Understanding structural evolution of nanostructures during deformation from 2D (3D) experiments

O Renk¹*, M W Kapp² and R Pippan²

¹ Montanuniversität Leoben, Department of Materials Science, Chair of Materials Physics, Jahnstrasse 12, 8700 Leoben, Austria
² Erich Schmid Institute of Materials Science, Austrian Academy of Sciences, Jahnstrasse 12, 8700 Leoben, Austria

* corresponding author email: oliver.renk@oeaw.ac.at

Abstract. Understanding how microstructures evolve during deformation and/or annealing treatments is of fundamental interest, as the underlying structure dictates the resulting property spectrum. This issue is even more important for nanomaterials, where interfaces and defect populations have a stronger effect on mechanical properties. In addition, for nanometals the grain architecture can also be an important parameter for improvement of mechanical properties. However, synthesis of bulk high strength materials by severe deformation with tailored and desired properties requires a thorough understanding about the processes governing their microstructural evolution at large and severe strains. Here, we summarize findings from 2D (3D) experiments uncovering these restoration processes. Interestingly, the main mechanisms seem to change with deformation temperature, causing formation of extremely elongated grain structures at intermediate deformation temperature regimes. Apart from discussing the reasons for this finding, we also present recent results focusing on the mechanisms triggering these restoration processes at low temperatures, highlighting the crucial role of plastic strain to facilitate these processes.

1. Introduction
Mechanical properties of metals are inherently determined by the underlying defect and microstructure. Accordingly, any changes occurring during deformation and/or annealing will greatly influence their performance. This dependency seems to become even more important for ultra-strong nanomaterials. In such confined volumes, properties become extremely sensitive to defect concentrations, as well as the interfaces themselves, causing phenomena not present in conventional grain size regimes. For instance, deformed or distorted boundaries possess enhanced diffusivity [1] and when required, can emit defects much more easily compared to boundaries in the relaxed state, which can lead to further strengthening if mobile defects and boundaries are recovered [2–4]. Apart from this, the nanograin architecture may also play an important role for mechanical properties. As cracks preferentially grow along interfaces or grain boundaries, experiments suggest that preferentially aligned grains or phases could provide a promising strategy to unite strength and exceptional damage tolerance in one or two loading directions [5–7]. Accordingly, to make use of this knowledge, in-depth understanding of how specific interfaces, defect structures or grain architecture can be produced or how they result from a synthesis process is required. Nevertheless, although bulk nanostructures have been successfully fabricated by severe plastic deformation (SPD) [8] for several decades, knowledge of how desired nanostructures can be generated...
is extremely limited. So far, the resulting nanostructures have been processed by trial and error methods, rather than based on an understanding what deformation and restoration processes determine the resulting nanostructure after severe strains. However, such knowledge is necessary to deliberately process nanomaterials with desired and exceptional property combinations.

In recent years, research has tackled these questions and provided a considerable understanding of the processes that govern microstructural evolution after severe strains, already allowing a qualitative understanding of these mechanisms. It is thus the aim of this work to summarize these results obtained from various 2D and 3D experiments (with the third dimension representing the evolution as a function of strain or temperature) at different scales, showing that migration of boundaries or junctions are the essential processes governing the resulting nanostructure. Further, the interplay between these processes results in an interesting temperature effect, causing in some cases enormous grain elongations. In the second part, we focus on the question of what triggers boundary migration at comparably low deformation temperatures, an important question where controversial results can still be found. Recent experimental insights on this topic will be presented, which clearly indicate that plastic strain rather than the applied stress drives their motion.

2. Restoration during severe straining and the (unexpected) effect of temperature on grain shape

During heavy plastic deformation dislocation cell and grain sizes decrease continuously with strain, although the initially rapid size reduction tends to stagnate with strain [9]. Already early investigations on severely deformed single phase materials showed indeed that microstructural features (e.g. grain size or shape, and misorientation distribution) do not change on average any further once a sufficiently high strain is applied ($\varepsilon \sim 10$ for torsion at ambient temperature) [10–12]. As materials deformed to such strain levels can still be deformed further, structural recovery or restoration mechanism are required to keep structural features on average at a constant level. Despite knowing for a long time that such a steady state or saturation regime exists for single phase metals, the underlying necessary restoration or recovery processes remained unknown and controversially discussed. Accordingly, various different mechanisms have been proposed, see for instance Refs. [13–16].

Several years ago, results from 2D (3D) experiments on submicron sized copper and aluminum, respectively, allowed for the first time to unambiguously identify the actual mechanisms at the grain scale [17,18]. The structural evolution of samples already cold worked to severe strains was monitored quasi in-situ during additional strain increments. The experiments unambiguously revealed that migration of grain boundaries (GBs) and/or triple junctions (TJs) provokes a local collapse of grains, allowing adjacent grains to grow and maintain their average size during deformation. Therefore, the notion of saturation or steady state might be misleading, as the structure is not stable at all, but rather a dynamic equilibrium between (local) coarsening and refinement is achieved. It should be noted that migration of boundaries is often not observed over the entire boundary length. Rather, local bulges form that split apart neighboring grains, before being in some cases totally consumed (see figure 1). In any case, both processes, grain fragmentation or migration of junctions, can effectively reduce the grain length and ensure that average microstructural features are maintained. Thus the processes may be considered to be similar to that of conventional dynamic recrystallization, but occurring at much lower deformation temperatures (below $0.3 \ T_m$). Due to the absence of significant thermal activation, the migration processes are thus often referred to as mechanically induced. Nevertheless, a clear thermal assistance can be determined down to extremely low deformation temperatures of $0.05 – 0.10 \ T_m$ [19].

As migration of GBs or TJs is certainly a thermally facilitated process, both mechanisms will occur at higher rates if the (deformation) temperature is increased. This is also reflected in an increase of the minimum grain dimensions for SPD at elevated deformation temperatures [19,20]. Additionally, more globular grain structures should evolve for an increase of the deformation temperature, a trend also observed in classic hot working experiments [9,21]. Similar behavior was also found for SPD deformed single phase materials, although mostly low melting point metals, such as aluminum alloys, were investigated in these studies, e.g. Refs. [22,23]. Interestingly, recent investigations on single phase
materials deformed by high pressure torsion (HPT) up to strains of \( \varepsilon \sim 100 \) at different temperatures (between 0.05 and 0.35 \( T_m \)), suggest a different but general behavior [24,25]. For both tantalum (99.95 % purity) and nickel (99.99 %), the grains became more elongated with an increase of the deformation temperature. This trend continued up to deformation temperatures of about 0.2 – 0.25 \( T_m \), before the generally expected trend of decreasing aspect ratios with deformation temperature could be observed. Interestingly, while the trends for nickel and tantalum were qualitatively similar, the absolute values of the resulting aspect ratios differed significantly.

Figure 1. Microstructural evolution of ultra fine grained (UFG) nickel during an additional strain increment of \( \varepsilon \sim 0.2 \) by rolling at ambient temperature. Grain boundary migration is one of the main restoration mechanisms at severe strains. Boundaries do not necessarily move homogenously over the entire boundary length. In many cases bulges (an example is highlighted with arrows) form locally and effectively reduce the length of adjacent grains.

While for nickel maximum aspect ratios of about 4.5 were measured, the tantalum grains became exceptionally elongated at 673 K and reached values of almost 10, see figure 2a. Investigations on several other materials suggest that the trend of the aspect ratio as a function of the deformation temperature observed for nickel and tantalum is indeed a general one (figure 2b). Presumably this has been overlooked so far, as research has been focused mainly on low melting point model materials for which room temperature deformation lies already well above 0.25 \( T_m \), i.e. the temperature range where for most other pure metals maximum grain elongation was observed. It should be noted, that although the results were obtained on HPT deformed samples, similar trends are observable for other deformation routes as well, although the values of the aspect ratio may change, being higher in case of rolling compared to torsion, see figure 2b. In addition, alloying shifts the deformation temperature causing maximum grain elongation towards larger values, figure 2b.

Certainly understanding of this unexpected behavior would yield valuable insights into the underlying mechanisms. Based on evaluation of the microtextures a shift of the deformation mechanism can be excluded, and dislocation-based plasticity prevails for all investigated deformation temperatures [24]. A closer look at the various microstructures confirmed that the increase of grain elongation at intermediate deformation temperatures results from a significantly faster increase of the long grain dimensions compared to the shortest ones (i.e. in the case of the HPT disk along the axial direction) [24,25]. It is also evident that at moderately elevated deformation temperatures most of the grain boundaries appear straight, and the frequently observed fragmentation events from bulging boundary segments (figure 1) become widely absent with an increase of the deformation temperature, see figure 2a. This suggests that the formation of the bulges is an important mechanism for grain subdivision that prevents significant grain elongation at low deformation temperatures.
Figure 2. a) Representative microstructures of pure nickel and tantalum deformed at room temperature and the temperature (418 K and 673 K for nickel and tantalum, respectively) where maximum grain elongation was determined. b) Results obtained on a variety of materials under two deformation modes (HPT and rolling) confirm that the observed trends are indeed general ones. Image b) is taken from Ref. [26].

Formation of these bulges could be explained by connecting the presented results with in-situ TEM observations of mechanically induced grain boundary migration [27,28]. These experiments revealed that mechanically induced migration proceeds by the movement of individual GB steps, so called disconnections. These steps need not be pure steps, but can be associated with a dislocation, thus their movement will also realize a shear strain, referred to as coupled motion of a GB [27,28]. Of course their Burgers vector does not necessarily have to lie in the GB plane but could contain a climb component, necessitating elevated temperatures for their migration [28]. Such steps, with comparably low mobility, were found to hinder more mobile ones, resulting in the formation of huge macro-steps [27], having a similar appearance to the bulges observed during deformation of UFG structures especially at low deformation temperatures (figure 1). The observed increase of the aspect ratio with deformation temperature can thus be rationalized by increasing mobility of the steps within the boundaries. At elevated temperatures, the individual step mobility is increased, resulting not only in increased minimum grain dimensions (vertical axis in figure 2a), but as well in a reduced probability for macro-step formation, preventing frequent grain fragmentation, in line with the observed increase of the aspect ratio.

Interestingly, the deformation temperatures causing maximum grain aspect ratios, were also found to be in excellent agreement with those where the severely deformed structures started to coarsen during static annealing treatments [24,25]. The onset of structural coarsening in severely deformed metals was found to originate from migration of triple junctions [29]. For this reason, the reduction of the grain elongation above certain deformation temperatures (> 0.25 \( T_m \) for pure metals expected, compare figure 2) seems to be a consequence of the onset of triple junction migration, which effectively reduces the grain length.

While the description above provides a qualitative but general view on the important mechanisms that govern microstructural evolution at severe strains, the differences between the individual materials are astonishing, as seen in figure 2b. While measurable for iron or nickel, the effect is significantly more pronounced for the tantalum samples. It can thus not be explained by, or related to, the crystal structure. However, the absolute values of the measured aspect ratios are found to scale with the grain boundary energies that can be expected for the various materials [30], with larger grain aspects for metals having
higher boundary energies. For pure metals the interfacial energies are also proportional to the grain boundary free volume [31,32], which in turn may explain why, for certain materials, intergranular mobility is enhanced. Nevertheless, despite being technically sound, this argument requires further research to be validated.

3. What triggers mechanically induced boundary migration?

In the previous section, we have summarized experimental findings, showing that the interplay between migration of GBs or the mobility of individual GB steps seem to determine the resulting microstructures after severe strains at moderate deformation temperatures (< 0.25 $T_m$). As diffusion processes are rather limited in this temperature regime, the processes are believed to be mainly mechanically triggered. While the description in the previous chapter provided a rather qualitative description of microstructural evolution and the responsible processes, quantitative predictions require a more fundamental understanding of what triggers the motion of grain boundaries. As described above, mechanically induced boundary migration proceeds by the movement of disconnections, which can couple with an applied stress field. Based on experimental evidence for shear coupling in bicrystal experiments and in-situ straining on UFG aluminium (e.g. Refs. [27,33]), most studies conclude that boundary migration is stress induced, becoming much easier in nanometals due to their enhanced yield stresses. Although technically sound, several studies also provide hints that migration is facilitated in highly strained regions, see for instance Refs. [34,35].

To provide further insights regarding this question, ultrafine-grained samples were subjected to cyclic loading at comparably large plastic strain amplitudes, as low cycle fatigue conditions are well known to provoke significant grain coarsening in nanostructured metals [36]. As severely deformed metals can be considered to deform in an almost ideally plastic manner (i.e. negligible work hardening), microstructural investigations on samples tested at different strain amplitudes, should allow for a conclusive statement regarding the effect of stress or strain on mechanically induced growth. However, in conventional fatigue experiments, tracking of the structural evolution is difficult as the position where grain growth and fatigue damage happens is hard to distinguish a priori. Due to these experimental constraints, a different testing routine was used. The nanostructured samples processed by HPT were directly subjected to cyclic torsion in the same setup [35]. After a fixed twist angle, the lower anvil is forced to change its rotation direction, mimicking LCF conditions, see the schematic in figure 3. The radial dependency of strain in torsion allows for the investigation of various strain amplitudes within one sample, while the applied hydrostatic pressure prevents sample failure and allows tracking of the microstructures up to an arbitrarily large number of cycles.

Experiments were conducted on ultra-fine grained nickel (99.99 %) at ambient temperature to avoid large thermal contributions and to probe the mechanically dominated regime. The nickel samples were subjected to a twist angle of five degrees for various numbers of cycles, and hardness and structural evolution were recorded continuously. As expected, the grains coarsened continuously with increasing number of cycles. However, within a couple of applied cycles, grain growth caused the formation of a shear band that had a wedge like shape spanning over the entire HPT disk, see figure 3. Interestingly, grain growth within the shear band was amplified compared to the regions outside the band, where the applied stresses are still high but where grain growth was only negligible. This unambiguously shows, that mechanically induced grain growth is promoted by the plastic strain. However, this finding may not be contradictory to the findings that GB motion proceeds by movement of disconnections. While nucleation of a disconnection at a GB would require rather high stress levels [37], they may be easily generated by interaction and absorption of lattice dislocations with/at GBs [28,38]. Thus it may not be surprising, that in highly strained regions, where such interactions also occur more frequently, GB migration rates are enhanced. Interestingly, the heavily coarsened grains inside the shear band region exhibited also preferred crystallographic orientations, different from the as HPT deformed (monotonic) state or outside the shear band [35], see figure 4. These orientations can accommodate cyclic slip on a single or two coplanar slip systems, while outside the shear band two non-coplanar systems are required.
[39], with the latter one presumably retarding dislocation-GB interactions and so eventually the generation of disconnections.

Figure 3. a) Schematics showing the experimental setup for cyclic HPT. b) Structure of ultra-fine grained nickel subjected to 50 cycles of cyclic HPT at a twist angle of five degrees at ambient temperature. A shear band developed throughout the HPT disk in which grain growth was amplified compared to the regions outside.

Figure 4. Inverse pole figure maps of the starting microstructure (monotonic HPT) and the grain structure outside and inside of the shear band (heavily coarsened region) plotted along the tangential (shear direction) and the axial direction (shear plane) of the HPT disk. It can be noticed that during cyclic HPT preferential crystallographic orientations develop, which can accommodate the cyclic strain on only one or two coplanar slip systems. Evaluated data is taken from Ref. [35].
4. Summary and conclusions
As a whole the experiments suggest that the main mechanisms controlling microstructural evolution at severe strains change with temperature. At low temperatures formation of macro-steps was found to be mainly responsible for grain fragmentation. Consequently, the enhanced step mobility at elevated deformation temperatures causes a significant increase of the resulting grain aspect ratios until frequent motion of triple junctions subdues this effect. Although at low deformation temperatures coupling with the applied stress field can force elementary boundary steps to migrate, experiments clearly show that boundary migration is amplified in highly strained regions, highlighting the importance of dislocation absorption at boundaries to initiate these processes.

Acknowledgements
Financial support by the European Research Council under ERC Grant Agreement No. 340185 USMS and by the Austrian Science Foundation (FWF) under project P25325-N20 is gratefully acknowledged.

References
[1] Divinski S V., Reglitz G, Golovin IS, Peterlechner M, Lapovok R, Estrin Y and Wilde G 2015 Acta Mater. 82 11
[2] Huang X, Hansen N and Tsuji N 2006 Science 312 249
[3] Rupert T J, Trelewicz J R and Schuh C A 2012 J. Mater. Res. 27 1285
[4] Renk O, Hohenwarter A, Eder K, Kormout K S, Cairney J M and Pippan R 2015 Scr. Mater. 95 27
[5] Pippan R and Hohenwarter A 2016 Mater. Res. Lett. 4 127
[6] Hohenwarter A, Völker B, Kapp M W, Li Y, Goto S, Raabe D and Pippan R 2016 Sci. Rep. 6 33228.
[7] Nikolić V, Wurster S, Firneis D and Pippan R 2018 Int. J. Refract. Met. Hard. Mater. 76 214
[8] Estrin Y and Vinogradov A 2013 Acta Mater. 61 782
[9] Hansen N 2001 Metall. Mater. Trans. A. 32 2917
[10] Valiev R Z, Ivanisenko Y V., Rauch E F and Baudelet B 1996 Acta Mater. 44 4705
[11] Wang Z C and Prangnell P B 2002 Mater. Sci. Eng. A 328 87
[12] Hebesberger T, Stüwe H P, Vorhauer A, Wetscher F and Pippan R 2005 Acta Mater. 53 393
[13] Mohamed F A 2003 Acta Mater. 51 4107
[14] Pippan R, Scheriau S, Taylor A, Hafok M, Hohenwarter A and Bachmaier A 2010 Annu. Rev. Mater. Res. 40 319
[15] Zhao Y H, Liao X Z, Zhu Y T, Horita Z and Langdon T G 2005 Mater. Sci. Eng. A 410–411 188
[16] Edalati K, Akama D, Nishio A, Lee S, Yonenaga Y, Cubero-Sesin J M and Horita Z 2014 Acta Mater. 69 68
[17] Renk O, Hohenwarter A, Wurster S and Pippan R 2014 Acta Mater. 77 401
[18] Yu T, Hansen N, Huang X and Godfrey A 2014 Mater. Res. Lett. 2 160
[19] Renk O and Pippan R 2018 Scr. Mater. 154 212
[20] Vorhauer A and Pippan R 2008 Metall. Mater. Trans. A 39 417
[21] Rollett A, Humphreys F, Rohrer G S and Hatherly M 2004 Recrystallization and Related Annealing Phenomena: Second Ed. (Amsterdam: Elsevier)
[22] Chen Y C, Huang Y Y, Chang C P and Kao P W 2003 Acta Mater. 51 2005
[23] Wang Y Y, Sun P L, Kao P W and Chang C P 2004 Scr. Mater. 50 613
[24] Renk O, Ghosh P and Pippan R 2017 Scr. Mater. 137 60
[25] Renk O, Ghosh P and Pippan R. 2017 IOP Conf. Ser.: Mater. Sci. Eng. 219 012037
[26] Renk O, Pippan R. 2019 accepted for publication in Mater. Trans. doi.org/10.2320/matertrans.MF201918
[27] Rajabzadeh A, Legros M, Combe N, Mompiou F and Molodov D A 2013 Philos. Mag. 93 1299
[28] Rajabzadeh A, Mompiou F, Lartigue-Korinek S, Combe N, Legros M and Molodov D A 2014 Acta Mater. 77 223
[29] Yu T, Hansen N and Huang X 2013 Acta Mater. 61 6577
[30] Vitos L, Ruban A V, Skriver H L and Kollár J 1998 Surf. Sci. 411 186
[31] Olmsted D L, Foiles S M and Holm E A 2009 Acta Mater. 57 3694
[32] Scheiber D, Pippan R, Puschnig P and Romaner L 2016 Model. Simul. Mater. Sci. Eng. 24 035013
[33] Winning M, Gottstein G and Shvindlerman L S 2001 Acta Mater. 49 211
[34] Fang T H, Li W L, Tao N R and Lu K 2011 Science 331 1587
[35] Kapp M W, Renk O, Leitner T, Ghosh P, Yang B and Pippan R 2017 J. Mater. Res. 32 4317
[36] Mughrabi H and Höppel H W 2010 Int. J. Fatigue 32 1413
[37] Rajabzadeh A, Mompiou F, Legros M and Combe N 2013 Phys. Rev. Lett. 110 265507
[38] Priester L. 2013 Grain Boundaries From Theory to Engineering 1st ed. (Dordrecht: Springer Netherlands)
[39] Toth L S, Gilormini P and Jonas J J 1988 Acta Metall. 36 3077