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Transition from thermally assisted to mechanically driven boundary migration and related apparent activation energies

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ABSTRACT

Grain boundary migration during deformation of nanostructured materials has been reported for a variety of materials and loading conditions. As boundary migration occurs even under cryogenic conditions, it is believed to be mainly mechanically driven, although results suggest thermal assistance. Yet, these contributions could not be separated and the underlying apparent activation energies remain uncertain. Here we present an approach that is capable to determine the average activation energies for mechanically driven boundary migration. While the apparent activation energy is increasing with temperature, below critical temperatures it becomes zero, indicating the existence of an entirely mechanically driven regime.

It is nowadays widely accepted that high angle grain boundaries (HAGBs) are not acting as static objects during deformation, but can migrate when mechanical loads are applied [1]. Especially for nanocrystalline (NC) and ultra-fine grained (UFG) materials, consisting of large quantities of HAGBs, their motion has been observed frequently upon various deformation conditions for a large variety of metals [2–10]. As in many of these studies grain coarsening was even observed at low homologous temperatures, the migration processes are supposed to be mechanically driven, although experimental data clearly suggest some thermal assistance. Increased testing times at fixed testing temperature during indentation creep or elevated testing temperatures and reduced strain rates during a tensile test unambiguously amplified the coarsening rate [2, 11, 12]. However, the extent of thermal assistance remains unclear and seems difficult to determine. Undoubtedly, determination of the apparent activation energies of boundary migration in nanostructured metals at low temperatures could help to quantify the role of thermal assistance. Additionally, knowledge of these values would allow also for more reliable modelling activities capable of forecasting mechanical stability of nanostructures. Clearly, for mechanically driven processes the apparent activation energy would be zero, while for thermally assisted ones any value between zero and those observed for thermally induced migration (i.e. the activation energy of grain boundary self-diffusion) could be expected. However, measuring such activation energies is challenging for NC or UFG specimens, as approaches used for measurement of stress induced migration in coarse grained crystals with well-defined boundaries are not applicable [1, 13]. An attempt in that direction was given by Vorhauer [14] and Ghosh et al. [15]. Both investigated for a variety of materials the relationship between the minimum crystallite size after severe plastic deformation and the Zener Hollomon parameter, Z, a parameter reflecting the temperature and strain rate compensated driving force for thermally activated processes. These studies clearly showed, that for high Z values, corresponding to low deformation temperatures and/or high strain rates, the grain size was only weakly depending on Z. Contrary, for low Z values, i.e. high deformation temperatures and/or low strain rates, the steady state grain size was strongly depending on Z, clearly suggesting the existence of a mainly athermal deformation regime. Nevertheless, for calculation of the Zener Hollomon parameter the activation energy for grain boundary self-diffusion, Q_{GB}, was used. However, upon closer inspection, these values are way too large.

The main mechanisms that occur during severe plastic deformation in order to restore a certain equilibrium grain size are the migration of grain boundaries and triple junctions (TJs), with grain boundary (GB) migration being dominant at low temperatures [16–18]. As this on average constant grain size results from a balance between grain refinement and the motion of GBs and TJs, for a given temperature, they have to move at a certain velocity v_{GB}, in order to sustain this dynamic equilibrium. Calculation of v_{GB} based on a diffusion based approach, i.e. activation energies for self-diffusion, yields for low homologous temperatures values being orders of magnitude too low. Typically for ambient temperatures the necessary v_{GB} is on the order of nanometers per second [12]. Additional grain fragmentation may occur under certain conditions due to formation of micro shear bands, which locally can split grains and reduce the aspect ratio.

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Based on experimentally observed boundary velocities at different temperatures, recently the kinetics of grain coarsening in nanogradient copper have been analyzed in detail [12]. Different to the experiments described above, the generally used Arrhenius type equation for v_{GB} was used [19], but the activation energy was assumed to be reduced by the presence of mechanical stresses. Above a critical stress σ_0 the apparent activation energy Q_0 was thought to decrease to zero, while for lower stress levels it was modelled with a power law, reaching values $Q_0 = Q_{GB}$, i.e. those for grain boundary self-diffusion, at zero load [12]. Although the coarsening kinetics of the nanograined copper could be well described with this approach, it is based on various assumptions. One of these, the stress dependency of the activation energy and so the direct correlation of the boundary velocity with the applied stress seems controversial. Despite most studies claim that the high shear stresses present in nanostructured materials during loading interact with disconnections at the boundary and provoke movement of the boundaries [1, 4, 7, 20, 21], many reports clearly show, that there is a direct correlation of coarsening and plastic strain [6, 10, 22]. These controversies motivated an unbiased approach determining the activation energies avoiding assumptions. Indeed, this is possible evaluating the necessary average boundary velocities of severely deformed specimens at various temperatures. Using nickel and tantalum as model materials, we will show, that with this approach the apparent activation energies for mechanically induced boundary migration can be reliably determined.

To obtain the necessary grain size data, pure nickel (99.99%) and tantalum (99.999%) samples were severely deformed using quasi constrained high pressure torsion (HPT). Disks having 8 mm in diameter and a thickness of 0.8 mm were deformed at various temperatures between 77 K and 473 K (nickel) or 673 K (tantalum) for 10 rotations at a rotational speed of 0.2 min^{-1}. Such high strains are sufficient to obtain a homogenous equilibrium microstructure throughout the disk, except for the very center. Temperatures of 77 K were realized immersing the whole setup into liquid nitrogen, while temperatures between 77 K and room temperature (RT) were adjusted using a double-wall container. The inner part of the container, which surrounded the HPT anvils, was filled with ethanol and cooled from outside to the selected temperature using liquid nitrogen, see schematics in Fig. 1. The temperature of the ethanol bath was controlled throughout the deformation procedure with a thermocouple. All microstructures were analyzed at a disk radius $r = 3$ mm in radial direction (see Fig. 2) as this viewing direction shows the minimum grain dimensions as well as the grain elongation of the HPT structure. Structural parameters were recorded using electron backscatter diffraction (EBSD) equipped with a Zeiss Leo 1525 field emission scanning electron microscope. The EBSD data were evaluated using a conventional OIM (orientation imaging micrograph) software package. HAGBs were defined as boundaries having a misorientation angle more than 15 deg. For analysis only data points with a confidence index (CI) being larger than 0.05 have been used. Representative colour coded inverse pole figure maps (IPF) of the equilibrium grain structures obtained at various temperatures are shown in Fig. 2. Along with the microstructure, microhardness was measured at the same disk radius ($r =$ 3 mm). The average grain dimensions as well as the room temperature hardness values for all deformation temperatures are summarized in Table 1. It should be noted, that the microstructures produced by HPT consist mainly of HAGBs (~ 80%), having almost random distributions of misorientation angle and axis [15]. As all structural investigations were carried out at room temperature, structural stability during warming of the samples deformed under cryogenic conditions (77 K, 173 K and 223 K) cannot be excluded a priori. Because structural observation of the structures under cryogenic conditions and during warming was not possible, the samples with the largest potential for coarsening (77 K) were subjected to additional isochronal annealing treatments (30 min) to identify their thermal stability. Prior to recrystallization of severely deformed metals, structural coarsening proceeds by continuous motion of triple junctions which progressively reduces the grain aspect ratio [23]. Therefore, if structural coarsening already took place during warming it will continue upon further annealing above room temperature. However, room temperature hardness measurements and corresponding structural analysis of the additionally annealed samples showed stable microstructures for tantalum and nickel up to 573 K and 373 K, respectively (see Supplementary section). Therefore, it is reasonable to assume that the structures synthesized under cryogenic conditions remain stable during warming to room temperature.

From Table 1 it is evident, that the dimensions of the short grain axis (minor axis) are decreasing with temperature, with the changes becoming progressively smaller towards lower deformation temperatures. Interestingly, below a certain temperature no reduction in grain size could be measured when deforming the samples at even lower temperature, compare Fig. 3 and Table 1. This indicates that below these temperatures, structural restoration processes such as boundary migration are occurring athermal. The critical temperatures to reach this mechanically driven migration regime were found to be about 0.05 T_m for tantalum and 0.10 T_m for nickel, Fig. 3. As mentioned before, at a given temperature and after sufficient strain, the velocity of the grain boundaries or triple junctions is in equilibrium with the refinement caused by the deformation path and the average grain size remains constant. It has been shown that at low homologous temperatures grain boundary migration is the dominant mechanism and significant contributions of triple junctions to the restoration process can only be expected above 473 K and 673 K for nickel and tantalum, respectively [18, 24], what are the maximum deformation temperatures applied to these materials within this study. Additionally, formation of micro shear bands was not observed for any of the samples. For this reason, grain boundary migration can be considered as the process responsible to maintain an equilibrium grain size. To fulfil this conditions, for an additional applied shear strain of $\gamma = 2$, which would refine the grains to half of their size, boundaries have to migrate on average by half of their thickness. Based on the experimental conditions the applied shear strain rate $\dot{\gamma}$ can be calculated to be $\dot{\gamma} = 0.07\ s^{-1}$ at a radius $r = 3$ mm, i.e. the position where all analysis was carried out. The time interval to impose a shear strain of $\gamma = 2$ to the sample ($r = 3$ mm) can be calculated to be 25.5 s. With half of the average grain size in axial direction being the necessary migration distance, one can calculate then the average grain boundary velocity v_{GB} for each temperature. These values are listed in Table 1. For both materials, at low homologous deformation temperatures, the velocity is in the order of 1 nm s^{-1}. As already mentioned, at these low temperatures such large values cannot be explained considering diffusion based

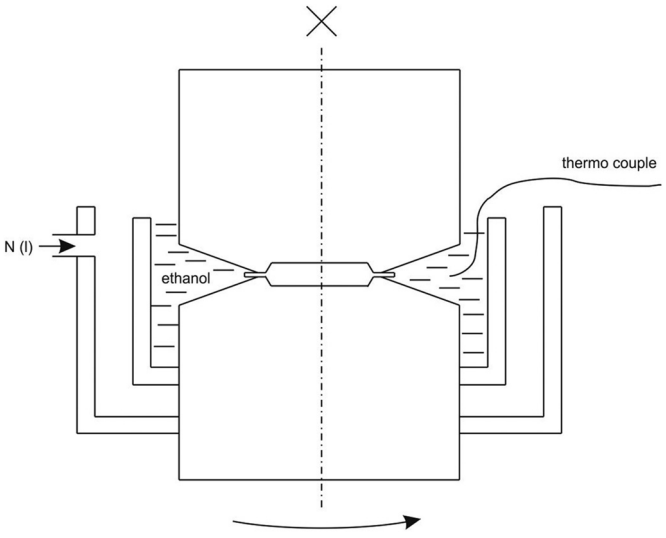

Fig. 1. Schematics, showing the HPT setup used to realize various cryogenic temperatures. Dimensions of the image are not to scale.