Effect of ECAP and extrusion on particle distribution in Al-nano-Al₂O₃ composite

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Abstract. In this study equal channel angular pressing (ECAP) and conventional extrusion were used as two different techniques for consolidation of attritioned aluminium powder (45 µm) with varying concentrations of nanoalumina powders (35 nm). The evolution of the homogeneity of the particle distribution in the material during ECAP and conventional extrusion was investigated by the quadrat method with image analysis software. The frequency histograms of the number of alumina particles per quadrat, N_q , for each investigated condition were plotted and the effect of particle distribution on fracture surface and wear resistance of the composite was investigated.

Keywords. Powder metallurgy; composite; ECAP; extrusion; particle distribution.

1. Introduction

Metal matrix composites (MMCs) are under attention for many applications in aerospace, defense and automotive industries. Among MMCs, aluminium MMCs are being considered as a group of new advanced materials due to its light weight, high strength, low thermal expansion coefficient and good wear resistance.¹ Reinforcing the ductile aluminium matrix with stronger and stiffer second phase reinforcements like oxides and carbides provides a combination of properties of the metallic matrix and ceramic reinforcement components.²

Conventional consolidation techniques are capable of consolidating nanopowders by using heat and pressure. However, they usually result in grain growth, which leads to loss of the nanostructure.³ Severe plastic deformation (SPD) techniques can consolidate the powders at room temperature, decreasing the potential for grain growth.⁴ Equal-channel angular pressing (ECAP), also termed as equal-channel angular extrusion is the most widely practiced among the various SPD techniques, in which a well-lubricated metallic billet or ingot is pressed by a ram through a die constrained within two intersecting channels which are placed at an abrupt angle that is equal to or close to 90°. ECAP induces intense plastic strains into massive billets without changing their cross-section.^{8,9}

The distribution of reinforcement should be homogeneous throughout the matrix in order to obtain isotropic property in the composite. Traditional secondary deformation processes such as rolling, forging and extrusion

are not very efficient in achieving uniform distribution of second phase particles. The problem becomes more aggravated in case of finer second phase particles, because it requires very high amount of strain to accomplish homogeneous dispersion in the matrix. As a huge amount of deformation can be induced, ECAP can be useful in synthesizing MMCs with uniform distribution of second phase particles in the matrix. In the regions of the particle clusters, high local stress triaxialities are found. It was proposed that the fracture processes are accelerated by these high local stresses leading to fracture initiation in particle clusters.^{10,11} According to Christman *et al*¹² the particle clustering leads to a decreasing composite flow stress, in comparison to a composite with homogeneous particle distribution. Based on the short overview, it is obvious that one of the most important microstructural features of MMCs is the spatial distribution of the particles. Hence it seems rational to use more efficient and reliable methods for better particle distribution in MMCs, which causes better utilization of materials. In this study, efforts were duly made in utilizing a novel method (ECAP) for better distribution of hard alumina reinforcement in the aluminium matrix and comparing with the conventional extrusion.

2. Experimental

High-purity aluminium powders with an average particle size of about 45 µm were used in this study. Scanning electron microscopy (SEM) micrograph of the as-received gas atomized aluminium particles, which exhibits an irregular shape, is presented in figure 1a.

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In order to make a MMC with Al_2O_3 reinforcement, $\alpha\text{-Al}_2\text{O}_3$ powder with an average particle size of 35 nm with purity of 99.9% was utilized. Then the powder mixtures were wet attritioned in pure ethanol slurry in a container with steel balls with a diameter of 5 mm and a rotational speed of 400 rpm for about 500 min. During the attritioning process the ethanol was removed from the mixture by a pump, and after the removal of the ethanol the powder mixture was dried in an oven at 150°C for 90 min and then sieved to eliminate the impurities arising from Teflon container. The sieving was conducted by different mesh sizes (100, 120, 140, 170, 200, 230, 270, 325, 400). Most of white Teflon impurities were larger than $120\text{ }\mu\text{m}$, which retain on the first two sieves, whereas the composite powder goes through these two sieves. Figure 1b shows the flaky shape composite powder after attritioning. The copper tubes were filled with aluminium composite powders. During hand pressing the powder was gradually added by a funnel and simultaneously pressed by hand and then new powder added to the tube to fill the empty volume. Then the tubes were subjected to equal channel angular pressing at 200°C for different ECAP passes with route ' B_C '. In route ' B_C ', the tube is rotated

90° in the same direction after each pass. The dimension of tubes before ECAP and the tubes after different ECAP passes is shown in figure 2. As it is seen in figure 3, the inner and outer angle of the ECAP die and the cross-section of the die was $\varphi = 90^\circ$, $\psi = 20^\circ$ and $\phi = 13\text{ mm}$, respectively.

Figure 1. SEM micrographs of the (a) as-received gas atomized aluminium particles and (b) attritioned composite powder.

Figure 2. Dimension of tubes before ECAP, and the tubes after different ECAP passes.

Figure 3. Inner and outer angle of the ECAP die and the cross-section of the die.

The ECAP results were also compared with the conventional extrusion for aluminium composite powder. Some of the attritioned powders were compacted uniaxially in a rigid die at compaction pressure of 200 MPa, and then the compacted powder with the initial diameter of 45 mm was sintered at 540°C in controlled atmosphere for about 60 min followed by extrusion in one step at the ratio of 20:1. In order to have a clear evidence for particle distribution besides investigating the polished surface of samples, the composite specimens were broken by an impact load using Izod test at room temperature and the fracture surfaces were investigated by SEM. The densities of the consolidated samples were determined using Archimedes' principle, according to specifications for density measurements for porous materials. Relative densities are based on the measured values and the theoretical density. Table 1 presents the relative density for green body before ECAP and extrusion and also after 4th pass of ECAP and extrusion.

The pin-on-disc test is a classical method commonly used for adhesive wear experiments. During the experiment, the sliding between the pin and disc may result in wear on both contact surfaces of the pair. In this study, the aluminium MMC was used as the pin and the heat-treated 4041 steel with a hardness of 58 HRC as the counterface. The test parameters were: normal loads on the pin, 10 N, sliding velocity of 0.03 m s⁻¹ and total sliding distance, 300 m.

To improve reinforcement distribution in particle MMCs, a reliable method for quantifying such a distribution needs to be identified, and a number of different approaches have been used in the past. For example, Lewandowski *et al.*¹³ studied the relation between particle size and spatial distribution on the damage accumulation which leads to fracture. The uniformity of Al₂O_{3p} particle distribution in the MMC was studied by the quadrat method.^{14,15} According to Karnezis *et al.*¹⁴ quadrat method is the most effective method for investigating the reinforcement distribution in MMCs with clustered particle distribution. Hence, it was chosen from many different methods.¹⁶

With the Quadrat method, the images were divided into a grid of square cells and the number of particles in each cell, N_q , was counted. Generally, a clustered distribution would be expected to produce a combination of empty quadrats, quadrats with a small number of particles, and

quadrats with many particles. A random distribution would be expected to produce results somewhere in between these two extremes.¹⁵ The major problem of the quadrat method is determining the optimal quadrat size, which is usually square for simplicity. Non-randomness is dependent on the size and shape of the quadrats. A very small quadrat will detect many particle-free areas, falsely indicating clustering, whereas a very large quadrat will tend to give the same number of particles in each, irrespective of clustering effects. A simple rule used in this study is that the appropriate square quadrat size should be approximately twice the size of the mean area per particle.¹⁷ To minimize the edge effects, the total number of particles completely inside and in contact with the left and bottom quadrat sides are counted. Thus, particles on the quadrat edge were not counted more than once. The quadrat method is evidently an effective method of detecting variations in the distribution of Al₂O₃ particles in MMCs induced by processing. The results from the quadrat analysis can also be compared with theoretical random or clustered distributions defined by statistical relation. Random and clustered distributions of particles can be expressed in mathematical terms if the particles are assumed to be points (i.e., their cross-sectional areas are zero). It has been proved mathematically that a random spatial distribution of points follows the Poisson model, a regular spatial distribution follows the binomial model, and a clustered spatial distribution follows the negative binomial model.

The images were divided into a grid of square cells and the number of particles in each cell, N_q , was counted. SEM images were taken at a magnification of 10,000 from the normal polished section of 10 and 15 vol% composites after the 1st and the 4th pass of ECAP. Each image was divided into 117 quadrats. The size of each quadrat was set to $a = 0.89 \mu\text{m}$, as the optimum quadrat size should be twice the size of the mean area per particle. The experimental particle number per quadrat distributions were compared to two theoretical distributions: (i) the Poisson distribution (equation 1), which should be valid for a homogeneous particle distribution

$$P(r) = \left(\frac{\mu^r}{r!} \right) \exp(-\mu). \quad (1)$$

Table 1. Relative density for the specimens before and after ECAP and extrusion.

	Relative density (%)		
	Al-10 vol% Al ₂ O ₃	Al-15 vol% Al ₂ O ₃	Extruded Al-15 vol% Al ₂ O ₃
Green body before ECAP/extrusion	45.5	43.8	71.5
After 4th pass of ECAP	97.8	92.7	—
After extrusion	—	—	99.4

(ii) The negative binomial distribution (equation 2), which corresponds to a clustered particle distribution

$$P(r) = \left(\frac{(k+r-1)!}{(k-1)!r!} \right) \left(\frac{p}{1+p} \right)^r \left(\frac{1}{1+p} \right)^k. \quad (2)$$

In the above equations, $P(r)$ denotes the probability, r is a variable (the number of particles per quadrat), μ the mean value of the number of particles per quadrat, k and p the two parameters determined according to Rogers.¹⁵

k measures the degree of clustering associated with the process. As k approaches to zero, the distribution converges on an exceptionally clustered distribution. In our study, after dividing the images into 117 quadrats, the number of particles in each cell was counted and the number of total particles and the mean number of particles was determined. The Poisson distribution and the negative binomial distribution were determined for each image.

The study of microstructures reveals that the shape of the N_q distribution varies significantly with the degree of clustering. Indeed, increasing the size of Al_2O_3 clusters leads to less symmetric N_q distribution. The degree of asymmetry of a statistical distribution around its mean can be quantified by its skewness, β , which is defined by

$$\beta = \frac{q}{(q-1)(q-2)} \sum \left(\frac{N_{q_i} - N_q^{\text{mean}}}{\sigma} \right)^3, \quad (3)$$

where q is the total number of quadrats, N_{q_i} the number of Al_2O_3 particles in the i th quadrat ($i = 1, 2, \dots, q$), N_q^{mean} the mean number of Al_2O_3 particles per quadrat and σ the standard deviation of the N_q distribution.

3. Discussion

Figure 4 presents the TEM micrograph of the composite after mixing. As it is seen, some clusters (shown by arrow) are present in the pre-processed composite powder with a non-homogeneous alumina particle distribution in the matrix.

The frequency histograms of the number of alumina particles per quadrat (N_q) after the 1st pass and after the 4th pass of ECAP are plotted in figures 5 and 6 for Al–10 vol% Al_2O_3 and Al–15 vol% Al_2O_3 composites, respectively. As it is seen, the frequency of empty quadrats decreases with the increase in the number of ECAP passes.

According to table 2, the skewness does not change significantly for 10 vol% composites where the amount of clustering is less. The skewness decreases from about 0.81 after the 1st pass to 0.53 after the 4th pass of ECAP for 15 vol% composite. This is a positive sign for particle declustering.

Figure 4. TEM micrograph of the composite after mixing and before consolidation.

Figure 5. Frequency histograms of the number of alumina particles per quadrat, N_q , for Al–10 vol% Al_2O_3 composite (a) after the 1st and (b) 4th pass of ECAP.

However, the histogram for Al-15 vol% Al_2O_3 composites seems to follow the negative binomial distribution (figure 6a,b), indicating a clustered particle distribution.

Figure 6. Frequency histograms of the number of alumina particles per quadrat, N_q , for Al-15 vol% Al_2O_3 composite (a) after the 1st and (b) 4th pass.

Table 2. Clustering skewness for the composites after each passes of ECAP.

Pass #	σ	β
Al-10 vol% Al_2O_3		
1st	0.959	0.79
2nd	0.945	0.72
3rd	0.942	0.715
4th	0.930	0.71
Al-15 vol% Al_2O_3		
1st	0.888	0.81
2nd	0.869	0.73
3rd	0.820	0.61
4th	0.814	0.53

The histogram of the particle distribution for Al-10 vol% Al_2O_3 after the 1st pass of ECAP (figure 5a) resembles the negative binomial distribution, but by increasing the number of ECAP passes the distribution converges to the Poisson distribution. Although four passes of ECAP is not enough for Al-15 vol% Al_2O_3 to fit the data to the Poisson distribution, the number of empty quadrats and the ones containing four particles decrease from 26 to 21 and 4 to 1, from the 1st pass to the 4th pass, respectively. This is a sign for better distribution of the particles. It can be concluded from these histograms that large alumina particles break to some extent, specially for Al-15 vol% Al_2O_3 composite during ECAP, and shear deformation can move some particles in the microstructure and contribute to better distribution. However, this effect diminishes as the amount and the size of alumina particles decreases, because of this fact that small particles need a great active plastic flow, which is not obtained during ECAPressing. In order to understand how much the alumina particles contribute to strengthening, image analysis with image tool software was conducted on the SEM micrographs of Al-15 vol% Al_2O_3 composite, and the frequency was depicted against alumina particle size. It is obvious from figure 7a that only about 23% of alumina particles are below 300 nm after the 1st pass, whereas after the 4th pass of ECAP (figure 7b) 54% of alumina particles lay in this category. This indicates higher contribution of the nanoalumina particles in composite strengthening. This analysis also proves the decrease in skewness and in turn improving alumina declustering by increasing the number of ECAP passes.

Figure 7. Frequency against alumina particle size for Al-15 vol% Al_2O_3 composite (a) after the 1st and (b) 4th pass.

Figure 8a and c presents the SEM micrograph of the Al–15 vol% Al_2O_3 composite after the 4th pass of ECAP, after the extrusion and after the 3rd pass of ECAP, respectively. As it is seen, better distribution of alumina particles (solid arrows) is achieved after four passes of ECAP in comparison to conventional extrusion, where clustered particles are still obvious in the microstructure. The dashed arrows in figure 8a show the porosity in the microstructure in the composite after the 4th pass of ECAP. Detailed microstructural investigations show that the alumina particles break during the ECAP process and move to free particle zones simultaneously with the elimination of porosity by increasing the number of ECAP passes. This is more obvious in 15 vol% Al_2O_3 composite due to higher amount of particle agglomeration in comparison to 10 vol% Al_2O_3 composite, which is less agglomerated. According to table 1, elimination of pores leads to density enhancement. This increase in density is more remarkable for the ECAPed specimens than the extruded ones.

It has been reported that¹⁸ in conventional extrusion, it is usually necessary to have an extrusion ratio (ER) greater than 10 to ensure a fully coherent product. Even higher ratios may be needed for better diffusion. The relationship between the ER (initial area/final area) and the homogenized strain resulting from that extrusion is a logarithmic one so that as the desired strain is increased the required ER must increase exponentially. As a consequence of this, the loads that are required also increase

dramatically if the final part size is to be maintained. ECAP is a relatively new process that can impart a high amount of strain (~ 1) for each pass, as material is deformed by simple shear in a thin layer as it passes from a vertical to a horizontal channel. According to Bettles,¹⁸ for achieving the same total strain in ECAP and extrusion with ratio of 20:1, one should use three passes of ECAP. SEM micrograph of the extruded sample and the one ECAPed for three passes are compared in figure 8b and c. One can see better alumina distribution and smaller clusters in figure 8c. This is a positive sign for the effectiveness of ECAP process than conventional extrusion.

Stress concentrations in local clusters origin from the repetitive loading of the tubes when they undergo shear deformation at the intersection of the entry and exit channels. So, it is rational to depict the required force for particle declustering during ECAP, which in turn produces stress and accumulated strain. Figure 9a and b shows the force–displacement curve for 15 vol% Al_2O_3 composite after the 1st and 4th pass of ECAP, respectively. As it is seen, the level of required force increases by increasing the number of passes due to strain hardening of the tube material and the consolidated powder. According to this figure, the maximum required force is nearly two times greater for the 4th pass than the 1st pass.

In order to investigate the alumina distribution, it is interesting to see the fracture surface of the Al–15 vol% Al_2O_3 composite after the 4th pass of ECAP and also

Figure 8. SEM micrographs of the Al–15 vol% Al_2O_3 composite (a) after the 4th pass of ECAP, (b) after the extrusion and (c) after the 3rd pass of ECAP.

Figure 9. Force–displacement curve for 15 vol% Al_2O_3 composite (a) after the 1st and (b) 4th pass of ECAP.

Figure 10. Fracture surface of the Al-15 vol% Al_2O_3 composite (a) after the 4th pass of ECAP and (b) after the extrusion.

after the extrusion in figure 10a and b. It is obvious that ECAP has a prominent effect on alumina distribution due to more homogeneous decohesion of alumina particles from the matrix. The fracture surface for the extruded

sample is not as homogeneous as the ECAPed one, which is a sign for random detachment of alumina particles from the matrix and in fact improper distribution of alumina in the matrix material.

Figure 11. Wear surface of Al-15 vol% Al_2O_3 composite (a) after the 4th pass of ECAP and (b) after the extrusion at 10 N load and 300 m sliding distance.

Figure 11a and b shows the wear surface of Al-15 vol% Al_2O_3 composite after the 4th pass of ECAP and after the extrusion at 10 N load and 300 m sliding distance. When alumina particles distribute homogeneously in the matrix, as it is seen in ECAPed sample, the adhesive area and large grooves disappear and a smoother surface is formed in comparison to the extruded sample, which is composed of plastic deformation and material detachment from the surface and also some grooving.

4. Conclusion

In this study the effect of distribution of particle reinforcement in the matrix on properties of Al/ Al_2O_3 composite was investigated by the quadrat method. Two different processes (ECAP and conventional extrusion) were applied on the attritioned composite powder. The results show that the ECAPed samples after four passes have better distribution of particles and in turn have higher wear resistance. Among the ECAPed samples, the variation in skewness is more remarkable for Al-15 vol% Al_2O_3 composite than Al-10 vol% Al_2O_3 composite. This is because of higher amount of alumina agglomeration in Al-15 vol% Al_2O_3 composite, which causes easier disintegration of alumina particles from the surface of the clusters and their movement to particle-free zones. Increasing the number of ECAP passes increases the contribution of particles below 300 nm in strengthening by more than two times. Fracture surface shows that ECAP has a prominent effect on alumina distribution than conventional extrusion due to more homogeneous decohesion of alumina particles from the matrix.

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