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Formation mechanism of Fe_3Al and FeAl intermetallic compounds during mechanical alloying

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Fe and Al elemental powder mixtures with composition $Fe_{75}Al_{25}$ and $Fe_{50}Al_{50}$ were mechanically alloyed in a planetary ball mill under different conditions. The structural changes of powder particles were studied by x-ray diffractometery and scanning electron microscopy. Mechanical alloying of $Fe_{75}Al_{25}$ and $Fe_{50}Al_{50}$ first produced a fine Fe/Al layered microstructure which transformed directly to the corresponding intermetallic compounds, Fe_3Al and FeAl, with nanocrystalline structure at longer milling time. No intermediate phase, i.e. solid solution, was formed during mechanical alloying as a precursor to the intermetallic phase. The rate of mechanical alloying process was found to be dependent on milling variables such as rotation speed of mill, ball-to-powder weight ratio and number of milling balls.

1. Introduction

Fe_3Al and FeAl intermetallic compounds exhibit unique physical and mechanical properties including high melting temperature, high hardness, low density and good oxidation and corrosion resistance [1–3]. Furthermore the yield strength of these intermetallic compounds increases with increasing temperature up to 600°C [4]. This combination of properties has made Fe_3Al and FeAl intermetallic compounds potentially useful for many structural applications including gas-metal filters, heating elements, heat treatment fixtures, high temperature dies and molds and cutting tools [1–4]. A wide variety of methods including casting, powder metallurgy and self-propagation high temperature synthesis (SHS) and mechanical alloying (MA) are introduced for the synthesis of intermetallic compounds [1–4].

The materials transfer by diffusion of components during MA provides the means to synthesize intermetallic compounds. This solid state technique is especially useful for fabrication of those compounds that are difficult to prepare by conventional processes due to high vapour pressure and/or the large differences in melting points of components. Additionally, materials production can take place at room temperature which can have advantages over high temperature synthesis. It has been shown that in MA experiments not only the milling time but also further parameters such as the type of mill, the milling tool, the rotation speed of mill and the ball-to-powder weight ratio affect the kinetic energy of colliding balls and thereby have significant influence on phase transformations occurring during MA and therefore, on the final structure [5, 6].

MA of Fe-Al system has been previously reported, nevertheless there is still some uncertainty regarding the phase formation sequence occuring during MA. Oleszak and Shingu [7, 8] studied the MA behaviour of Fe-Al powder mixtures containing 10–85 at.% Al using a low energy ball mill. For alloys containing 10–50 at.% Al a disordered solid solution was formed. However MA of Fe-Al powder mixtures containing 60–85 at.% Al resulted in an amorphous structure. Bonetti *et al.* [9] studied MA behaviour of $Fe_{75}Al_{25}$ powder mixture in a high-energy ball mill. They observed that an extended solid solution of Al in Fe was obtained after 1 h of milling. No amorphization reaction occurred even after 32 h MA. Differential scanning calorimetry (DSC) upto 600°C (at heating rate of 20 $K\cdot min^{-1}$) showed that the Fe(Al) solid solution transformed to the disordered Fe_3Al intermetallic at ~380°C. The scope of this work was to study the development of Fe_3Al and FeAl intermetallic compounds during MA. The effect of process variables on the kinetics of the MA process was also investigated.

2. Experimental methods

99.8% pure Fe powder and 99.9% pure Al powder were mixed to give nominal compositions of $Fe_{75}Al_{25}$ and $Fe_{50}Al_{50}$ (at.%). Fig. 1 shows morphology of as-received Fe and Al powder particles. As seen the Fe particles had a nearly uniform size of ~300 μm with a

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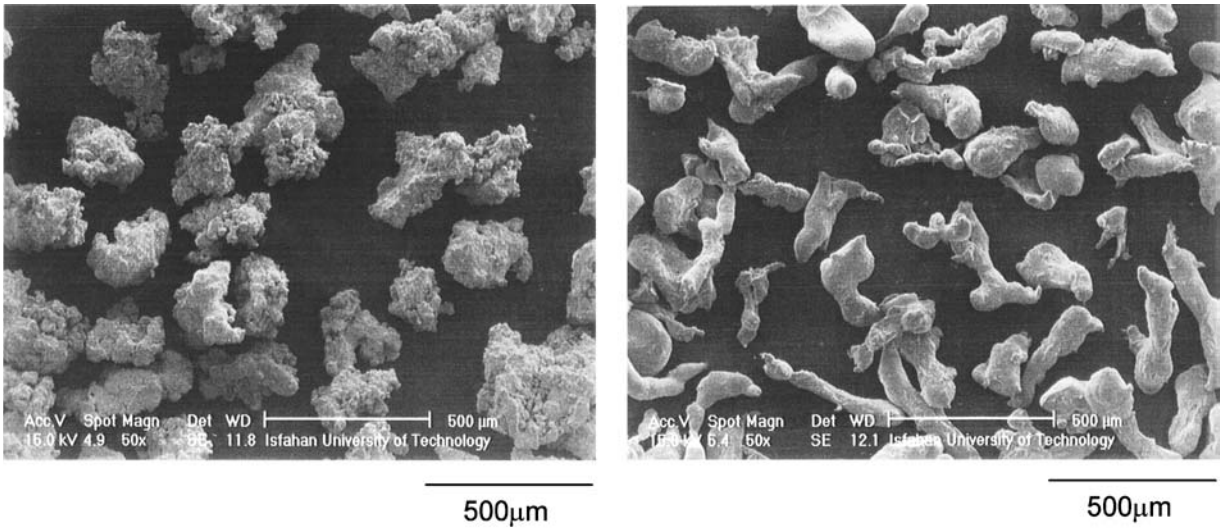
TABLE I Mechanical alloying conditions

Series	Composition	Ball-to-powder weight ratio	Rotation speed (rpm)	Number of balls	Diameter of balls (cm)	Weight of powders powders mixture (gr)
I	$Fe_{75}Al_{25}$	5.2	390	4	2	25
II	$Fe_{75}Al_{25}$	5.2	195	4	2	25
III	$Fe_{75}Al_{25}$	10.4	390	4	2	12.5
IV	$Fe_{75}Al_{25}$	5.2	390	32	1	25
V	$Fe_{50}Al_{50}$	5.2	390	4	2	25



(a) (b)

Figure 1 SEM micrographs of elemental: (a) Fe and (b) Al powder particles.

sponge morphology. The Al particles were irrgular in shape with a size distribution of 100–400 μm. Mechanical alloying was carried out, nominally at room temperature using a laboratory planetary ball mill under Ar atmosphere. Mechanical alloying was done under different milling conditions (Table I). Samples were taken at selected time intervals and characterized by X-ray diffraction (XRD) in a Philips X'PERT MPD diffractometer using filtered Cu K_α radiation ($\lambda = 0.1542$ nm) and scanning electron microscopy (SEM) in a Philips XL30 SEM. Isothermal annealing was carried out to study the thermal behaviour of milled powders. MA samples were sealed and then annealed in a conventional tube furnace. The structural transitions occurred during annealing were determined by XRD.

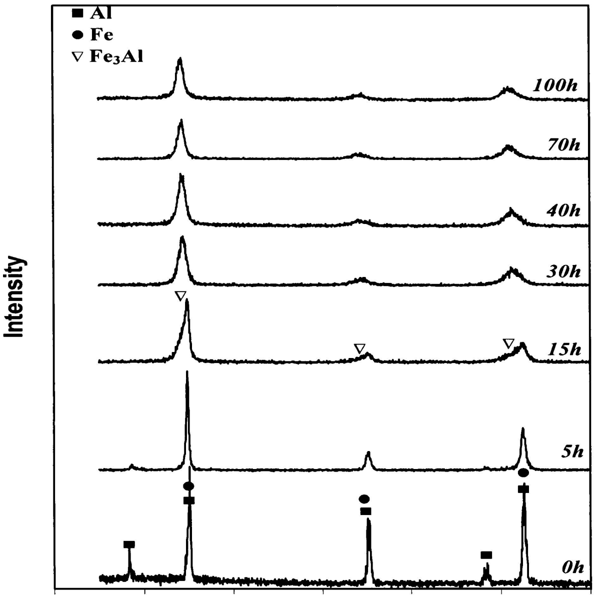


Figure 2 XRD traces from $Fe_{75}Al_{25}$ powder particles as-received and after different milling times (series I).

3. Result and discussion

3.1. $Fe_{75}Al_{25}$

Fig. 2 shows XRD traces for $Fe_{75}Al_{25}$ powders (series I) as-received and after different MA times. During MA the sharp crystalline diffraction peaks of the as-received powder broadened progressively with increasing processing time associated with accumulated internal strain and refinement of grain size. After ~15 h of milling time traces of Fe_3Al phase appeared on XRD traces. As MA proceeded, the fraction of Fe_3Al phase gradually increased, accompanied by a progressive decrease in intensity of crystalline Fe and Al diffraction peaks until XRD traces taken after 40 h MA suggested a completely Fe_3Al structure without any indication of additional phases. Increasing milling time to 100 h led to no further structural changes of powder. Lack of superlattice diffraction peaks suggests that the crystalline Fe_3Al phase has probably a disordered structure. The average Fe_3Al grain size after 40 h MA, estimated from broadening of XRD peaks using Scherrer's equation [10], was about 14 nm and appeared to remain almost unchanged upon further milling. The grain size

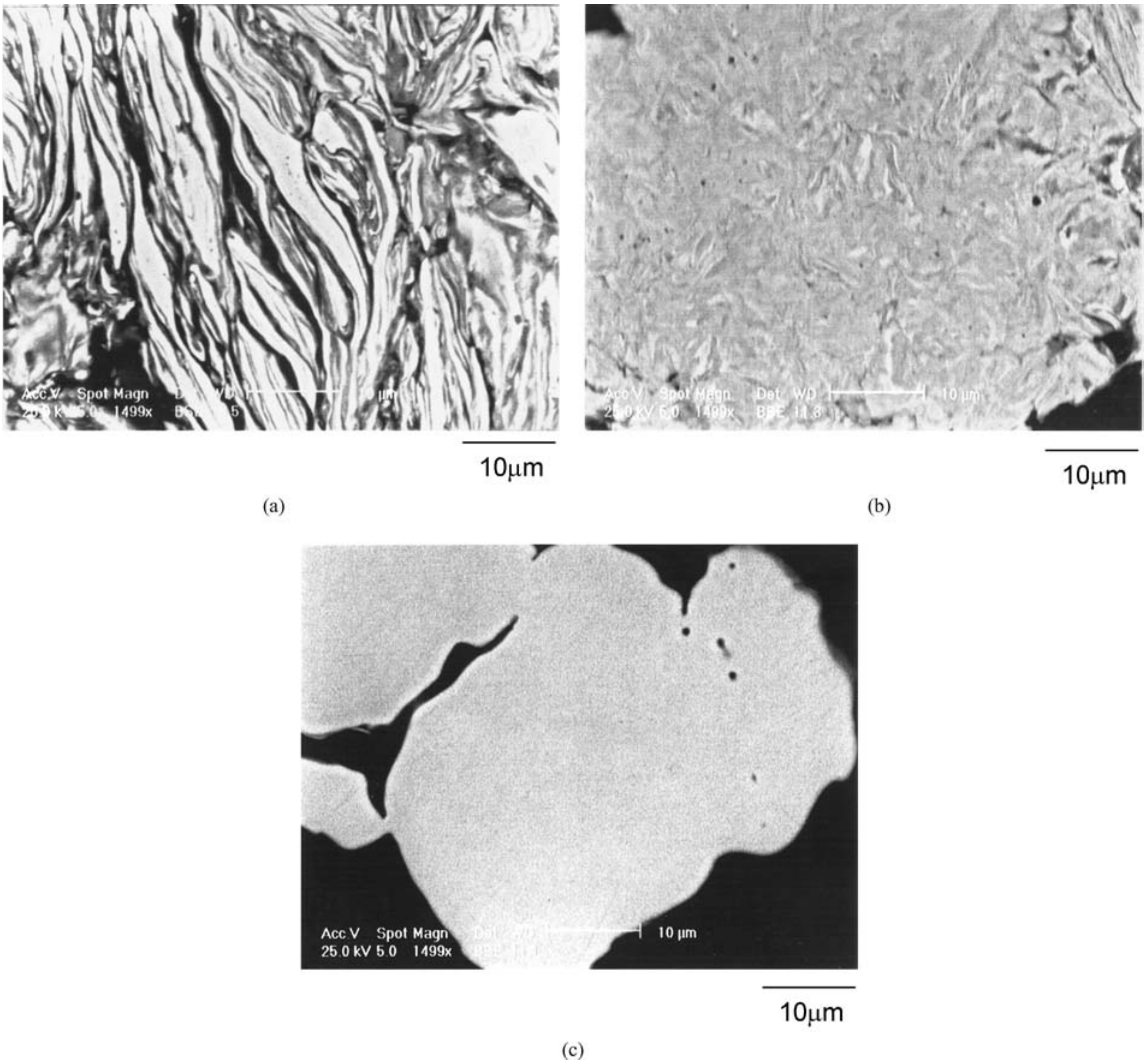


Figure 3 Cross-sectional SEM images of $Fe_{75}Al_{25}$ powder particles after (a) 5 h, (b) 15 h and (c) 80 h of milling time.

obtained here is in good agreement with the previous results from the same alloys [9].

Fig. 3 shows cross-sectional SEM images of $Fe_{75}Al_{25}$ powder particles after different milling times. MA for 5 h produced a layered microstructure consisting of cold welded Fe and Al layers. At this stage the layers are coarse and vary in thickness over the particle cross sections. As MA proceeds further, the layer thickness progressively refined and became more uniform due to the repeated cold welding and fracturing of particles. Such a fine layered structure provides an extensive Fe/Al interface suitable for any potential reaction between Fe and Al. As shown in Fig. 3c the microstructure of powder particles after 80 h MA was featureless in the SEM in consistent with the XRD results.

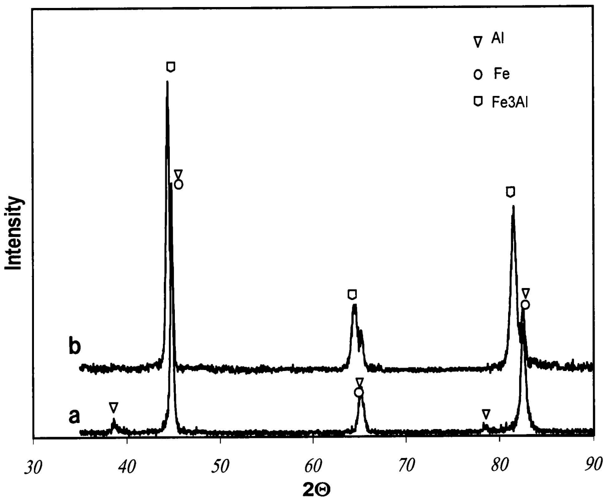


Figure 4 XRD traces from $Fe_{75}Al_{25}$ powder particles: (a) as-milled for 5 h and (b) after subsequent annealing at 550°C for 1 h.

Formation of Fe_3Al phase during MA process was further comfirmed by heat treatment experiments. Fig. 4 shows XRD traces from $Fe_{75}Al_{25}$ powders as MA for 5 h and also after subsequent anealing at 550°C for 1 h. As shown in Fig. 4b, after isothermal annealing the XRD traces included additional peaks which were indexed as the Fe_3Al intermetallic phase which is caused by the reaction of Fe and Al during annealing. The presence of the Al and Fe peaks indicates that Fe and Al were not entirely consumed. Fig. 5 shows XRD traces from $Fe_{75}Al_{25}$ powders as MA for 15 h and also after subsequent anealing at 550°C for 1 h. After annealing only the Fe_3Al diffraction peaks were seen on the XRD traces suggesting the complete consumption of

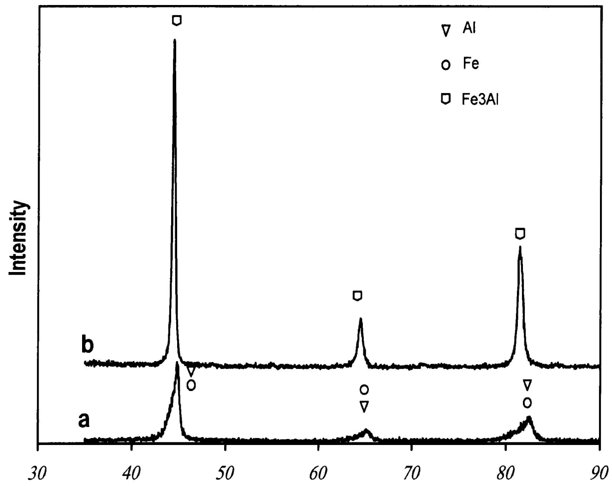


Figure 5 XRD traces from $Fe_{75}Al_{25}$ powder particles: (a) as-milled for 15 h and (b) after subsequent annealing at 550°C for 1 h.

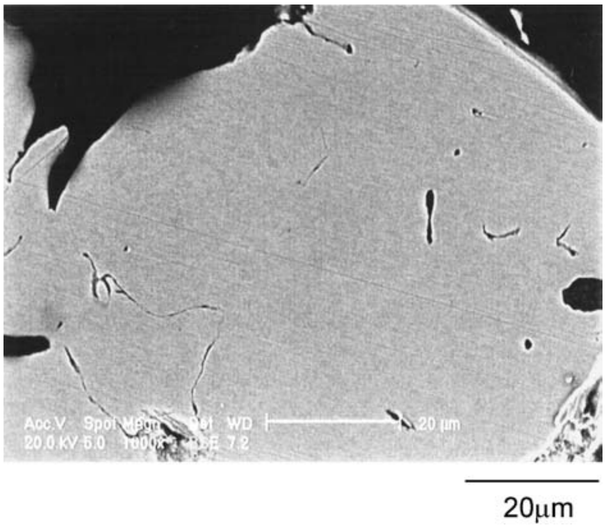


Figure 6 Cross-sectional SEM image of $Fe_{75}Al_{25}$ powder particles MA for 15 h and then annealed at 550°C for 1 h.

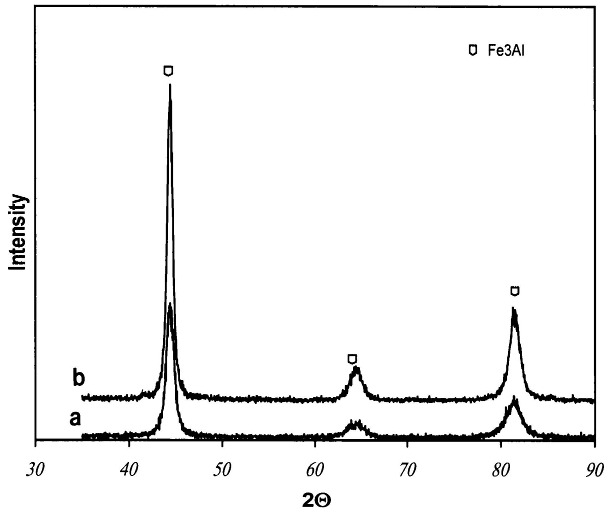


Figure 7 XRD traces from $Fe_{75}Al_{25}$ powder particles (a) as-milled for 40 h and (b) after subsequent annealing at 550°C for 1 h.

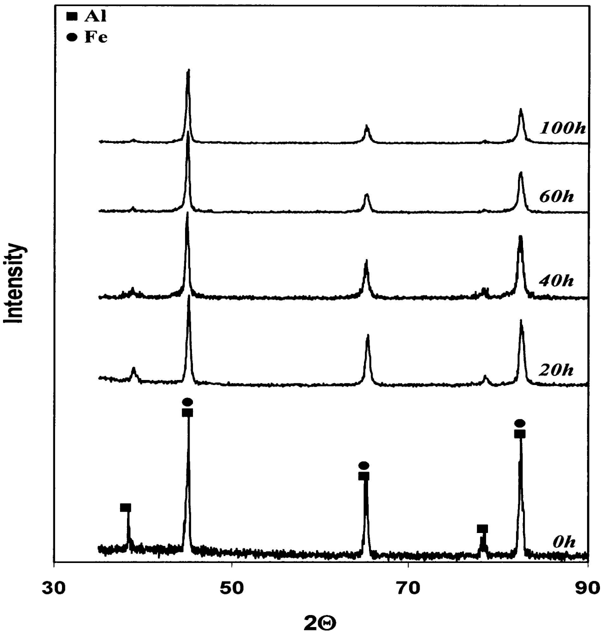


Figure 8 XRD traces from $Fe_{75}Al_{25}$ powder particles as-received and after different milling times (series II).

Fe and Al by the Fe_3Al reaction. Consistently, the layered structure virtually disappeared on the SEM cross-sectional images as shown in Fig. 6. The finer Fe/Al layered structure as well as the higher density of dislocations and grain boundaries in sample MA for 15 h aid the kinetics of the Fe-Al reaction by accelerating the diffusion process. As shown in Fig. 7 annealing of powders MA for 40 h led to no new phase indicating that the resulting structure after 40 h MA is the Fe_3Al phase. It is worthnoting that the superlattice diffraction peaks are still lacking on XRD taces taken after heat treatment. This can be related to the low intensity and spread of the superlattice diffraction peaks into the background rather than the absence of ordering in the annealed structure. This limitation did not allow verification of whether the Fe_3Al phase formed during MA as well as during annealing has an ordered structure or not.

To study the effect of milling parameters on the formation of Fe_3Al compound, MA of $Fe_{75}Al_{25}$ powder mixture was repeated with different milling parameters (Table I). As shown in Fig. 8 decreasing the rotation speed of mill from 390 to 195 rpm (series II) led to only the broadening of Fe and Al diffraction peaks. No evidence of Fe_3Al phase was observed on XRD traces even after 100 h of MA. Moreover the average layer thickness in series II of MA experiments was significantly larger than that observed in series I after the same milling time. The kinetic energy of the balls on impact with the powder particles and hence, the extent of plastic deformation and work hardening reduce as the rotation speed of mill decreases. This causes the grain size to decrease and the dislocations density to increase at a slower rate during MA, which in turn retard the formation of Fe_3Al phase kinetically. Increasing ball-to-powder weight ratio from 5.2 to 10.4 (series III) accelerates the rate of formation of Fe_3Al phase. XRD traces (Fig. 9) sugessted that the Fe/Al layered structure is fully transformed to the Fe_3Al phase after 20 h of MA which is shorter than 40 h observed in series I of experiments. MA at higher ball-to-powder weight ratio also causes the layered structure to refine

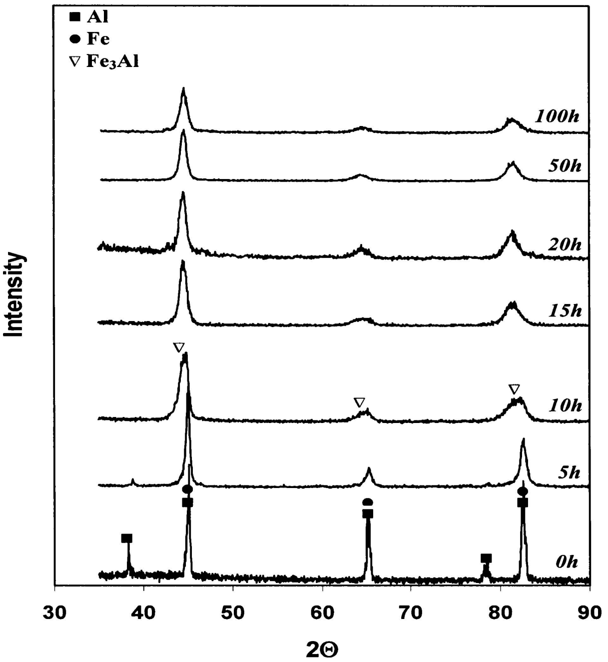


Figure 9 XRD traces from $Fe_{75}Al_{25}$ powder particles as-received and after different milling times (series III).

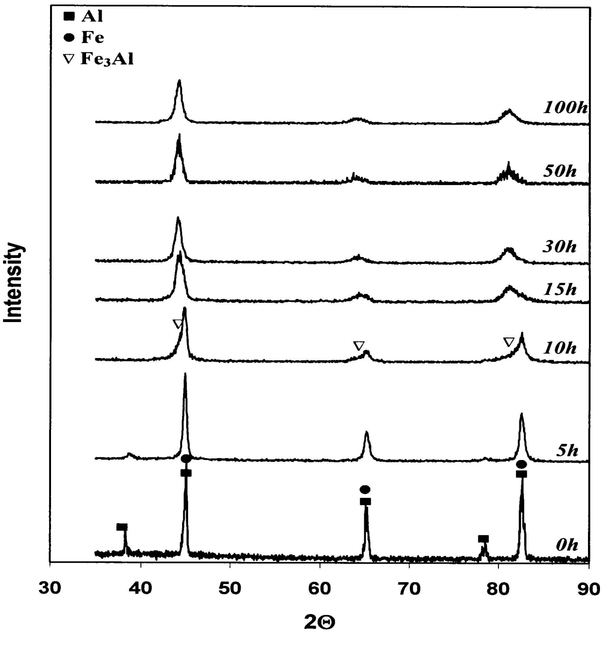


Figure 10 XRD traces from $Fe_{75}Al_{25}$ powder particles as-received and after different milling times (series IV).

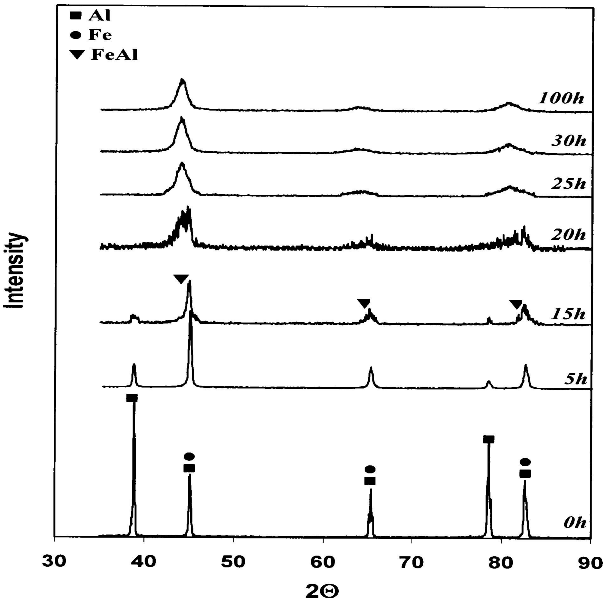


Figure 11 XRD traces from $Fe_{50}Al_{50}$ powder particles as-received and after different milling times (series V).

faster. These results imply that at higher ball-to-powder weight ratio the number of ball-powder-ball collisions per unit time and therefore the kinetic energy of the ball mill charge per unit mass of powder particles are higher. Increasing number of balls, while the ball-to-powder weight ratio and weight of powder mixture kept constant (series IV), was also found to increase the rate of refinement of layered structue as well as the formation of Fe_3Al phase. As seen in Fig. 10 a fully Fe_3Al structure was formed after 30 h which is shorter than 40 h observed in series I of MA experiments. Higher number of balls with a smaller size provide more collisions per unit time although the energy of each collision is lower due to the reduced mass of each ball. The former effect appeared to predominate during MA of $Fe_{75}Al_{25}$ powder mixture. Consequently the rate of MA process increases.

3.2. $Fe_{50}Al_{50}$

Fig. 11 shows XRD traces for $Fe_{50}Al_{50}$ powders (series V) as-received and after different MA times. As seen, the MA behaviour of $Fe_{50}Al_{50}$ alloy was in general similar to $Fe_{75}Al_{25}$ alloy, i.e. the progressive broadening of the crystalline Fe and Al diffraction peaks of as-received powders, the subsequent development of FeAl intermetallic compound after 15 h of MA time and the gradual growth of FeAl phase and the concurrent vanishing of the Fe and Al diffraction peaks with increasing MA time. The XRD traces after 30 h of MA time suggested a fully FeAl structure. XRD peak broadening suggested a grain size of 10 nm for FeAl compound after 30 h of MA time which remained almost constant for longer milling times. Once again no intermediate phase was observed on the XRD traces prior to the formation of intermetallic compound during MA. It should be noted that annealing (at 550°C for 1 h) of $Fe_{50}Al_{50}$ powders milled for times shorter than 30 h resulted in the formation of the FeAl phase.

Like $Fe_{75}Al_{25}$ alloy MA of $Fe_{50}Al_{50}$ alloy led to the development of a coarse, non-uniform layered structure after 5 h of MA time. The layered structure progressively became finer and more uniform with increasing milling time. The microstructure was completely featureless on SEM after 30 h of milling time, suggesting a fully FeAl structure in agreement with the XRD data.

4. Conclusions

Elemental Fe-Al powder mixtures containing 25 and 50 at.% Al were mechanically alloyed in a planetary ball mill under different conditions. In early stages of milling a Fe/Al layered structure was formed, with a progressively refined layer thickness with increasing milling time. In this stage isothermal annealing of powder particles led to the reaction of Fe and Al in layered structure to form intermetallic compound. Those samples milled for longer times exhibited a greater fraction of the intermetallic phase after the same heat treatment due to the increasing density of lattice defects and also the continuing refinement of layered structure. As mechanical alloying proceeded further the $Fe_{75}Al_{25}$ and $Fe_{50}Al_{50}$ layered structure transformed to the corresponding intermetallic compounds, Fe_3Al and FeAl, with a nanocrystalline structure. No intermediate phase developed prior to the formation of intermetallic phases. Decreasing rotation speed of mill retarded the rate of structural evolutions during mechanical alloying. Conversely, increasing either ball-to-powder weight ratio or number of balls (with a constant ball-to-powder weight ratio) accelerated mechanical alloying process.

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