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TRIPLE JUNCTION MOTION IN ALUMINUM TRICRYSTALS

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Abstract—The results of an investigation of the steady-state motion of grain boundary systems with triple junctions in high-purity aluminum are presented. In particular, the migration of systems with <111> and <110> tilt boundaries was studied. The experimental results demonstrate that the motion of grain boundary systems with triple junctions in aluminum can be controlled by slowly moving triple junctions. The influence of triple junctions depends on temperature, and it is particularly strong at low temperatures. In the high-temperature regime the motion of a connected grain boundary system is less affected by the triple junction, and, therefore, effectively controlled by the grain boundary mobility. The experiments reveal a drastic difference between activation enthalpy of grain boundary and triple junction motion. Therefore, there is a temperature below which triple junctions govern the motion of the connected boundary system. This temperature was found to depend on the particular grain boundary and triple junction geometry. *© 2001 Acta Materialia Inc. Published by Elsevier Science Ltd. All rights reserved.*

Keywords: Triple junction; Mobility; Grain boundaries

1. INTRODUCTION

The role of triple junctions in grain boundary motion has been discussed for many years in interface science. In the 1950s the commonly accepted viewpoint was that of Mullins, who assumed that triple junctions do not affect grain boundary motion and that their role in grain growth is reduced to preserve the thermodynamically prescribed equilibrium angles at the lines where boundaries meet. One of the important consequences of this theoretical approach is the well-known Von Neumann–Mullins relation, which predicts for 2D polycrystals that grains with less than six neighbors (topological class $n<6$) will shrink while those with $n>6$ will grow. Accordingly, grain boundaries constitute the dominating elements of the microstructure with respect to grain growth phenomena, and the mobility of grain boundaries is the intrinsic material parameter to control the change in the mean grain size during grain growth.

The selection of grain boundaries as the controlling element of cellular microstructures, however, was arbitrary and dictated by the fact that the properties of grain boundary junctions had not been determined experimentally. Hence, there were neither independent measurements of grain boundary energies at the point where the boundaries meet, nor were the mobilities of grain boundaries (m_b) and triple junctions (m_{tj}) measured and, therefore, compared. However, the number of triple junctions in polycrystals is comparable in order of magnitude with the number of boundaries. Therefore, triple junctions could have been chosen equally as structural elements to determine grain growth. The mobility of triple junctions in this case could have been derived from a change in the mean displacements of junctions, similar to how grain boundary mobility is extracted from grain growth experiments.

To determine the effect of triple junctions on the kinetics of grain growth quantitatively, the mobility of a triple junction has to be measured. However, a steady-state motion of a grain boundary system with a triple junction, where the curved boundaries maintain their shape during motion so that the entire system moves with the same and constant velocity V, is only possible in a very small set of geometrical boundary configurations. One of them is shown in Fig. 1. This model grain boundary system can move steadily. For 2D systems, assuming that all grain boundaries possess equal properties, irrespective of grain misorientation and crystallographic orientation of the bound-

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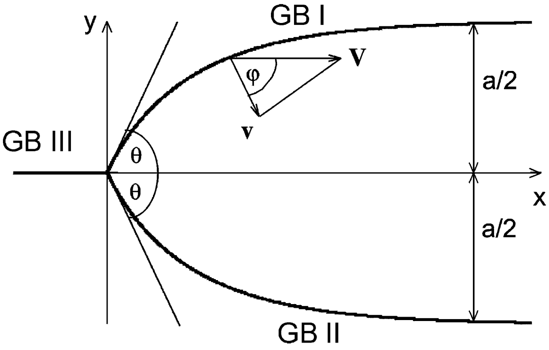

Fig. 1. Geometry of the grain boundary system with triple junction during steady-state motion.

aries (so-called uniform grain boundary model), the shape and velocity of moving grain boundaries with a triple junction can be calculated analytically. The complete analysis of the motion of such grain boundary system is given in [1–3].

The most important parameter of the motion of a boundary system with a triple junction in Fig. 1 is the angle θ at the point where the three boundaries meet. The magnitude of this angle θ defines the shape of the steadily moving grain boundary system [2]. Because of symmetry we need to determined only the shape of GB I in Fig. 1

$$y(x) = \xi \arccos\left(e^{-\frac{x}{\xi} + C_1}\right) + C_2, \tag{1}$$

$$\xi = \frac{a}{2\theta}, \; C_1 = \frac{1}{2}\ln(\sin\theta)^2, \; C_2 = \xi\left(\frac{\pi}{2} - \theta\right)$$

as well as the steady-state velocity V of the entire system (Fig. 1):

$$V = \frac{2\theta m_b \sigma}{a}, \tag{2}$$

where σ is the grain boundary surface tension.

The role of the angle θ becomes clear from the relation between the steady-state value of θ and the dimensionless criterion $\Lambda = m_{tj}a/m_b$ which describes the influence of the triple junction on the motion of the entire boundary system:

$$\Lambda = \frac{m_{tj}a}{m_b} = \frac{2\theta}{2\cos\theta - 1} \tag{3}$$

When Λ is small, i.e. $\Lambda \sim 1$, the angle θ tends to zero and the steady-state velocity is controlled by the mobility of the junction:

$$V = m_{tj}\sigma \tag{4}$$

For $\Lambda \gg 1$ the junction easily adjusts to the motion of the boundary system, and the angle θ tends to its equilibrium value $\theta = \pi/3$.

The motion of a grain boundary system with a triple junction with a geometry as shown in Fig. 1 was measured in zinc tricrystals [4]. From a considerable number of experiments on samples with different grain boundary geometries it was found that triple junctions can effectively retard grain boundary motion. This result is of principal importance for our fundamental understanding of microstructure evolution. In particular, as shown in [4, 5], in the case of a finite mobility of triple junctions there is no unique dividing line between vanishing and growing grains with respect to their topological class any longer, like $n = 6$ in the Von Neumann–Mullins approach.

The current study for the first time addresses the effect of triple junctions on the grain boundary motion in aluminum (Al) by measurement of the motion of specific grain boundary systems with triple junctions in tricrystals. Since the mobility of grain boundaries in Al has been well investigated in bicrystal experiments, such measurements permit a direct comparison of grain boundary and triple junction properties.

2. EXPERIMENTAL

The experiments were carried out on tricrystals of high purity (99.999%) aluminum with a grain boundary geometry as shown in Fig. 1. The crystallographic characteristics of the tricrystals studied are given in Table 1. The orientations of the three contiguous grains of each sample were determined by the Laue technique and electron backscatter diffraction (EBSD).

The tricrystals were grown by directional crystallization. Prior to the measurements the samples were electrolytically polished to improve the surface quality and to remove any grain boundary grooves. For measuring the migration rate of the grain boundary system an X-ray diffraction technique was used. This method makes use of the strong gradient of the diffracted X-ray intensity at a grain boundary, if one crystal is in the Bragg condition while the other is not. A feedback mechanism moves the specimen holder to maintain a constant intermediate intensity. This is the basis of the XICTD system as discussed in detail in [3, 5]. Figure 2 shows the time–displacement diagrams for the motion of one of the investigated grain boundary systems at different temperatures. For each temperature the velocity V, the angle θ, and the width of the shrinking grain a were determined. For this the sample was rapidly cooled down after measurements of the migration rate at each temperature. The position of the grain boundary system and the angle θ were recorded from the grain boundary grooves. Before subsequent heating to measure the migration rate, the specimen was polished to remove the grooves.

Table 1. Misorientation of the tricrystals investigated

Sample	GB I	GB II	GB III
SI	21° <111>	18° <111>	3° <111>
SII	27° <110>	22° <110>	5° <110>
SIII	44° <110>	29° <110>	15° <110>

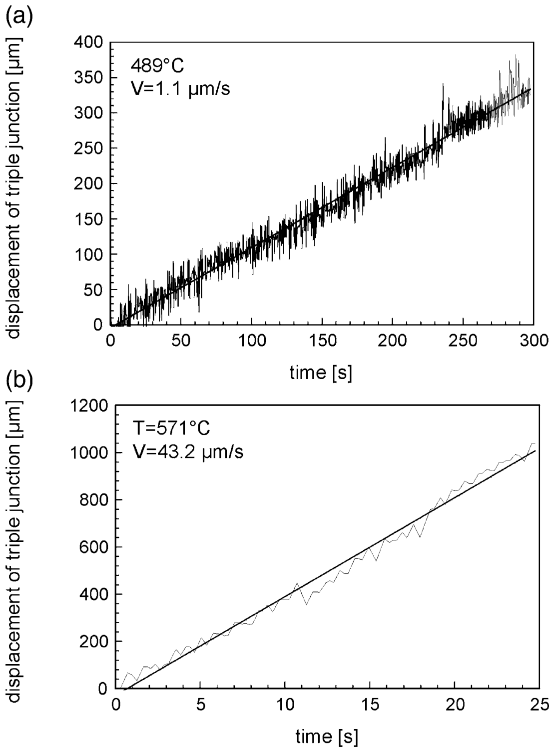

Fig. 2. Triple junction displacement vs. time for different temperatures for sample SII.

3. RESULTS

The motion of three grain boundaries systems with triple junctions (Table 1) was investigated in the temperature range 400 to 590°C. The triple junctions investigated consisted of two high-angle tilt grain boundaries (GB I and GB II) and a low-angle tilt boundary (GB III). Owing to the different properties of low-angle and high-angle grain boundaries and under the assumption that the properties of high-angle grain boundaries vary only slightly with changing misorientation, the grain boundary systems investigated can be regarded as symmetrical junctions.

As can be seen from Fig. 1, equations (1)–(4) can be applied not only to the uniform grain boundary model but also to the symmetrical configuration with two identical curved boundaries and a different straight boundary. In fact, this approach represents the general theoretical background of the measurements of the velocity of motion of a grain boundary system with a triple junction.

The respective surface tensions and mobilities of the boundaries are:

$$\sigma_1 = \sigma_2 \equiv \sigma \neq \sigma_3,\ m_{b1} = m_{b2} = m_b \neq m_{b3} \qquad (5)$$

For the situation given in Fig. 1 the velocity of the triple junction V_{tj} can be expressed as [2, 3]

$$V_{tj} = m_{tj}(2\sigma\cos\theta - \sigma_3) \qquad (6)$$

In the case of steady-state motion of the entire boundary system the velocity of the triple junction equals the velocity of the grain boundaries (see equation (2)). Therefore, the steady-state value of the angle θ is determined by equations (2) and (6):

$$\frac{2\theta}{2\cos\theta - \dfrac{\sigma_3}{\sigma}} = \frac{m_{tj}a}{m_b} = \Lambda \qquad (7)$$

The criterion Λ, as mentioned before, reflects the drag effect of the triple junction on the motion of the system. For a low mobility of the triple junction ($\Lambda \to 0$) the motion of the system is controlled by the mobility of the triple junction. For the opposite limiting situation ($\Lambda \to \infty$) the motion of the system is governed by the grain boundary mobility. The velocity of motion of the boundary system is then given by

$$V = \frac{2\theta_{eq} m_b \sigma}{a} \qquad (8)$$

where the equilibrium triple junction angle θ_{eq} is equal to

$$\theta_{eq} = \arccos\left(\frac{\sigma_3}{2\sigma}\right) \qquad (9)$$

The state of motion of the entire grain boundary system can be determined experimentally for a given ratio σ_3/σ by measuring the contact angle θ.

For all three samples the velocities V (Fig. 2) were found to remain constant during an experiment at a given temperature over the entire temperature range investigated. The angle θ was seen to increase with increasing temperature (Table 2). Owing to the temperature dependence of θ, the criterion Λ, determined by equations (3) and (7), was found to increase with increasing temperature as well. For a calculation of Λ for the motion of grain boundaries in the sample SI the ratio σ_3/σ was determined under the assumption that for temperatures near the melting point the

Table 2. Parameters of the motion of grain boundary systems investigated with triple junctions in Al-tricrystals

Sample	T (°C)	θ	Λ	V (m/s)	A_b (m^2/s)	A_{tj} (m^2/s)	H_b (eV)	H_{tj} (eV)
SI $\sigma_3/\sigma = 0.261$	398	58.5	2.6	0.58×10^{-6}		8.7×10^{-10}		
	418	62	3.19	1.1×10^{-6}		2.0×10^{-9}		3.2
	418	62	3.19	1.17×10^{-6}		2.5×10^{-9}		
	438	67	4.5	5.52×10^{-6}	2.8×10^{-9}	1.3×10^{-8}		
	454	72.5	7.4	1.48×10^{-5}	7.0×10^{-9}	5.2×10^{-8}	1.4	
	459	78	16.6	6.4×10^{-6}	3.4×10^{-9}	5.7×10^{-8}		
	479	80	32.4	1.45×10^{-5}	7.6×10^{-9}	2.5×10^{-8}		
SII $\sigma_3/\sigma = 0.286$	469	55	2.2	5.8×10^{-7}		6.6×10^{-10}		
	489	61.5	3.2	1.13×10^{-6}		1.7×10^{-9}		2.7
	510	61.5	3.2	4.79×10^{-6}		6.0×10^{-9}		
	530	62	3.3	1.85×10^{-5}	7.4×10^{-9}			
	551	63.5	3.7	3.3×10^{-5}	1.4×10^{-8}			
	571	70	6.1	4.32×10^{-5}	1.9×10^{-8}		1.4	
	591	72	7.4					
SIII $\sigma_3/\sigma = 0.735$	530	61.5	9.8	5.11×10^{-6}	2.7×10^{-9}			
	551	62	10.9	1.0×10^{-5}	5.3×10^{-9}			
	571	64	16.4	1.86×10^{-5}	9.8×10^{-9}		1.3	
	591	67	50.7	2.06×10^{-6}	9.4×10^{-9}			

value of θ reaches the thermodynamic equilibrium value† (see equation (9)). The measurement of θ near the melting point was obtained by the following experiment. A sample with grain boundaries in configuration SI was annealed at the temperature $T = 655$°C for 5 min. To avoid the complete disappearance of the shrinking grain between GB I and GB II the sample was notched as shown in Fig. 3a. After annealing, the sample was rapidly cooled down, and the position of the grain boundaries and the angle $2\theta \approx 2\theta_{eq} = 165°$ (Fig. 3b) were measured by optical microscopy. To determine Λ for the samples SII and SIII the data of the calculation of grain boundary energy calculated by Hasson and Goux were used [7]. The values of the ratio σ_3/σ for all systems investigated are given in Table 2.

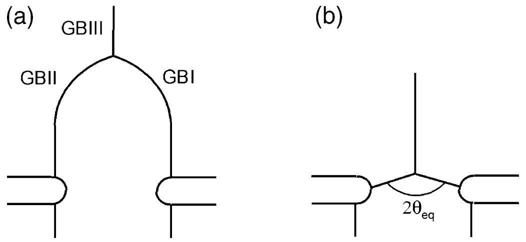

Fig. 3. Sketch of an experiment to determine θ_{eq}: (a) grain boundary system with triple junction and notches to avoid the complete disappearance of the grain enclosed by GB I and GB II; (b) the same system after annealing at 655°C.

4. DISCUSSION

The measured quantities in the current experiments were the migration rate V of the grain boundary system with a triple junction and the triple junction angle θ. Using the measured values of θ, the dimensionless parameter Λ was calculated for each annealing temperature (equation (7)), and the temperature dependency of Λ was determined. Figure 4 shows this dependency for the samples SI and SII. As follows from the approach given above, at low temperatures, where Λ is of the order of unity, the motion of the grain boundary system is controlled by the mobility of the triple junction. An increase in Λ with increasing temperature reflects the transition from triple junction to grain boundary controlled motion of the grain boundary system.

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† It is obvious that $\theta \rightarrow \theta_{eq}$ when the kinetics of the system become grain boundary controlled. In other words, $\theta_{measured} \approx \theta_{eq}$ can be expected near the melting point.

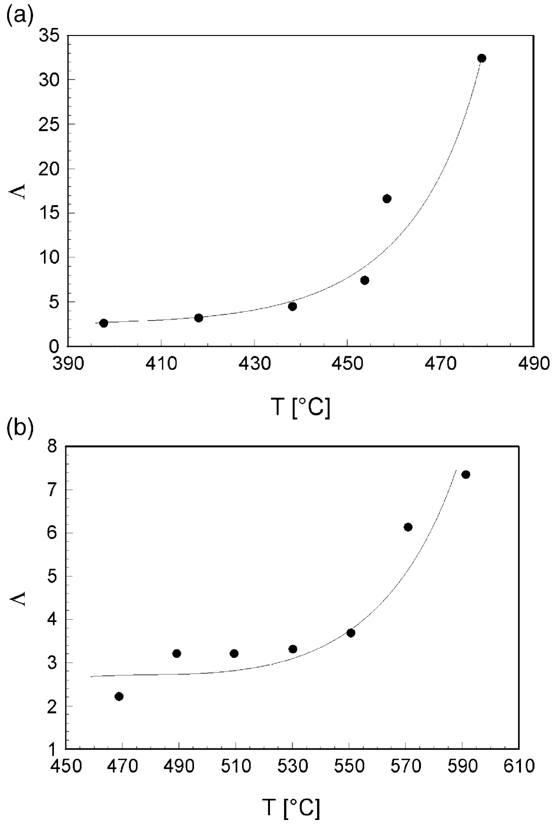

Fig. 4. Temperature dependence of the criterion Λ for triple junctions in samples SI (a) and SII (b).

The technique used for measuring the angle θ raises the important question of whether the angle measured after the sample was cooled down corresponded to the real triple junction angle at the annealing temperature. There are two main factors that might affect the angle θ: a thermodynamic influence, which reflects the temperature dependence of the grain boundary surface tension, and a kinetic influence, defined by the relation between grain boundary and triple junction mobilities and the width of the vanishing grain a. The thermodynamic effect is negligible, since the boundary surface tension depends only slightly on temperature, although how slightly is not known exactly. In any event, if the grain boundary surface tensions and their temperature coefficients are comparable, then the angle θ should be essentially temperature independent. The kinetic factor can really affect the magnitude of the angle θ. Taking into account the fact that the triple junction kinetics predominates at relatively low temperatures, as observed in [3, 4], the observed value of θ can be reduced compared to the real angle at the annealing temperature. However, because of the strong temperature dependence of the grain boundary mobility and the high cooling rate of the sample this change of the real value of θ is likely to be sufficiently small.

As Fig. 4 shows, the measurement of the temperature dependency of Λ can be used as a method to analyze the experimental data and to separate different regimes of motion of the system, i.e. triple junction and boundary controlled kinetics. At relatively low temperatures, up to 430°C for sample SI and up to 510°C for sample SII, the value of Λ is of the order of unity. Since the criterion Λ specifies the ratio of triple junction mobility to grain boundary mobility (equation (7)), low values of Λ mean that the triple junction mobility is comparable to the grain boundary mobility. Therefore, the motion of the system in the low-temperature regime can be interpreted as a triple junction controlled motion. The increase in Λ with increasing temperature indicates that the system motion becomes increasingly less affected by the triple junction, and boundary kinetics become predominant.

For a comparison of the current results with measured mobilities on single grain boundaries we determined the temperature dependence of the reduced mobility A of the triple junctions and grain boundaries involved (Fig. 5). Using equations (6) and (8) we obtain

$$A_{\mathrm{b}} \equiv m_{\mathrm{b}}\sigma = \frac{V\cdot a}{2\theta} = A_{0\mathrm{b}}\,\mathrm{e}^{-H_{\mathrm{b}}/kT} \qquad (10)$$

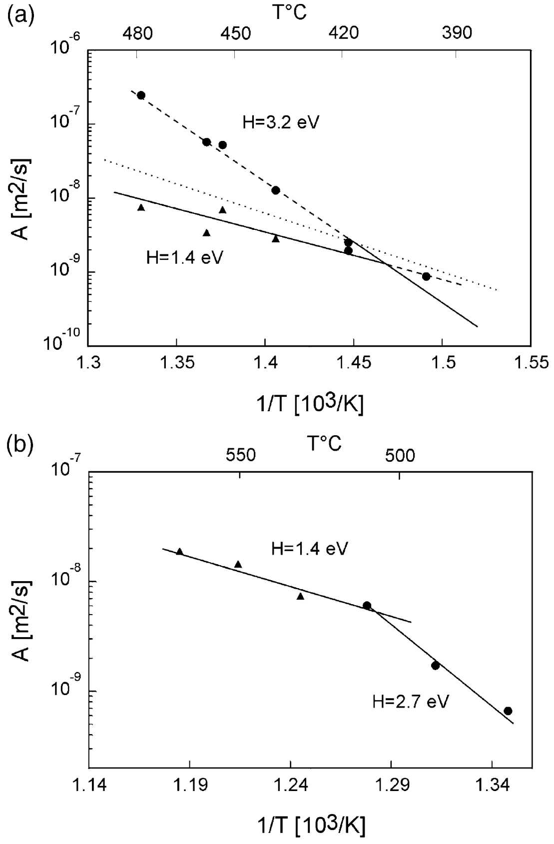

Fig. 5. Temperature dependence of triple junction (●) and grain boundary mobility (▲) in samples SI (a) and SII (b). The dotted line represents the mobility of a 21° <111> tilt boundary in Al as reconstructed from the literature data [3] ($H = 1.6$ eV; $A_0 = 10^3$ $\mathrm{m^2/s}$).

$$A_{tj} \equiv m_b \sigma a = \frac{V \cdot a}{2\cos\theta - \sigma_3/\sigma} = A_{0tj}\, e^{-H_{tj}/kT} \quad (11)$$

From velocity data and the temperature dependence of the contact angle 2θ as expressed in terms of $\Lambda(T)$ in Fig. 4, the respective grain boundary and triple junction mobility can be calculated according to equations (10) and (11). The results are given in Fig. 5 where the triangles represent the (reduced) grain boundary mobility as determined by equation (10), while the circles denote the respective triple junction mobility determined by equation (11). The system will obviously be controlled by the slowest moving constituent, i.e. the triple junction at low temperatures and the grain boundary at high temperatures. According to their respective linear behavior in the Arrhenius plot, the activation enthalpy can be determined as indicated in the figure. Obviously, there is a distinct transition from triple junction kinetics at low temperatures to grain boundary kinetics at elevated temperatures and the activation enthalpy for triple junction motion H_{tj} is considerably higher than for grain boundary migration (H_b). The behavior of the grain boundary controlled branch in Fig. 5 compares well with measurements of the reduced grain boundary mobility obtained from literature data [3] of independent bicrystal experiments. The corresponding evaluation for system SI is given in Fig. 5a and yields comparable behavior.

The measurements of grain boundary motion in the sample SIII were performed at a relatively high temperature. The persistently higher values of Λ and the constant but comparatively low activation enthalpy in the entire temperature range investigated (Fig. 6) indicate that the system always moves in the boundary controlled regime. Obviously, this has to be attributed to the structure and property of the rectilinear grain boundary in sample SIII which is practically a high-angle boundary contrary to the respective boundaries in the samples SI and SII, which are low-angle boundaries. It is worth noting that the motion of system SII, also consisting of <110> tilt boundaries, in this temperature range is controlled by the boundary motion as well.

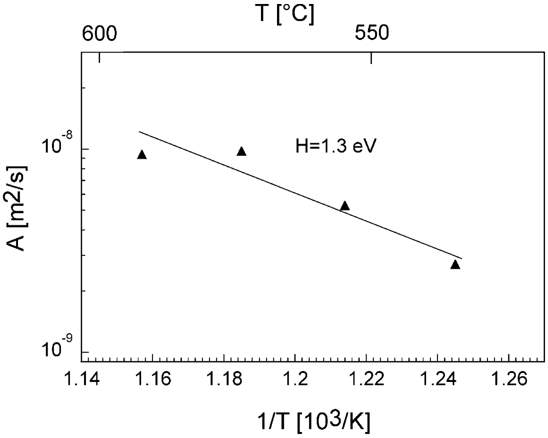

Fig. 6. Temperature dependence of the reduced mobility of grain boundaries in sample SIII.

5. CONCLUSION

The experimental results demonstrate that the motion of grain boundary systems with triple junctions in Al can be controlled by slowly moving triple junctions. The influence of triple junctions depends on temperature. It is particularly strong at low temperatures and small grain size. In the high-temperature regime the motion of a grain boundary system is less affected by the triple junction, and, therefore, effectively controlled by the grain boundary mobility.

A drastic difference between the activation enthalpy of grain boundary and triple junction motion was revealed by the current experiments. Therefore, there is a temperature below which triple junctions govern the motion of the connected boundary system and grain boundaries simply follow the triple junction. In other words, triple junctions play a role similar to a “source of dry friction” with respect to boundary motion. It seems that grain boundary motion in polycrystals is affected by their connectivity, i.e. by concurrent triple junction movement, and at relatively low temperatures the triple junctions act as slowly moving pinning centers.

As only a few triple junction systems have been investigated so far, it is an open question how the transition range between grain boundary and triple junction control of migration kinetics depends on the grain boundary and corresponding triple junction geometry. In particular, grain boundary systems consisting of structurally different grain boundaries (<111> and <110> tilt boundaries in the current study) need to be addressed to probe the influence of triple junctions and their geometry on texture development during grain growth.

It is finally noted that the results of the current study again demonstrate that the determination of an average grain boundary mobility by measurement of the mean grain size evolution during grain growth in polycrystals can be deceptive and, in particular, different for measurements in the high and low temperature range.

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