in 1960s by Benjamin [6], mechanical alloying was successfully used for preparation of intermetallics [2], extended solid solutions [7] and amorphous phases [8] of powders. A number of studies have been reported on the coating by mechanical alloying methods, such as Ti-Al coatings [9], Al coating [5], FeAl coating [10], TiN coating [11] and Cr coating [12]. The result showed that the mechanical alloying technique can be used to fabricate of thick multi-component structures with high bond ability on different substrates. However, there are limited studies on the Fe-Al coating with Fe-Al powders as starting coating material using a mechanical alloying method. In the



present paper, MA process was used to prepare Fe-50%Al coatings on low carbon steel. The effects of milling time and heat treatment on the coating structure were studied and discussed.

2. Experimental procedures

Fe-50%Al coating was deposited on low carbon steel plates using a mechanical alloying method. The approximate dimension of steel plate used in this study is $10 \times 8 \times 3 \text{ mm}^3$. Before coating process, the surface of the steel plates was polished using SiC paper for up to No. 1200 and cleaned with standard cleaning method to remove contaminants from the surface. After that, it was dried using air flush.

The commercial Fe (99,5% purity) and Al (99,5% purity) powders were used as raw material. In order to deposit Fe-Al coating on the surface of low carbon steel, the two steps of milling were carried out using high speed shaker mill with oscillation frequency of 700/min. First, Fe (50 at %) and Al (50 at %) powders were introduced into cylindrical steels vial with ball to powder ratio of 10:1 and mixed in an air for 1 hour. Second, the low carbon steel samples and mixed powder were charged into vials and then mechanical alloyed for several period of times as 0.5, 1, 2, and 3 hours. The heat treatment was carried out in a vacuum of 5.6 Pa at 600 °C for 2 hours. Then, it was cooled down in the furnace to minimize the internal stresses [5]. The morphology and coating thickness of Fe-50%Al coating was determined from the cross-sectional sample using a scanning electron microscopy (SEM-Hitachi High-Tech Co. Ltd, Japan). X-ray diffraction (XRD) (SmartLab-Rigaku Co. Ltd., Japan) was used to evaluate the phase structure of the coating layer.

3. Results and Discussion

Figure 1 shows the cross-sectional microstructures of Fe-50%Al coating on low carbon steels with various milling times. The surface of the Fe-50%Al is rough and the coating interface can be clearly distinguished. In addition, as shown in Figure 1, the thickness of Fe-50%Al coating is not uniform. The coating thickness increases with increase in milling time, see Figure 2. It is about 58% when the milling time increases from 0.5 h to 1 h. At longer milling time ($> 1 \text{ h}$), however, an increase in the coating thickness is less than 30%. Increasing milling time enhances the collision between ball, powder and substrate. This affects the deposition of the powder on the steel substrate. The powder particles trapped between the ball and substrate became cold-welding to the substrate surface [5], increasing the coating thickness [12]. Besides the change in the coating thickness, the evolution of Fe-50%Al coatings as function of milling time is also shown in Figure 1. The elongated lamellar with gray color can be observed in the microstructure of coating when the milling time was increased from 0.5 h to 2 h. In addition, the structure of the coating is dominated by large particle. Formation of the elongated lamellar structure is caused by the plastic deformation and cold-welding process during mechanical alloying [13]. The contrast different of the backscatter electron SEM images indicates their difference in chemical compositions. The region around the lamellar structure seems to be porous and density of the particles is low. The more uniform coating structure is accomplished after 3 h in milling time. This indicates that Fe and Al form mainly Fe(Al) solid solution. The result will be confirmed by XRD characterization and discussed latter.

Typical X-ray diffraction (XRD) patterns of the substrate and Fe-50%Al coating at varying milling times are shown in Figure 3. The diffraction peaks of Al (111) can be clearly seen at the milling time of less than 2 h. However, its reflection is likely to decrease with increase in milling time. This indicates that Fe and Al are not completely reacted. These results are in good agreement with the results of SEM characterization as shown in Figure 1. After 3 h of milling time, the Al (111) reflections completely disappear. Meanwhile, the reflections of Fe (110) becomes broadening and their intensity tends to decrease and shift toward lower than 2θ angles. This suggests that the Fe(Al) solid solution is formed by replacement of the separate Fe and Al phases [14]. The coating shows uniform microstructure and the elongated lamellar structure disappears.

Figure 4 shows XRD patterns of the surface of deposited Fe-50%Al coatings at various milling times after heat treatment at 600°C for 2 h. Structural evolution occurs after heat treatment. The peak intensities of Fe and Al shift to low angels as seen at diffraction peak of $2\theta = 30.88^\circ$. These peaks are

attributed to a new Fe-Al intermetallic phase, which is formed due to the transformation of Fe and Al [3]. The results reveal that all XRD pattern correspond to the peak of the FeAl (100); FeAl (110), FeAl (200) and FeAl (211) [ICDD-00-045-0983]. The reflections of Al, Fe, or oxidation product of Fe and Al are not detected in the XRD patterns. The FeAl intermetallic compound has a cubic structure with strong peak intensity of FeAl (110).

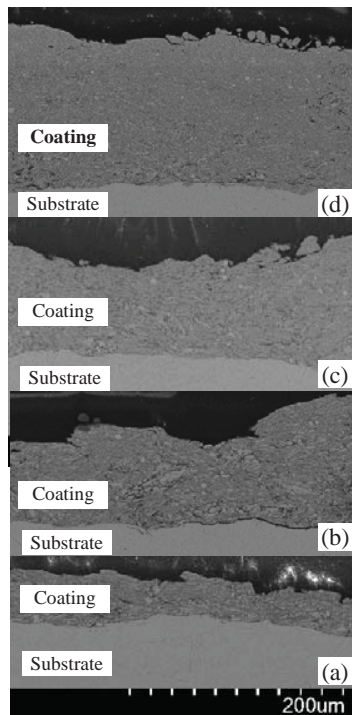


Figure 1. SEM images of the cross-sectional Fe-50%Al coatings with various milling times of (a) 0.5 h, (b) 1 h, (c) 2 h, and (d) 3 h.

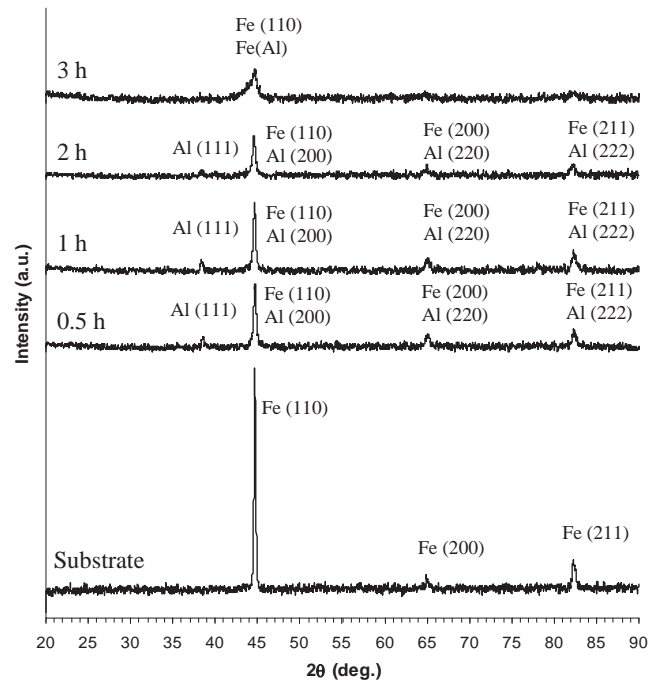


Figure 3. XRD patterns of Fe-50%Al coatings prepared at various milling times.

The full width half maximum (FWHM), crystalline size, lattice strain, dislocation density (ρ) and lattice parameter (a) of the FeAl (110) intermetallic are listed in Table 1. A change in the FWHM and lattice strain (ε) can be seen in the Table 1. This can be caused by the effect of change in the crystalline size, as reported by Krasnowski et al., [15]. In our study, the milling time and heat treatment are believed to have contribution in the structure evolution. As reported by Shi et al., [1], the lattice strain increase steady with increasing milling time and reach the final value of 1.7%. In the mechanical alloying, the generation and movement of dislocations cause plastic deformation which is believed to affect the lattice strain [16]. However, heat treatment after mechanical alloying in the intermetallic coating has a high affects in the evolution of crystalline size and lattice strain. Several researchers reported that the lattice strain decreases and crystalline size increases after heat treatment. Haghighi et al., [3] reported that the lattice strain of FeAl coating decreases from 2.2% to 1.2% with increasing the annealing temperature from 300°C to 700°C. Another result shows that the lattice strain decreases from 1.85% to 0.5% after heat treatment at 610°C [16]. In our study, the lattice strain of Fe-50% Al coating after heat treatment is approximately 0.18-0.27%. This result is smaller than the results of previous researchers. The mixing of Fe and Al powder before coating process seems to contribute in reducing the lattice strain. In addition, the duration time of heat treatment process in this study also greatly affects the diffusion process and reaction between Fe and Al in the coating. The

time is available for recovery and recrystallization processes. The appropriate time for heat treatment allows further ordering and reducing the defect from the mechanical alloying process as dislocation and severe plastic deformation. Reducing the crystal defects are characterized by a small value of dislocation density, 0.413×10^{16} - 0.620×10^{16} line/m².

Table 1. FWHM, crystalline size, lattice strain (ϵ), dislocation density (ρ) and lattice parameter (a) of FeAl (110) intermetallic compound.

Milling time (h)	Position 2θ	FWHM	Crystalline size (\AA)	ϵ (%)	ρ (line/m ²) $\times 10^{16}$	a (\AA)
0.5	44.160	0.290	5.16373	0.1787	0.413	2.9004
1	44.068	0.306	4.89214	0.1890	0.437	2.9061
2	44.148	0.435	3.44234	0.2682	0.620	2.9011
3	44.040	0.304	4.92384	0.1879	0.435	2.9079

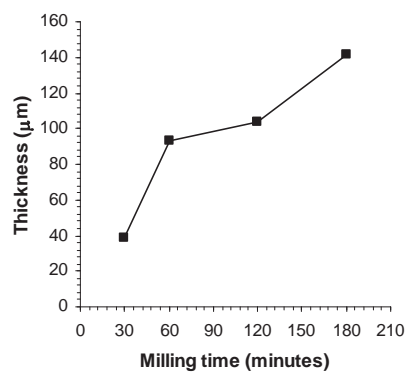


Figure 2. Thickness of Fe-50%Al coatings as function of milling times.

Table 2. Element compositions from the EDS-spots of the Fe-50%Al coating after 1 h milling time annealed at 600°C, 2 h.

Element	Spots-1 (at%)	Spots-2 (at%)	Spots-3 (at%)
Al	52.87	52.23	42.04
Fe	47.13	47.77	57.96
Totals	100	100	100

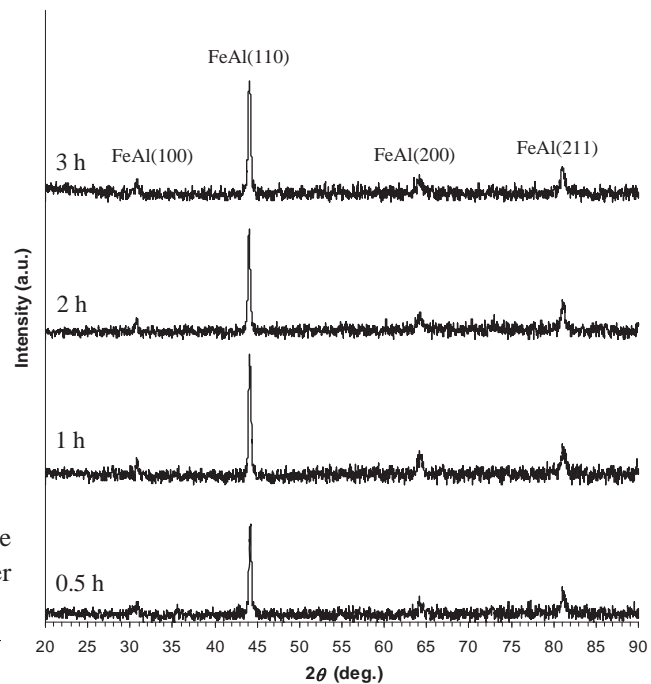


Figure 4. XRD patterns of Fe-50%Al coatings at various milling times after heat treatment at 600°C for 2 h.

Based on XRD characterization as presented in Figure 4 and Table 1, Fe-50%Al coating milled for 3 h and heat treated at 600°C for 2 h has a narrower FWHM compared to the other samples. This indicates that the crystallinity of sample is high. Table 1 also shows that the lattice parameter of the FeAl (110) in this experiment is lower than theory ($a = 2.9125 \text{ \AA}$ from ICDD-00-045-0983) with the variation difference in the range of 0.0046-0.0121 \AA . The decrease in the lattice parameter is probably due to the presence of compressive stress fields within the non-equilibrium grain boundaries inside of crystallite [17] during mechanical alloying process. In the heat treatment process, the movement of excess vacancies in the lattice leads to the ordering of the structure [3] and the reaction between Fe and Al to form FeAl intermetallic. This result is supported by the result of SEM-EDX analysis of

cross-sectional Fe-50%Al coating after heat treatment at 600°C for 2 h (as shown in Figure 5). It can be seen that Fe and Al are distributed in the coating. The composition of the FeAl layer from EDS point analysis is shown in Table 2. The ratio of Fe and Al are closed to atomic ratio of 1:1 (FeAl). This result is in good agreement with the result of phase characterization by XRD analysis which shows that the FeAl intermetallic compound was formed after heat treatment.

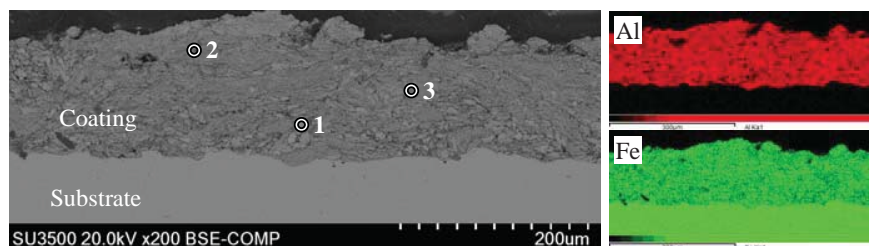


Figure 5. SEM and EDS elemental mappings images of the cross-sectional Fe-50%Al coating (1h milling time) after heat treatment at 600°C for 2 h

4. Conclusion

Fe-50%Al was successfully coated on the low carbon steel by using a mechanical alloying method. The coating thickness increased with increasing the milling time. The Fe(Al) solid solution was formed after mechanical alloying process for 3 h. Heat treatment process led to the the formation of FeAl intermetallic phase. These suggest that the structure evolution of the Fe-50%Al coating was affected by the milling time and heat treatment process.

Acknowledgment

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