Proceedings of the 9th International Conference on Aluminium Alloys (2004)
Edited by J.F. Nie, A.J. Morton and B.C. Muddle

Influence of Mechanical Alloying and Sn and TiCN Additions on the Liquid Phase Sintering of a 2xxx Series Alloy

M.L. Delgado[1], E.M. Ruiz-Navas[1], J.M. Torralba[1], W.A.P. Mcphee[2], G.B. Schaffer[2]

[1] Departamento de Ciencia de los Materiales e Ing. Metalúrgica, Universidad Carlos III, Spain
[2] Division of Materials, The University of Queensland, Australia

Keywords: aluminum alloys, trace addition, reinforcement addition, mechanical alloying, liquid phase sintering

Abstract

In this work the influence of mechanical alloying on the liquid phase sintering of a 2xxx aluminum alloy is studied. Mechanical alloying is a solid state processing technique where by means of welding and fracturing processes, the morphology of the powder particles is modified. The surface of the powder is improved, resulting in improved liquid phase sintering through liquid diffusion. In this work, the influence of Sn and TiCN on the liquid phase is also studied.

1. Introduction

Trace elements are widely known as agents that can affect the processing of materials, including the sintering process. Specifically, Sn additions can enhance the final properties of sintered aluminium materials, as Sn diffuses into the Al matrix ahead of the copper. This ties up vacancies and delays the transient aspect of the liquid phase during sintering [1-5]. Improvements in material properties can also be achieved through the addition of, ceramic elements. In previous studies [6] it has been concluded that the presence of TiCN enhances thermo-mechanical properties of Al-Mg_2Si cast alloys. . Processing the materials by mechanical alloying can also improve the materials properties [7]. Mechanical alloying, which involves welding and fracturing processes, changes the particle morphology. The change in the external morphology results in an increase in particle surface area. This particle surface enhancement could be very useful in liquid phase sintering due to the improvements in liquid phase diffusion. In this paper the influence of Sn and TiCN additions on the liquid phase during sintering of an Al-Cu-Mg-Si alloy are analysed. The effect that combining Sn additions with mechanical alloying has on the liquid phase during sintering has also been examined.

2. Experimental

The aluminum alloy employed in this work is the Alumix 123 grade, provided by Ecka Granulate (Germany). The trace element is Sn and the ceramic addition is TiCN. The powder details are shown in Table 1.

Table 1: Characteristics of the elemental powders.

Powder	Source	Grade	Particle size distribution		Composition	
			Size fraction (µm)	%	Element	%
ECKA Alumix 123	Ecka Granulate	AS 91/S	>200	3 max.	Cu	4.2-4.8
			>160	8 max.	Mg	0.4-0.6
			>100	15-35	Si	0.5-0.7
			>63	40-55	Lubr.	1.4-1.7
			>45	5-20		
			<45	15 max.		
TiCN	H.C. Starck	C	1.0-1.3	100	C_{TOTAL}	9.3-10.3
					C_{FREE}	0.05 Max.
					N	10.3-11.3
					O	1.5 Max
Sn	Goodfellow		<45	100		99.75

Powder alloys were prepared by mixing for 30 minutes in a tubular powder mixer, and also, by mechanical alloying in a horizontal high energy ball mill (attritor), Zoz Gmbh. Stearic acid was mixed with the powder in 1.5wt% as PCA. Milling time was 10 hours and the ratio of balls to the powder was 20:1 (in weight). The alloys used for this work are summarised in the table 2.

Table 2: Composition of powder alloys, and pressing conditions.

Alloy	Powder condition	Compaction pressure (MPa)	Lubricant for compaction
Alumix 123	Mixed	117	No
Alumix 123 + 0.15wt% Sn	Mixed	117	No
Alumix 123 + 0.15wt% Sn + 5wt% TiCN	Mixed	117	No
Alumix 123	Mechanically alloyed	800	Yes
Alumix 123 + 0.15wt% Sn	Mechanically alloyed	800	Yes
Alumix 123 + 0.15wt% Sn + 5wt% TiCN	Mechanically alloyed	800	Yes

Specimens were obtained by cold uniaxial pressing using different pressures and percentages of lubricant. Mixes from raw powders (used in “as received” state) were compacted using a pressure of 117 MPa and lubricant was not added. Mechanically alloyed powders were compacted using a pressure of 800 MPa, and stearic acid (1wt%) was added as lubricant. Compaction of the specimens was performed using a hand operated hydraulic press and a floating cylindrical die. Specimens were pressed to a green density of ≈80%. These conditions produced specimens with dimensions of ≈10.1×7.5 mm.

Green density was calculated by measuring specimen dimensions and mass to an accuracy of 0.001 mm and 0.0001 g, respectively. Sintered density was determined using the MPIF Standard42, by Archimedes´ method, although ethanol was used instead of water.

Densification,Ψ was calculated to determine the amount of shrinkage or expansion:

$$\psi = \frac{\rho_s - \rho_g}{\rho_t - \rho_g} \qquad (1)$$

where ρ_s, ρ_g and ρ_t are the sintered, green and theoretical density, respectively. A positive value of Ψ indicates shrinkage; Ψ approaches unity as full density is attained.

Delubrication and sintering was carried out in a vertical furnace with a high purity nitrogen atmosphere. The nitrogen flow rate was ≈2 l·min^{-1}. The heating rate from dewaxing to sintering temperature was 10 ºC·min^{-1}, and 5ºC·min^{-1} from this temperature to 590ºC. The furnace was held at 590ºC for 60 minutes. Samples were water quenched after being sintered for 60 minutes.

3. Results and Discussion

The sintering process is analysed by means of densification and microstructure. The densification parameter depends strongly on the milling step of the mechanical alloying process. This is shown in figure 1, where densification is represented for each alloy, after 60 minutes of sintering time.

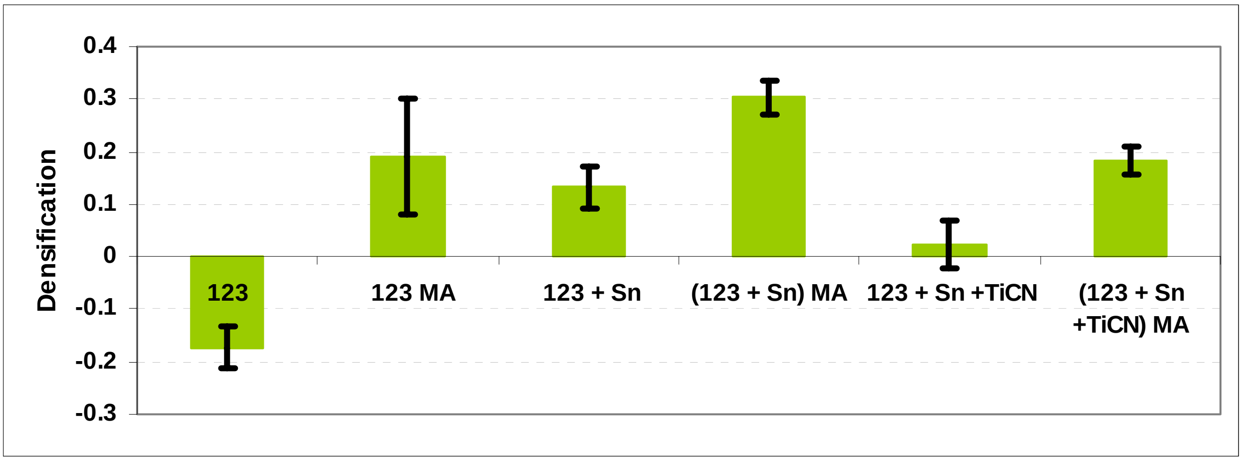


Figure 1: Effect of the different additions and the mechanical alloying process on the alloys densification.

As can be seen in Figure 1, the addition of Sn and the mechanical alloying process both have a beneficial effect on the alloys' densification. Expansion is only observed for the Alumix 123 alloy.

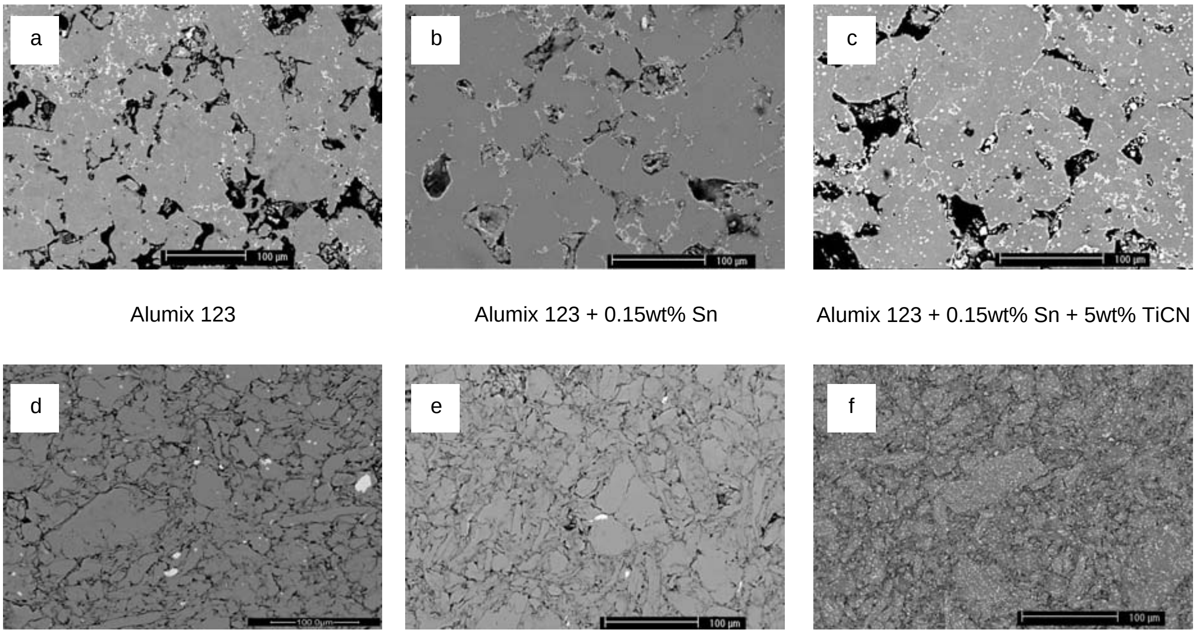
The Sn addition improves the densification of the material. Alumix 123 + Sn and Alumix 123 + Sn + TiCN densification values are increased compared to Alumix 123 alloy.
Besides milling the powder, higher densification values have also been reached. All the alloys that have been mechanically alloyed show enhanced densification values. Alumix 123 + Sn mechanically alloyed increases the higher densification value.

It has to be also noted that the addition of TiCN influences properties. The addition of ceramic reinforcement to the base alloy decreases the material densification.

In order to analyse with more detail the effect of Sn, it is very important to consider the main characteristic of the Al-Cu system: transient liquid phase sintering. However it is interesting to note that depending on the sintering temperature and alloying elements, the system can be fully transient or have a large persistent aspect [2]. The liquid phase, basically consists of the melting of Al_2Cu eutectic and, due to the solubility of Cu in Al, the volume fraction of liquid decreases with the time until the equilibrium level is attained. One way to increase sintering density is by hindering or decreasing transient aspect of the liquid phase. With Sn addition, liquid is slightly slower to form, but higher quantities persist for longer times [1, 8]. Sn has both vacancy binding and higher diffusivity in aluminum than copper does. As a result, the liquid persists for longer times, which results in improved sintering [2].

Mechanical alloying process affects the final properties of the material in a different way. Due to the specific advantages in the production of powders by mechanical alloying, after the powders milling each particle is a fully prealloyed particle [15-21]. Sintering of mechanically alloyed powders does not present a conventional liquid phase, but the behaviour could be considered as super solidus liquid phase sintering. As soon as the powders are mechanically alloyed, the liquid phase appears not only between particles, but also within grains, since the elements that form the eutectic melt are present within the individual particle. The mechanically alloying process not only changes the morphology of the particles. In this way, it should be considered that mechanical alloying provides more specific surface to the powder, which could enhance the liquid phase diffusion beyond the particles.

The influence of the additions and high energy milling can be analysed through the microstructure.



Alumix 123 | Alumix 123 + 0.15wt% Sn | Alumix 123 + 0.15wt% Sn + 5wt% TiCN

Alumix 123 MA | (Alumix 123 + 0.15wt% Sn) MA | (Alumix 123 + 0.15wt% Sn + 5wt% TiCN) MA

Figure 2: Sintered alloy microstructures, water quenched after 60 minutes.

If the Sn addition is analysed in first place, it should be noted that the porosity is reduced in Alumix 123 + Sn alloy compared to Alumix 123 (Figures 2a and 2b respectively). If the Sn binds preferentially with the Al and delays the transient aspect of the liquid, this liquid persists for longer times, and as a consequence, the result is a higher sintered density [2]. The TiCN addition influence, decreasing the material densification, can also be explained by the microstructure (figure 2c). The location of TiCN particles, between the aluminum particles, would reduce the rearrangement of the last during sintering process, limiting their contact, diminishing densification and increasing the porosity of the sintered sample.

The mechanically alloyed microstructures (Figures 2d-f) show important differences compared to the alloys that have not been milled. The porosity in the mechanically alloyed has been significantly reduced. As a consequence of the increase in the specific surface, mechanically alloyed Alumix 123 shrinks compared to Alumix 123 as a result of the decreasing of the porosity. Together with the Sn addition, it is possible to enhance the material densification to obtain a higher value by the maximum decrease in porosity.

Besides, Alumix 123 + Sn + TiCN mechanically alloyed porosity (figure 2.1) has also been reduced (compared with the unmilled alloy). It is due to the milling process. At the beginning of the milling, the balls crack the TiCN particles, and when the milling is concluded, these pieces are inside the final particle. The new size of the ceramic particles reduces the interparticle spacing, providing more contact between aluminum particles.

According to these results, it is clear that milling in an high energy ball mill (attritor) can affect to final properties of the sintered materials. Also these results confirm that trace additions of Sn, enhance sintering properties of aluminum alloys [8].

4. Conclusions

1. Addition of Sn improves alloy densification. Sn additions help to increase the permanence of the liquid phase.
2. Adding TiCN decreases alloy densification. TiCN particles reduce the ability of particles to approach the of aluminum particles during the sintering.
3. Densification reaches higher values when the powder is milled by mechanical alloying. Mechanical alloying gives each particle more surface area, due to the change in morphology of the powder, which then helps with liquid phase diffusion.

References

[1] G.B. Schaffer, S.H. Huo, J. Drennan, G.J. Auchterlonie, Acta Mater 49 (2001) 2671-2678
[2] T.B. Sercombe, G.B. Schaffer, Acta Mater., Vol. 47-2 (1999) 689-697
[3] R.N. Lumley, G.B. Schaffer, Scripta Mater., 39 (1998) 1089-1094
[4] D.P. Bishop, J.R. Cahoon, M.C. Chaturvedi, G.J. Kipouros, W.F. Caley, Mater. Sci. Eng., A290 (2000) 16-24
[5] T.B. Sercombe, G.B. Schaffer, Mater. Sci. Eng , A 268 (1999) 32-39
[6] G. Levi, W.D. Kaplan, M. Bamberger, Mater. Sci. Eng., A326 (2002) 288-296
[7] C. Suryanarayana, E. Ivanov, V.V. Boldyrev, Mater. Sci. Eng., A304-306 (2001) 151-158
[8] T. Watanabe, K. Yamada, Int. J.of Powder Metall., 4 (1968) 37
[9] T.B. Sercombe, Mater. Sci. Eng., A341 (2003) 163-168
[10] William A.G. McPhee, Liquid pool formation and the influence of iron on sintering of Al-Cu alloys, PhD Thesis, The University of Queensland, 2002
[11] J. Lacaze, G Lesoult, I. Ansara, Mater. Sci. Forum, (1996) 171-176
[12] W.T. Kim, B. Cantor, Acta Mater., (1994) 3045-3053
[13] G.B. Schaffer, S.H. Huo, J. Drennan, G.J. Auchterlonie, Acta Mater 49 (2001) 2671-2678
[14] T.B. Sercombe, G.B. Schaffer, Mater. Sci. Eng , A 268 (1999) 32-39
[15] J.B. Fogagnolo, E.M. Ruiz, M.H. Roberts, J.M. Torralba, Scripta Mater., 47 (2002) 243-248
[16] R. M. German, Int. J.of Powder Metall, 26 (1990) 23-34
[17] R. Tandon, R. M. German, Int. J.of Powder Metall., 33 (1997) 54-60
[18] J. Liu, A. Lal, R.M. German, Acta Mater., 47 (1999) 4615-4626
[19] Z.Y. Liu, N.H. Loh, K.A. Khor, S.B. Tor, Mater. Sci. Eng, A293 (2000) 46-55
[20] R. Tandon, R. M. German, Int. J.of Powder Metall., 30 (1994) 435-443
[21] A. Lal, R.G. Iacocca, R.M. German, J. Mater. Sci. 35 (2000) 4507-4518