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High Temperature Behavior of Nanostructured Al Powders Obtained by Mechanical Alloying under NH_3 Flow

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Abstract. Aluminium powder was mechanically alloyed under ammonia gas flow for different times (1-5 h) in order to produce a second-phase reinforcement, mainly by aluminium nitride (AlN). After milling, powders were consolidated by cold uniaxial pressing and vacuum sintering. A small amount of copper powder was added to the Al milled powder to improve its sintering behavior. Hardness and indirect tensile test were carried out at room and high temperature to evaluate the mechanical properties evolution. Results showed an remarkable hardness increase with the second phases content, even at high temperature (up to 229 HB at 400 ºC). However, the high content of second phases of ceramic nature decreases the ductility, resulting in low values of tensile strength (lower than 160 MPa).

Keywords: Al powder, mechanosynthesis, nitriding, mechanical properties.
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INTRODUCTION

Mechanical alloying is a solid-state powder processing technique carried out in a high-energy ball mill. It is normally a dry process which allows the production of homogeneous materials starting from elemental powder mixtures [1]. This technique has been employed to develop a variety of commercially useful and scientifically interesting materials [2-6]. Traditionally, in the powder metallurgy field, aluminium has been reinforced with a variety of directly-added particles [7,8]. However, it was found that the reinforcement of aluminium powder is more effective when second phases are formed through direct reaction, obtaining finely dispersed phases in the matrix [9,10].

On the other hand, it is known that, in general, mechanical properties of aluminium alloys undergo an important degradation after heating above 100 ºC. Nevertheless,

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aluminium composites reinforced by ceramic dispersoids strongly improve high-temperature mechanical properties of sintered compacts [11,12].
The aim of this work is to increase mechanical properties of the as-received aluminium powder (AR Al). In this sense, AR Al powder was milled under ammonia gas flow to produce a nitrides dispersion strengthening. Powders were consolidated by uniaxial cold pressing and vacuum sintering.

EXPERIMENTAL PROCEDURE

Starting material was atomised elemental aluminium powder (AS 61, Eckart) with purity higher than 99.7 %, and mean particle size of 80.5 μm. Aluminium powder was milled in a high-energy attritor ball-mill. The water-cooled stainless steel vessel had a capacity of 1400 cm^3. A 3 wt.% of etilen bis-stereamide (EBS) micropowder organic wax was used as process control agent (PCA). Mill charge contained 72 g of powder and 3600 g of balls (charge ratio 50:1). All milling experiences were carried out under ammonia gas flow (1 cm^3/s) with a purity higher than 99.96 % (Air Liquide).

With the aim of studying the influence of milling time, experiences were carried out for 1, 3 and 5 hours, with a rotor speed of 500 rpm. Powders morphology was analysed by scanning electron microscopy (SEM, PHILIPS XL-30). X-ray diffraction (XRD, Bruker D8 Advance, CuKα radiation) of milled and sintered powders (650 ºC, 1 h) in vacuum (5 Pa) was used to identify and quantify the formed phases. To test powders compressibility, a universal testing machine, INSTRON 5505, with a charge cell of 100 kN, was employed.

All milled powders were mixed with a 2.5 wt% Cu powder (CH-L10, Eckart) before their consolidation by a single cycle of cold uniaxial pressing (1300 MPa) and vacuum (5 Pa) sintering (650 ºC, 1 h), followed by furnace cooling. EBS wax was also used as die-wall lubricant during cold pressing. The hardness and indirect tensile tests were determined over cylindrical compacts (diameter: 12 mm; mass: ca. 4g) using a EMCO M4U-025 hardness tester and a INSTRON 5505 machine, respectively.

RESULTS AND DISCUSSION

Milled Powder Characterization

Figure 1 collects milled-powders SEM micrographs. As can be seen, powders morphological evolution is remarkable for short increases of milling time. After 1 h milling, powder particles have flakes shape (Figure 1a); thereafter, particle size is further reduced, whereas its geometry tends to be more rounded (Figure 1b and 1c). However, as a consequence of the small particle size reached after milling for 3 and 5 h, powders tend to agglomerate. Thus, their actual size distribution is distorted when using laser diffraction to measure the granulometry. In any case, particle size is always lower than 10 μm for a milling time of 3 h or higher, as determined by SEM measurements.

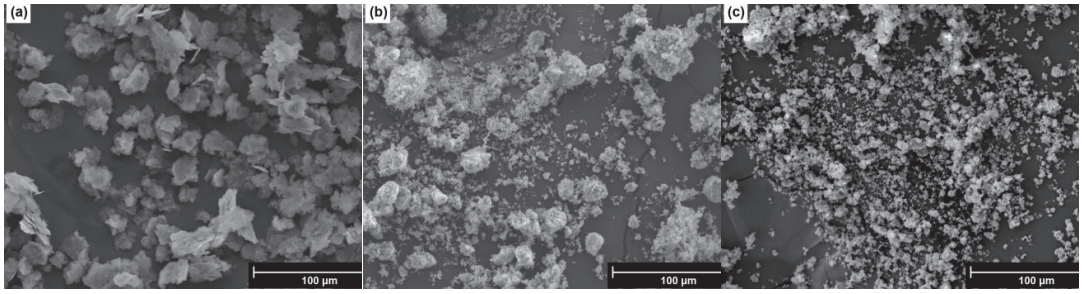

FIGURE 1. SEM-SE images of milled powder under ammonia gas flow for (a) 1 h, (b) 3 h and (c) 5 h.

In order to study the powders densification during compaction, and to indirectly [10] estimate their hardness, compressibility curves have been determined (Figure 2). Furthermore, for comparison purposes, the compressibility curve of as-received aluminium powder (AR Al) has been included.

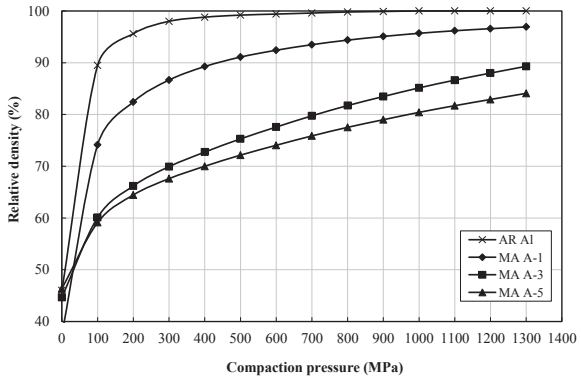

FIGURE 2. Compressibility curves of AR Al and mechanically alloyed Al powders in ammonia flow atmosphere.

Considering a pressure of 1000 MPa, powder milled for 1 h (MA A-1) has a relative density of 95 % (Figure 2). On the other hand, relative density strongly decreases when the milling time is 3 and 5 h (MA A-3 and MA A-5, respectively), reaching values around 85 and 80 %, respectively, at the same pressure of 1000 MPa.

XRD patterns of all milled powders only show Al reflections (MA A-5 is shown in Figure 3). After sintering, XRD patterns also show reflections corresponding to aluminum nitride (AlN) and aluminum oxynitride (Al_5O_6N). Qualitatively, it is observed that peaks corresponding to AlN and Al_5O_6N grow with milling time, whereas the Al reflections decrease. Therefore, nitrogen coming from the ammonia gas is incorporated, in solid solution, to the Al powders during milling [9]. So, solid solution hardening is added to work hardening caused by the repeatedly cold deformation of powder particles, therefore causing the compacts relative green density to be reduced by increasing milling time.

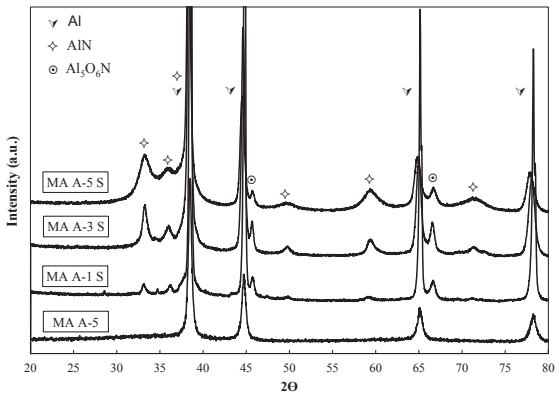

FIGURE 3. X-ray diffraction patterns, after sintering, of powders milled for 1, 3 and 5 h. Pattern of powder milled for 5 h, before sintering, is also included.

Quantification results, obtained via Rietveld refinement, corresponding to phases formed during sintering, show a considerable amount of AlN formed after the first hour of milling (Table 1). It should also be highlighted a significant increase with milling time: AlN content increases from 8 to 30 wt.%, and from 30 to 63 wt.%, when milling time was extended from 1 to 3 h and from 3 to 5 h, respectively.

TABLE 1. Phases formed (wt.%) in sintered Al powders, after being milled for 1, 3 and 5 h in ammonia gas flow atmosphere.

	Material (wt.%)		
Phase	**MA A-1 S**	**MA A-3 S**	**MA A-5 S**
Al	87	58	30
AlN	8	30	63
Al_5O_6N	5	12	7

Sintered Compact Properties

In order to improve the compacts relative density after sintering, a 2.5 wt% of copper was added to Al milled powders before pressing them at 1300 MPa. This addition allowed, for all powders, achieving relative densities above 90% after sintering. Then, cylindrical compacts were subjected to a heating of 400 ºC, for 100 h, to evaluate the subsequent effect on mechanical properties. Table 2 shows the indirect tensile strength (ITS) and Brinell hardness (HB) results of sintered compacts, as well as the hardness at different temperatures after being subjected to such heating of 400 ºC for 100 h.

Regarding the hardness (HB), it is not observed an important difference between the values obtained before and after the long-term heating. Thereby, hardness is not significantly affected by the prolonged heating of the samples. Furthermore, hardness increases with milling time as a consequence of the higher weight percentage of ceramic second phases formed during sintering, mainly AlN (the contribution to the mechanical properties of Al-Cu phases, formed due to Cu addition, can be neglected). In this way, compacts from unmilled powder (AR Al) have much lower hardness than that of powder milled under ammonia gas. It is remarkable the important increase in

hardness produced in samples from milled powders in ammonia for 3 and 5 h (MA A-3 Cu S and MA A-5 Cu S, respectively). As expected, hardness decreases with increasing temperature, although the high content of ceramic phase allows keeping high values of hardness. Thus, compacts from powders milled 5 h reach, at 400 ºC, hardness values even higher than those of the MA A-3 Cu S compacts at room temperature.

TABLE 2. Indirect tensile strength and hardness of sintered materials.

	ITS (MPa)	HB (kp/mm^2)					
Sample	**25 ºC**	**Previous to HT**	**25 ºC**	**100 ºC**	**200 ºC**	**300 ºC**	**400 ºC**
AR Al S	67	21	28	22	13	10	-
MA A-1 Cu S	-	74	78	72	51	34	21
MA A-3 Cu S	159	226	244	191	157	99	71
MA A-5 Cu S	96	303	335	290	268	254	229

Regarding indirect tensile strength (ITS), measured values are also higher for compacts from powder milled under ammonia gas. Nevertheless, indirect tensile strength decreases for too high AlN contents (159 and 96 MPa for MA A-3 Cu S and MA A-5 Cu S, respectively), since the material becomes harder and more brittle. In any case, compacts from powders milled in ammonia reach indirect tensile strength values much higher than that of the as-received aluminium powder.

CONCLUSIONS

Aluminium compacts from powder milled under ammonia gas flow during different times were produced and studied. Hardness at different temperatures, as well as indirect tensile strength at room temperature were evaluated. Results are as follow:

1. Relative density is reduced as the weight percentage of second phases (AlN and Al_5O_6N) is increased. However, if a 2.5 wt% Cu is added to milled powder, relative densities above 90 % after sintering are achieved.
2. Hardness increases with milling time as consequence of ceramic second phases formed during sintering, mainly AlN. Furthermore, hardness is not significantly affected by the prolonged heating of the samples due to the ceramic nature of AlN and Al_5O_6N phases.
3. Indirect tensile strength is increased for powder milled under ammonia gas flow, although it is reduced with milling time, since the material becomes harder and more brittle.

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