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Microstructural transitions in an RS Al–4La alloy

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The use of rapid solidification (RS) techniques to increase the thermal stability of aluminium and aluminium alloys is considered a promising processing route. The Al–La equilibrium binary system tends to form a high melting temperature intermetallic compound on the aluminium-rich side. The limited solid solubility of solute, high solubility in the liquid and the relatively low solid state diffusivity, in general, of rare earth (RE) elements in aluminium suggests this system (as well as other Al–RE systems) as a suitable lightweight, dispersion-strengthened alloy candidate for elevated homological temperature applications. RS processing promotes the formation of a relatively finer microstructure in an Al–4La alloy, as well as the formation of a finely dispersed metastable phase. This metastable phase, is fairly stable up to 400° C. Lanthanum concentration in the ribbon bulk matrix, is considerably increased via RS processing, and the formation of a heavy eutectic is suppressed.

1. Introduction

The role of strengthening additive elements from the lanthanide group in aluminium and aluminium alloys is the topic of some current research [1–4]. Introducing rapid solidification (RS) techniques as a means of increasing the thermal stability of aluminium and aluminium alloys is considered a promising processing route. One of the general strategies involves internal introduction of hard and stable intermetallic compounds into aluminium. Owing to the prospect of dispersoids and supersaturated refined microstructure formation [5, 6], via RS processing, improved mechanical and thermal stability properties, in aluminium–rare earth (RE) alloys are assumed.

The Al–La equilibrium binary system, as shown in Fig. 1 [7], tends to form a high melting temperature intermetallic compound on the aluminium-rich side. The limited solid solubility of solute, high solubility in the liquid and the relatively low solid state diffusivity, in general, of RE elements in aluminium suggests this system (as well as other Al–RE systems) as a suitable lightweight, dispersion-strengthened alloy candidate for elevated homological temperature applications.

2. Experimental details

Ribbons from a nominal aluminium–4 wt % lanthanum master alloy were prepared by the melt spinning technique. A clean, sand blasted, master alloy ingot was placed in a high-purity quartz crucible with a 1 mm nozzle diameter. Following r.f. induced melting, the charge (typically 50 g) was then expelled through the nozzle by an applied 27 kPa argon overpressure. The metal stream was directed on to a 300 mm diameter polished, clean, water-cooled copper wheel, rotating at 1400 r.p.m. (tangential velocity of 22.4 m sec^{-1}). The ribbon received has 3 mm × 0.07 mm cross-section average dimensions.

Samples cut from a continuous ribbon were heat treated for 2 h at 100, 200, 300, 400, 500° C in an argon protective atmosphere and was designated T1, T2, T3, T4, T5, respectively. These samples and samples in the as-received condition, designated T0, and the master alloy, designated MA, were studied by several experimental techniques.

Specimens were subjected to scanning electron microscopy (SEM) and electron dispersion of X-rays (EDX) examination and analysis and scanned in the following different positions (relative to the scanning beam) and conditions.

(i) T0: flat, contact surface on top.

(ii) T0: flat, free surface on top.

(iii) T0, T1, T2, T3, T4, T5: longitudinal transverse cross-section, mounted in a "cold" acrylic resin, polished only down to 1 μm alumina powder. Polished and then etched with a standard Keller solution.

(iv) MA: from the original master alloy thin polished, and polished and etched.

For transmission electron microscopy (TEM) studies, specimens were punched from a T0 ribbon and were mounted in a sample holder having a hot-stage installation. Locations in the specimen being transparent to the electron beam were examined. In general the contact surface was kept perpendicular to the incident electron beam. Samples were studied at: room temperature, 400° C for 2 h, or 500° C for 2 h. Changes were constantly monitored, and recorded at 40 min intervals.

X-ray diffractometry (XRD) was performed on short pieces which were cut from T0, T1, T2, T3, T4, T5 ribbons. Using a thin amorphous double-sided adhesive, these pieces were respectively glued to a glass slide, contact surface in direct exposure to the X-ray beam.

Relatively thick and flat pieces from the MA were also X-rayed in same conditions as the ribbons.

3. Results and discussion

A scanning electron micrograph from longitudinal

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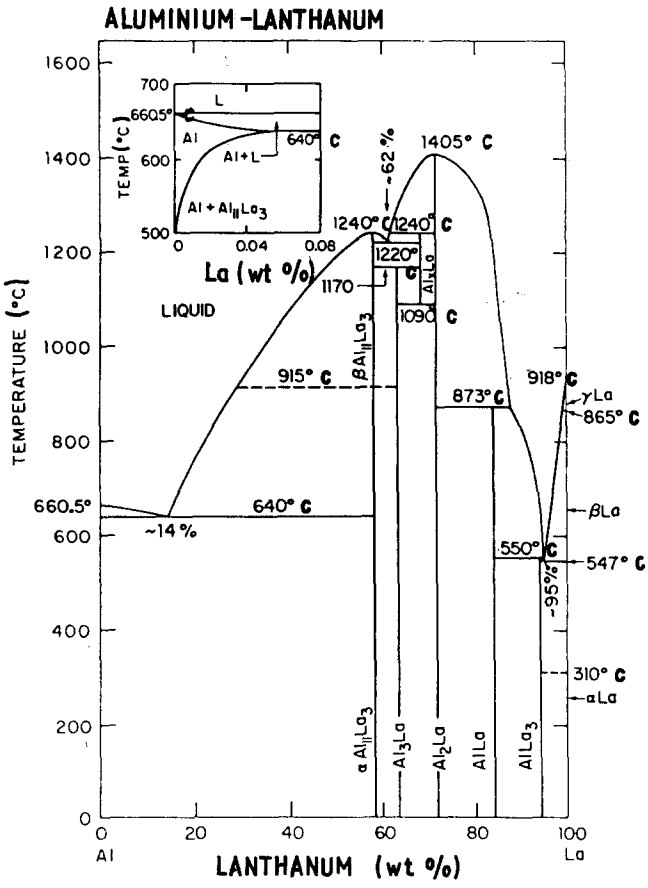


Figure 1 Phase diagram of the Al–La binary system [7].

transverse cross-sections, as shown in Fig. 2, indicates the existence of two distinct types of micromorphologies:

(i) a columnar structure adjacent to the contact surface followed by a transient interface; and

(ii) equiaxed grains having a degenerated dendritic substructure closing from the interface up to the free surface.

The relative proportions across ribbon occupied by each structure varies from a well-developed columnar structure occupying about 50% of the cross-section, to an equiaxed grains structure entirely occupying the cross-section, Fig. 3.

The different types of micromorphologies as observed across the ribbon's smallest dimension arise from varying solidification rates across the ribbon. These variations are probably due to the local fluctuations in conditions during the manufacturing process; variations in contact efficiency between ribbon and wheel, nonuniform liquid metal supply, etc. These fluctuations resulting in the formation of a mixed mode of micromorphologies across the ribbon. The columnar structure can be related to higher cooling rates (near the contact) as compared to the relatively slower cooling rates which promote the formation of the equiaxed structure.

Lanthanum macroconcentration along the cross-section, in the T0 condition, varies slightly from contact surface to free surface but is on average 3.7 wt % La. After etching, the average lanthanum

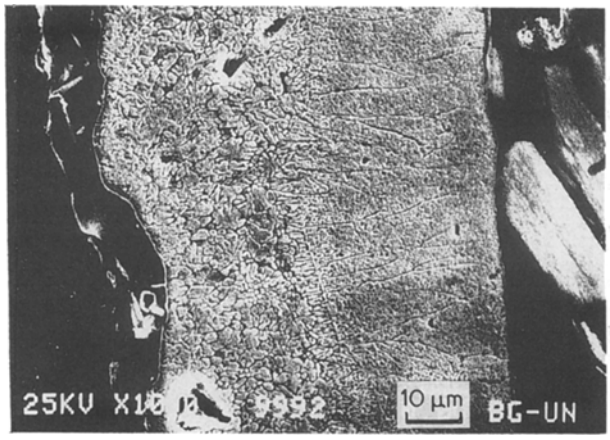


Figure 2 SEM longitudinal transverse cross-section micrograph of the ribbon, showing the two distinct types of micromorphologies. Contact surface is on the right.

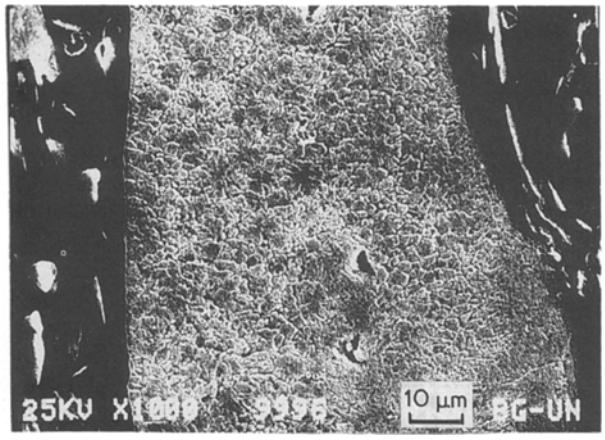
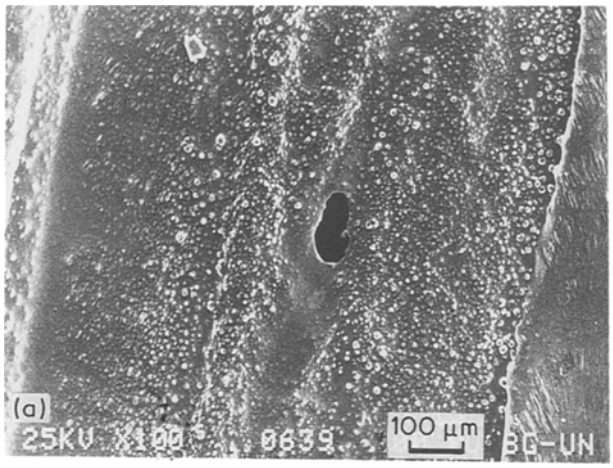
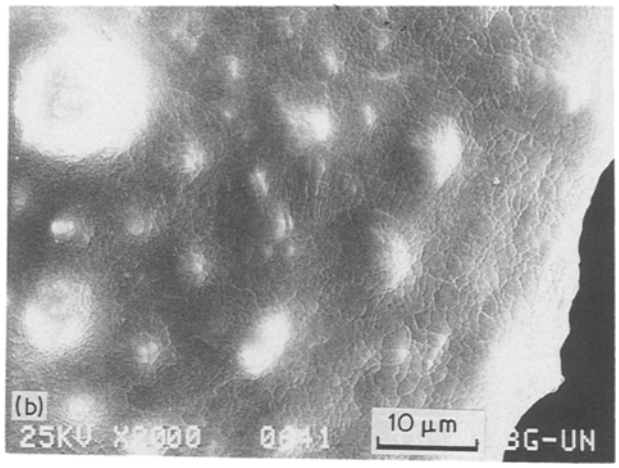


Figure 3 SEM longitudinal transverse cross-section micrograph of the ribbon, showing a single type of micromorphology. Contact surface is on the left.





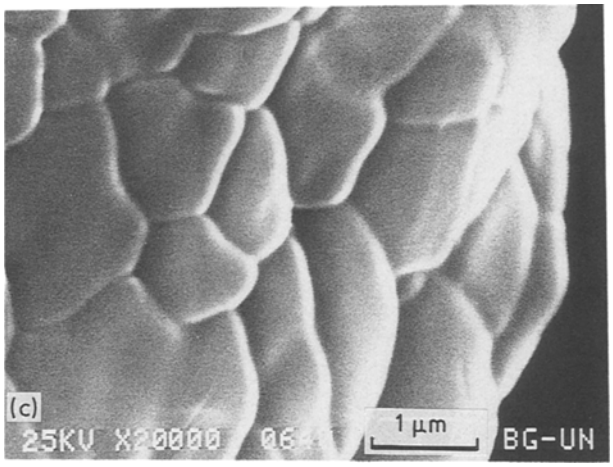


Figure 4 (a, b, c) Scanning electron micrographs of a T0 flat, free surface on top of a ribbon, showing the edge of a hole at different magnifications. Similar locations were studied by TEM.

macroconcentration in a mixed mode, cross-section, micromorphology was 3.6 wt % La in columnar structure; 2.6 wt % La in equiaxed structure. The average lanthanum macroconcentration sampled from etched ribbons having an equiaxed structure only, was nearly uniform at 2.7 wt % La across the ribbon.

High cooling rates can widen the lanthanum concentration range in an aluminium matrix either by increasing miscibility, or by the formation of finely dispersed, lanthanum-rich secondary phases. Lanthanum excess, which was not trapped in the matrix during solidification, segregates to the grain boundaries. In spite of selective etching, which removed lanthanum or lanthanum-rich phases from grain boundaries, the lanthanum concentration in the RS columnar and equiaxed structures was much higher than dictated by the equilibrium phase diagram, suggesting lanthanum, or lanthanum-rich phase entrappment in the matrix.

The differences in lanthanum concentration between columnar and equiaxed structures is due to the fact that in the equiaxed structure, grain-boundary density is relatively high and the total amount of lanthanum migrating to these boundaries is higher than in the columnar. Selective etching of the equiaxed structure resulting in a lower overall lanthanum concentration than in the etched columnar structure.

Figs 4a to c are scanning electron micrographs of a T0, flat, free surface of the top of a ribbon, showing the edge of a hole, at different magnifications. Similar locations, transparent to the TEM electron beam, are shown in the transmission electron micrograph in Fig. 5. The existence of a circular cross-section phase, not uniformly distributed, with the smaller diameter particles occupying the space near the hole's edge can be observed, and as the location moves into the thicker part of the ribbon, the particle diameter gradually increases.

Upon heating to 500° C in the TEM hot-stage, the very fine particles rapidly disappear, and as the exposure to 500° C is continued, a more significant volume reaction takes place, shown by the sequential micrographs in Figs 6a to d. The circular phase dissolves and an irregular-shaped phase starts to form. The earliest recorded evidence of the formation of the irregular phase can be seen in Fig. 6b as a needle-like streak at the upper right corner in the micrograph. A 400° C TEM hot-stage exposure results in similar volume reactions, but apparently at a lower rate.

The combined XRD partial diffractograms shown in Fig. 7 show the as-received intensities of samples at different conditions, including the master alloy. Comparing the T0 condition to the MA, at the low angles of the diffractograms, an additional peak is present in T0. This single extra intensity gradually decreases in the T1, T2, T3, T4, and T5 conditions. All other peaks are congruent with the peaks present in the master

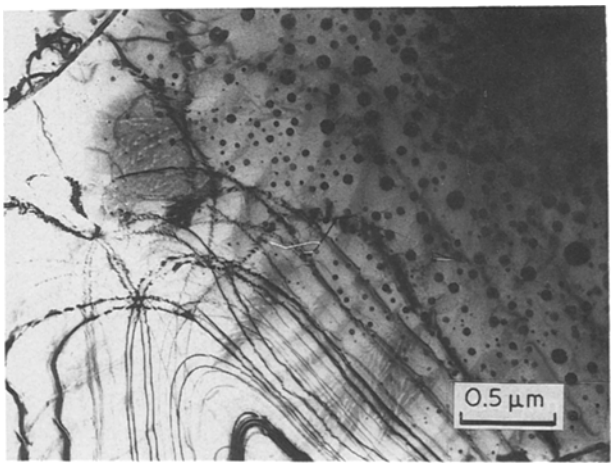


Figure 5 Transmission electron micrograph of a T0 ribbon location transparent to the electron beam.

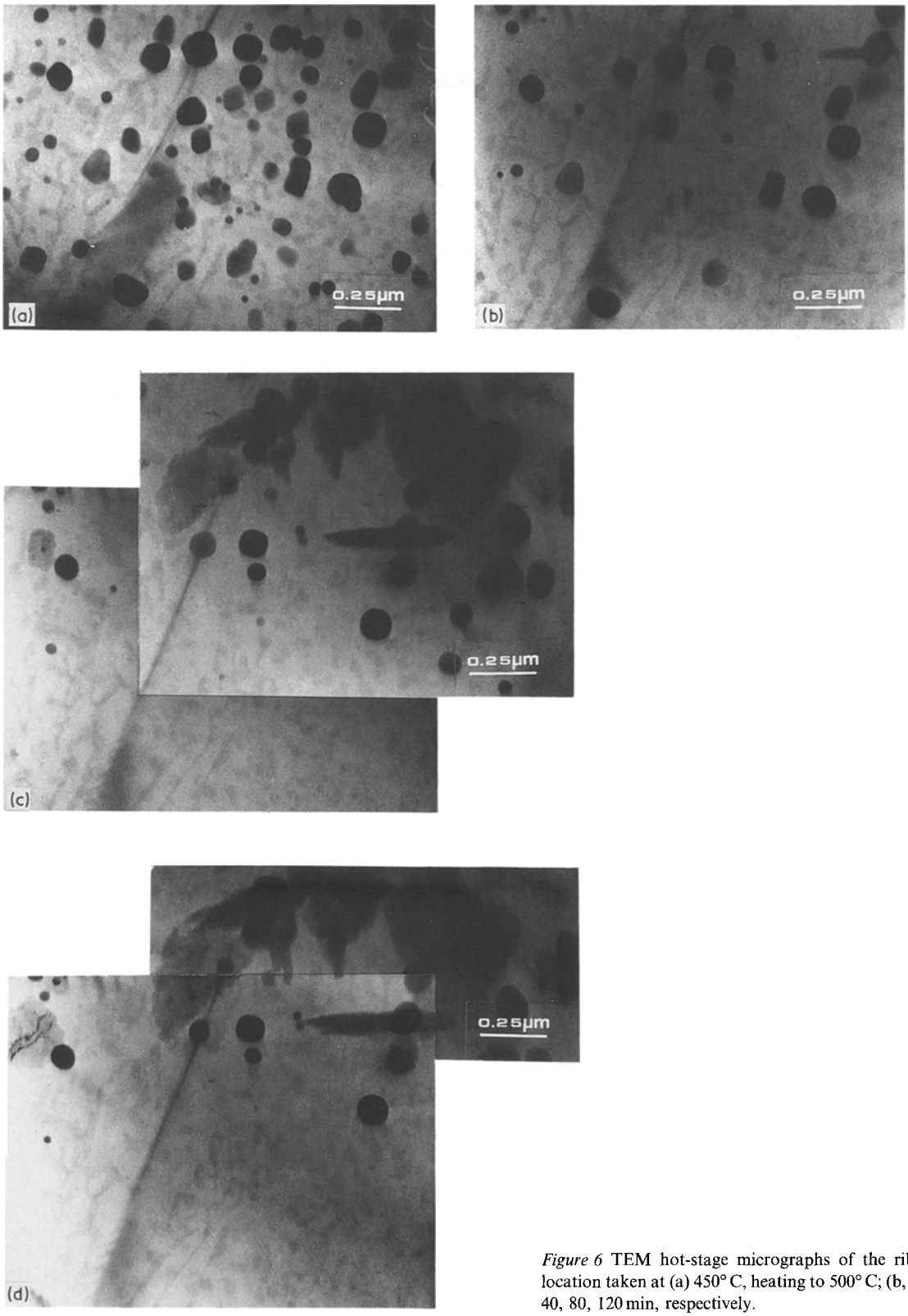


Figure 6 TEM hot-stage micrographs of the ribbon transparent location taken at (a) 450° C, heating to 500° C; (b, c, d) 500° C, after 40, 80, 120 min, respectively.

alloy. TEM and XRD indicate the existence of a metastable phase which upon heating undergoes a gradual decomposition. This phase has not yet been identified or indexed. The other phase has been identified as the equilibrium $Al_{11}La_3$ phase.

It appears that the formation of the metastable phase, in terms of size and distribution, is strongly cooling-rate dependent. The fact that the very small dispersoids form in the thinner section of the transparent location, where the cooling rate is the highest, supports this assumption. From TEM and XRD results it is evident that the metastable phase, up to 400° C (2 h exposure), is fairly stable, but at 500° C this phase decomposes rapidly, and the formation of a coarse, stable $Al_{11}La_3$ phase is observed.

The fact that no substantial angle shifts of aluminium matrix peaks in the RS XRD diffractograms were observed, indicates that lanthanum solid solubility in the matrix was low, not effecting any significant, relative or absolute changes in the aluminium lattice parameter.

A scanning electron micrograph taken from the master alloy, Fig. 8, shows a rounded elongated grain structure and a heavy intergranular structure of a fine

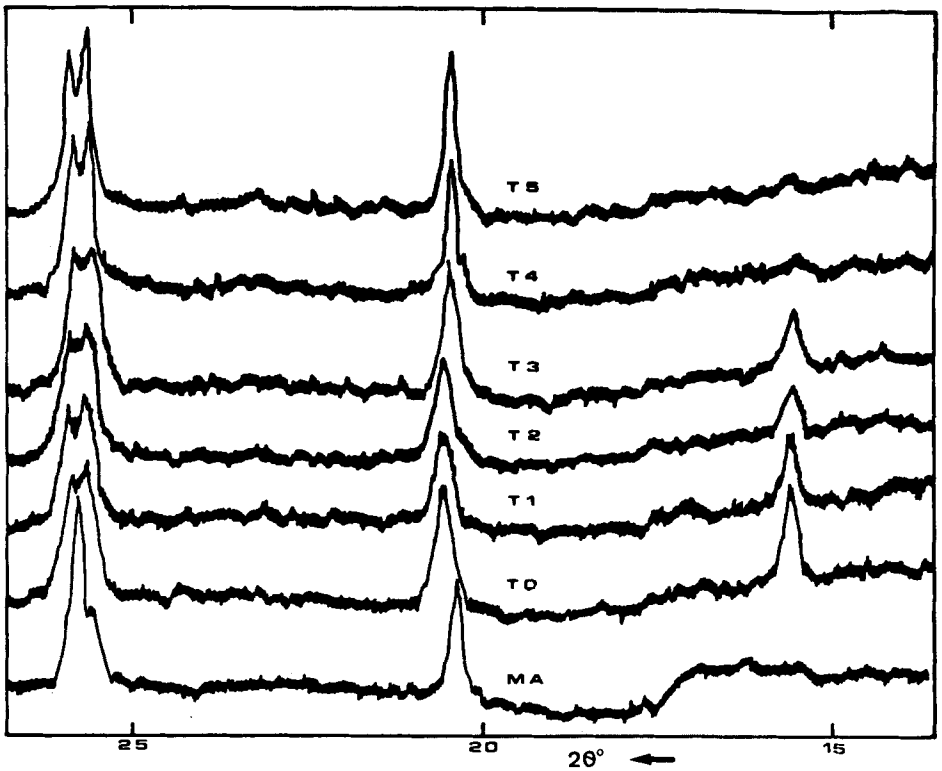


Figure 7 X-ray diffraction combined partial diffractograms from the master alloy and from the ribbon at different conditions.

lamellar eutectic. The overall average lanthanum macroconcentration was 4 wt % La, while the average lanthanum concentration in the grain centres and in the eutectic was 0.5 and 11 wt % La, respectively.

The RS products in terms of microstructure, solute distribution and grain size, are clearly different from the master alloy. While the MA, in general, can be considered as a conventionally solidified alloy, having relatively large grains and a well-developed eutectic, the RS ribbons, on the other hand, have a finer microstructure, a metastable phase and no evident traces of eutectic formation.

4. Conclusions

RS processing promotes the formation of a relatively finer microstructure in an Al–4La alloy, as well as the formation of a finely dispersed metastable phase. This metastable phase, as indicated by TEM and XRD studies, is fairly stable up to 400° C, 2 h exposure. The lanthanum concentration in the ribbon bulk matrix, compared to the conventionally solidified MA, is considerably increased on using RS processing, and the formation of a heavy eutectic is suppressed.

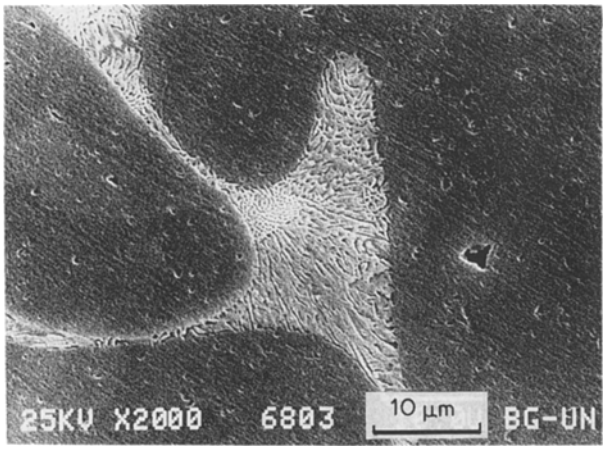


Figure 8 Scanning electron micrograph of the Al–4La master alloy (MA).

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