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Evolution of manufacturing parameters in Al/Ni_3Al composite powder formation using blending and mechanical milling processes

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Abstract Aluminu–matrix composites produced by Ni_3Al intermetallic particles are increasingly used in aerospace and structural applications because of their outstanding properties. In manufacturing of metal–matrix composites using powder metallurgy blending and milling are important factors. They control the final distribution of reinforcement particles and porosity in green compacts which in turn, strongly affect the mechanical properties of the produced PM materials. This paper studies different conditions for producing composite powders with uniform dispersion of Ni_3Al particles in aluminum powders and improved physical and mechanical properties. The results indicated that an intermediate milling time for fabrication of composite powder, better than prolonged and shortened ones, causes better microstructure and properties. It was shown that addition of 5 vol.% Ni_3Al particles, produced by 15 h mechanical alloying to aluminum powders, and then 12 h blending operation provides an appropriate condition for producing Al–Ni_3Al composite powder.

Introduction

Ni–Al intermetallic compounds possess special combination of characteristics such as superior oxidation and corrosion resistance, high-temperature strength and stiffness, and relatively low density. However, the intermetallic compounds are brittle so that they cannot serve as structural materials, lonely. Attempts have been made to compensate the brittleness by embedding the particles in a ductile matrix material. Some papers confirm that intermetallics of the Ni–A1 system are excellent candidates as reinforcement in Al-base composites [1–4].

The intermetallic compounds may be produced either by mechanical alloying (MA) or gas atomization. The interfacial characteristics in metal–matrix composites play an important role in determining the resultant composite properties. Fracture surface analysis of the composites indicated that the addition of mechanical alloyed intermetallics to the aluminum matrix, better than atomized ones, can cause good adherence in the particle–matrix interfaces [5, 6].

Effective parameters in MA are time, speed of milling, ball-to-powder-weight ratio (BPR), atmosphere, etc. [7–9]. To obtain appropriate characteristics of MA intermetallic powder, the process parameters should be selected properly. Mechanical alloying time, as well as the interfacial reaction between matrix and reinforcement, is one of the important parameters affecting properties of composites.

The first requirement for a composite material to show its superior performance is the homogeneous distribution of the reinforcing phase [10]. Therefore, in powder metallurgy, blending process of matrix and reinforcement powders is the critical step toward a homogeneous distribution throughout the consolidated composite material. Differences in particle size, density, geometry, electrical charge, and the tendency of the particles to agglomeration all contribute to particle agglomeration [10]. The agglomeration of the reinforcement particles deteriorates mechanical properties of composites.

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Mechanical milling (MM) is a dry, high-energy ball-milling (BM) process for producing composite metal powders with a controlled fine microstructure [11–14]. Each composite particle includes a uniform distribution of the reinforcement phase in the whole particle. The reinforcement particles are larger in the low-energy mixed powders than in the composite mechanically milled particle. These reinforcement particles also show defects, such as cracks. The high-energy milling process reduces the reinforcement size, and tends to eliminate reinforcement defects and sharp edges, producing a rounded reinforcement morphology, which will result in improvement of composite properties.

The general characteristics and mechanisms of mechanical milling and alloying have been studied, but the influence of milling parameters is not yet fully known. This paper reports a study on determining optimum condition for producing composite powder with homogeneous dispersion of fine Ni_3Al particles in aluminum powders and improved physical and mechanical properties.

Experimental procedure

Raw materials

Elemental Al powder (Merck-1056—99%, <160 μm) and Ni (Merck-112277—99.5%, <10 μm) were used as raw materials. The morphologies of both powders were studied using SEM. The as-received aluminum powder was flake-like, while nickel powder was polygonal in shape.

Mechanical alloying process

Mechanical alloying of the elemental Al and Ni powders (with 1:3 at.% ratio of Al to Ni) was conducted using a planetary apparatus (model FP2) to produce Ni_3Al powder particles as reinforcement of composite. The powder and hardened steel balls with 20 mm in diameter were sealed in a hardened stainless steel vial and operation was undertaken at room temperature. The BPR and rotational speed were 20:1 and 550 rpm, respectively. In order to avoid oxidation, the entire process was performed in argon atmosphere. After 15-h MA, the powders were analyzed for morphological and structural studies as well as XRD pattern of nanostructure of the reinforcement.

Blending operation

Further, 5 vol.% (12.8 wt%) Ni_3Al, produced by MA process, was added to Al powder. After a short blending process in a planetary high-energy ball-mill, Al–5 vol.% Ni_3Al composite powder was prepared. Two different blending times were examined to obtain an optimum condition for producing Al/Ni_3Al composite powders with more uniform distribution of Ni_3Al in Al powders. The blending process was carried out at room temperature in argon atmosphere using hardened steel vials and steel balls with 10 and 20 mm in diameter.

In order to investigate the effect of other blending parameters, various BPR (6:1 and 15:1), and rotational speed (200 and 300 rpm) were chosen. To avoid significant temperature rise, 3 h of milling was alternated by 30 min of cooling through the built-in fans in the system.

Powder characterization

In order to measure the crystallite size and lattice strain of particles the powders were characterized by Bruker; Advance-D8 X-ray diffractometer. Full experimental details regarding the XRD methods used can be found elsewhere [15]. To study morphological changes scanning electron microscope model LEO 440 i, was used. Also, size distribution of powder particles was evaluated by Clemex image analyzer.

Powder compaction

Aluminum and Al-5 vol.% Ni_3Al powders were uniaxially cold pressed in a cylindrical die in the range of 400–800 MPa. Two major stages of compaction, i.e., rearrangement and plastic deformation happened. The green density of each specimen was determined by measuring of its mass and volume. The inner surface of the cylindrical die was lubricated using oil graphite to reduce the effect of friction forces during the consolidation process.

Sintering process

The compacted specimens such as pure aluminum and mixed powders were sintered at 620 °C in a vacuum furnace for 30 min followed by furnace cooling.

Physical and mechanical properties

Densities of the sintered parts were determined using Archimedes principle (DIN ISO 3369). To determine hardness, Vickers tests were performed in the carefully sectioned and polished specimens. Vickers hardness values were the average of at least 10 indentations applying 30 kg load for 10 s. To investigate the effect of production process on mechanical properties of composites, quasi-static uniaxial compression experiments at strain rate of 10^{-4} s^{-1} were performed on specimens consolidated at different condition.

Results and discussion

The initial morphologies of both powders, Al powder and Ni powder, are shown in Fig. 1. Also, their XRD patterns are indicated in Fig. 2. The figure shows only the peaks of pure Al and Ni before milling but with increasing milling time up to 15 h, the powders are consumed and Ni_3Al is formed.

The lattice parameter of 15 h mechanical alloyed powder was calculated from the XRD peaks according to the Bragg's law as 3.98 Å which is equal to the lattice parameter of Ni_3Al. Figure 3 shows SEM images of the powder at the milling time of 15 h, which is a sign of its purity. Point analysis at different and random sites demonstrates 75:25 at.% ratio of Ni to Al. Back-scattered electron image of powder confirms the results of XRD analysis because no pure Al and Ni are detected.

As reported elsewhere [15], the optimum time of MA to produce Ni_3Al reinforcement particles with desirable properties is 15 h. In this research, the minimum size of particle and grain in the produced Ni_3Al powder under the mentioned MA parameters was obtained after 15 h of processing.

Effective parameters in blending process of aluminum and Ni_3Al reinforcement particles are time, speed of milling and BPR. The effect of these parameters on composite powders properties is discussed separately.

Effect of milling time

Figure 4 demonstrates the x-ray diffraction of mixed and milled composite powder at different times which can be used to investigate the grain size and lattice strains. It is shown that in this process the variation of compound does not change and only peak width is increased.

As Table 1 indicates increasing milling time causes reduction in grain size. However, internal lattice strains remain almost constant. Figure 5 shows the morphology of milled powder at different times. Long time blending of aluminum and reinforcement powders (Ni_3Al), e.g. 12 h, causes more uniform distribution of the reinforcement. The largest size of Ni_3Al particles is about 50 μm. The particles are dispersed heterogeneously among aluminum powders. Impact of milling balls on the powders causes plastic deformation of soft flake-like aluminum powders

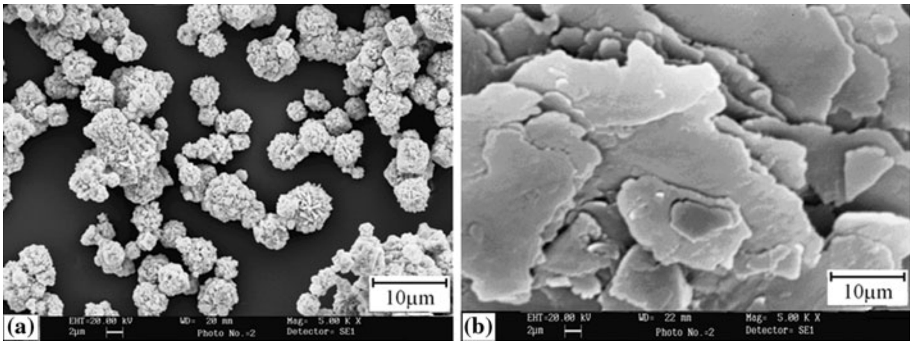


Fig. 1 SEM images of Al powder and Ni powder

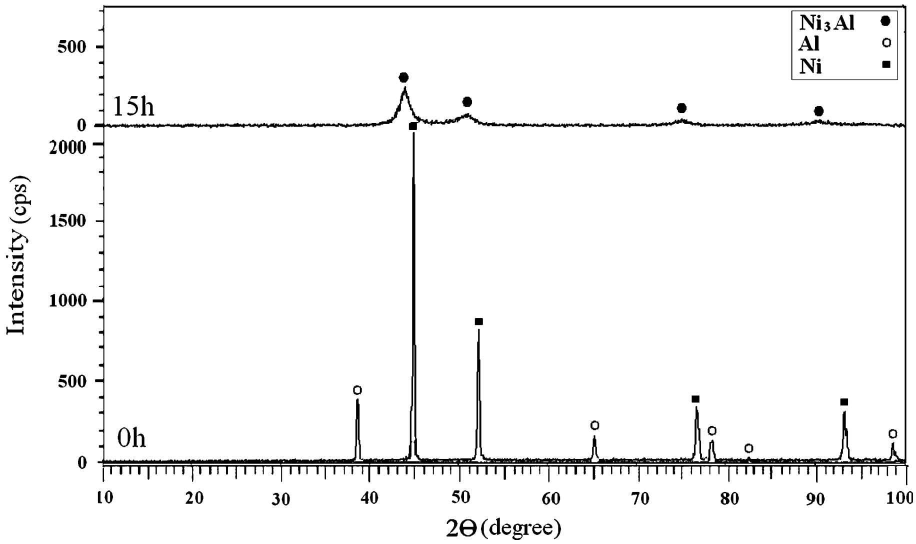


Fig. 2 XRD pattern of nanostructure reinforcement

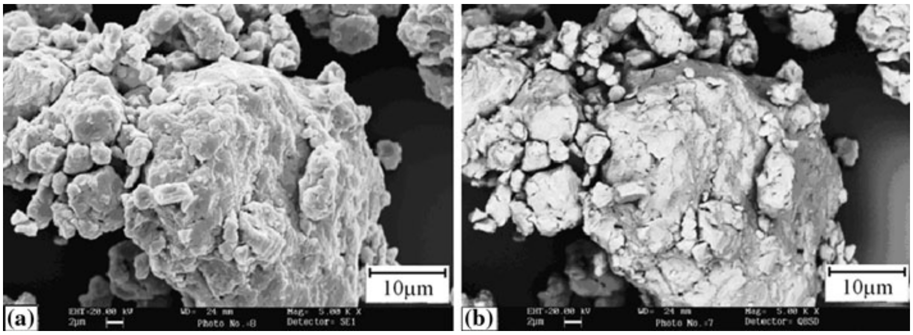


Fig. 3 SEM image of the obtained powder after 15 h MA; **a** SE and **b** QBSD

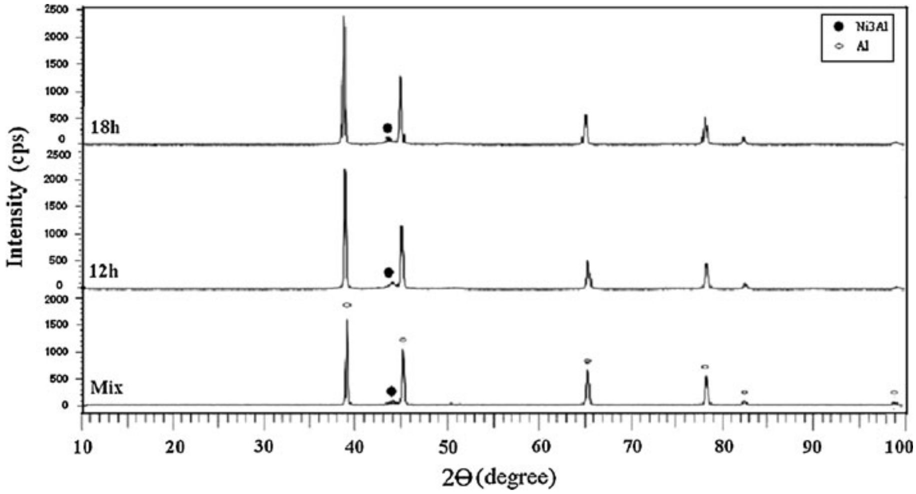


Fig. 4 X-ray diffraction of mixed and milled composite powder at different times

Table 1 Grain size and strain of milled powder at different time (0 h milling means just blending)

Milling time (h)	0	12	18
Grain size (nm)	69	68	54
Strain %	0.13	0.14	0.13

and brittleness of Ni_3Al particles because of fracturing phenomena.

During milling, the fine reinforcement particles are cold welded to the aluminum flakes. Continuance of milling causes cold welding of these flakes together, as a result composite powder includes aluminum and Ni_3Al is formed. The microstructure resulted from this phenomenon is shown in Fig. 6.

With increasing mechanical milling time from 12 to 18 h, the morphology of composite powder does not change while in this condition contamination, impurities and strain hardening of powders are enhanced.

Effect of milling speed

To study influence of milling speed on mechanical milling process, mixed powders were milled at 200 and 300 rpm. Figure 7 shows morphology of powders milled for 18 h at different speeds.

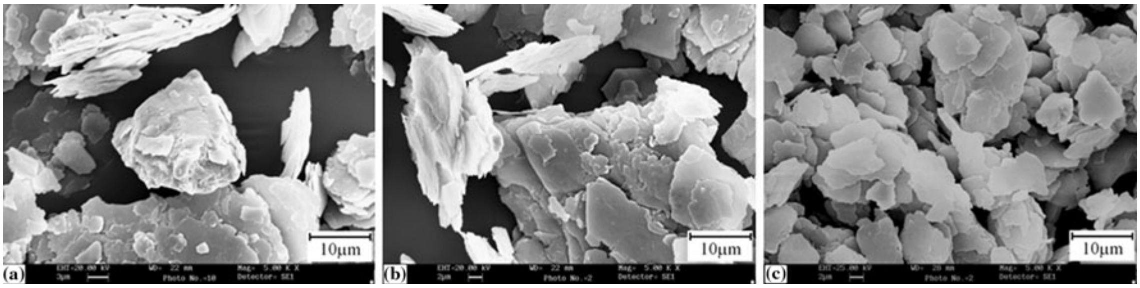


Fig. 5 Morphology of milled powder at different times

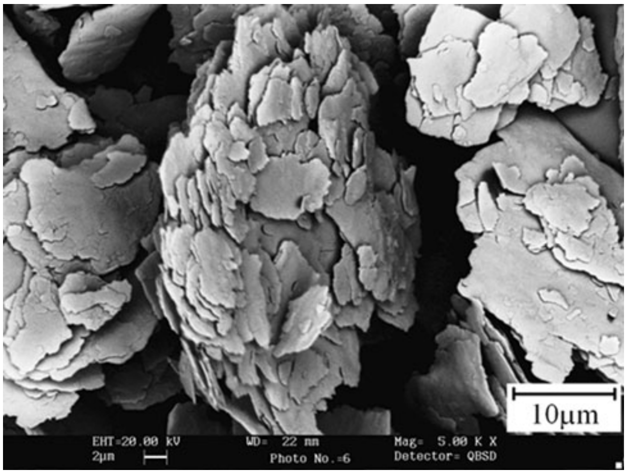


Fig. 6 Morphology of 12 h milled powder

Fracture and welding between powders rise with increasing milling speed considerably, because entered energy within the system in unit time increases.

At 300 rpm reinforcement particles penetrate in aluminum flakes and composite powders are formed. Morphology of powders is smaller, and distribution of reinforcements in matrix is complete. However, at 200 rpm aluminum powders are larger and cold welding between reinforcement particles and aluminum powder is not completed.

The present experimental results indicated that at higher speed of milling cold welding overcomes fracturing mechanism therefore, particle size of powders becomes bigger. It is estimated that at milling speed of more than 300 rpm reinforcement and aluminum particles will react together so instead of Ni_3Al other intermetallics will be formed.

Effect of ball-to-powder ratio

Figure 8 indicates morphology of powders milled in 6:1 and 15:1 ball-to-powder ratio. Increasing ball-to-powder ratio increases the number of impacts applied by the balls in unit time. Therefore, mechanical milling with higher ball-to-powder ratio causes the fracture processes overcome cold welding, so refinement of powders is occurred.

Comparison of powder morphology after blending and after high-energy mechanical milling process explains that blending process alone cannot completely remove agglomerates. Mechanical milling process improves reinforcement distribution throughout the whole Al-particle. Optimum intermediate milling time should be applied for producing Al/Ni_3Al composite powder. In current investigation, uniform distribution of Ni_3Al particles in aluminum powders was obtained after 12 h of milling with 300 rpm and 15:1 BPR.

Metallography study of microstructure of sintered parts shows agglomeration of intermetallic particles under blending process while mechanical milling improves the distribution of the reinforcement particles in the matrix.

Fig. 7 Morphology of powders milled at 18 h at different speeds

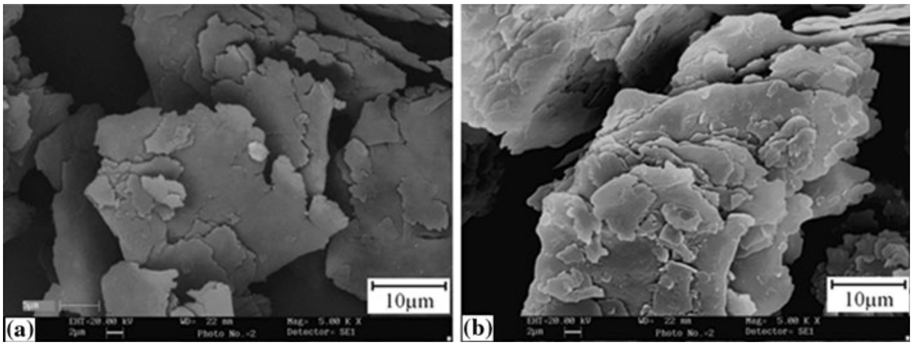
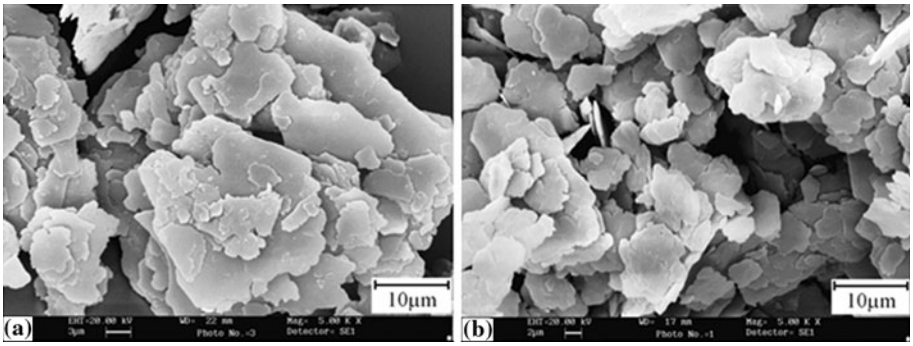


Fig. 8 Morphology of powders milled in 6:1 and 15:1 ball-to-powder ratio



The Ni_3Al intermetallic compound becomes ultrafine so that the size of the individual particles is <10 µm in diameter at the end of ball milling.

The study of reaction between matrix and reinforcement and other produced phases during blending of Al/Ni_3Al composite, under different time of MA and sintering conditions was reported in a previous work performed by the authors [16]. Mechanical milling increases the reactivity of reinforcement particles with matrix through the following parameters such as: fragmentation of Ni_3Al particles, high degree of deformation, high density of dislocations, and reinforcement particles dispersed in the matrix.

XRD patterns of mixed and ball milled (BM) composite, after heating at 620 °C for 30 min, are displayed in Fig. 9. XRD patterns were obtained using a Cu Kα radiation ($\lambda = 0.15406$ nm). Both composites show peaks of Al, Ni_3Al, and Al_3Ni with different intensity.

Dissolution of Ni_3Al particles in milled samples occurs with a higher diffusion rate of Ni from intermetallic particles to matrix when compared to mixed samples. On the other hand, ball milling of composite powder results more homogenization in the microstructure of sintered composite (Fig. 10).

Physical and mechanical properties

In the first stage of compaction, the powders slide on each other and rearrange to decrease the interparticle spacing. If during this stage, the applied pressure is eliminated, powder orientation will return to its original form without compaction. This phenomenon is due to the elastic deformation of powder particles. However, by increasing compressive pressure, plastic deformation, and then densification occurs.

The milled powder has a more flaky morphology. Continuous impacts of balls result in strain hardening. This type of morphology and resulted strain hardening causes incompactness of milled powder in pressure up to 400 MPa whereas the green density of just blended powder reaches 89% of the theoretical density. With increasing pressure from 400 to 800 MPa, the green density of blended powders is 95% of the theoretical density. The green density of

Fig. 9 X-ray diffraction of mixed and milled composite sintered at 620 °C

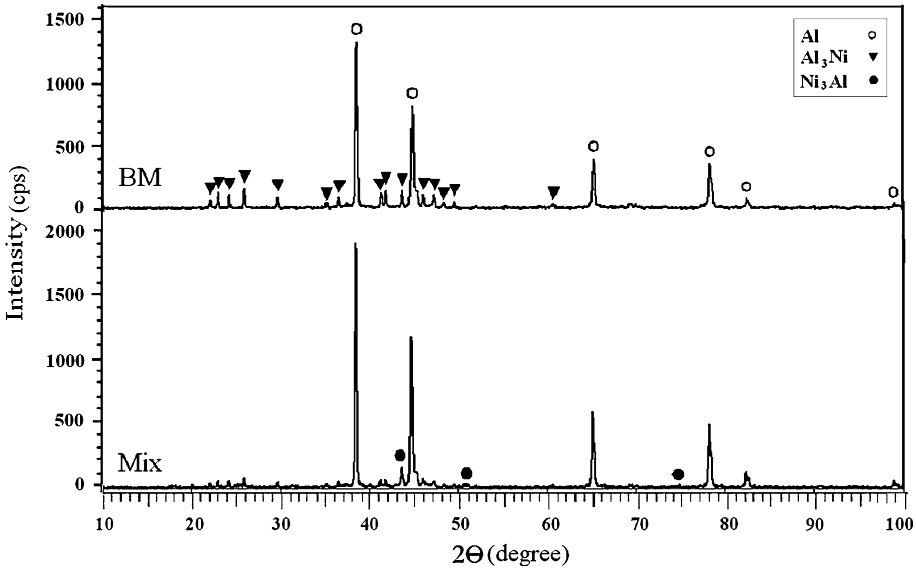
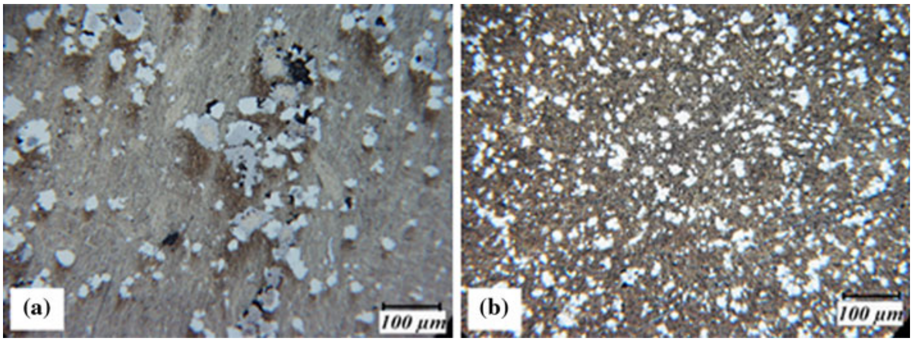


Fig. 10 Microstructure of **a** mixed and **b** milled composite sintered at 620 °C



milled powders is 90% of the theoretical density at 800 MPa which is less than the blended sample compressed in the same pressure.

Because of the flaky morphology of the milled powder, higher pressure causes shear stress between these particles which in turn results in cold welding between them, and densification is attained. Such a behavior cannot be observed at 400 or 600 MPa and needs higher pressure value. Actually, the cold welding between milled powders, which is associated with high pressure compaction, can partly compensate for the incompactness due to the increase in the strain hardening of the powder.

In general, due to work hardening, hardness of the milled powders increases which results in difficulty of cold compactness of powders. Prolong ball milling duration caused microhardness to increase due to work hardening. In summary, due to the work hardening and increase in hardness, densification of the hardened powders by cold compact becomes more difficult resulting in decrease in density. The same results have been reported by Lti et al. [17].

Since the morphology of the blended, or mixed, powders is nearly equiaxed, these powders are more compressible. Therefore, its packing is more convenient and compaction occurs at lower pressures. There exist some differences between milled and blended powders. Packing of milled powder is less convenient compared to nonmilled powder; thus, higher pressure is required for densification. Generally, it is believed that density increases with rising of compaction pressure.

The effect of mechanical milling on the compressibility of Al6061 alloy reinforced by micrometric AlN particles has recently been reported by Fogagnolo et al. [18]. They have shown that mechanical milling changes the morphology of the composite powder particles, leading to less densification upon uniaxial compaction.

The densities of blended and sintered samples decrease to 88% and 92% of the theoretical density (relative density) in 400 and 800 MPa, respectively. The decrease in density is generally due to vaporization of lubricant during the sintering process. In the contrary, the decrease in density does not observe in the milled samples. Because milled powders have higher ability to be sintered due to refinement of particles during milling process, the density of these specimens has slightly increased in comparison with its green density.

Table 2 summarizes values of relative density (*D*), Vickers hardness (HV), compressive strength, and elongation (El) for consolidated materials. Samples designated as pure Al, included for comparison, were consolidated from unreinforced elemental Al powder under equivalent conditions.

Table 2 Physical and mechanical properties of pure aluminum and aluminum–matrix composite

	Relative density (%)	Hardness (HV 30)	Strength (MPa)	Elongation (%)
Al 400	93.45	43 ± 3.8	166	17.2
Al 800	95.31	49.3 ± 6.3	140	12.6
Mix 400	87.91	29 ± 5.7	217	18.9
Mix 800	92.18	44 ± 7.4	186	14.3
BM 800	90.54	95 ± 5.7	167	6.3

With increase in compaction pressure, hardness of sintered specimens increases from 29 to 44 HV in mixed composites in comparison with hardness of pure aluminum (from 43 to 49 HV). Strength and elongation of the samples decrease at higher pressure due to more work hardening, brittleness, and density. Meanwhile, hardness of BM composite at 800 MPa pressure is 95 HV, which is 50% higher than that of the mixed composite consolidated in the same pressure. Mechanical milling through the finer microstructure, high degree of deformation, high dislocation density and oxide and reinforcement particles dispersed in the matrix increases the hardness of composites materials. Conversely, milling process resulted in lower compressive strength.

Conclusions

An Al/5%Ni_3Al composite was produced using a planetary ball mill at different time with a rotational speed of 200 and 300 rpm. The ball/powder ratio was selected 6:1, 15:1. The following conclusions were obtained:

1. The addition of intermetallic reinforcement particles to the blended, or mixed, and compacted composites do not influence their properties.
2. Prolonged milling procedure increases oxidation probability of aluminum powders besides, more contamination is obtained such as Fe, in consequence of balls and container wearing.
3. Conversely, short-time operation would not be suitable with regard to insufficient distribution of Ni_3Al in Al powders as well as their cold welding to each other.
4. Optimum intermediate milling time of 15 h should be applied for producing Al/Ni_3Al composite powder.
5. Uniform distribution of Ni_3Al particles in aluminum powders is obtained after 12 h of blending time with 300 rpm speed of milling and 15:1 BPR.
6. Using of milling process to attain composite powder can improve properties of composites.
7. With increase in compaction pressure, hardness increases although, compressive strength and elongation of samples decrease at higher compaction pressure due to more work hardening, brittleness, and density.

8. It was shown that addition of 5 vol.% Ni_3Al particles, produced by 15 h MA to aluminum powders, and then 12 h blending operation provides an appropriate condition for producing Al–Ni_3Al composite powder.

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