ON THE FEASIBILITY OF COMBINING MECHANICAL ALLOYING AND SPLAT COOLING TECHNIQUES FOR THE PRODUCTION OF Al-Fe ALLOY POWDERS

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الملخص

تم تصنيع جهاز يجمع بين ملامح كل من تقنيتي التسبيك الميكانيكي والتجميد الفائق السرعة واستخدامه لإنتاج مسحوق من سبيكة الالومنيوم المحتوية على 5.2% حديد مباشرة من المصهور، بين التحليل الغرابيلي أن نسبة الدقائق التي يقل مقياسها عن 2 ملم تشكل 71% من المسحوق المنتج. عرضت عينات من المسحوق المنتج ذات أحجام مختلفة لطحن ميكانيكي إضافي لفترات زمنية متزايدة باستخدام نفس الجهاز مع إضافة نسبة من كربيد السليكون (SiC) أو بدون تلك الإضافات وبين الفحص المجهرى لدقائق المسحوق أنه بزيادة الطحن يتحول شكل الدقائق من البيضاوي المستطال إلى الشكل القرصي المتراكب الطبقات مع زيادة كبيرة في الأبعاد المسطحة للأقراص . كما بينت دراسة حيود الأشعة السينية وأطياف موسباور أنه خلال الفترة الزمنية الابتدائية للطحن تتعرض دقائق المسحوق إلى تشوه ميكانيكي شديد مصحوباً بترسب بعض الدقائق الفائقة الصغر مشتته داخل دقائق المسحوق . وباستمرار عملية الطحن تتزايد نسبة ذرات الحديد الذائبة في المحلول الجامد ذي الأساس الألومينى ما يؤدى إلى تناقص في أبعاد الشبيكة البلورية مصطحباً بظهور دلائل على انفصال بعض الأطوار المغناطيسية على أطياف موسباور.

ABSTRACT

An apparatus, combining the features of both splat cooling and mechanical alloying, has been fabricated and utilized to produce an Al-5.2% Fe alloy powder directly from the melt. Sieve analysis of the produced powder revealed that the fractions with particle sizes below 2 mm constitute 79% of the powder. Powder samples of different sizes have been subjected to further mechanical milling with and without additions of SiC powder for increasing periods of time. Microstructure observation revealed that with increasing the milling time the powder morphology gradually changes form oblong to irregular layered disc-shaped with a marked increase in the disc planar dimensions. X-ray diffraction analysis and Mössbauer spectroscopy revealed that within the initial period of milling intensive mechanical deformation has taken place, probably accompanied by dispersion precipitation. With further milling, dissolution of Fe atoms into the Al-base solid solution takes place, giving rise to a decrease in the lattice parameter and magnetic splitting indications in Mössbauer spectra.

KEYWORDS: Splat cooling - Mechanical alloying - Al-alloy powders - Application of Mössbauer spectra.

INTRODUCTION

Equilibrium and near-equilibrium phases constitute the majority of the commonly used metallic materials [1]. Modern technology, however, requires new combinations of properties, that cannot be achieved by commonly produced alloys. Some of the proper candidates are non-equilibrium phases, which either may be stable for time intervals exceeding the service life of industrial articles, or may be stabilized by external means to increase their lifetime.

Mechanical milling utilizes high energy ball impacts to achieve structural modifications on an atomic scale. These may include dispersion-strengthening [2], widening of solid solubility range, ultra refining the basic structural size [3], creation of metastable inter-metallic compounds [4-6] and/or amorphous phases [7-8]. Recent applications of this technique include the production of nano-crystalline metals and alloys [9-10], formation of quasi- crystals, in- situ nitride and carbide formation and fabrication of micro- and nano-composites [9-10] .

Splat cooling is another technique in which solidification of alloy melts takes place under cooling rates exceeding 10^5-10^6 °C/sec, giving rise to high degrees of under cooling to obtain highly super-saturated solid solutions, frequently of very fine grain size, meta-stable intermediate phases or amorphous phases [11].

The aim of this work is to introduce a modified milling attritor, which combines the principles of both splat cooling and mechanical milling, to be used for powder production and milling from the liquid state. The preliminary results of utilizing this technique for producing Al-6%Fe alloy powder directly from the melt and some characteristics of this powder are also given.

EXPERIMENTAL

The modified attritor mill

An experimental rig, capable of receiving the alloy melt, performing its solidification at high cooling rate, transforming it into powder and further mechanically milling it, was designed, manufactured and assembled. As a basis for the rig attritor ball mill principle [2] was utilized, because it constitutes the highest dynamic effect, while may be easily modified to receive the alloy melt and effect splat cooling.

The ordinary attritor mill is composed of a massive stationary container tank with a firmly, sitting cover, through which a rotating impeller passes. The impeller is drived by a motor via a drive shaft. The impeller is equipped with two pairs of arms, welded perpendicular to the impeller axis at a fixed distance from each other and from the impeller free end, to which a scraper is welded. The axis of each pair of arms is at right angle to that of the neighbour pair or scraper. During impeller rotation the arms agitate balls of proper size and quantity to perform the milling action inside the container, while the scraper prevents the powder particles from settling at the container bottom.

The rotating impeller and the cover had to be modified to allow receiving the melt, transforming it into droplets that are directed towards the agitated impacting balls, where they solidify at high cooling rate.

Figure 1 shows the details of the modified impeller, where a truncated cone (3) with a cone angle in 60° is added, with its larger base downward and situated at a distance of 24 mm above the axis of the uppermost arms (2). While the impeller is rotating this cone repels the poured melt, spreads and scatters it into fine droplets which are then squeezed between the impacting cold balls to be cooled at a high rate.

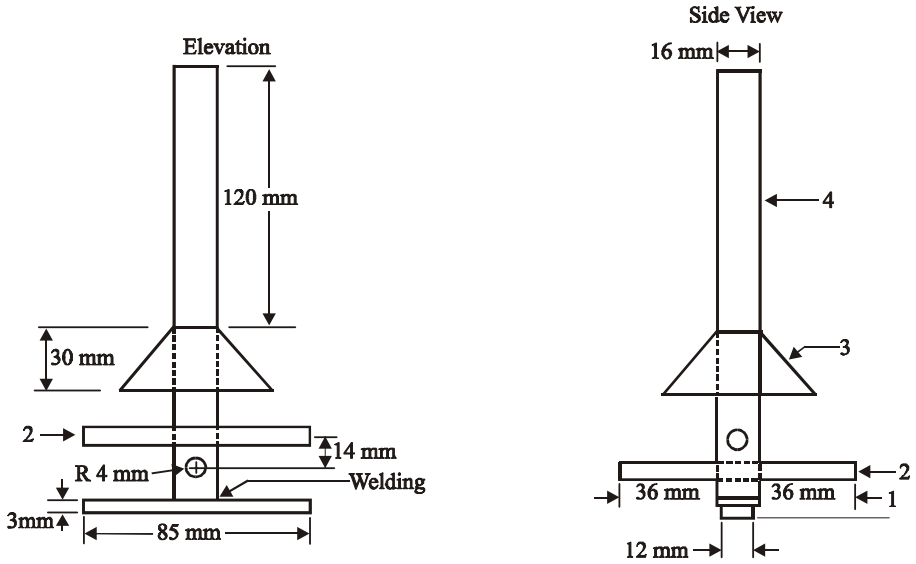


Figure 1: The Modified Rotating Impeller, (1) Scraper, (2) Pairs of arms, (3) Truncated cone, (4) Impeller shaft

To allow pouring the alloy melt into the container, while preventing any melt droplets or powder particles from coming out, a central hole 50 mm in diameter has been performed at the outer upper surface of the closing cover. This hole (Figure 2) widens downward to become 65 mm in diameter at the lower inner surface of the cover. To allow the cover to sit firmly on the upper container wall it was designed with an inner lip of the same diameter as the inner diameter of the container.

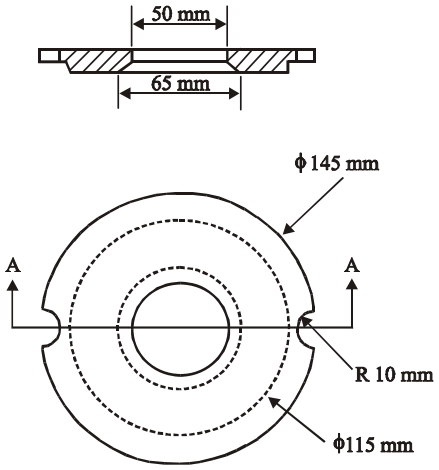


Figure 2: The Container Cover

The container proper has a cylindrical form with 102 mm inner diameter and 120 mm outer one. The total inner working highet of the container was taken 105 mm. The container, its cover and the impeller were fabricated of carbon steel, while high carbon, chromium ball bearing steel was utilized for the impeller arms and the impacting balls.

Experimental Procedure

To check the utility of the modified attritor mill for the production and milling of highly non-equilibrium powders directly from the melt Al-6%Fe alloy was utilized. The alloy was prepared by melting the proper amounts of commercially pure aluminum and mild steel turnings under a fluoride- chloride protecting flux [12] in a quenched graphite crucible by means of an electric resistance furnace. The melt was poured into a square steel mould, of which rectangular blocks $20 \times 20 \times 30$ mm were cut for further remelting. Table 1 introduces the chemical composition of these blocks.

Table 1: The chemical analysis of the as-cast alloy (wt %)

Cu	Fe	Mg	Mn	Ni	Si	Pb	Sn	Zn	Al
0.03	5.2	0.005	0.04	0.01	0.40	0.02	0.05	0.05	Bal

The modified attritor mill was assembled with the scraper bottom 2 mm above the bottom of the container. Forty seven bearing balls, each 17 mm in diameter, were charged into the container before closing and fixing it. The volume of the balls constituted 15% of the internal container volume.

Al- 6%Fe blocks were remelted in sintered alumina crucibles at a temperature of 890°C (i.e. 70°C above the liquidus temperature)

The mill was switched on at a rotating speed of 800 rpm for two minutes during which the melt was poured.

The product of this process had a powder form. It was weighed and subjected to sieve analysis between sieves of 8 mesh (2000 μm) up to 100mesh (150 μm). Table 2 shows the result of this sieve analysis.

Table 2: Sieve analysis of the produced powder

Mesh No.	Mesh size (μm)	Mass of powder (gr)		Yield%
		Retained	Passed	
8	2000	67.0	252	79
16	1000	85.0	167	52.35
30	500	94.2	72.8	22.82
50	300	33.4	39.4	12.35
100	150	23.8	15.6	4.89

Relying on the high speed of the flying droplets and the squeezing effect performed on them by the cold impacting balls, it was thought that the as-solidified powder product consisted mainly of supersaturated solid solution of iron in aluminum. This

effect was monitored qualitatively by x-ray Diffraction. However, no attempt has been performed to quantify the degree of super-cooling.

To check the capability of the equipment for mechanical milling, half the powder with particle size 500 μm < d < 1000 μm has been subjected to further milling for 18 minutes. Half the powder product of the later step was further milled for 20 minutes; thus we had three powder products of approximately the same starting size subjected to milling for 2, 20 and 40 minutes, respectively. Samples of these products were characterized by optical microscopy, X-ray diffraction analysis as well as Mössbauer spectroscopy.

The powder fraction of size 150 μm < d < 300 μm was utilized to investigate the possibility of mechananical dispersoid alloying of the produced powder by SiC particles. 90% of this powder was mixed with 10 % (by weight) of SiC powder sieved to below 200 mesh size (~75 μm). The blended powder was subjected to further 10 minutes of milling. Two thirds of this powder was further milled for 20 minuets. Half of the last product was milled for additional 30 minuets. These three products were also characterized by the same techniques as above.

RESULTS AND DISCUSSION

As-solidified powder characterization

The results of the sieve analysis given in Table 2 indicate that the as- solidified powder may be classified as an aggregate of coarse powder with 20% granules (above 2 mm) .The powder coarseness may be attributed to the large size of the balls used in the attritor mill. The larger the balls, the larger are the voids between them, available for droplets solidification. This gives the opportunity to control the powder average size via the selection of the suitable ball size.

Figure 3 introduces representative micrographs of the powder fraction (+500-1000 μm) which constitutes ~30% of the as-solidified powder. The particles are generally of oblong morphology with some cracks at their surfaces. Occasionally, some evidence of impact welding of more than one particle may be observed. This is probably the result of the two minutes of milling, following the melt pouring into the mill. The phase content of the powder was followed by X-ray diffraction. Part of the diffraction pattern, representing the powder fraction (+500-1000 μm) is shown in Figure 4. Peaks at 2Θ = 38.7°and 44.9° correspond to {111} and {200} reflections of the Al-base solid solution, respectively. The peaks at 2Θ =34.2° and 44.4°are two of the three strongest reflections of Fe Al_3 [13] .The powder, thus, includes Al- base solid solution and Fe Al_3. Calculation of the lattice parameter of the solid solution, extrapolated to Θ = 90° [14] indicate an average distance of nearest approach between atoms in 2.8588 Å. Taking into account that the corresponding distance in pure Al is 2.8636 Å and in the equilibrium saturated solid solution of Fe in Al is 2.8634 (the radius of Fe atom =1.241Å is much smaller than that of Al = 1.43 Å) it can be concluded that the solid solution constituent of the powder aggregate is highly super-saturated with Fe atoms, as is always the case with splat-cooled alloys. X-ray diffraction patterns, representing other powder size fractions, showed identical results, indicating that super- saturation does not depend on the particle size.

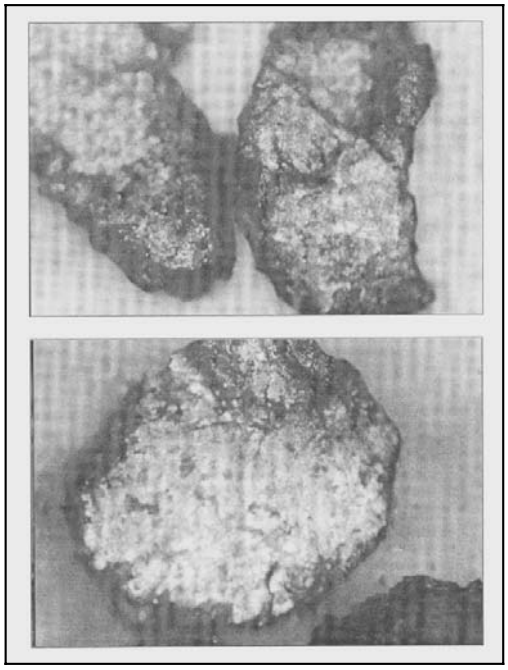


Figure 3: Typical Micrographs of Powder size (+ 500 -1000 µm) x100

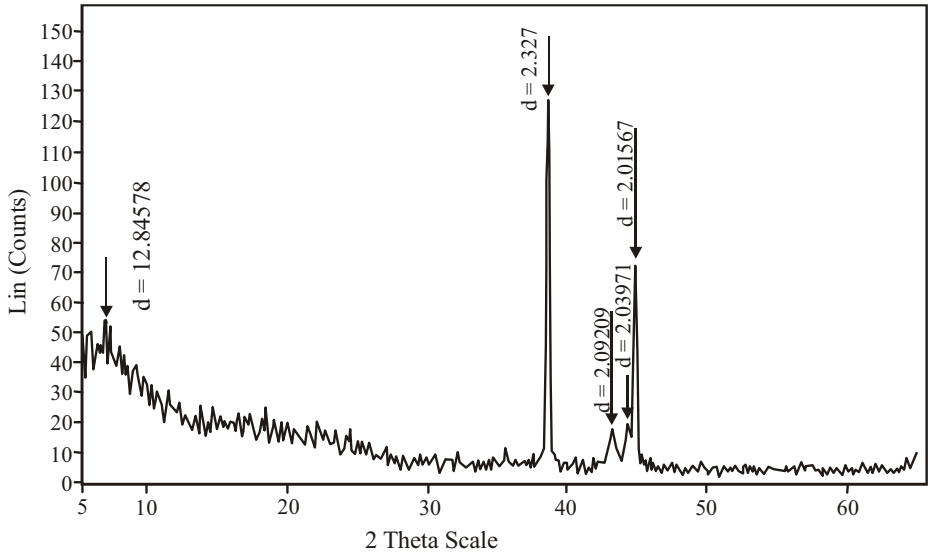


Figure 4: Portion of x-ray diffraction chart of the as produced powder (Fraction + 500 -1000 µm)

To determine the state of iron atoms in as-solidified powder and its evolution with milling Mössbauer spectra of absorption of the 14.4 gamma rays of Co^{57} diffused into Cr-plate was measured at room temperature. Figure 5 shows such a spectrum for the as-solidified powder. It consists of one doublet of isomer shift 0.215 mm/s and quadropole splitting 0.321 mm/s. These data are typical of Fe atoms in the Fe^{+3} state [15 .It may be thought that Fe atoms can occupy extra positions in the Al- base solid solution only if they give three electrons to the electron gas (as do Al atoms) to retain the stability of the structure. This supports the conclusion, that the solid solution is highly super-saturated with Fe atoms due to the splat cooling effect.

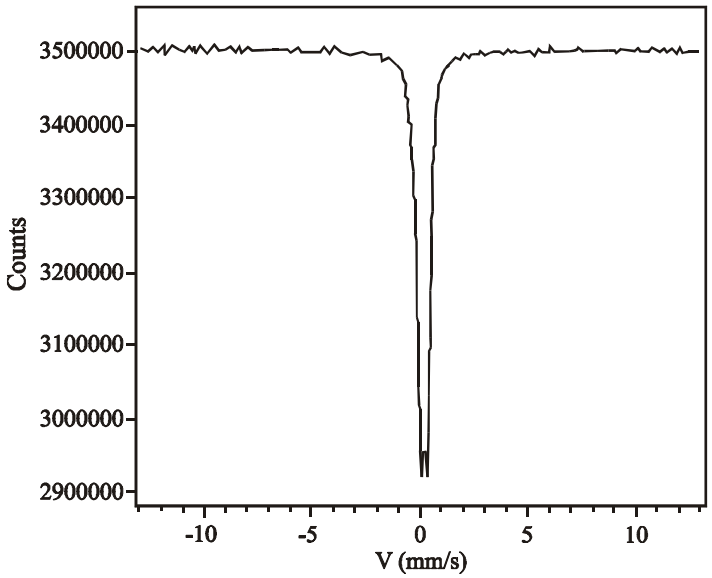
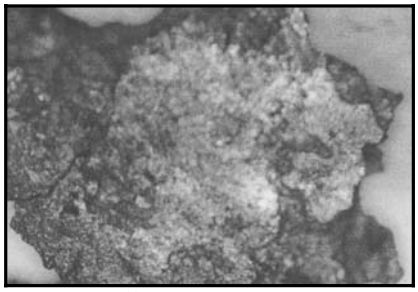


Figure 5: Mössbauer Spectrum of the as produced powder (Fraction +500-1000 µm)

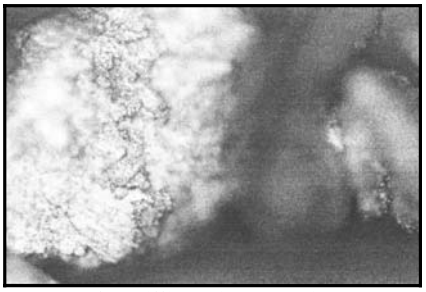
Effect of further mechanical milling

During subsequent milling the powder particles markedly change both their shape and size. Figure 6 presents typical photographs of particles milled for 20 and 40 minutes. After 20 minutes of milling the particles are flattened and impact welded together forming disc- shaped layered particles of large size with numerous cracks (Figure 6-a). With increasing the milling time to 40 minutes the particles become multi-layered much smaller in size (Figure 6-b).The evolution of the particles morphology with milling time is indicative of the repeated processes of deformation, flattening, welding, cracking and fracturing, which take place during multi- body impacts.

The interplanar spacing of the indexed diffracting planes of the identified phases on similar portions of the diffraction patterns of powder samples milled for 20 and 40 minutes are shown in Table 3. The same peaks are observed for the Al- based solid solution, but with a continuous decrease in the average nearest distance of approach between atoms. This is evident of continuously increasing replacement of smaller Fe atoms for the larger Al ones, i.e. increasing supersaturation of the solution with Fe atoms.



(a) Milling Time 20 min. x 50



(b) Milling Time 40 min. x 100

Figure 6: Representative particles of milled powder (Fraction + 500 - 1000 μm)

Table 3: Change in the lattice parameter and nearest distance of approach in the Al- base solid solution with milling time.

Milling time, min.	Interplaner distance, Å		Average lattice parameter, Å	Average nearest distance of approach, Å
	d_{111}	d_{200}		
2	2.3270	2.0159	4.0436	2.8588
20	2.3235	2.0133	4.0403	2.8565
40	2.3165	2.0055	4.0315	2.8503

The Mössbauer spectrum of the powder subjected to 40 minutes of milling showed the same doublet as for the as-solidified powder, but with very weak indications of some magnetic effects at the base line, indicating a decreasing distance between Fe atoms in the highly super-saturated solid solution, leading to a tendency for residual magnetic alignment. This supports the result of the X-ray diffraction analysis, concerning the increase of super-saturation with milling time.

Mechanical dispersed alloying

Milling of the as-solidified powder fraction of size(+150 μm-300 μm), blended with SiC powder for increasing time intervals led to flattening, with numerous welding of two or more, frequent cracking and indications of embeded SiC particles. The dimensions of the resulting disc-shaped particles (Figure 7) reached 2 mm after 30 minutes of milling and 3mm after 60 minutes. The interplaner spacings of {111} and {200} of the Al-base solid solution, calculated from the diffraction patterns of the milled blended powders, are given in Table 4, together with the lattice parameters and the nearest neighbors separation distances. The first 30 minutes of milling lead to a gradual increase of the interatomic spacing. This may be attributed to intensive plastic deformation and/or precipitation of some excess super-saturating atoms (i.e. decreasing super-saturation). However, with increasing the milling time to 60 minutes a drastic drop of the nearest neighbors approach distance takes place, indicating intensive redissolution of Fe atoms into the solid solution, effecting a high degree of super-saturation. The difference in behavior between this case and that of mechanical milling of the unblended coarser particles is not enough understood and should be looked for in the deformation behavior and the presence of SiC particles.

Error!

Figure 7: Features of a large disc- Shaped particle x200 showing cracked layered structure with indication of SiC

Table 4: Summery x-ray of diffaction data for powder samples milled with SiC for various time intervals.

Milling time, min.	D values, Å		Calculated lattice parameter, Å	Nearest nighbour separation, Å
	d_{111}	d_{200}	4.0481	2.8620
10	2.3307	2.0211		
30	2.3450	2.0317	4.0525	2.8651
60	2.3094	2.0081	4.0280	2.8478

Mössbauer spectra of the milled dispersoid blended powder undergo corresponding evolution. After 10 minutes of milling the spectrum shows the same doublet as that of the as- solidified powder. After 30 minutes a singlet has replaced the doublet, indicating wide separation of the Fe atoms approaching the equilibrium solubility limit.(Fe^{+3} ions disappear). On increasing the milling time to 60 minutes the spectrum (Figure 8) changes drastically: the doublet is restored and a clear indication of a magnetic effect appears at the base line.

These changes may be attributed to the high supersaturation of the solid solution with Fe atoms, with the separation between them becoming so small that their residual magnetic moments may aligne.

The zones, where such alignment takes place, constitute ~7% of the volume of the solid solution. These observations and conclusions are completely in agreement with the results of the X-ray diffraction analysis reported above.

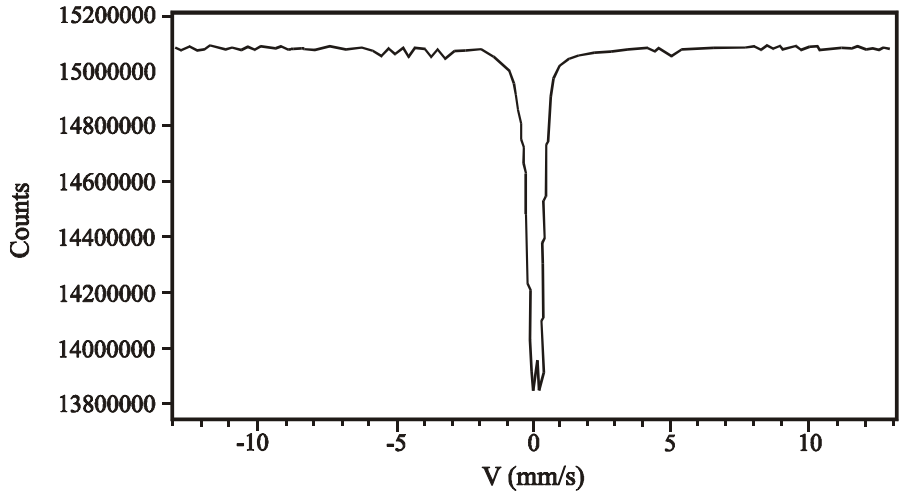


Figure 8: Mössbauer Spectrum of blended powder, milling time – 60 min.

CONCLUSIONS

A solidification-milling apparatus, combining splat cooling and mechanical alloying has been fabricated and utilized to transform Al- 5.2% Fe alloy melt diretly into super-cooled coarse powder aggregate, ready for further milling and alloying. The yield constitutes 79% of powder size below 2 mm of oblong morphology. With further milling the powder changes its morphology into layered disc-shape with gradual increasing the super-saturation of its Al-base solid solution with Fe. Mechanical alloying of the produced powder with SiC powder slightly altered the initial phase of powder evolution.

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