JOURNAL OF MATERIALS SCIENCE **37** (2002) 4603–4607

# Extrusion of mechanically milled composite powders

J. B. FOGAGNOLO
*Department of Manufacturing Engineering, Mechanical Engineering Faculty, State University of Campinas, Cidade Universitária, 13083-970, Campinas-SP, Brazil; Materials Science and Engineering Department, Universidad Carlos III de Madrid, Avenida de la Universidad, 30, E-28911, Leganés, Spain*
*E-mail: jfoga@ing.uc3m.es*

M. H. ROBERT
*Department of Manufacturing Engineering, Mechanical Engineering Faculty, State University of Campinas, Cidade Universitária, 13083-970, Campinas-SP, Brazil*

E. M. RUIZ-NAVAS, J. M. TORRALBA
*Materials Science and Engineering Department, Universidad Carlos III de Madrid, Avenida de la Universidad, 30, E-28911, Leganés, Spain*

Mechanical alloying has come to the fore in fields such as the production of intermetallic compounds, supersaturated solid solutions, amorphous materials and metal matrix composites. With this process, composite powders are obtained with characteristics that are impossible to achieve employing conventional powder metallurgy techniques. In this work, aluminium powder AA6061 is mixed with silicon and aluminium nitrides in a conventional mixer and in a high-energy ball mill to obtain composite powders that are subsequently uniaxially cold pressed and hot extruded. The necessary pressure to extrude the composite cold pressed powders varies with the powder condition, being lower for the composite powder conventionally mixed, higher for the composite powder after a short time of milling, and intermediate after a longer time of milling, due to the morphological and structural changes typical of the mechanical alloying process.


## 1. Introduction

The synthesis of materials by high-energy ball milling of powders was first developed by John Benjamin and his co-workers [1] at the International Nickel Company. They successfully produced fine, uniform dispersions of oxide particles ($Al_2O_3$, $Y_2O_3$ and $ThO_2$) in nickel-base superalloys, which could not be made by conventional powder metallurgy methods. This process is now being used to produce numerous materials and alloys, including supersaturated solid solutions, amorphous materials, intermetallic compounds and metal matrix composites [2–6].

The process of mechanical alloying consists of repeated welding-fracture-welding of a mixture of powder particles in a high-energy ball mill. The central event is that the powder particles, trapped between the colliding balls during milling, undergo deformation and/or fracture. In mechanical alloying of at least one ductile component, there is an initial stage when deformation dominates the process, followed by a stage in which welding is predominant. After a certain period of milling, the powder is hard enough for fracture to dominate. At the end of the process, the powder reaches a steady state characterised by equilibrium between welding and fracture [7–9].

The use of hot extrusion in powder metallurgy avoids the sintering process and results in a full density final product. Hot extrusion of powders allows a high shear strain rate, providing a high strength bonding between particles and a microstructure very similar to that of a wrought product. In the case of aluminium and its alloys, hot extrusion breaks the typical oxide layer that coats the powder, providing better bonding of the particles [10]. In MMCs, hot extrusion tends to eliminate the clustering of reinforcement particles and therefore achieves a more uniform distribution of particles within the metal matrix [11].

This work investigates the extrudability of an aluminium AA6061 matrix composite reinforced with particles of silicon and aluminium nitrides, mixed in a conventional and a high-energy ball mill for different lengths of time.

## 2. Experimental

AA6061 prealloyed aluminium alloy powder (of 75 $\mu$m average particle size) was used as the matrix. Aluminium nitride (of 8.0 $\mu$m average particle size) and silicon nitride (of 8.6 $\mu$m average particle size) were used as reinforcement particles. Three different compositions were performed: 5% and 15% by weight of AlN and 5% by weight of $Si_3N_4$.

The aluminium alloy and the reinforcement powders were firstly mixed in a conventional low energy ball mill for 1.5 hours and, subsequently, in a high-energy centrifugal ball mill (*Fritshc Gmbh*, *Pulverisette 6*) for 1.5, 3, 4.5, 6, 8 and 10 h. The ball's powder weight ratio was 6 to 1 and the rotation speed was 700 rpm. 1% by weight of Microwax C (Hoechst) was used as the process control agent (PCA).

The powders were then characterised by scanning electron microscopy (SEM). Also, the particle size distribution was determined for selected powders by LASER diffraction (Malvern, model Master Sizer S, England); three measurements were carried out for each sample.

The composite powders were then uniaxially cold compacted at 300 MPa followed by hot extrusion without canning and degassing. The extrusion ratio was 25 : 1, the temperature was 500°C, and the extrusion rate 30 cm/min. The necessary pressure to extrude the material was monitored. Reported values of the required extrusion pressure are the mean of eight tests.

## 3. Results and discussion

Fig. 1 shows the morphology of the 5% silicon nitride reinforced composite powder after 3 hours of high-energy milling. The spherical morphology of the as-received aluminium powder particles has changed to flat, typical of the beginning of the high-energy milling process [12]. At this stage, characterized by the predominance of deformation and welding, whenever reinforcement particles come between two or more aluminium particles, they get trapped on the interfacial welding boundary and, consequently, get incorporated into the matrix.

After 10 hours of milling, the process reaches a steady state in which welding and fracture are in balance and the particle morphology is approximately equiaxed, as shown in Fig. 2. At this stage, each particle is a

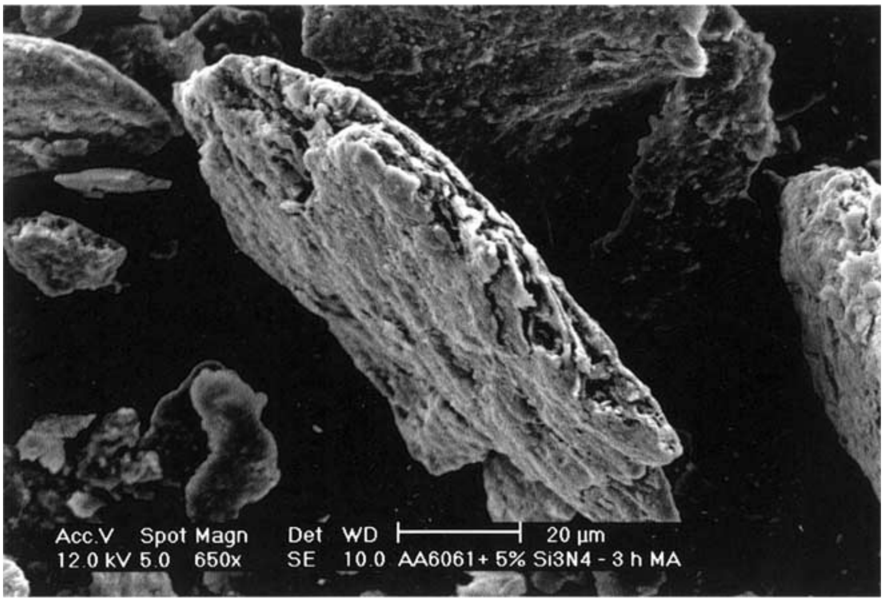


*Figure 1* Morphology of the 5% silicon nitride reinforced composite powder after 3 hours of high-energy milling.

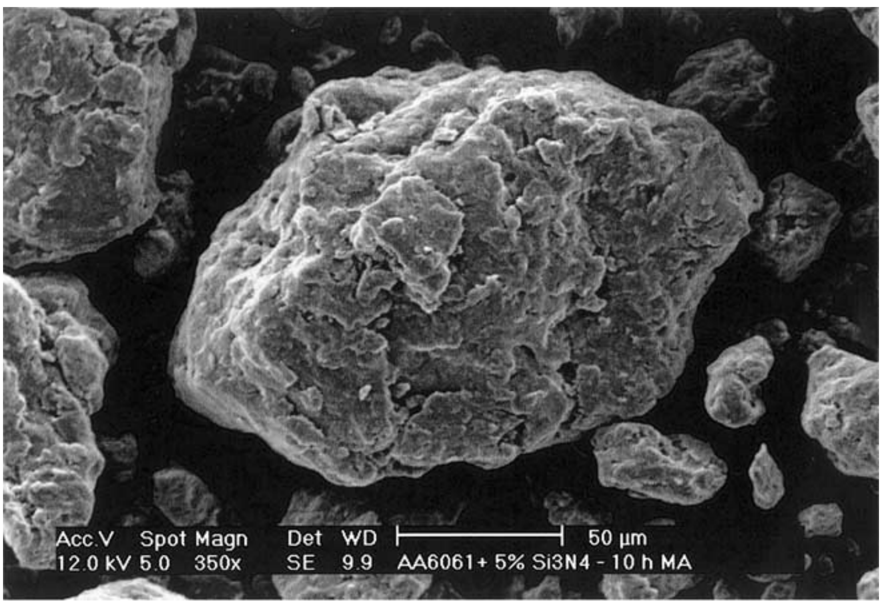


*Figure 2* Morphology of the 5% silicon nitride reinforced composite powder after 10 hours of high-energy milling.

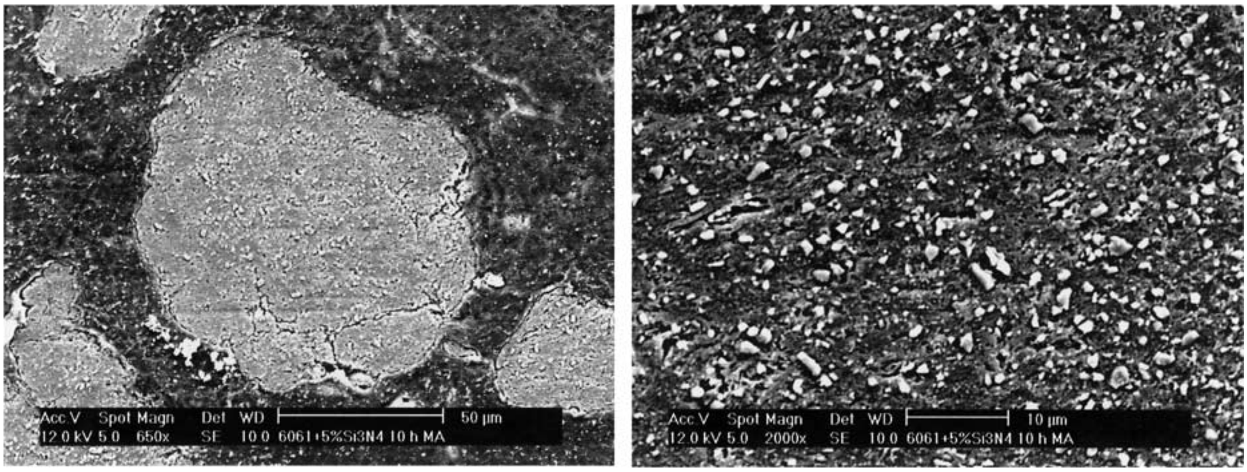


*Figure 3* Microstructure of the 5% silicon nitride reinforced composite powder after 10 hours of high-energy milling—higher magnification in the right side.

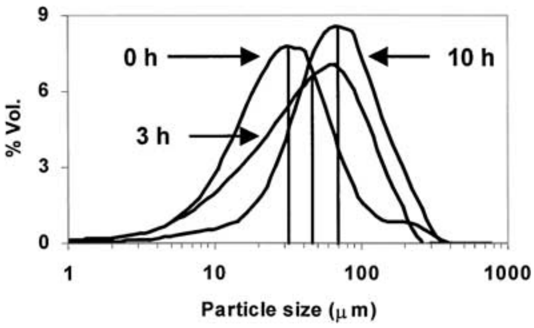


*Figure 4* Particle size distributions for PM6061 reinforced with 5% $Si_3N_4$ for 0, 3 and 10 hours of high-energy milling. Vertical lines refer to the equivalent diameter size ($D_{50}$).

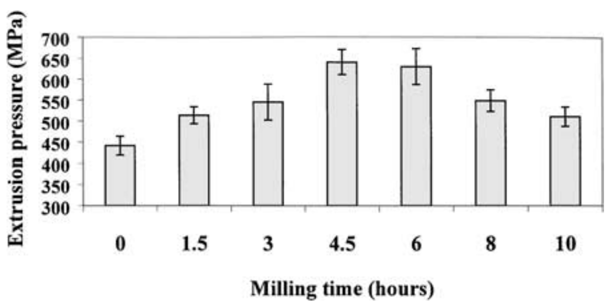


*Figure 5* Extrusion pressure versus high-energy milling time for the aluminium matrix composite reinforced with 5% of silicon nitride.

composite one produced by mechanical alloying [13], displaying a fine, homogeneous distribution of the reinforcement within the particle. Fig. 3 shows, with 2 different magnifications, the cross section of 5% $Si_3N_4$ reinforced composite particles obtained by 10 hours of mechanical alloying. In the left side, the darker gray background is the mounting material.

Fig. 4 shows the particle size distributions for 6061 reinforced with 5% $Si_3N_4$ for 0, 3 and 10 hours of high-energy milling. Vertical lines refer to the equivalent diameter size ($D_{50}$). The particle size distribution of the composite powder mechanically milled for 3 hours is wider than the unmilled one, with a significant increase of $D_{50}$. This wider shape of the curve is mainly owed to how the LASER diffraction analyser measures the sizes of flattened particles. Different values can be obtained depending on the angle formed between the laminar particle and the LASER beam. On the other hand, it can be assumed that the average diameter is really related to the equivalent diameter size, due to the high number of particles taken into account for each measurement and the same probability for all the angles. The wilder shape of the size distribution curve reveals the laminar morphology of the particle at the beginning of the milling process. A significant deviation to the right side can also be observed. This deviation and the increase of $D_{50}$ are related to the predominance of welding in this stage of milling.

The particle size analysis of the reinforced powder milled for 10 hours shows a more symmetric size distribution, which indicates the equilibrium between fracture and welding mechanisms, and the fact that fracture consumes larger particles while welding consumes the smaller ones. Note that the mechanical alloying process produces, in this case, a significant increase in the particle size. The particle size analyses of the other powders show the same behaviour.

Fig. 5 shows the extrusion pressure versus high-energy milling time for the aluminium matrix composite reinforced with 5% of silicon nitride. Zero hours of milling time refers to the low-energy mixed powders, which are used for comparison. The low-energy mixed powders show the lowest extrusion process. A maximum in the extrusion pressure in found for powders subjected to 4.5 hours of high-energy milling. Further increases in milling time reduce the required extrusion pressure.

The pressure required to extrude a monolithic alloy is a function of material flow stress, temperature, friction, strain rate, and deformation. The following equation expresses the extrusion pressure $P$ [14]:

$$P = \mathrm{K} \ln R \quad (1)$$

where $R$ is the extrusion ratio $A_i/A_f$ (the ratio of initial to final cross-sectional area), and K the extrusion constant for the material. This constant combines flow stress, friction, and redundant work due to non-uniform deformation of the material [15]. Theoretically, the pressure needed to extrude can be divided into three components: the homogenous work of deformation, the work needed to overcome the friction, and the redundant work.

In powder extrusion, the presence of porosity and unbonded particles changes the deformation mechanism significantly from that of a monolithic material. In this case, Equation 1 does not hold since the yield strength of the materials changes markedly during the extrusion process. Roberts and Ferguson [14] propose the following equation for powder extrusion:

$$P = a + b \ln R \qquad (2)$$

where $a$ is the component that relates to the redundant work (non-uniform deformation) and $b$ is the component that relates to the homogenous work of deformation.

The redundant work also includes the process of forming welds between the particles, breaking welds and rewelding, during deformation. These phenomena are closely related to the surface area of the particles, which depends on the size, size distribution, morphology, and surface roughness of the particle powder.

In this work, the temperature, friction, strain rate, and deformation are constants (the extrusion ratio, rate and temperature are constant parameters), but not the redundant work. The morphological changes in the powder particles due to the high-energy milling influence strongly the powder surface area: the spherical as-received powder particles have the lowest powder surface area, which increases when the particles are flattened, and is then reduced when the mechanical milling reaches the steady state with equiaxed morphology of the particles.

Although other powder characteristics that influence the powder surface area could be acting in the opposite way, such as the particle size distribution, the difference in the surface area caused by the morphological changes is high enough to make this characteristic the predominant one. The differences between the particle size distributions of the unmilled powder and the powder submitted to 3 hours of milling are highly influenced by the technical of measure. The increasing of the $D_{50}$ with increasing milling time should produce a decrease in the surface area of the powder, which could contribute to the decreasing of the pressure required to extrude the powder submitted to longer milling times, but never would contribute to the increasing of the pressure for the shorter milling times.

Besides, the high-energy milling greatly increased the particle hardness but it seems not to have the same influence as the particle morphology on the pressure required to extrude the powder billet. For short milling times, both effects, the increasing surface area and the increasing hardness, contribute to increase the pressure required to extrude. However, for milling times longer than 4,5 hours, the decreasing surface area with increasing milling time tends to decrease the extrusion pressure, while the increasing of the powder hardness, studied as an isolated phenomenon, would increase the extrusion pressure. The results demonstrate that in the investigated composite system, the effect of the surface area on the extrusion pressure is greater than the effect of hardness of the powder. Lieblich and co-workers [16] observed that average particle size (related to the powder surface area), as well as the extrusion temperature, strongly influences the extrusion pressure. Less influence was observed with the extrusion ratio and rate.

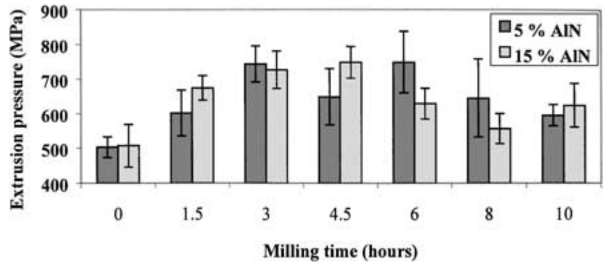

*Figure 6* Extrusion pressure versus high-energy milling time for the aluminium matrix composite reinforced with 5% and 15% of aluminium nitride.

Other parameters could also influence the pressure required during powder pressing, and in the case of the composite powders analysed in this investigation, the powders submitted to different milling times reached different densities after cold pressing at the same pressure, as related elsewhere [17]. However, during the extrusion of metal powders, a high densification of the pressed powder is produced in the initial stage of the process, similar to a hot pressing process, giving to a material with a nearly full density, but with weak bonding between the particles [18]. In the subsequent step, a higher pressure is necessary to flow the material. Since the pressure involved in the densification of the pressed powders is significantly smaller than the pressure required to flow the material, the differences between the densities of the cold pressed powders are eliminated during the initial stage of the extrusion and should not influence the level of pressure required to extrude the material. Preliminary studies confirmed this assumption, in which powders were pressed at 200, 300, 400 and 500 MPa, and extruded at constant parameters. No relations were found between the pressed densities (from 75 to 90% of theoretical density) and the pressure required to extrude.

Fig. 6 shows the extrusion pressure versus high-energy milling time for the aluminium matrix composite reinforced with 5% and 15% of aluminium nitride. The tendency observed with the composite powders reinforced with silicon nitride is also observed in the composite powders reinforced with aluminium nitride. As shown in a previous work [19], a higher reinforcement percentage produces equiaxed powder morphology in a shorter milling time. After 8 hours of milling, the composite powder reinforced with 5% of AlN has a predominantly equiaxed shape, while that reinforced with 15% of AlN needs only 6 hours of milling to display the equiaxed morphology. The greatest difference between the pressure required to extrude the composite powders reinforced with 5% and those with 15% of AlN, submitted to the same milling time, occurs at 6 hours, when the composite powder reinforced with 5% of AlN still shows laminar morphology, while the powder reinforced with 15% of AlN already shows equiaxed morphology. This corroborates the significant influence of the powder morphology on the pressure required to extrude the mechanical-milled powder.

## 4. Conclusions

– The mechanical alloying method can produce 5% silicon nitride and 5% and 15% aluminium nitride reinforced aluminium alloy AA6061 matrix composite powders with a fine distribution of the reinforcement particles within the matrix.
– The extrusion pressure is low for the composite powder mixed by the conventional process, due to the spherical morphology and the lower yield strength, in comparison with the milled powders, of the as-received aluminium powder alloy.
– The extrusion pressure increases for the composite powder after short mechanical alloying times, due to the flattened morphology of the particles at this stage of the process.
– The extrusion pressure decreases after longer times of mechanical alloying, due to the equiaxed morphology of the particles typical of the final stage of the process.


## Acknowledgements

The Authors thank CAPES–Brasil (Fundação Coordenação de Aperfeiçoamento de Pessoal de Nível Superior) and CICYT–Spain (ref.: MAT2000-0442-C02-01) for financial support.


## References


1. J. S. BENJAMIN, *Metall. Trans.* **1** (1970) 2943.
2. A. BOSE, "Advances in Particulate Materials" (Butterworth-Heinemann, Newton, MA, 1995).
3. L. LU and M. LAI, "Mechanical Alloying," edited by Kluwer Academic Plublishers, 1998.
4. Y. B. LIU, S. C. LIM, L. LU and M. O. LAI, *J. Mater. Sci.* **29** (1994) 1999.
5. C. SURYANARAYANA, *Progress in Materials Science* **46** (2001) 1.
6. C. E. COSTA, W. C. ZAPATA, J. M. TORRALBA, J. M. RUIZ PRIETO and V. AMIGÓ, *Mat. Sci. Forum* **217** (1996) 1859.
7. J. S. BENJAMIN and T. E. VOLIN, *Met. Trans.* **5** (1974) 1929.
8. J. S. BENJAMIN, *Modern Developments in Powder Metallurgy* **21** (1988) 397.
9. B. J. M. AIKIN and T. H. COURTNEY, *Met. Trans. A* **24** (1992) 647.
10. G. B. SCHAFFER, T. B. SERCOMBE and R. N. LUMBEY, *Mater. Chem. Phys.* **67** (2001) 85.
11. M. J. TAN and X. ZHANG, *Mat. Sci. Eng. A* **244** (1998) 80.
12. R. SANKAR and P. SINGH, *Mat. Letters* **36** (1998) 201.
13. L. LU, M. O. LAI and C. W. NG, *Mat. Sci. Eng. A* **252** (1998) 203.
14. P. R. ROBERTS and B. L. FERGUSON, *International Materials Reviews* **36**(2) (1991) 62.
15. S. KALPAKJIAN, Manufacturing Process for Engineering Materials, World Student Series Edition, Addison-Wesley Publishing Company, Inc., p. 634.
16. M. LIEBLICH, G. CARUANA, M. TORRALBA and H. JONES, *Powder Metall.* **39**(2) (1996) 143.
17. J. B. FOGAGNOLO, E. M. RUIZ-NAVAS, M. H. ROBERT and J. M. TORRALBA, *Mat. Sci. Eng. A*, submitted for publication in *Mat. Sci. Eng. A*.
18. J. ZHOU, PhD Thesis from Delft University of Technology, Holland, 1991.
19. J. B. FOGAGNOLO, F. J. VELASCO, M. H. ROBERT and J. M. TORRALBA, accepted for publication in Mat. Sci. Eng. A.



*Received 2 November 2001*
*and accepted 3 July 2002*