An Investigation of Cast Structures in Al-Hf-(Sc)-(Zr) Alloys and Their Subsequent Effect on Recrystallisation Resistance after Cold Rolling

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Abstract

In order to develop aluminium alloys with a high structural stability, dispersoid-forming elements are added. Examples of such elements are scandium (Sc) and zirconium (Zr). In this work Hf, which is a relatively untried element with respect to recrystallisation resistance, was added to pure aluminium in combination with Sc and Zr. The results revealed that twinned columnar grains (TCG's) formed during solidification at a high Hf-content (1.1 wt%) in both Al-Hf-Zr and Al-Hf-Sc. However, when the Hf-content was kept low (0.2 wt%) a more equiaxed structure was obtained. The effect of TCG's on the recrystallisation resistance after cold rolling was also examined. However, despite the presence of twinned columnar grains a remarkably high recrystallisation resistance was found for Al-Hf-Sc.

1. Introduction

During cold-rolling, deformed, "pancake"-shaped grains with a high interior dislocation density do form, and the stored energy in such deformation structures provide the driving force for recrystallisation [1]. As recrystallisation is connected with a considerable loss in strength, dispersoid-forming elements are added to aluminium alloys in order to inhibit this process [2-3]. Dispersoids exert a drag force, often referred to as the Zener-drag, on the moving subgrain boundaries during annealing, and prevent the subgrains from becoming potential nuclei for recrystallisation. In order to obtain the largest possible drag force, a high volume fraction of small and homogeneously distributed dispersoids is necessary. It is also important that the interfacial energy between the particle and matrix is low and that the dispersoids are thermally stable in order to avoid excessive coarsening during heat exposure [4].

Examples of elements, which form dispersoids with such characteristics, are Sc and Zr, which forms Al_3X-dispersoids (X=Sc,Zr) that increases the recrystallisation resistance during high temperature annealing. However, one of the major drawbacks by using these elements, is that they may lead to very coarse grained as-cast structures. In this work Hf, which is a relatively untried element with respect to recrystallisation resistance, was added to aluminium in combination with Sc and Zr. One of the objectives of this work has been to investigate how such additions affect the as-cast structures. Another objective was to study how the as-cast structures in turn influence the recrystallisation resistance after cold rolling.

2. Experimental

2. 1 Casting.

The alloys were produced by mixing appropriate amounts of Vigeland metal (99. 999% Al) and master alloys of Al-5wt%Hf, Al-10wt%Zr and Al-2wt%Sc. The alloys were directionally solidified in order to reduce porosity and solidification contraction at the top. A well stirred melt was poured at 760^oC into a cylindrical fibre tube mould (Ø40mm x150mm) and cooled in the bottom by a large copper cylinder. The actual temperature was monitored by thermocouples during solidification, and the cooling rate was found to vary between 5-10^oC/s. Finally, it should be mentioned that no grain refiner was added during casting.

2.2 Homogenisation and Cold Rolling.

All alloys were homogenised in a Heraeus K750 air-circulating furnace for 20hr at 475^oC. A heating rate of 50^oC/hr was applied. An external reference thermocouple was used in order to monitor the temperature in the oven during the heat treatment. After homogenisation the samples were quenched in water and subsequently cold rolled 90% to a thickness of approximately 1.1 mm. Between each pass the sample were rotated 180^o.

2.3 Annealing.

In order to investigate the structural stability, i.e. recrystallisation resistance, samples were cut from the cold rolled material and annealed from room temperature in a Heraeus K750 air-circulating furnace using a heating rate of 50ºC/hr. The samples were removed from the furnace at temperatures between 100ºC and 600ºC and subsequently quenched in water.

2.4 Hardness and Conductivity.

Vickers Hardness measurements were carried out in order to follow the softening reactions during annealing. The hardness was determined as the average of 4 measurements at a load of 1 kg. A Foster-Sigmatest D 2.068 was used in order to measure the conductivity, i.e to monitor the precipitation reactions occurring during annealing. A frequency of 60kHz was applied.

2.5 Optical Microscopy.

Samples were anodised at an applied voltage of 20VDC using an electrolyte consisting of 0.5L of H_2O, 2.3ml fluoboric acid and 3.5g boric acid. Polarised light was then used in the investigation of the grain structure in a Leica MEF4M.

2.6 TEM

A Jeol 2010 operated at 200kV was used for all TEM work. TEM samples were prepared by electro-thinning in 75% methanol/25% nitric acid in a Struers Tenupol. A temperature of -25^oC and a voltage of 15V were used.

3. Results and Discussion

3.1 Alloy Selection

Table 1 The chemical composition (wt%) of the alloys.

Alloy 1	Al-0.2Hf-0.15Zr
Alloy 2	Al-1.1Hf-0.15Zr
Alloy 3	Al-0.2Hf-0.2Sc
Alloy 4	Al-1.1Hf-0.2Sc

3.2 Cast structures

One of the objectives of this investigation was to study how the various additions of Hf, Sc and Zr influenced the as-cast structures and Figure. 1-2 show that highly different microstructures were obtained. In alloys 1 and 3, i.e. the variants with a low Hf-content (0.2 wt%), a fairly equiaxed structure was observed. However, feather crystals or twinned columnar grains (TCG's) were observed in alloys 2 and 4, which had a Hf-content of 1.1 wt% (see Figure. 3-4). Later investigations have shown that the Hf content should be kept below approximately 0,7wt% Hf in order to prevent TCG's from forming in Al-Hf-Zr. It is likely that a transition from an equiaxed structure to a structure containing TCG's exists for Al-Hf-Sc as well, but this has not yet been investigated.

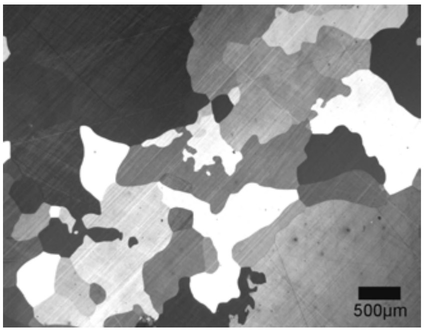

Figure 1: Anodised alloy 1 sample.

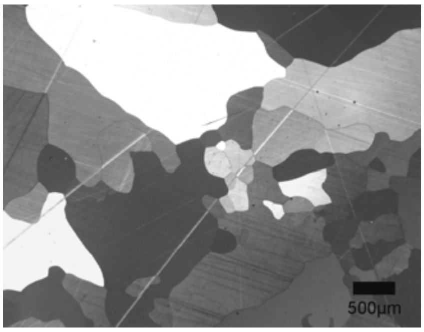

Figure 2: Anodised alloy 3 sample.

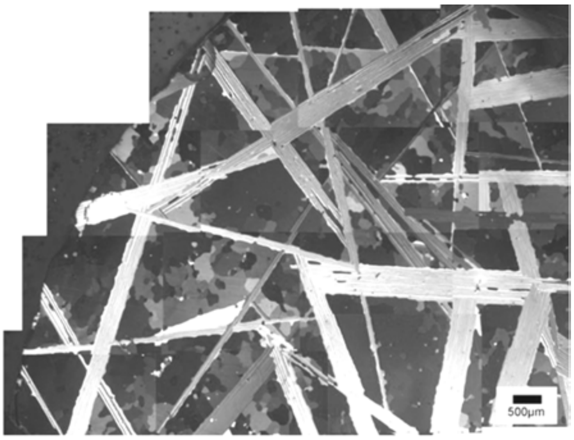

Figure 3: Alloy 4 micrographs showing feather grains (TCG).

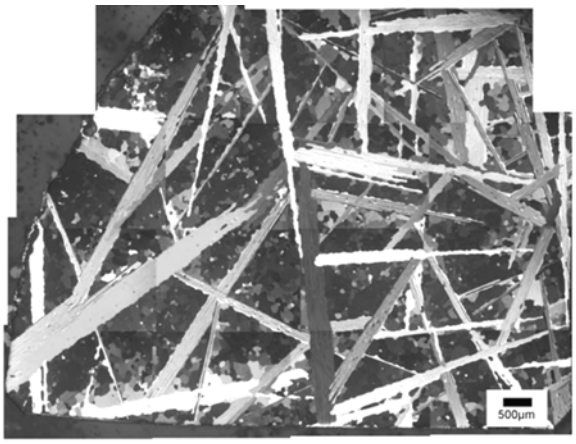

Figure: 4 Alloy 2 micrographs showing feather grains (TCG).

TCG's have often been found to form in cases where the grain refinement has been poor [5]. As no grain refiner was used during casting of these alloys, this is a likely reason for TCG-formation also for the present alloys. However, previous investigations [6] and the above results clearly show that the formation of such crystals also must be connected to the amount of Zr and Hf. In alloy 2 and 4 a large amount of primary phases were observed, while few of these were observed in the variants with a lower Hf-content. It is possible that the formation of TCG's in alloys 2 and 4 can be related to the presence of these phases, but a more thorough investigation is needed before a clear conclusion can be made.

3.3 Precipitation annealing

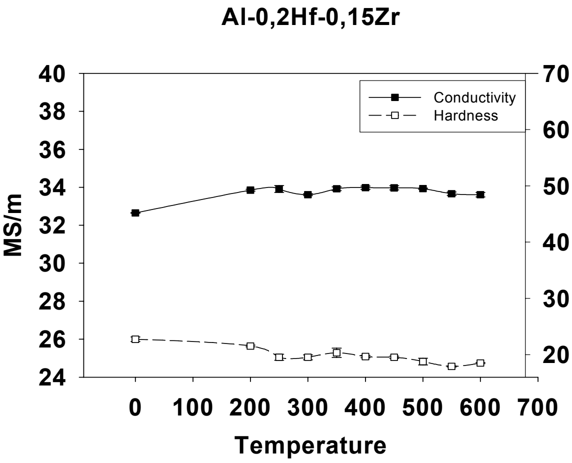

Figure 5: Alloy 1 slowly heated (50°C/hr) from room temperature to 600°C in order to determine the optimum precipitation annealing temperature.

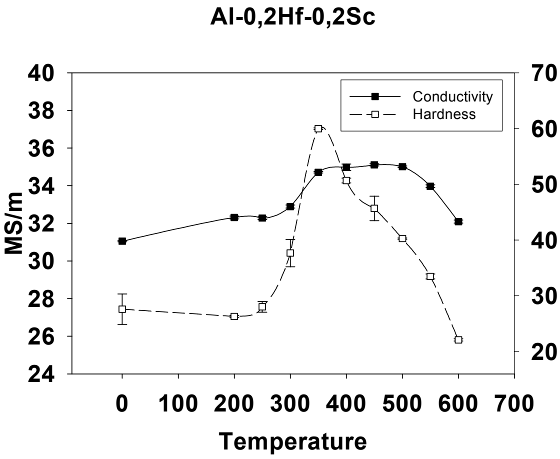

Figure 6: Alloy 3 slowly heated (50°C/hr) from room temperature to 600°C in order to determine the optimum precipitation annealing temperature. The peak in hardness is due to the formation of $Al_3(Hf,Sc)$-dispersoids.

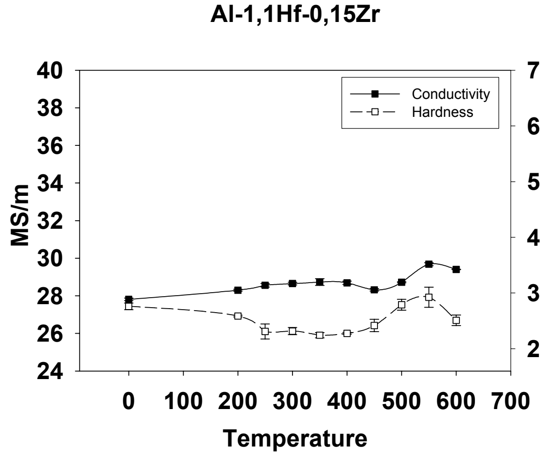

Figure 7: Alloy 2 slowly heated (50°C/hr) from room temperature to 600°C in order to fine the optimum precipitation annealing temperature.

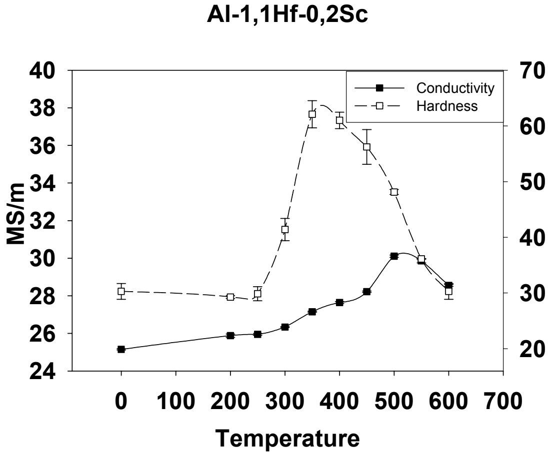

Figure 8: Alloy 4 slowly heated (50°C/hr) from room temperature to 600°C in order to determine the optimum precipitation annealing temperature. The peak in hardness is due to the formation of $Al_3(Hf,Sc)$-dispersoids.

Previous investigations have shown that Al_3Zr does not form if the heating rate during precipitation annealing is too high [7]. As the effect of heating rate on the formation of $Al_3(Hf,Zr)$ is unknown, a heating rate of 50°C/hr was consequently applied for all alloys. Furthermore, in order to find the optimum precipitation annealing temperature, the conductivity and hardness were first measured during slow heating from room temperature to 600°C as described in the previous section.

Figure. 6 and 8 show that the precipitation of $Al_3(Hf,Sc)$-dispersoids begins at 200-250°C in the Al-Hf-Sc-variants, and the formation of these phases is accompanied by a significant increase in strength, which reaches a maximum at ~350°C. However, as maximum conductivity was obtained at 450°C-500°C, a precipitation annealing temperature of 475°C was chosen.

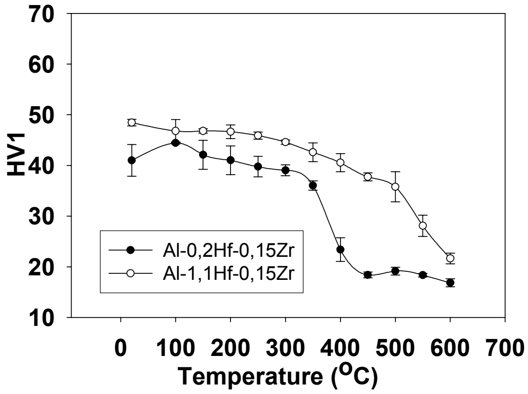

Figure 9: Softening curves for the Al-Hf-Zr-variants, (alloys 1 and 2).

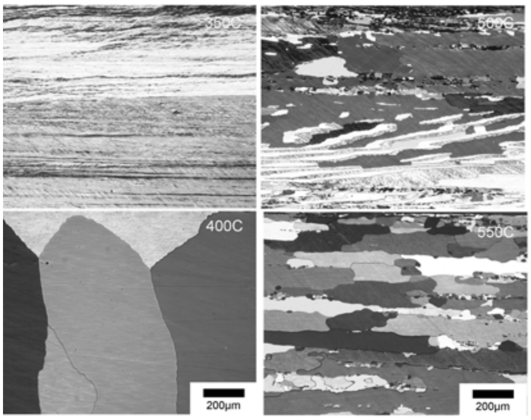

Figure 10: The micrographs to the left show the Al-0.2Hf-0.15Zr alloy at 300^{o}C and 400^{o}C, respectively, while the micrographs to the right show Al-1.1Hf-0.15Zr at 450^{o}C and 550^{o}C.

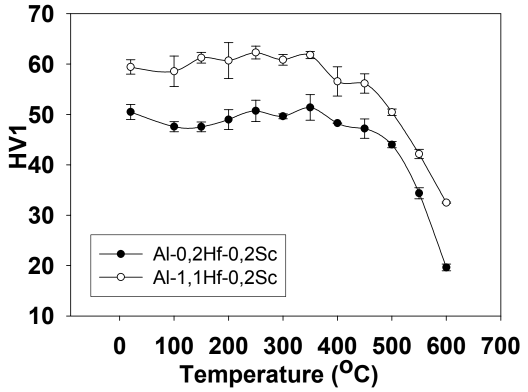

Figure 11: Softening curves for Al-Hf-Sc-variants, (alloys 3 and 4).

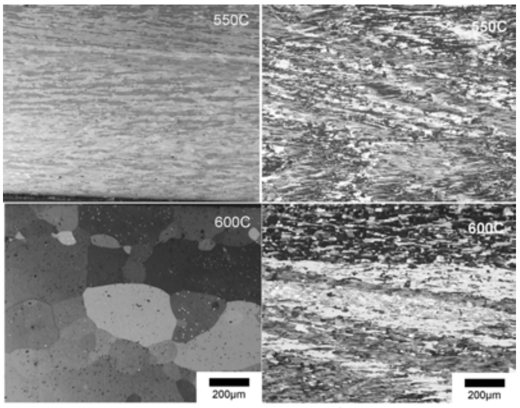

Figure 12: The micrographs to the left show the Al-0.2Hf-0.2Sc alloy at 500^{o}C and 600^{o}C, respectively, while the micrographs to the right show Al-1.1Hf-0.2Sc at 500^{o}C and 600^{o}C.

In the Al-Hf-Zr-alloys the changes in both hardness and conductivity were relatively small (Figure. 5 and 7). This is in accordance with the TEM-observations, which have shown that few and relatively coarse $Al_3(Hf,Zr)$-dispersoids formed in alloy 1, while a heterogeneous distribution of these phases was found in alloy 2. This explains the small hardness increase in this alloy compared to alloy 1. However, the dispersoid number and density in alloy 2 was far lower than what was observed in alloy 3 and 4.

The slight increase in alloy 2 took place between 400^{o}C and 550°C, and in order to compare directly with the Al-Hf-Sc-variants , 475°C was chosen as the precipitation annealing temperature also for the Al-Hf-Zr-alloys.

3.4 Softening after cold rolling

After annealing the as-cast variants for 20 hours at 475°C, the alloys were cold rolled to 90% reduction. Figure 9 displays the softening curves for the Al-Hf-Zr alloys. These curves show that while alloy 1 only resist recrystallisation up to 400^{o}C, alloy 2 was able to resist recrystallisation up to 500^{o}C (see Figure 10). The higher recrystallisation resistance can be attributed to the higher Hf content, which led to the formation of more $Al_3(Hf,Zr)$ dispersoids.

As expected from previous investigations [2,7], a higher recrystallisation resistance was obtained when Sc was used instead of Zr in combination with Hf (Figure 11).

However, once again a higher Hf-content is beneficial with respect to recrystallisation resistance as the first signs of recrystallisation were observed at 550°C and 600°C in alloy 3 and alloy 4, respectively (Figure. 12). The high recrystallisation resistance of these variants can of course be attributed to the dense and homogeneous distribution of small Al_3(Hf,Sc)-dispersoids (r~12nm), see Figure 13. It is important to notice that alloy 4 displays an equally high recrystallisation resistance as other Sc- and Zr-containing alloys with a more eutectic-like as-cast grain structure [8]. This indicates that a possibly negative effect of TCG's on the recrystallisation resistance is overshadowed by the positive effect from the dispersoids.

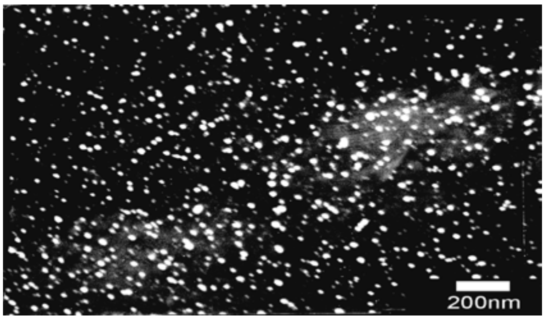

Figure 13 Dark Field TEM micrographs of dispersoids Al_3(Hf,Sc) in alloy 3 after homogenisation.

4. Conclusions

1. Twinned columnar grains (TCG's) were observed in the as cast structure of the alloys with a high Hf-content (1.1wt%).
2. Homogeneous and dense dispersoid distributions were obtained in the Al-Hf-Sc-alloys. In the Al-Hf-Zr-variants, however, few relatively coarse and heterogeneously distributed dispersoids were obtained. The Al-Hf-Sc-alloys consequently displayed a far better recrystallisation resistance than Al-Hf-Zr.
3. The presence of TCG's in the as cast structure is not believed to have a great negative influence the recrystallisation resistance. In the alloys studied here a possible negative effect seems to be overshadowed by the positive effect from the dispersoids.

Acknowledgement

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