Study of twin-roll cast Aluminium alloys subjected to severe plastic deformation by equal channel angular pressing

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Abstract. Aluminium alloys prepared by twin-roll casting method become widely used in industry applications. Their high solid solution supersaturation and finer grains ensure better mechanical properties when compared with the direct-chill cast ones. One of the possibilities how to enhance their thermal stability is the addition of zirconium. After heat treatment Al₃Zr precipitates form and these pin moving grain boundaries when the material is exposed to higher temperatures.

In the present work twin-roll cast aluminium alloys based on AA3003 with and without Zr addition were annealed for 8 hours at 450 °C to enable precipitation of Al₃Zr phase. Afterwards they were subjected to severe plastic deformation by equal channel angular pressing, which led to the reduction of average grain size under 1 µm. During subsequent isochronal annealing recovery and recrystallization took place. These processes were monitored by microhardness measurements, light optical microscopy and in-situ transmission electron microscopy. The addition of Zr stabilizes the grain size and increases the recrystallization temperature by 100 °C.

1. Introduction
Nowadays, aluminium is due to its superior properties the most widely used non-ferrous metal in many branches of industry. Because the grain size is one of the principal tools for controlling mechanical properties of polycrystalline aluminium [1], many studies have been devoted to investigation of methods of grain refinement [2-4]. Severe plastic deformation (SPD) of bulk billets, which achieves ultra-fine grained structure by imposing a very high plastic strain [5], seems to be the most promising processing route for manufacturing of nanostructured aluminium alloys for high range of industrial applications [1]. The most common SPD technique is equal channel angular pressing (ECAP). The sample of required dimensions is pressed through a die which consists of two channels of equal crosssection that intersect at an angle Φ [5], see fig. 1. The main advantage of this technique is the possibility of repeating the pressing several times to induce a required level of strain into the material.

Twin-roll casting (TRC) is one of the methods of casting metallic strips [6-7]. Metal or alloy is melted in a holding furnace and the melt flows through a launder and a cast box to a nozzle, which directs the melt between two water-cooled rolls. As compared to conventional casting, twin-roll casting is less demanding on the amount of energy and material used during manufacturing; thus the production of strips is more economic. However, the initial microstructure of the TRC strips shows some differences from conventionally cast materials, therefore it has been carefully studied in recent years [8-12]. The high solidification rate of around 500 °C/s results in a microstructure refinement, formation of finely distributed primary particles and high solid solution supersaturation.

Many works have been devoted to the investigation of microstructure evolution during and after ECAP processing in conventionally cast aluminium alloys, e.g. [13-15]. However, the application of the ECAP on TRC alloys is still to be investigated.

It is a well known fact that the addition of small amount of zirconium into aluminium alloys leads to the enhancement of mechanical properties [16-17]. After heat treatment at 450 °C [18-20] coherent precipitates of cubic Al₃Zr phase form in the aluminium matrix. These particles pin moving grain boundaries, and thus shift the onset of recrystallization to higher
temperatures and refine the grain size [21-22]. This effect has been observed in both conventionally cast and twin-roll cast aluminium alloys after various methods of deformation like cold-rolling [23]. The aim of the present study is to evaluate the role of $\text{Al}_3\text{Zr}$ precipitates on the recrystallization and grain growth processes in TRC alloys processed by ECAP.

2. Experimental

Two aluminium alloys from AA3003 series with nominal composition 1.0-1.5 wt.% Mn, ≤ 0.7 wt.% Fe, ≤ 0.6 wt.% Si and 0.05-0.2 wt.% Cu were studied. The first one as a reference denoted “Al” and the second one “Al+Zr” with addition of 0.16 wt.% Zr.

Both alloys were twin-roll cast in industrial conditions to strips 8 mm thick in the normal direction (ND) and 1 m wide in transversal direction (TD). The as-cast materials were annealed in an air furnace. They were heated to 450 °C with a heating rate of 0.5 K/min, held at 450 °C for 8 hours and subsequently quenched into cold water.

The sheets were cut to billets with length 120 mm in the rolling direction (RD) and subjected to severe plastic deformation by equal channel angular pressing at room temperature. The cross section of the channel was 10 x 10 mm$^2$ and the intersecting angle of the two channels was $\Phi = 90^\circ$. The equivalent strain induced into the material after one pass was $\varepsilon \sim 1$ [5]. The total number of passes was four and the billet was rotated by 90° around its axis (RD) after each pass (so called route $B_c$ [24]). The pressing speed was 10 mm/min. The orientation of the TRC materials during the odd ECAP passes is depicted on fig. 1; as the route $B_c$ was used, during even passes the ND and TD are switched.

The evolution of the microstructure and mechanical properties during isochronal annealing after ECAP with regard to the content of zirconium in the alloy were explored by microhardness measurement, light optical microscopy and transmission electron microscopy (TEM). The isochronal annealing was performed with steps of 50 °C/50 min. The Vickers microhardness was measured with a load of 100 g; for light optical microscopy in polarized light the samples were electrolytically etched by Barker solution to visualize the grains. The TEM observations were done in JEOL JEM 2000FX equipped with an in-situ heating unit operated at 200 kV.

3. Results

The as-cast materials have grains of size in the order of 100 µm and contain primary particles of $\alpha$-Al(Mn,Fe)Si phase [17] with an average size of 1 µm. The initial annealing at 450 °C led in both “Al” and “Al+Zr” alloys to the formation of secondary particles of $\alpha$-Al(Mn,Fe)Si phase with an average size of 100 nm. Additionally, coherent $\text{Al}_3\text{Zr}$ precipitates with a diameter of about 10 nm precipitated in the Zr-containing material (Figure 2).

After four ECAP passes the average grain size dropped under 1 µm in both materials (Figure 3) and the microhardness increased significantly, from 50 MPa to 80 MPa in the reference alloy and from 60 MPa to 90 MPa in the alloy with zirconium addition.
During isochronal annealing the values of Vickers microhardness (HV0.1) first decreased only slowly, afterwards a significant drop was observed in both materials and finally at temperatures above 450 °C the microhardness stagnated at values around 45 MPa (see Fig. 4). The addition of Zr significantly influenced the main microhardness drop. Concerning the Zr-containing alloy “Al+Zr”, the decrease occurred between the temperatures of 400 °C and 450 °C. At 400 °C first recrystallized grains were observed in the central part of the billet in the vicinity of large primary particles. After further annealing to 450 °C the material was fully recrystallized with grain size in the order of 100 µm (Fig. 5). Further annealing to 600 °C led only to subtle grain coarsening. However, in the reference material “Al” the drop of microhardness was observed at annealing temperatures by 100 °C lower. After annealing up to 300 °C the material is only partially recrystallized, while annealing to 350 °C resulted in almost full recrystallization. The average size of the recrystallized grains was much larger than in the “Al+Zr” material, the diameter of the largest grains approached 1 mm (Fig. 5).

The in-situ heating experiments in TEM revealed substantial microstructural changes in the course of annealing (Fig. 6 and 7). In both alloys the dislocation substructure recovered at temperatures below 200 °C. At higher annealing temperatures the grain boundary migration and grain coalescence was observed accompanied by a significant grain growth. During annealing after ECAP no new precipitates of α-Al(Mn,Fe)Si phase were formed; only those already present in the matrix after deformation first slightly coarsened and then partially dissolved at temperatures above 450 °C (Fig. 3).

Figure 2: Detail of Al$_2$Zr precipitates in the Zr containing alloy after annealing at 450 °C. Left: dark field image in TEM, right: diffraction pattern in [001] zone axis with additional spots from coherent Al$_5$Zr phase.

Figure 3: Zr containing material after 4 ECAP passes. Microstructure with submicrometric grains just after ECAP (left) and recrystallized material isochronally annealed to 450 °C with particles of the primary phase (indicated by an arrow) and precipitates of α-Al(Mn,Fe)Si phase (right).
Figure 4: Evolution of Vickers microhardness during isochronal annealing 50 °C/50 min for the reference material “Al” and the material with zirconium addition “Al+Zr”.

Figure 5: Grain structure in the reference material after annealing up to 350 °C (top) and in the Zr-containing material at 450 °C (bottom).
4. Discussion

The initial heat treatment at 450 °C led to the precipitation of dense dispersion of coherent Al₃Zr particles in the Zr-containing alloy. These were found to have a major influence on the thermal stability of the deformed material.
The severe plastic deformation introduced into the material by four passes of equal channel angular pressing caused significant grain segmentation resulting in submicrometric grain size. This effect was also responsible for augmentation of the microhardness values. During isochronal annealing with a heating rate of 50 °C/50 min a softening of the material indicated by a decrease of the microhardness was caused by substructure recovery and recrystallization. The in-situ transmission electron microscopy showed that the recrystallization occurred in a continuous manner. During the continuous recrystallization the highly deformed alloy may transform during annealing by relatively localized boundary migration to a microstructure of approximately equiaxed defect-free grains, which are predominantly bounded by high angle grain boundaries [4]. It has been shown that the continuous recrystallization is promoted by large strains induced into the material and second-phase particles [4, 25], which is the case of the materials investigated in this study, because a dense dispersion of secondary particles of α-Al(Mn,Fe)Si phase was achieved by annealing at 450 °C prior to the ECAP processing. The Al\textsubscript{3}Zr precipitates beneficially contributed to the shift of the recrystallization and the main microhardness decrease to the temperature range of 400-450 °C in the Zr-containing material; as compared to the reference material which recrystallized between 300-350 °C. The average grain size after isochronal annealing was much lower in the Zr-containing material, which is another positive effect of the Al\textsubscript{3}Zr particles – they pin moving grain boundaries and inhibit grain growth [26].

5. Conclusion
The influence of zirconium addition on the evolution of microstructure of twin-roll cast aluminium alloys at elevated temperatures was studied. After four ECAP passes both materials with and without Zr contained grains of submicrometric size. In the course of isochronal annealing recovery of deformed substructure and continuous recrystallization caused significant softening of materials. The recrystallization in the alloy with Zr addition took place at temperatures by approximately 100 °C higher than in the Zr-free alloy; moreover, the observed grain size was lower in the Zr-containing alloy. This shift in recrystallization resistance and stabilization of the grain size was attributed to the presence of coherent Al\textsubscript{3}Zr precipitates in the aluminium matrix.

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