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MOBILITY OF <111> TILT GRAIN BOUNDARIES IN THE VICINITY OF THE SPECIAL MISORIENTATION Σ=7 IN BICRYSTALS OF PURE ALUMINIUM

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Introduction

There is experimental evidence that grain boundary mobility depends on misorientation and that this dependency controls the texture and microstructure evolution during recrystallization and grain growth. Studies of the mobility of grain boundaries in aluminium bicrystals have shown that tilt grain boundaries with <111> rotation axis have the highest mobility [1-2]. It was also found that grain boundary mobility depends on the angle of misorientation in a non-monotonic fashion and that the mobility maxima are obtained for low Σ boundaries.

On the other hand, it is known from growth selection experiments of Lücke et al. [3-5] that Θ<111> boundaries with $\Theta \geq 40°$, i.e. distinctly different from the Σ7 (38.2°<111>) coincidence misorientation move fastest.

The aim of the current study was to investigate the mobility of <111> tilt grain boundaries in aluminium in the vicinity of the Σ7 special misorientation in order to settle the conflict mentioned above.

Experimental

Grain boundary motion under a constant driving force **p** was investigated in aluminium bicrystals. The driving force was provided by the surface tension of a curved grain boundary and reads (per unit volume) $\mathbf{p} = \sigma/\mathbf{a}$, where σ is the grain boundary surface tension, and **a** the width of the grain to be consumed (Fig. 1) [6]. Pure <111> tilt boundaries with misorientation angles in the vicinity of the special misorientation Σ7 ($\varphi = 38.2°$) were studied (Table 1). The bicrystals were grown by directional crystallization, using high purity (99.999at%) aluminium. The orientation of the crystallographic axis of the monocrystal seeds and the fabricated bicrystals was measured with an accuracy of ±1°, using the characteristic reflection pattern of a laser beam from the specially prepared crystal surface. For this, the samples were etched in a solution of 18 ml HCl, 9 ml HNO_3 and 2 ml HF. The misorientation of the grains in the bicrystals was determined with an accuracy of ±0.4° by measuring with high accuracy the misorientation of etch pits in both grains of the bicrystal under an optical microscope. In-situ measurement of grain boundary motion was conducted with an X-ray continuous tracking goniometer. The method employed X-ray diffraction to determine the grain boundary position and, therefore, did not interfere with the grain boundary migration process itself. Details of measuring procedure, device and accuracy are given elsewhere [7]. Owing to the

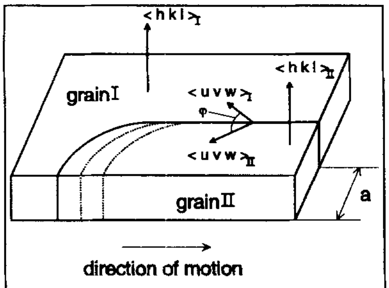

Fig.1: Used bicrystal geometry for grain boundary motion measurements under a constant driving force.

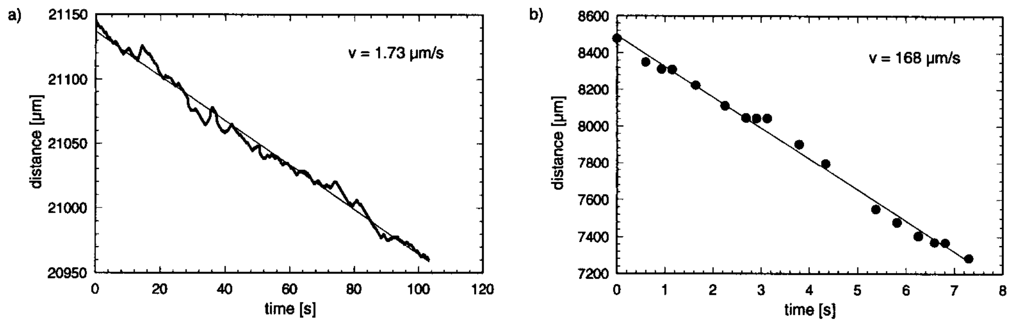

Fig. 2: Distance-time diagrams of grain boundary migration at a) low and b) high velocity. The diagrams comprise a) 429 and b) 17 measuring points.

constant driving force, the boundary was expected to displace at a constant rate, and this was actually observed (Fig. 2). From the displacement versus time diagram the velocity was determined by linear regression with an accuracy in the order of 1%. During isothermal experiments the temperature remained constant within ±0.3°C. To avoid thermal grooving, the sample was exposed to a nitrogen gas atmosphere during measurement.

Results

The mobility of a grain boundary is given by the ratio of velocity v and driving force p

$$m = \frac{v}{p} = \frac{v}{\sigma / a} \tag{1}$$

For convenience we use the reduced mobility

$$A \equiv va = m\sigma = A_0 \exp\left(-\frac{E}{kT}\right) \tag{2}$$

where **E** is the activation energy of migration and A_0 a pre-exponential factor. The reduced mobility depends on temperature, as evident from Eq. (2), but also on misorientation (Fig. 3). The activation energy was found to be constant for a given misorientation over the entire investigated temperature range as obvious from straight line Arrhenius plots (Fig. 4). Activation energy and pre-exponential factor, however, do depend on the angle of misorientation in a non-monotonic fashion (Figs. 5 and 6).

Discussion

The orientation dependence of grain boundary mobility in the small angular interval of misorientation investigated in the current study revealed that for a given temperature, there is an angle of misorientation, for which the grain boundary mobility attains a maximum. The respective misorientation, however, depends on temperature. At lower temperatures, strictly speaking below 430°C, the grain boundary mobility is the highest for an angle of misorientation of 38.2°. Above this temperature grain boundaries with misorientation of 40.5° have the absolutely highest mobility. This is an interesting result and very surprising according to the current understanding of grain boundary mobility. Generally it is believed that low Σ boundaries exhibit a high growth rate owing to a proposedly lower propensity for

Misorientation Angle	Activation Energy	Pre-exponential Factor
	E [eV]	$\log(A_0\ [\mu m^2/s])$
35.2° ± 0.4°	2.05 ± 0.08	18.8 ± 1.1
37.1° ± 0.4°	1.69 ± 0.07	16.3 ± 1.0
37.4° ± 0.4°	1.32 ± 0.05	13.5 ± 0.8
37.7° ± 0.4°	1.28 ± 0.05	13.4 ± 0.8
37.9° ± 0.4°	1.23 ± 0.05	13.3 ± 0.8
38.2° ± 0.4°	1.29 ± 0.05	13.6 ± 0.8
38.7° ± 0.4°	1.56 ± 0.06	15.2 ± 0.9
39.3° ± 0.4°	1.75 ± 0.07	16.4 ± 1.0
39.9° ± 0.4°	1.80 ± 0.07	16.8 ± 1.0
40.5° ± 0.4°	2.19 ± 0.09	20.2 ± 1.2
41.7° ± 0.4°	1.73 ± 0.07	16.6 ± 1.0
42.5° ± 0.4°	1.56 ± 0.06	15.4 ± 0.9
43.0° ± 0.4°	1.48 ± 0.06	14.8 ± 0.9

Table 1: Misorientation and mobility parameters for the investigated <111> tilt grain boundaries in bicrystals of pure Al (99.999 at.%).

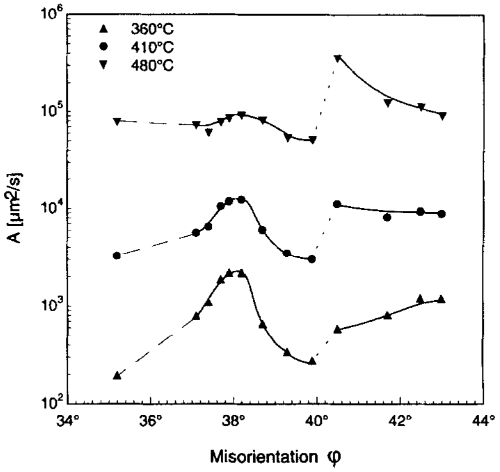

Fig. 3: Misorientation dependence of the grain boundary mobility at different temperatures.

segregation. Such an interpretation was strongly supported by the early experiments on bicrystals by Aust and Rutter [8-10] and the results of Shvindlerman et al. [1-2] on aluminium bicrystals with much larger spacing of misorientation angle than in the current investigation. At lower temperatures the mobility maximum at the exact Σ7 misorientation is very pronounced, and there is no need to modify our current understanding of misorientation dependence of mobility. With increasing temperature, however, the mobility maximum at the exact Σ7 misorientation tends to flatten, while the absolute mobility of the 40.5° misoriented grain boundary increases drastically. Actually, the mobility dependency on misorientation changes almost discontinuously close to 40.5° at high temperatures. It is

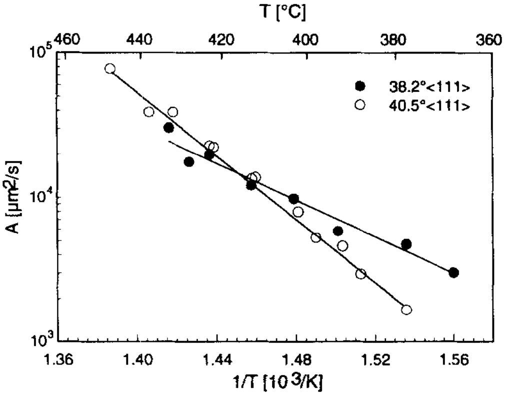

Fig. 4: The temperature dependence of the mobility for 38.2° and 40.5°<111> tilt grain boundaries.

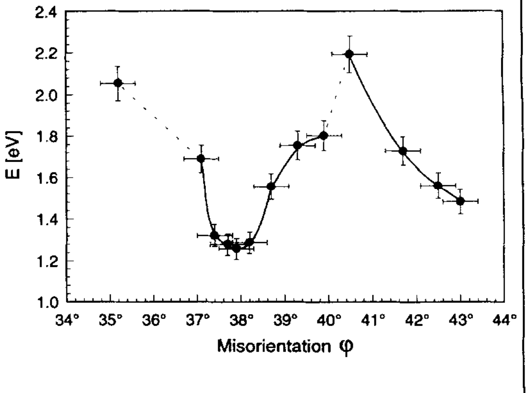

Fig. 5: The misorientation dependence of migration activation energy for <111> tilt grain boundaries.

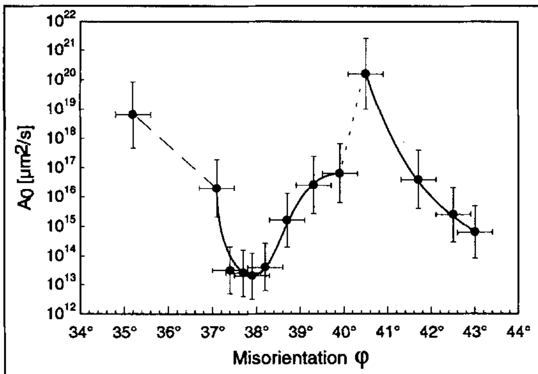

Fig. 6: The misorientation dependence of the pre-exponential mobility factor for <111> tilt grain boundaries.

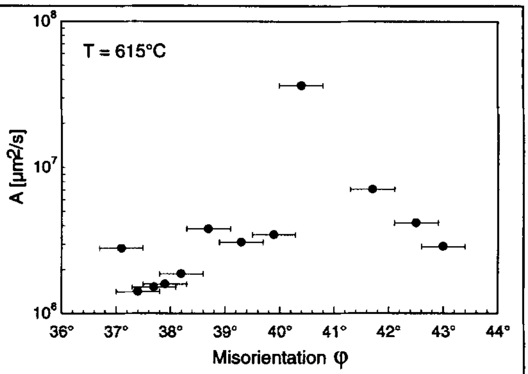

Fig. 7: The mobility of investigated <111> tilt grain boundaries in Al at 615°C (obtained by extrapolation of the measured temperature dependence to 615°C)

not clear from the current investigation what may be the cause of this temperature dependence of mobility on orientation, but we surmise that this behavior is related to a transformation of the grain boundary structure from a longe range periodic atomic arrangement to a random boundary structure, which is referred to in literature also as transition from a special to a random grain boundary [11]. The angular difference of 2.3° between the exact Σ7 misorientation and the maximum growth rate orientation at high temperatures corresponds to about 15°/Σ [12]. If this orientation distance indicates the maximum angular range where a periodic secondary grain boundary dislocation arrangement preserves a long range periodic coincidence structure in the boundary, then obviously the criterion 15°/Σ is a more realistic approximation for this range than the commonly used Brandon criterion $15°/\sqrt{\Sigma}$ [13].

The result, however, is of particular importance in the field of crystallographic textures. For the first time it reconciles measurements of grain boundary mobility in bicrystals with growth selection experiments. As already stated by Lücke [5], the growth selection experiments on aluminium clearly yield a maximum growth rate at an angle of rotation of ≥40° and definitely not at the Σ7 misorientation. Growth selection experiments are by nature conducted at very high temperatures [14-16]. The numerous growth selection experiments conduced by Lücke, Masing and coworkers on aluminium [3,5] were carried out at 615°C. The temperature interval of the current investigation did not extend to these temperatures, but if we extrapolate the results from the lower temperature regime - which is obviously allowed, since there is a unique Arrhenius type dependency for the entire investigated temperature regime and for each specific misorientation - we find the misorientation dependence of grain boundary mobility at 615°C as given in Fig. 7. Evidently, the mobility at a misorientation of 40.5° is at least an order of magnitude higher than the mobility at any other misorientation in the interval between 36° and 44°. There is no further need to explain why growth selection experiments at that temperature yield such a distinct and pronounced preference of 40°<111> grain boundaries as observed by Lücke, Ibe and coworkers [14-16]. At high temperatures the grain boundary mobility in this angular interval is obviously not dominated by the segregation behavior of low Σ boundaries, but by other structural and compositional effects of the boundaries yet to be determined.

The results obtained by growth selection experiments have been successfully used to relate recrystallization textures to deformation textures by 40°<111> rotations. According to the results of the current investigation, there should be slightly different texture developments in the temperature regime above and below 430°C, since the maximum growth rate shifts from a misorientation of 40.5° to 38.2°. Because of the large scatter of experimental texture data, sophisticated texture analysis will be needed to show the existence of such a difference and, therefore, this remains to be demonstrated.

A more detailed analysis of the activation parameters reveals that the reason for the high growth rate of the 40.5° misoriented boundary is a high pre-exponential mobility factor despite a high activation energy. As a matter of fact, there is an interdependence of activation energy and pre-exponential factor, also referred to as "compensation effect". A high energy of activation means also a high pre-exponential factor, such that the activation energy changes linearly with log A_0 (Fig. 8).

Such dependency can be expressed as

$$E = kT_c \ln A_0 + B, \qquad (3a)$$

where **B** is a constant.

For $T = T_c$

$$A = A_0 \exp(-E/kT_c) = \exp(-B/kT_c), \qquad (3b)$$

which defines the critical temperature T_c, the compensation temperature. In the current case $T_c = 430°C$. Above this temperature the process - or in this case the respective migrating boundary - with a higher activation energy will exhibit the higher rate, as evident from Fig. 4 in conjunction with Figs. 5 and 6. The compensation effect is very generally observed in thermally activated grain boundary processes, like grain boundary diffusion or, in this case, grain boundary migration. Evidently, it represents a more fundamental principle of thermally activated grain boundary phenomena as discussed elsewhere [17].

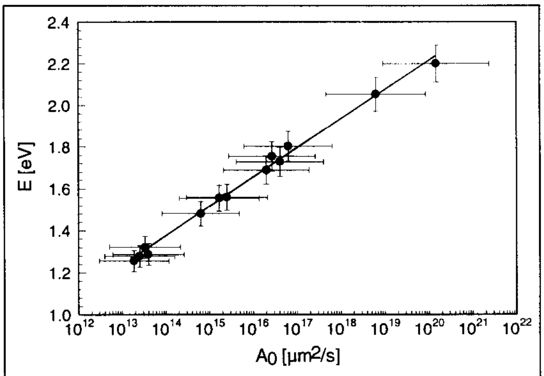

Fig. 8: The dependence of migration activation energy on pre-exponential mobility factor for the investigated <111> tilt grain boundaries.

Conclusions

The temperature dependence of the mobility of <111> tilt boundaries in aluminium with angle of misorientation between 35° and 43° was investigated in aluminium bicrystals in the temperature regime between 370°C and 500°C. The following results were obtained:

1. The grain boundary mobility depends on the angle of misorientation in a non-monotonic fashion.
2. The orientation dependence of grain boundary mobility is subject to change with increasing temperature such that the fastest moving boundary at lower temperatures is the exact Σ7 boundary, while at very high temperatures the boundary with 40.5° misorientation moves at the highest rate. The cross-over temperature was found to equal 430°C.
3. This experimental finding is related to and can be expressed by the compensation effect, which means that the activation energy increases linearly with the logarithm of the pre-exponential factor. The compensation temperature T_c (cross-over temperature) is proportional to the slope of the corresponding straight line.
4. The exact Σ7 boundary moves with the lowest activation energy, but has also the smallest pre-exponential factor. In contrast, the activation energy for the 40.5° misoriented boundary is the highest, but also associated with the highest pre-exponential factor. Correspondingly, this boundary is the fastest at temperatures above the compensation temperature.
5. The observed misorientation dependency of grain boundary mobility reconciles the contradictory results obtained in bicrystal (and recrystallization) experiments and growth selection experiments. Due to the high temperature used in growth selection experiments, the 40.5° boundary will dominate growth competition and, therefore, the result of growth selection.

6. The sharp rise of activation energy and pre-exponential factor on approaching the 40.5° boundary is interpreted as phase transition from a special to a random boundary structure. If this interpretation holds, the relation $15°/\Sigma$ would yield a more accurate measure for the allowed misorientation range of a long range structured low Σ boundary than the conventionally used Brandon criterion $15°/\sqrt{\Sigma}$

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