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ULTRAFINE-GRAINED MATERIALS

Grain growth in ultrafine grained aluminium processed by hydrostatic extrusion

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Abstract Ultrafine grained materials can be produced by a number of techniques among which one can distinguish hydrostatic extrusion. In aluminium, this method can be used to obtain a structure with the grain size of 300 nm and high fraction of HAGBs (more than 70%). During annealing this structure undergoes significant changes which were evaluated quantitatively. Annealing for 1 h at temperatures up to 200 °C results in normal grain growth whereas at higher temperatures or for longer annealing times a transition from normal to abnormal growth is observed. The activation energy for low temperature regime is 43 kJ/mol whereas for high temperature annealing—128 kJ/mol. The former corresponds to grain boundary diffusion whereas the latter is close to activation energy of self diffusion in aluminium. The change in activation energy well corresponds to the transition in grain growth mechanism from normal to abnormal.

Introduction

Ultrafine grained (UFG) metals obtained via grain refinement induced by severe plastic deformation (SPD) are promising engineering materials of XXI century. Their microstructure results from the transformation of coarse into ultrafine grains via accumulation and re-arrangement of defects (in particular dislocations which contribute to the formation of new grain boundaries). The SPD microstructures exhibit, in general, features partly characteristic for deformed and partly for recrystallized state containing relatively large fraction of high angle grain boundaries (HAGB) as well as low angle grain boundaries (LAGB) [1]. UFG materials possess also unique properties such as very high strength combined with acceptable ductility and improved fatigue, corrosion and wear resistance. This makes them prospective for many industrial applications.

UFG materials may be produced by a number of severe plastic deformation techniques, including: equal channel angular pressing (ECAP), high pressure torsion (HPT) and accumulative roll bonding (ARB). These techniques were developed to allow for unusually high strains necessary for microstructure transformation. Depending on processing technique and its parameters, in the same material one can obtain different grain size/shape distributions, grain boundary structure and different properties [2, 3]. More recently, hydrostatic extrusion was shown to be also an efficient method of grain refinement in metals and alloys [4, 5].

One of the challenges related to application of UFG materials is their high stored energy and high driving force for grain growth. On the other hand, the microstructural features such as a fraction of HAGB and grain size uniformity may significantly influence the grain growth kinetics as presented in Humphreys model [6] and confirmed experimentally in [7]. The aim of the present study is to show that hydrostatically extruded materials exhibit similar behaviour upon annealing as SPD processed counterparts and to give insight into the mechanism of grain growth in UFG materials. To this end, the changes in grain size and grain boundary characteristics during annealing at various temperatures were evaluated quantitatively for technically pure aluminium processed by hydrostatic extrusion.

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Experimental

Technically pure (99.5%) 1050 aluminium was hydrostatically extruded to 3 mm wire in four extrusion passes: 50 → 20, 20 → 10, 10 → 5, 5 → 3 corresponding to a total true strain of 5.4. The samples were water cooled at the exit of the die to minimize the effect of adiabatic heating during the processing. Samples of the deformed material were subsequently annealed at a temperature range from 100 °C to 350 °C. Specimens for TEM observation were cut perpendicularly to the extrusion axis. The foils have been examined in a Jeol JEM-1200 electron microscope operated at 120 kV. The microstructures were evaluated quantitatively using computer aided image analysis. The grain size changes during annealing were described using the equivalent grain diameter d_2 (defined as the diameter of a circle of equal area to the surface area of a given grain) and variation coefficient $CV(d_2)$ defined as a ratio of standard deviation $SD(d_2)$ to the mean value. The misorientation angles were determined for a population of randomly selected grain boundaries. Crystallographic orientations of individual grains were calculated from Kikuchi lines patterns obtained in TEM by convergent beam diffraction. These patterns were subsequently used to calculate misorientations across boundaries. For each annealing temperature, a population of 100 grain boundaries was analysed. Although misorientation measurements in TEM is time consuming limiting the number of grain boundaries analysed, the advantage is that all grain boundaries present in the observation field are revealed and taken into account. In EBSD technique, which allows to analyse larger population of grain boundaries, some information can be lost (especially those related to LAGBs) due to limited resolution of this technique.

Results

Deformation microstructure

Figure 1 shows a typical TEM micrograph and grain size distribution of the as-deformed material. The microstructure exhibits an ultrafine grain structure with the average grain equivalent diameter of 300 nm. The grains are almost free of dislocations and fairly equiaxial. Grain orientation analyses have shown that the grains have either <111> or <100> direction parallel to the extrusion direction, as illustrated by an insert in the micrograph. The grain boundary misorientation distribution is given in Fig. 2 which clearly demonstrates two peaks in the histogram. The fraction of high angle grain boundaries (HAGB), defined as having misorientation angle greater than 15, was calculated to be 72%. This and the bimodal distribution of misorientations agree with other studies on severely deformed Al alloys [7–9]. However, it should be noted that in the case of hydrostatically extruded material, similar microstructural parameters were achieved for a relatively low value of true strain (~5). The advantages of hydrostatic extrusion also include: fast processing (the strain rates are often higher than 10^2 s^{-1}) and the opportunity of obtaining products of large dimensions, in the form of rods or wires with various cross sections.

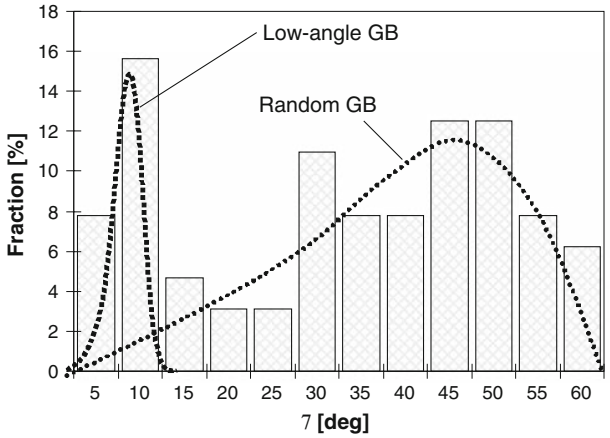

Fig. 2 Distribution of grain boundary misorientation in the as-deformed material

Microstructure changes during annealing

The microstructures of 1050 aluminium after annealing for 1 h are shown in Fig. 3. The average values and variation

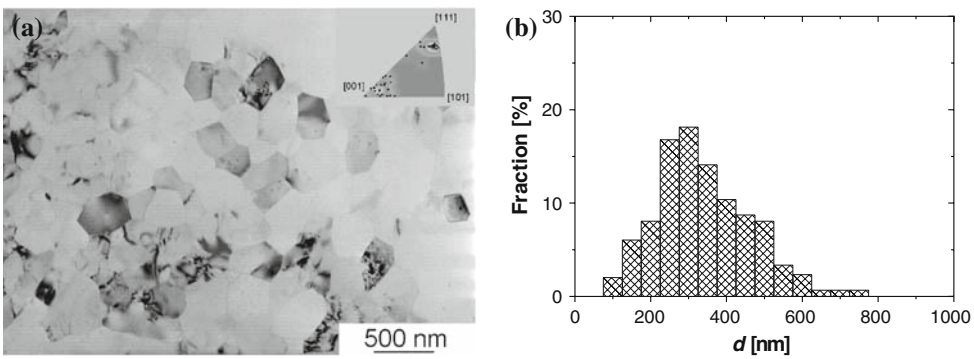

Fig. 1 TEM micrograph and grain size distribution of the as-deformed material

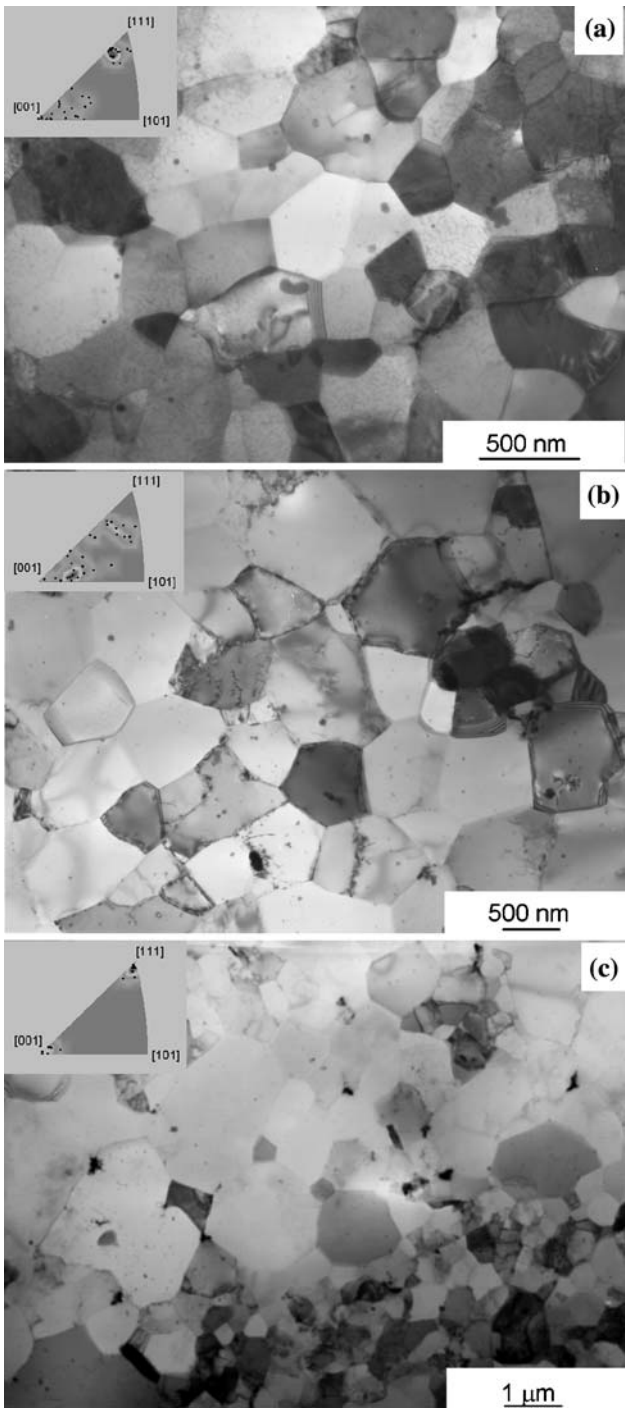

Fig. 3 Microstructures of 1050 aluminium after annealing for 1 h at: 100 °C (**a**), 200 °C (**b**) and 300 °C (**c**)

coefficients of grain size, d_2, and grain boundary misorientation, θ, as a function of annealing temperature are presented in Table 1. It can be seen that annealing at 300 °C results in an increase in grain size by an order of magnitude (from 300 nm to 4 μm). However, at lower temperatures (up to 200 °C) a low rate of grain coarsening is observed.

It can be also noted that up to 200 °C uniform grain growth occurs. Such a growth does not alter the grain size distributions which are characterized by fairly unchanged variation coefficient $CV(d_2)$ of grain population. Furthermore, there is no significant change in the fraction of HAGBs which remains constant at ~70%. Also, grain orientations remain similar to as-deformed state (as seen in inserts in the micrographs).

After annealing at 300 °C, two populations of grains, which significantly differ in diameter (Fig. 3d), can be distinguished. The grain size distribution becomes bimodal and variation coefficient has much higher value. It is also worth noting that larger grains have <100> whereas smaller ones <111> direction parallel to extrusion direction. Grain boundaries of larger grains are mostly of high angle type whereas those of small grains have rather small misorientation angle. This temperature can be hence considered as an onset of transition from normal to abnormal grain growth for 1 h annealing. One can also consider the large grains in the specimen as "recrystallized". However, the microstructure evolution occurring during annealing does not fit into the classical recrystallization process with distinct stages of nucleation and growth. First, a significant fraction of HAGBs (more that 70%) already exists in as-deformed state. Second, there is no preferential site of large grains—they appear randomly in the structure. Therefore, it seems better to consider annealing behaviour of hydrostatically extruded aluminium as grain coarsening or grain growth.

To analyse grain growth transition in hydrostatically extruded aluminium, annealing at 200 °C and 300 °C was carried out for up to 4 h. The results of grain size measurements are presented in Fig. 4a. It should be noted that although 1 h annealing at 200 °C does not result in grain growth, a longer time of annealing leads to a significant increase of grains which average diameter approaches 2 μm for 4 h. It should be also noted that the grain growth proceeds in a discontinuous way (the value of $CV(d_2)$ significantly increases as a function of annealing time (see Fig. 4b). The kinetics of grain growth is much faster during annealing at 300 °C—the mean grain diameter achieves the value of 100 μm for 3 h annealing. The analysis shows that CV value grows significantly for 1 h annealing (Fig. 4b) but for higher annealing times it returns to values characteristic of normal grain growth. It means that when all small grains disappear (i.e. the "recrystallization" is completed) the grain growth becomes normal again.

Discussion

Ultrafine grained microstructures formed during hydrostatic extrusion in pure aluminium have limited stability and

Table 1 The average values and variation coefficients of grain size, d_2, and grain boundary misorientation, θ, as a function of annealing temperature

Temperature (°C)	E (d_2) (μm)	CV (d_2)	E (θ)	CV (θ)	Fraction of HAGBs
20	0.30	0.38	31.4	0.70	72%
100	0.32	0.40	28.5	0.69	68%
150	0.38	0.43	–	–	–
200	0.45	0.50	32.8	0.57	77%
300	4.7	0.62	14.0 (small)	1.20 (small)	18% (small)
			34.8	0.54 (large)	90% (large)
350	17.4	0.53	–	–	–

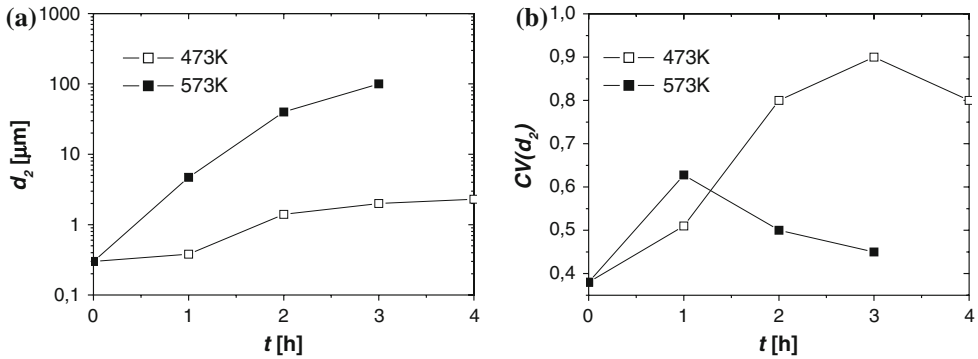

Fig. 4 Mean grain diameter (**a**) and variation coefficient (**b**) as a function of annealing time

undergo significant changes during annealing. The main reason of this instability is high stored energy. This energy is stored mainly in the form of grain boundaries. Taking into account only the difference in relative grain boundary area, ($S_v \approx 2/l$, where l is the mean intercept length), between microstructures with mean grain diameter of 300 nm and 30 μm, the energy stored at grain boundaries is two orders of magnitude higher in the case of ultrafine grained material, providing high driving force for grain growth.

Despite the high driving force for grain growth, there is remarkable grain size stability in UFG aluminium at temperatures up to 200 °C which corresponds to 0.5 T_m. At this temperature range, the microstructure coarsening is relatively slow with no significant changes in grain size diversity, material texture and misorientation distribution. Such a homogenous microstructure evolution is often termed continuous (normal) grain coarsening [7]. At higher temperatures ($T > 200$ °C) or for extended annealing times ($t > 2$ h), the transition from normal to abnormal growth is observed. It should be noted that in the case of conventionally deformed materials such a transition occurs at significantly lower temperature [10]. HE processed aluminium exhibits improved thermal stability which is similar to that found in UFG aluminium processed by SPD methods [11–13]. The thermal stability of aluminium may be additionally improved by the presence of second phase dispersoids as evidenced for Al-Sc alloy [8].

Improved thermal stability of SPD materials (including aluminium processed by hydrostatic extrusion) can be attributed to a large content of HAGBs, which is one of the parameters influencing grain growth. The analytical mean field theory developed by Humphreys [6] predicts that single phase microstructures with the large fraction of LAGBs are highly unstable and undergo discontinuous grain coarsening whereas continuous coarsening is predicted for microstructures containing a large fraction of HAGBs (usually higher than 0.6).

To have a better insight into the role of HAGB, grain growth process has been modelled by Monte Carlo methods described elsewhere [14]. For efficient investigation of 3D structures, image-based algorithms of VORONOI tessellations have been applied [15]. The grain structures modelled in the present study differed in the distribution of the grain boundary misorientations, as evidenced in Fig. 5. Calculated changes in the average value of the equivalent diameter and the coefficient of variation are presented in Fig. 6a and b, respectively. The results show that the grain growth rate is significantly higher for the structure with non-homogeneous distribution of the grain boundary misorientation. The two initial structures analysed here trigger grain growth which proceeds along different routes also in coefficient of variation-time coordinates, as shown in Fig. 5b. In particular, the structure with non-homogeneous distribution of the grain boundary misorientations has a tendency to abnormal grain growth whereas those highly homogeneous are prone to normal grain growth.

Another important parameter, which can provide insight into the nature of the grain growth, is its activation energy.

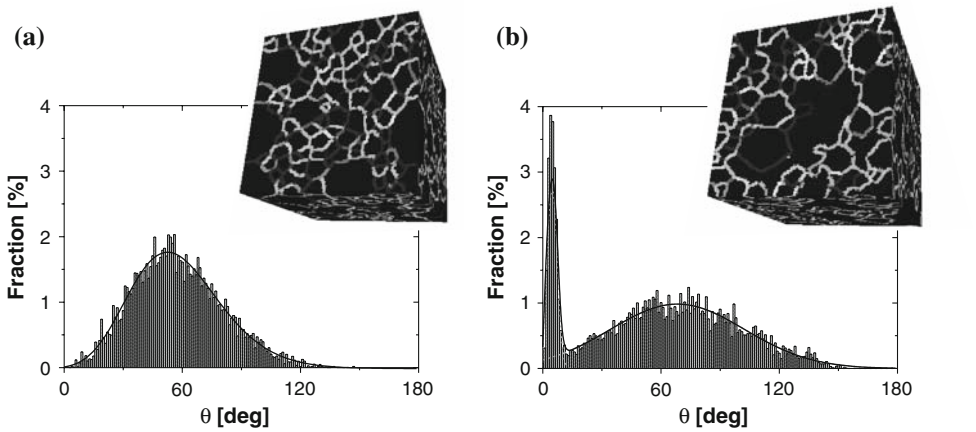

Fig. 5 Modelled structures with highly homogeneous and non-homogeneous distribution of the grain boundary misorientations

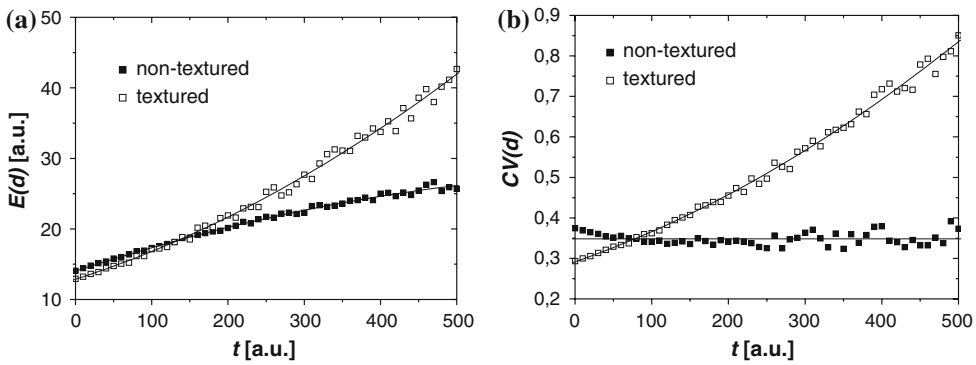

Fig. 6 Calculated changes in the average value of equivalent diameter (**a**) and variation coefficient (**b**) for structures with highly homogeneous and non-homogeneous distribution of the grain boundary misorientations

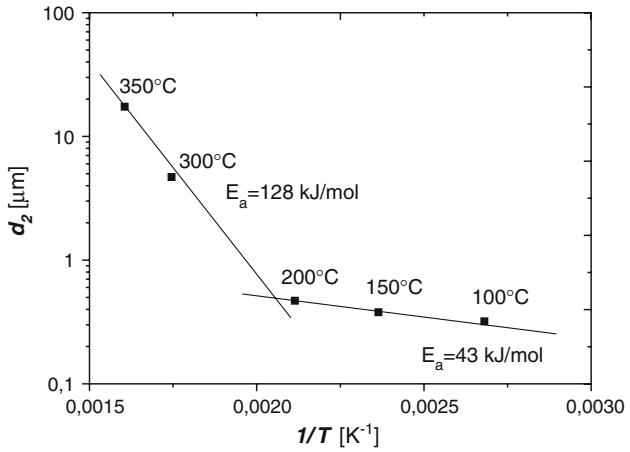

Fig. 7 The plot of grain size against the reciprocal of absolute temperature

To determine the activation energy for grain growth in pure aluminium processed by hydrostatic extrusion, the results of grain size measurements were plotted against the reciprocal of absolute temperature, $1/T$ (Fig. 7). It can be seen that the plot can be fitted with two straight lines, one below 0.5 T_m and another above it. At low temperature regime, the activation energy for grain growth is 43 kJ/mol whereas at higher temperatures—128 kJ/mol. Such two regimes of grain growth were also reported in nanocrystalline nickel obtained by electrodeposition [16], UFG Al–3%Mg alloy and 1050 aluminium both processed by ECAP [12, 17]. The reason of low activation energy for the low temperature grain growth, although observed previously, is still unclear. Some authors suggest [18] that the low value of activation energy results from the presence of strong texture and highly unstable grain boundaries, which need only a small amount of energy for re-ordering. Others [12] indicate that during low temperature annealing heavily deformed materials may undergo simultaneous recovery of dislocation structure, re-ordering of non-equilibrium grain boundaries and coarsening of grain structure. These overlapping processes influence the measured value of the activation energy which does not reflect the characteristic energy of any of the contributing mechanisms.

On the other hand, the change in activation energy of grain growth at a certain temperature is similar to the change in activation energy of diffusion when one considers lattice and grain boundary diffusion. At low temperatures, grain boundary diffusion (for which activation energy is small) is faster whereas at higher temperatures lattice diffusion is dominant. In this context, one should underline that the activation energy of grain growth in hydrostatically extruded aluminium at high

temperature regime is close to the activation energy of volume diffusion in aluminium whereas those at low temperature regime are only slightly lower than activation energy of grain boundary diffusion in aluminium. It suggests that grain growth at high temperatures is controlled by lattice diffusion whereas at low temperatures by grain boundary diffusion. It is also worth noting that the change in activation energy well corresponds to the transition in grain growth mechanism from normal to abnormal.

Conclusions

The changes induced by annealing in hydrostatically extruded aluminium were studied by TEM. It was shown that:

1. Hydrostatic extrusion generates an ultrafine grained structure with a high fraction of HAGBs (more than 70%). However, misorientation distribution is bimodal with a peak also for 10 misorientation angle.
2. Annealing for 1 h at temperatures up to 200 °C results in normal grain growth whereas at higher temperatures or for longer annealing times a transition from normal to abnormal grain growth is observed.
3. The activation energy for low temperature regime is 43 kJ/mol whereas for high temperature annealing—128 kJ/mol. The former corresponds to grain boundary diffusion whereas the latter is close to the activation energy of self diffusion in aluminium. The change in activation energy well corresponds to the transition in grain growth mechanism from normal to abnormal.

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