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http://dx.doi.org/10.1080/10426910701235991

Published in Materials and Manufacturing Processes,

Volume 22, Issue 4 May 2007 , Pages 492 – 496

SYNTHESIS OF ALUMINIUM-CEMENTITE METAL MATRIX COMPOSITE BY MECHANICAL ALLOYING

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Abstract

Cementite powder was prepared from elemental iron and graphite powder by mechanical alloying in a specially built dual drive planetary mill. The phase evolution, particle size distribution and morphology of particles were studied during 40 hours grinding period. X-ray diffraction shows formation of Cementite and other iron carbides alongwith elemental iron after milling, whereas after annealing only Cementite is present. Initially particle size increases with milling due to ductility of iron powder and then reduces with further milling.

Al-Cementite composite was synthesized by mixing Cementite with Al powders, and then by hot pressing or cold compaction and sintering. XRD analysis of Al-Fe_3C composite shows Fe_3C, FeAl, Al and other iron carbides along with Al_4C_3 after sintering. SEM micrograph of hot pressed samples shows excellent compatibililty between Al matrix and Cementite particles.

Keywords: Al-Fe_3C composite; Cold compaction and sintering; Dual-drive planetary mill; Hot pressing; Mechanical alloying; Metal matrix composite; Particle size analysis; XRD

INTRODUCTION

Although the very large body of information available on metal matrix composites (MMC), there is limited published work, to the best of the author's knowledge on MMC with reinforcing agents such as Cementite. The current interest in research and development is focused on the light structural material such as Al based MMC. Al-Fe_3C composite finds place in automobile industry and aerospace applications. It also can be used in other structural purposes.

Mechanical alloying is normally a dry, high-energy ball milling technique and has been employed to produce a variety of commercially useful and scientifically interesting materials. Mechanical alloying can produce a variety of materials, starting from supersaturated solid solutions, stable, metastable crystalline, quasicrystalline intermediate phases, and amorphous alloys to composites.

Fe-C system has received much attention due to its weight in steel industry. This has led to the synthesis of high carbon content carbides such as Fe_3C by mechanical alloying. The main limitation of mechanical alloying is contamination from milling media (e.g. iron from steel balls and vials) and carbon. When milling Fe-C materials, at least two of these contaminants are no longer concern. Since these high temperature carbides in metastable state are difficult to produce (Campbell et al. [1]), mechanical alloying is one of the best techniques to produce homogeneously distributed composite particles.

The reaction milling or mechano-chemical synthesis of metal carbides was proposed by Le Caer et al. (1990) [2]. They prepared carbides of Ti, V, Cr, Mn, Fe, Co, Ni, Zr, Nb, Mo, Ta, W, Re, Al, and Si by milling the above elements with graphite powder in either a SPEX 8000 shaker mill or a Fritsch P-7 planetary mill for a fixed amount of time (Matteazzi and Caer, 1991) [3]. Carvalho et al. [4]

prepared Cu-Fe_3C nanocomposite by mechanical alloying of Cu, Fe and graphite powders in a planetary mill. They observed that Fe_3C and Fe_7C_3 were present in the as milled material, whereas only Fe_3C present after annealing. Goodwin et al. [5] synthesized Fe-85 vol. % Cementite nanocomposite by mechanosynthesis of elemental iron and graphite powders in a planetary mill. The crystallize size in the mechanosynthesis powder was 7 nm for Fe_3C and 8 nm for iron. Then the powders were densified by field assisted sintering at 450°C for 3 min in air under a pressure at 63 MPa. Ryszard Nowosielski et al. [6] milled Fe-6.67 mass % C in SPEX mill for 100 hours. They obtained fine dispersion of carbides in ferrite matrix. Hidaka et al. [7] studied the milling of Fe- (0-6.3) mass %C alloy powders containing a small amount of Cr (0.2 mass%), Si (0.06 mass%) and Mn (0.23 mass%) for 100 hours. They showed that depending on the carbon content, it was possible to produce full ferrite, ferrite + cementite and full cementite. Halil et al. [8] produced Fe-Fe_3C composite by mechanical alloying of elemental Fe-1 wt% C in Ar atmosphere by using a high-energy ball mill for various milling time. The mechanically alloyed powders were then compacted and sintered at different temperatures to study compatibility and densification behaviour. N. T. Rochman et al. [9] did planetary milling of Fe, C and Si powder at various milling temperatures (252, 293 and 323 K). To inhibit the milling temperature change during milling, liquid nitrogen was flowed around the vial. The mechanically alloyed powders were then consolidated and hot isostatic pressing (HIP) was carried out. They showed that formation of super saturated iron solid solution depends on the milling temperature. The alloying mechanism is affected by the changes in plastic deformation, the diffusion rate of carbon and the refinement rate of the particles due to milling temperature.

From the past study, it is clear that Cementite can be produced from elemental iron and graphite powders by mechanical alloying using various types of mills. But these types of mills take more time and powder production is very less. From this standpoint, we have fabricated a dual drive planetary mill to produce large amount of powder in less milling time.

In the present investigation, initially Cementite powder was produced from elemental iron and graphite powder by mechanical alloying in a dual drive planetary mill. Then Al-Fe_3C composite was prepared; using Cementite and other iron carbide as second phase particles and Al as metal matrix. On the metal matrix composite, a few studies have been conducted on Fe–Fe_3C nanocomposites. But the studies have been very specific, for a particular fraction of the dispersoids, sintering conditions etc. and the matrix has been iron in almost all the cases. But no attempt has been made so far to characterize these composites. The concept behind the development of the composite is that their strength should be much higher than that of conventional materials (pure metals, steels etc), and should maintain toughness. From these standpoints, composites composed of metal matrix and Fe_3C (and other iron carbide) are very promising. In order to design such composites, it is necessary to understand the variation in their mechanical properties with temperature, volume fraction of carbide dispersoids, pressure etc.

EXPERIMENTAL PROCEDURE

Experimental set up

The dual drive planetary mill consists of a gyratory shaft and two cylindrical steel jars, both are rotated simultaneously and separately at high speed. Such high-speed rotation of both jars and the shaft makes the balls to move strongly and violently, leading to large impact energy of balls that improves the grinding performance. This type of mill has been used not only for grinding but also for mechanical alloying (MA) and mechanochemical (MC) operations (McCormick and Froes, 1998) [10]. The dual drive planetary mill developed specifically for the synthesis of nanocarbides has a rotating shaft that sweeps a circle of diameter 750 mm. The two steel jars of 13.5 mm diameter (2500 ml each) rotate about their own axes around the common axis of the main shaft. The planetary mill is powered by two motors. A 5 HP motor works on the main rotating shaft and 3 HP motor drives the jars. The rotating speed of both motors can be varied independently and continuously by a frequency controller.

Synthesis of cementite

Elemental iron and graphite powder were mixed in stoichiometric proportion to produce cementite. Total 250 grams of powder and 2.5 kg steel balls of 6 mm diameter were taken in a jar. A dispersant known as Licowax 1% by weight was taken to prevent agglomeration. After few hours of milling, powder was taken out from the mill to study the progress of reaction. No further milling was carried out, when it was found that; there was no increase in intensity of XRD peaks after milling. Milling was carried out for a total period 40 hours in a specially built dual drive planetary mill. The powder was characterized by XRD (Cu Kα), Laser particle size analyzer and SEM. To study stability of Cementite, 40 hours milled powder was vacuum-sealed in a quartz tube and annealed at 600, 700 and 800°C for 1 hour.

Synthesis of* Al- Fe_3C *composite

40 hours milled final powder and annealed powder (600°C, 1 hour) were mixed separately with aluminum powder and cold pressed. The cold compacted pellets were then sintered at 400, 500 and 600°C for 1 hour in controlled atmosphere. In another set of experiments, milled powder and annealed powder were mixed separately with Al powder and hot pressed at 400, 500 and 600°C for 30 min. The

sintered samples were characterized by XRD, SEM with inbuilt EDAX. Vickers micro hardness of the composites was carried out. A test load of 20 gm was applied for a period of 10 seconds. A minimum of 3 readings was taken for each specimen.

RESULTS AND DISCUSSIONS

Characterization of cementite

In order to determine the phase change of the particle mixtures during the mechanical alloying process, a small sample of milled product was repeatedly picked up at regular intervals for X-ray diffraction analysis. The XRD patterns of the end product (as milled powder) were shown in Fig.1 for different intervals of milling time. Several interesting observations were made. First, the graphite peak disappeared after about 20 hours of milling. This is a general observation in this study, which may be due to amorphization of graphite during milling. Second, it was observed that the peak positions of iron shift to a lower angle with increasing milling time. The shift of the iron peaks position to the lower angle means that carbon atoms dissolved into the iron lattice. It was also observed that iron peaks are broadened and their intensity decreases with milling time. This suggests the accumulation of lattice strain and reduction of crystal size. Finally, after 40 hours of grinding, Fe_3C and other iron carbides are present with elemental Fe and α Fe.

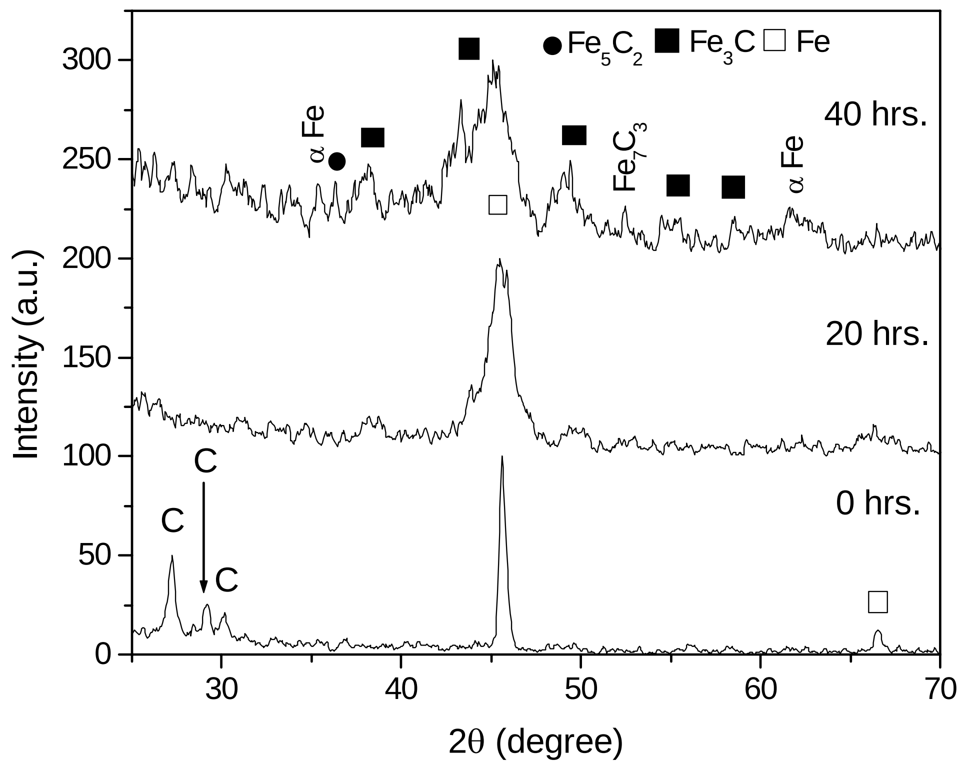

Figure 1. XRD patterns of milled powder at selected milling times.

To study the stability of cementite, 40 hours milled powder was vacuum-sealed in a quartz tube and annealed at 600, 700 and 800°C for 1 hour. Figure 2 shows the XRD patterns of powder milled 40 hours and then annealed at different temperatures for 1 hour. Several conclusions can be drawn from the XRD patterns. First, the XRD pattern shows the peaks of cementite at 600 and 700°C. But at 800°C no cementite peak is visible. At 800°C Cementite decomposes into iron and graphite and becomes unstable. Second, XRD pattern shows only cementite peaks, other iron carbides are not present.

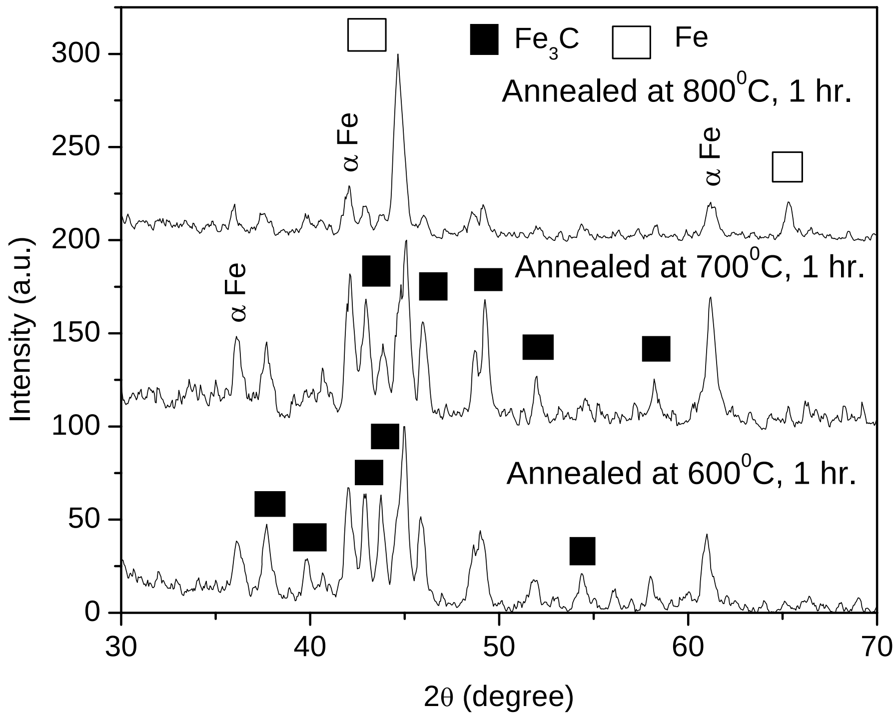

Figure 2. XRD patterns of powder milled for 40 hours and then annealed at different temperatures.

At the early stage of milling, the powder particles were flattened and flakes were formed due to the ductility of particles. Figure 3 shows particle size distribution of powder particles at different intervals of time. After 2 hours of milling, particle size is larger than before milled powder. The SEM picture also clearly reveals the cold welding of powder particles at the initial milling period. At later stage, due to plastic deformation during milling, the particles become brittle and size reduction takes place. After 40 hours of milling 70% of the particles below 10μ.

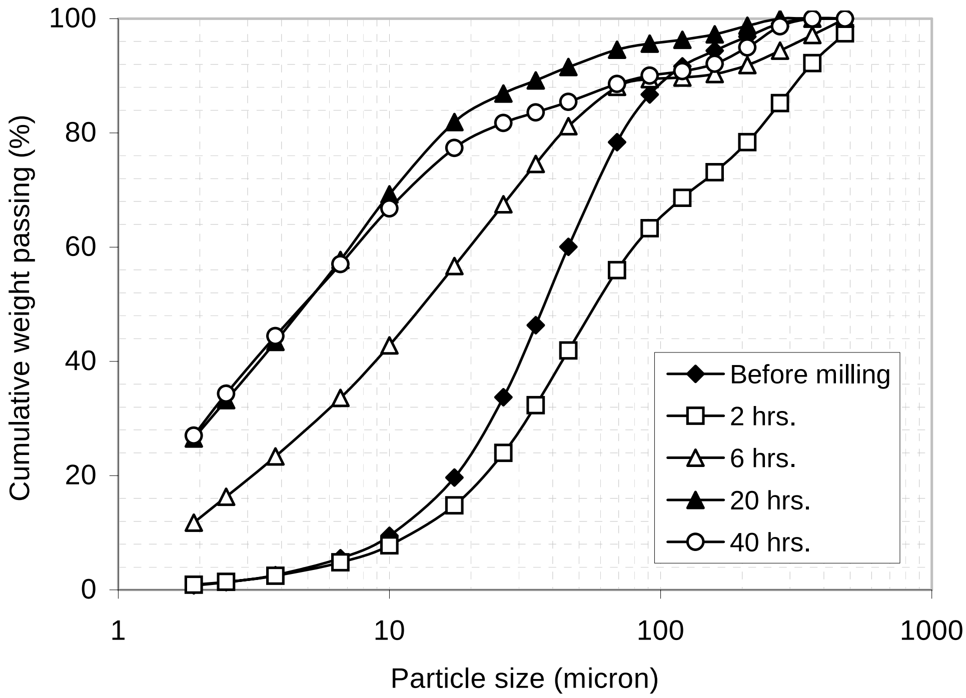

Figure 3. Particle size distribution of powder milled for different periods.

The SEM was used to study changes in shape and size of the milled powder during different stages of the reaction milling process. Figure 4 shows the SEM images at different milling times. After 2 hours of milling, particles aggregated to flake like powders. The powder particles undergo cold welding, binding to each other and their morphologies changes to flake shape. With further milling, the fracture and cold welding of the particles are repeated. As milling progresses the particles become spherical. The particle size also decreases gradually with increasing milling times, which are clear from SEM images.

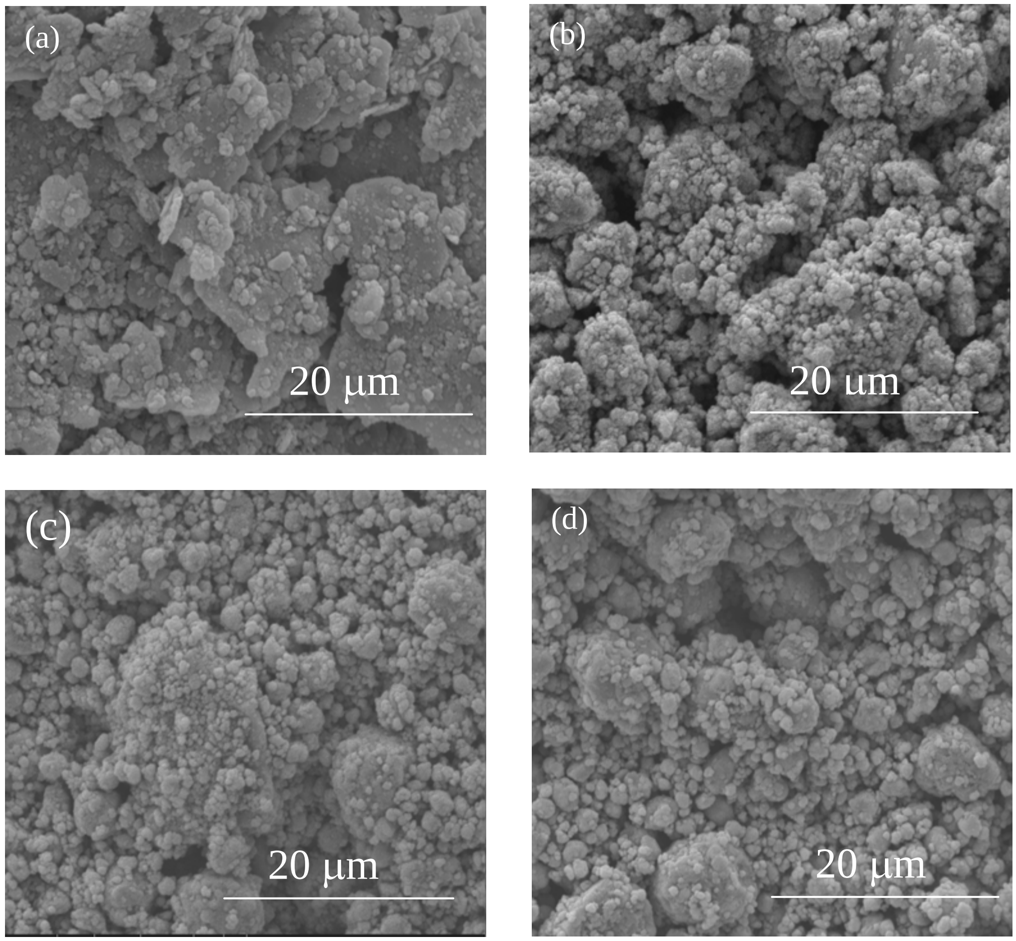

Figure 4. SEM micrographs of powder milled for different periods.

Characterization of **Al-Fe_3C** *composite*

To study the different phases after sintering, XRD (CuKα) was carried out. Figure 5 shows the XRD pattern of Al-20 vol. % Fe_3C hot pressed at 400°C for 30 min. After sintering, Al, Fe_3C, AlFe and other iron carbides are present. The ordered intermetallic phases FeAl and other aluminides are of interest as high-temperature structural materials due to their very high melting temperatures, good oxidation resistance, and low mass density. Al reacts with Fe_3C particles and Al_4C_3 was formed. The reaction at the carbon-aluminum interface to form aluminum carbide (Al_4C_3) has long been considered to affect critically the strength of Al-Fe_3C composites.

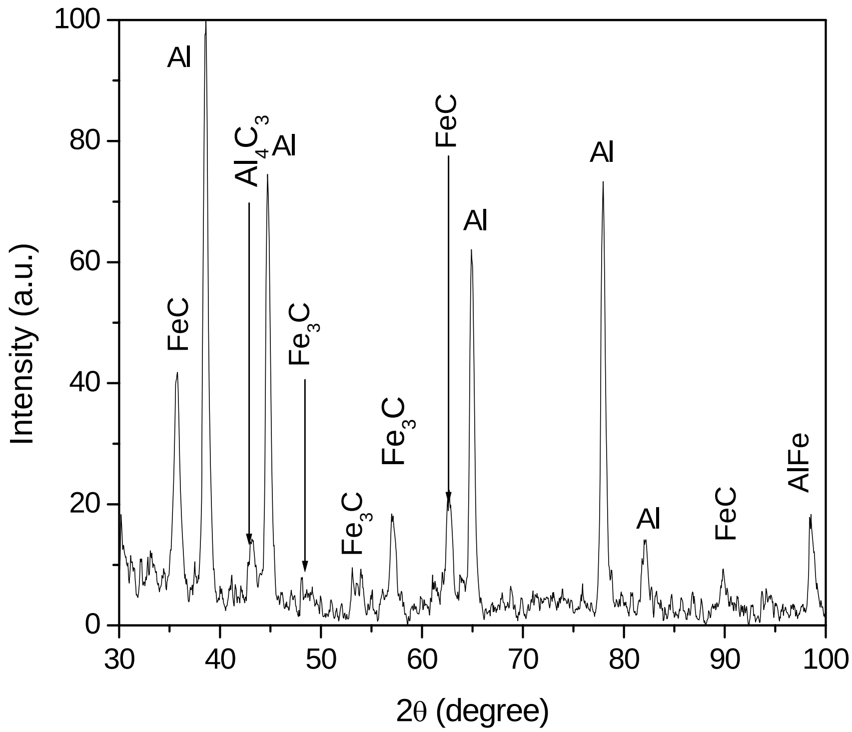

Figure 5. XRD pattern of Al-20 vol. % Fe_3C hot pressed at 400°C for 30 min.

To study the morphology and distribution of cementite particles in the Al matrix, SEM was used. Figure 6 shows the representative SEM picture and the EDS spectra of Al-20 vol. % Fe_3C composite that were hot pressed at 400°C for 30 min.

The EDS of embedded particle shows Fe and C while matrix shows Al as major constituent. In the EDS spectra some O_2 is there, which comes due to oxidation during milling. The SEM micrograph also shows that cementite particles are reasonably well distributed in Al matrix. There is a growth of particles composites hot pressed at 500 and 600°C.

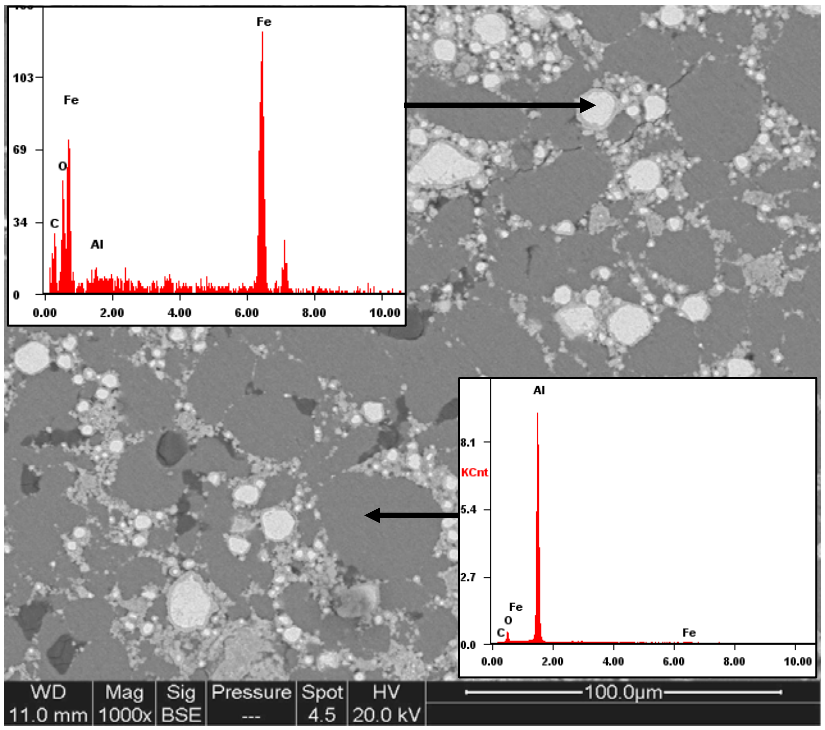

Figure 6. Representative SEM picture and EDS spectra of Al-20 vol. % Fe_3C hot pressed at 400°C for 30 min.

Interface study

The interface between the metal matrix and the particle plays a critical role in determining the overall properties of metal matrix composites. Strengthening by the hard particle reinforcement depends critically on the interfacial bond. A well-bonded interface facilitates the efficient transfer and distribution of load from matrix to the reinforcing phase, which lead to improved composite strengths. Conversely, load transfer becomes less effective with a weak bond, limiting the amount of strengthening that can take place. Therefore, it is concluded that a strengthened interfacial bond is responsible for the improvement in mechanical properties. Figure 7 shows the interface between Al and cementite particles of Al-20 vol. % Fe_3C composite hot pressed at 400°C for 30 minutes. It shows the good interfacial bonding between Al and Fe_3C particles.

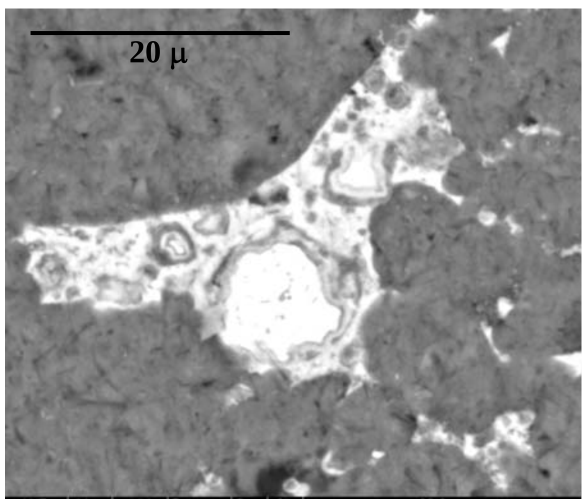

Figure 7. SEM picture of Al-20 vol. % Fe_3C hot pressed at 400°C for 30 min showing the interface between Al and Fe_3C particle.

Figure 8 shows the representative SEM micrograph of Al-20 vol. % Fe_3C composite cold compacted and sintered at 500°C for 1 hour. It was observed that composites prepared from milled and annealed powder by cold compaction and sintering shows interparticle cracks. It is clear from the micrograph that, there is poor compatibility between Al and Fe_3C particles. Whereas there is good compatibility between Al matrix and Fe_3C particles, due to simultaneous application of pressure and heat.

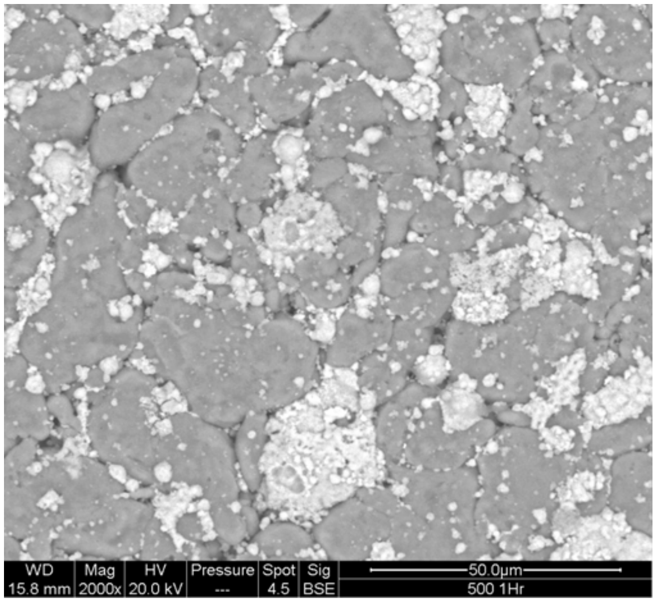

Figure 8. SEM micrograph of Al-20 vol. % Fe_3C cold compacted and sintered at 500°C for 1 hr.

Elemental mapping

Elemental mapping was used for displaying information about the elemental distribution in Al-Fe_3C composite. Figure 9 shows the elemental mapping of Al-20 vol. % Fe_3C composite. It shows the distribution of major components like Al and Fe in the matrix and second phase particles respectively.

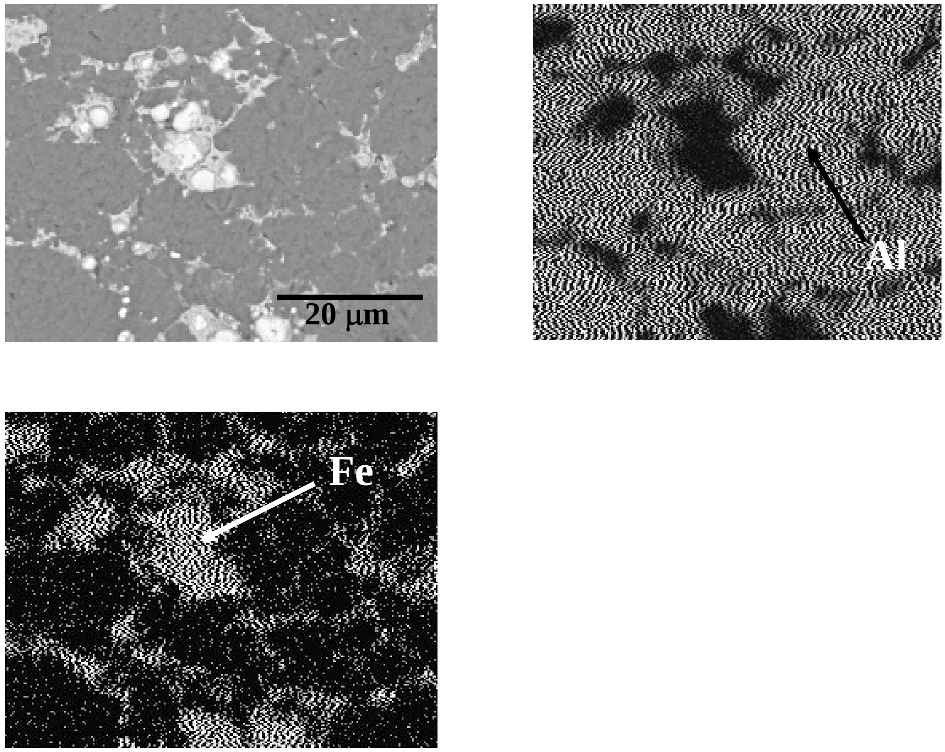

Figure 9. Elemental mapping of Al-20 vol. % Fe_3C composite hot pressed at 400°C for 30 min.

Hardness study

Table 1 shows the different processing schemes of sintering and corresponding hardness values of different samples. In case of Al-10 vol. % Fe_3C and Al-20 vol. % Fe_3C composites, hardness increases with increasing temperatures and with addition of Cementite. But hot pressing produces higher hardness than conventional sintering due to simultaneous application of pressure and heat.

Table 1. Different processing schemes of sintering and corresponding hardness values of Al-Fe_3C composite samples.

Samples	Processing schemes	Vickers Hardness
Al-10 vol.% Fe_3C	Sintered at 600°C for 1 hr.	43
Al-10 vol.% Fe_3C	Sintered at 600°C for 30 min.	31
Al-10 vol. % Fe_3C	Sintered at 500°C for 1 hr.	35
Al-10 vol. % Fe_3C	Sintered at 500°C for 30 min.	28
Al-20 vol.% Fe_3C	Hot pressed at 400°C for 30 min.	52
Al-20 vol.% Fe_3C	Sintered at 400°C for 1 hr.	30

CONCLUSIONS

The following conclusions can be drawn from the present investigation:

- Dual-drive planetary mill produces cementite and other iron carbides only in 40 hours, whereas commercially available planetary mills take more than 100 hours, as dual-drive planetary mill is more dynamic than commercially available mills.

- After 40 hours of milling, cementite and other iron carbides are present with elemental iron. After annealing only cementite is present, other iron carbides have undergone phase transformation. It is also observed that cementite becomes unstable at 800°C and decomposes into Fe and C.

- At the initial milling time, particles are flattened and flakes formed due to ductile nature of particles; with further milling particle size reduces and become spherical.

- It is made possible to synthesize Al-Fe_3C composite by hot pressing, and cold compaction and sintering. Hot pressing is better option than cold compaction and sintering. Hot pressing gives higher hardness due to better densification and good compatibility.

- There is good compatibility between Al matrix and cementite particles of hot pressed samples; no voids or cracks are present; which is clear from SEM micrograph. It is also observed that Fe_3C is reasonably well dispersed in Al matrix.

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