Deformation mechanisms in ARB processed aluminium alloy AA6016 at low temperatures

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Abstract. The deformation behaviour of the aluminium alloy AA6016 at low temperatures was investigated in the initial state and after 4 and 8 cycles of accumulative roll bonding (ARB). Tensile tests at 25 K, 77 K, 180 K and 296 K were performed at constant strain rate $10^{-4}$ s$^{-1}$. Stress relaxation experiments performed during the tensile tests were used to determine the experimental rate sensitivity $\lambda$ as a function of the flow stress $\sigma$. In all cases $\lambda(\sigma)$ is found to be linear revealing that the Cottrell-Stokes law holds. The effect of grain size on $\sigma$ can be adequately described through an additive athermal stress contribution $\sigma_d$, which is the higher the higher the degree of pre-deformation is. Moreover, the temperature dependence of the strain rate sensitivity $m(T)$ indicates that the rate controlling mechanism in the initial state is local single slip. The ARB states deviate from the single slip behaviour already at 25 K. The reason probably is the occurrence of additional thermally activated slip processes in the ARB states.

1. Introduction
In the last decade severe plastic deformation by accumulative roll bonding (ARB), first introduced by Saito et al. [1], turned out to be a very promising technique regarding “large scale production” of ultrafine grained (UFG) metals and alloys with grain sizes ranging from 1 µm to about 100 nm [2 - 4]. Compared to their conventional large grained counterparts, UFG materials show a markedly enhanced strength (e.g. [5]) with only a slightly decreasing and in some aluminium alloys even increasing ductility [6 - 8].

The influence of ARB processing on the tensile behaviour at ambient temperatures has been described for various UFG materials, e.g. for the technically relevant age-hardening aluminium alloy AA6016 [9]. Investigations at low temperatures are less frequent. Principally, investigations at low temperatures permit to study quantitatively the influence of ARB processing on the temperature dependent transition from pure dislocation slip controlled plasticity to diffusion controlled plasticity in aluminium AA6016.

This study aims at a qualitative investigation of the relevant deformation mechanisms in AA6016 at and below room temperature. For this purpose samples taken from three ARB sheets with different degree of deformation were subjected to tensile tests with stress relaxation experiments at temperatures between 25 K and 296 K. An application of thermal activation theory (e.g. [10]) then permits to distinguish between pure athermal (“Hall-Petch” like) and thermal (“nano” deformation mechanisms) effects of ARB processing on the flow stress [11].
2. Experimental

In this investigation samples of the initial state and after four and eight ARB cycles were tested. Prior to ARB the plates with a thickness of 1 mm were solution heat treated in a furnace for 1h at 520°C and subsequently quenched in water. The ARB-process with a thickness reduction of 50% per rolling pass was applied to aluminium plates with a width of 100 mm and a length of approximately 300 mm using a four high rolling mill (Carl Wezel, Mühlacker). Prior to rolling the plates were pre-warmed at 230°C for four minutes. After each cycle the surfaces of the air-cooled bonded plates were wire brushed to remove the oxide layer. The ARB process was done up to a maximum of eight cycles, which corresponds to a von Mises strain of 6.4 and leads to a plate with \( 2^8 = 256 \) bonded layers each having a thickness of about 3.9 µm. The conditions of the ARB processing are described in more detail in [12]. The chemical composition of the aluminium alloy AA6016 is listed in Table 1.

Table 1. Chemical composition of AA6016 (mean values)

|   | Si    | Cu  | Fe  | Mn  | Mg   | Cr  | Zn  | Ti  | other | Al   |
|---|-------|-----|-----|-----|------|-----|-----|-----|-------|------|
| wt.% | 1.0-1.5 | 0.52 | 0.5 | 0.2 | 0.25-0.6 | 0.1 | 0.2 | 0.15 | 0.15  | balance |

Figure 1 shows the microstructures of the sheets investigated, determined by electron back scatter diffraction (EBSD) measurements in a Zeiss Ultra 55 scanning electron microscope using 40 nm step size. The average grain size determined by the line intersection method is about 16 µm in the initial state. After four and eight ARB cycles the grain size is \( d_{RD} = (660 \pm 170) \) nm and \( (380 \pm 40) \) nm along the rolling direction (RD) and \( d_{ND} = (400 \pm 70) \) nm and \( (180 \pm 40) \) nm along the normal direction (ND), respectively. In all cases grain boundaries with misorientation angles \( \geq 3° \) were taken into account.

Figure 1. Aluminium alloy AA6016 in the initial state (left) and after 4 (upper right) and 8 (lower right) ARB cycles. Rolling direction (RD) and normal direction (ND) are indicated.
From the ARB sheets flat specimens with a gauge length of about 15 mm and a cross section of 3.5×1 mm² were cut by spark erosion. The tensile direction was parallel to RD. The deformation experiments were carried out in an open continuous flow cryostat (He coolant) permitting to keep the temperature constant within less than 0.05 K [13]. The cryostat was mounted in an Instron 4502 load frame. The experiments were performed at constant crosshead displacement rate providing an initial strain rate of 9×10⁻⁴ s⁻¹. The tensile tests were performed at 25K, 77K, 180K and 296K until necking or fracture occurred. To calculate the flow stress \( \sigma \) and total strain \( \varepsilon \) the diminution of the cross section of the specimens was taken into account.

The thermal activation analysis is based on stress relaxation experiments performed during the tensile tests. Details about the data acquisition and evaluation as well as the underlying theory are described elsewhere [11].

![Stress-strain curves for samples deformed at different temperatures: (a) initial state, (b) four and (c) eight ARB cycles.](image)

**Figure 2.** Stress–strain curves for samples deformed at different temperatures: (a) initial state, (b) four and (c) eight ARB cycles.

3. Results and Discussion

The stress–strain curves are shown in Figure 2. The stress drops are the result of the stress relaxation experiments. First of all, the curves show pronounced temperature dependence. Generally, it holds that for a given material the flow stress and the ductility are the higher the lower the deformation temperature is. On the other hand, at constant temperature the flow stress increases and the ductility decreases with increasing number of ARB cycles. All deformation experiments were terminated if pronounced necking took place. However, a special behaviour is observed at 25K. Here the specimens
of the ARB processed material fractured suddenly after the onset of necking. Some other parameters obtained from the true stress–true strain curves are discussed in detail in [14].

Figures 3a-c show the corresponding hardening coefficient $\theta = \left(\frac{\partial \sigma}{\partial \epsilon}\right)_T$ as function of flow stress. In all cases the $\theta$ values were determined by linear extrapolation of the stress–strain curves in front of a stress relaxation experiment. In the initial state $\theta$ almost linearly decreases with increasing stress, at least at high stresses. This clearly indicates so-called stage III strain hardening of face-centred cubic polycrystals [15]. The upper limit $\theta = \sigma$ of the Considère criterion [16] for plastic instability valid for rate-insensitive materials is illustrated by the dashed line in figures 3a-d. Actually, in the initial state necking starts at all temperatures at this point.

![Figures 3a-d](image)

**Figure 3.** Hardening coefficient $\theta$ vs. flow stress $\sigma$ at different temperatures: (a) initial state, (b) four and (c) eight ARB cycles, and (d) $\theta(\sigma)$ at 25 K in different ARB states. The dashed line marks $\theta = \sigma$.

The ARB samples deformed at 77 K and 25 K behave similar if compared with the initial state. Obviously, ARB processing of aluminium AA6016 at elevated temperatures (about 200°C) is not sufficient to exhaust the strain hardening capacity of the material at much lower temperatures. Figure 3d for all three ARB states shows the relationship $\theta(\sigma)$ at 25 K, that is at maximum ductility. It is interesting to note, that the $\theta(\sigma)$ curves after four and eight ARB are shifted to much higher stress levels, where they coincide extremely well. This indicates that most of the athermal Hall-Petch like hardening occurs during the first four ARB cycles, where most of the obstacles for dislocation motion were initially introduced (new grain boundaries, bonding layers, agglomerates of vacancies, etc.). Naturally, Taylor hardening by dislocation storage and grain fragmentation plays an important role at any stage of the ARB process. However, the balance between "pure" Taylor hardening and dynamic
recovery processes may be the reason for the “moderate” microstructural refinement between four and eight ARB cycles where, probably, all structure lengths were further downsized simultaneously until saturation is reached. It seems that saturation will be reached in aluminium AA6016 not far beyond eight ARB cycles.

At 296 K and 180 K the ARB samples show a rather limited ductility, but the region of stable homogenous deformation seems to be somewhat extended beyond $\theta = \sigma$. This can be attributed to an increased strain rate sensitivity of the ARB states, which delays the onset of necking. In this case the Hart criterion [17] for plastic instability $\theta \leq \sigma (1 - m)$ is valid. $m$ is the strain rate sensitivity of the material and at 296 K is about 0.01 in the initial state, but 0.4 after eight ARB cycles (cf. figure 5).

Figures 4a-c show the experimental rate sensitivity $\lambda$ as function of flow stress $\sigma$ of all materials investigated. There is a strictly linear relationship in all cases where strain hardening is sufficiently pronounced. In the ARB states this is the case only at 77 K and 25 K; at higher temperatures there is a lack of strain hardening. The linear relationship clearly indicates so-called Cottrell-Stokes behaviour, i.e. the increase of stress with strain during the experiment is totally caused by dislocation multiplication, movement, and storage.

**Figure 4.** Experimental rate sensitivity $\lambda$ vs. $\sigma$ for samples deformed at different temperatures: (a) initial state, (b) four and (c) eight ARB cycles. (d) shows the offset stresses (here called $\sigma_d$, see text) together with the flow stress after 0.2% strain at different temperatures as a function of the inverse square root of the arithmetic average of grain size $d = (d_{RD} + d_{ND})/2$. 

[Diagrams showing the relationship between $\lambda$, $\sigma$, and the inverse square root of the grain size at different temperatures and cycles.]
In all three materials the straight lines intersect more or less at one point at the stress axis. The resulting offset stress is 24 MPa in the initial state and 270 MPa and even 360 MPa after four and eight ARB cycles, respectively. Obviously, the offset stress is athermal by nature and adds to the Cottrell-Stokes stress caused by dislocation multiplication, movement, and storage mentioned above. The sum of both is the total flow stress $\sigma$ measured in the tensile tests [11].

In the initial state the small athermal offset stress of 24 MPa is probably due to slight precipitation hardening [18, 19]. During the first four ARB cycles the athermal stress component increases rapidly and reaches already 270 MPa. This strong increase of the athermal offset stress during ARB can be attributed mainly to the influence of grain size $d$ on the flow stress $\sigma$ and is, therefore, called $\sigma_d$ in the following.

Even between four and eight cycles, microstructural refinement still proceeds, but the increase of $\sigma_d$ is not as pronounced as in the first four cycles (from 270 MPa up to 360 MPa only). The impression of a moderate strengthening during higher ARB cycle numbers is confirmed by observations of the process of grain refinement performed by EBSD studies [9]. It is remarkable, that in aluminium AA6016 after eight ARB cycles more than 50% of the flow stress measured in tensile tests at 25 K is due to $\sigma_d$. For comparison, in pure ECAP nickel the contribution of $\sigma_d$ to $\sigma$ at 25 K is only 15% [11]. In this respect ARB processing seems to be very efficient for strengthening of aluminium alloys.

It cannot be excluded, however, that the ARB processing also affects the volume fraction, size, and distribution of precipitates and, thus, the athermal stress component. The incorporation of oxides from the (former) surface layers in the material may also be an important athermal strengthening mechanism in ARB aluminium alloys like AA6016 [20]. In this sense $\sigma_d$ is an upper limit for the athermal influence of grain size in ARB processed AA6016.

Figure 5. (a) Strain rate sensitivity $m$ after different numbers of ARB cycles as a function of deformation temperature $T$. The right figure (b) is a blow-up of the lower part of (a).

Figure 4d shows $\sigma_d$ (the offset stress in the initial state is included) together with the flow stress after 0.2% strain at different temperatures as a function of the inverse square root of the arithmetic average of grain size $d = (d_{RD} + d_{ND})/2$. The full line illustrates the expected Hall-Petch like behaviour of the ARB states after four and eight cycles assuming the same amount of oxide and precipitation.
hardening in both states. Probably, the initial state deviates to lower stresses (24 MPa) since here the precipitation hardening is not fully developed if compared with the ARB states. It is very likely that the too small offset stress in the initial state is the result of under-aging due to the missing pre-warming treatment (230° for five minutes prior to ARB, see e.g. [20]) and, of course, due to the lack of ARB induced oxide incorporation and precipitation hardening.

As can be seen from figure 4d the flow stress determined at 0.2% strain is only a poor estimate for the Hall-Petch stress since it always contains a thermal stress component that is the higher the lower the deformation temperature is. Thus, $\sigma_d$ is a lower bound for the pure athermal grain size induced stress and, probably, is a better quantity for the “true” Hall-Petch stress. The slope of the full straight line in figure 4d is $k_{HP} = 0.17$ MPa m$^{1/2}$.

Figure 5 shows the temperature dependence of the strain rate sensitivity $m$, that is, of the slope of the straight lines in figure 4. In the initial state the usual behaviour expected from forest cutting theory is observed (e.g. [11, 21, 22]). $m$ first strongly increases at low temperatures, passes a maximum at intermediate temperatures and then decreases towards high temperatures. This behaviour is typical for dislocation dominated plasticity [22] and may be attributed to local single slip [11, 23].

The ARB states deviate towards much higher strain rate sensitivities already at 25 K. As for pure ECAP nickel [11] it holds that the deviations are the more pronounced the higher the degree of pre-deformation is. The deviations indicate the appearance of additional, unusual deformation mechanisms in ARB aluminium AA6016 already at 25 K where, probably, diffusion plays no role in the deformation process. Therefore, we suppose that the thermally activated interaction of dislocations with grain boundaries is responsible for the increase of $m$ at low temperatures. However, at 296 K $m$ is up to forty times (after eight cycles) higher if compared with the initial state. There is no doubt that this is a hint for the dominance of diffusion controlled (recovery) processes taking place at room temperature [24, 25]. The behaviour of $m$ above room temperature is discussed by other authors, e.g. in Ref. [26] for UFG aluminium of commercial purity and in Ref. [27] for the UFG aluminium alloy AA6061, both processed by equal channel angular pressing.

4. Conclusions
   
i) ARB processing of aluminium AA6016 is a very efficient technique to produce a microstructure of sub-micron scale exhibiting unique unusual deformation behaviour.
   
ii) After ARB processing at about 200°C the aluminium AA6016 is heavily pre-deformed and, therefore, shows a limited ductility at room temperature. Towards lower temperatures the ductility strongly increases due to an increase of strain hardening capacity. At constant temperature the remaining strain hardening capacity decreases with increasing number of ARB cycles.
   
iii) In AA6016 most of the Hall-Petch like hardening during ARB processing occurs within the first four ARB cycles. At higher cycle numbers, the ongoing microstructural refinement by Taylor hardening competes with dynamic recovery processes.
   
iv) The microstructurally saturated state in aluminium AA6016, where strain hardening and dynamic recovery balance each other, seems to be not far beyond eight ARB cycles.
   
v) The athermal stress component $\sigma_d$ obtained from the $\lambda(\sigma)$ relationship represents the “true” temperature independent Hall-Petch stress.
   
vi) ARB processing strongly increases the strain rate sensitivity of aluminium AA6016 at room temperature and moderately at very low temperatures. This clearly indicates the appearance of unconventional deformation processes in ARB AA6016 at all temperatures. However, whereas at low temperatures probably dislocation–grain boundary interaction only slightly affects the plastic behaviour of ARB processed AA6016, diffusion controlled deformation mechanisms probably govern the plasticity of the material at room temperature.

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