20 years of INT KIT-USATU collaboration in the area of SPD

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Abstract. The field of Severe Plastic Deformation (SPD) has provided the scientific community with ultrafine grained and nanocrystalline materials. In contrast to other techniques for the synthesis of nanostructured materials, SPD processes allow the preparation of bulk quantities of metals and alloys. Technical applications have become a reality because commercial alloys can be transformed into novel nano- and microstructures, leading to superior mechanical properties, such as high strength and high ductility, resulting in many application areas. The field was pioneered at the Ufa State Aviation Technical University and has created worldwide interest. The report emphasizes some results of the close collaboration between Ufa State Aviation Technical University and the Institute of Nanotechnology at Karlsruhe Institute of Technology (KIT).

1. Introduction
The discovery of Severe Plastic Deformation (SPD) in its many different technical variations has opened a new field of materials research by allowing the preparation of many metastable metallic alloy systems in bulk form with ultrafine and nanometer grain sizes. The original idea to use severe plastic deformation to modify metallic materials and the establishment of the concept of non-equilibrium interfaces has been developed by Ruslan Valiev at Ufa State Aviation Technical University (USATU) [1], during the early period of nanocrystalline materials research [2]. The term SPD encompasses many well-established deformation processes, all leading to large plastic deformation and enormous modifications of the microstructure of the materials, resulting in large changes of the properties. Among these deformation processes are ECAP (Equal Channel Angular Pressing) [3], HPT (High Pressure Torsion) [4,5], ARB (Accumulated Roll Bonding) [6], and more. After years of mainly basic research the different processes have also been scaled up to fabricate larger amounts of materials, such as the continuous ECAP process (ECAP-Conform) [7], or more recently, the High Pressure Torsion Extrusion (HPTE) process [8]. The interesting basic science of severely deformed materials, the possibility to produce bulk samples with internal nanostructures with substantially larger volume compared to samples produced by other techniques such as Inert Gas Condensation (IGC) [9], and the opportunity to transfer such materials to industrial applications all led to a fast growth of this field in the past 20 years. Among the most exciting and pioneering scientific results that established a worldwide interest was the possibility to create materials with high strength and high ductility, not possible by conventional materials structures [10].

As a result of the complementary expertise at both institutions and the intensive exchange of scientists, the Institute of Nanotechnology at Karlsruhe Institute of Technology (INT KIT) collaborates with the group of Ruslan Valiev at Ufa State Aviation Technical University for the past twenty years, a good time to summarize the results of this collaboration. In this respect the groups were successful in
acquiring substantial research funding on both sides, leading to several joint publications. On the German side, the Deutsche Forschungsgemeinschaft (DFG) funded the project entitled “Plasticity in nanocrystalline metals and alloys” from 2006 to 2012 and the project “Basic Mechanisms of Mechanical Property Enhancement for Carbon Steels through Controlled Nanostructure Formation” from 2009 to 2012, leading to several joint publications \[11-13\]. On the other hand, the ideas in the area of SPD born at USATU were creatively developed at INT KIT, and the most prominent results are presented in this paper.

2. Instrumented High Pressure Torsion – a new tool to study deformation behavior of materials with limited ductility in a wide range of strains

As mentioned before, high pressure torsion (HPT) straining is a method of severe plastic deformation and it is conventionally applied as a technique for making ultrafine-grained materials, when a microstructure of coarse-grained metals and alloys is refined down to the grain size of 100–300 nm \[4,5\]. On the other hand, the application of HPT in a combination with torque measurement can be used as a method of mechanical testing. This is especially interesting for nanocrystalline materials with a grain size of 10–20 nm, because they show very limited ductility in tension, which restricts the experimentally accessible parameter space, whereas torsion testing under high pressure \[14,15\] allows practically unlimited shear strain without fracture. This technique was applied to a nanocrystalline Pd and Pd-10 at.% Au alloy prepared by IGC \[16\]. Figure 1 shows the graphs for IGC Pd with an initial grain size of 14 nm (as determined by TEM) deformed at three different strain rates. It is seen that the samples exhibit more rapid initial work hardening compared with coarse-grained Pd. Furthermore, the torque in the initial deformation stage (for \(e < 1\), figure 1) is significantly rate dependent, with a clear trend for more torque at faster strain.

![Figure 1. Shear Strain vs. Torque curves of coarse grained (CG) Pd deformed at 0.8×10\(^2\) s\(^{-1}\) and NC Pd deformed at different shear strain rates.](image)

The TEM observations of the microstructure after HPT revealed interesting changes resulted from severe straining. First of all, significant grain growth to 40 nm at the strain \(e = 15\), more than twice the initial value. At the same time, the grains appear still equiaxed at this strain. The most remarkable microstructural features are structures such as those shown in figure 2a,b, when several neighbouring segments of grain boundaries are seen to align in parallel. These features were typically observed quite frequently in samples deformed to \(e = 15\) at all studied strain rates, and were clearly absent in the as-IGC state. The formation of such arrays of coplanar grain boundaries confirms the predictions in Ref. \[17\], of a type of cooperative grain boundary sliding (GBS). Also significant strain rate sensitivity (figure 1) suggests that a thermally activated process like grain boundary sliding controls the deformation of nanocrystalline Pd. The subsequent studies of crystallographic texture development \[18,19\] confirmed that random texture persisted when the mean grain size was less than 20 nm. Such features as random texture and formation of mesoscopic shear planes extending over several grain boundaries represent a clear evidence of GBS at room temperature deformation in nanocrystalline Pd. And it could be revealed in material with limited ductility only thanks to application of instrumented HPT.
3. HPT-driven phase transformations

HPT not only leads to grain refinement but also drives phase transformations that might be distinguished as diffusion-controlled and diffusionless (martensitic) ones.

3.1. HPT-induced diffusion-controlled phase transformations

HPT processing allows an increase of the strain up to the extreme values without fracture. At steady state of deformation, the dynamic equilibrium between deformation-driven production of crystal defects and their relaxation (annihilation) is established. Such a strong influence on the material leads to accelerated mass transfer and, therefore, to various phase transformations \([1,20-30]\) e.g. the formation or decomposition of a supersaturated solid solution, dissolution of phases, disordering of ordered phases, alloying in immiscible systems, low-temperature synthesis of new nanocrystalline alloys.

Closer look at steady state processes revealed that the competition (and dynamic equilibrium) between the dissolution of precipitates and decomposition of supersaturated solid solution with precipitation of a second phase during HPT takes place in two-phase Cu-based alloys. As a result, a dynamic equilibrium between these two processes appears, and a certain steady-state concentration in a solid solution is reached (figure 3). This concentration is *equifinal* and equivalent to that if the sample annealed at the effective temperature \(T_{\text{eff}}\). Therefore, the equivalent diffusion coefficient for the HPT-driven mass transfer \([23,25]\) is comparable with the conventional diffusion coefficient at (elevated) \(T_{\text{eff}}\). This coincidence additionally supports the view point about deep physical links between *equifinal* amount of defects appearing in steady-state during HPT at low temperature \(T_{\text{HPT}}\) and defects *equilibrium* existing in the same material at elevated temperature \(T_{\text{eff}}\). At the same time, this fact nicely correlates with the estimations of non-equilibrium vacancy concentrations during HPT which are as high as that values close to melting temperatures \([31]\).

**Figure 2.** Dark field images using the 111 reflections of nc Pd microstructure after HPT for \(\dot{\gamma}=15\) at \(\dot{\gamma} = 0.8 \times 10^{-2} \text{s}^{-1}\) (a) and \(\dot{\gamma} = 0.8 \times 10^{-1} \text{s}^{-1}\) (b): (a) alignment of boundaries of 10 grains with formation of a mesoscopic shear plane; (b) another example of mesoscopic shear plane. Reprinted from [16] with permission of Elsevier.

**Figure 3.** The Cu-rich part of the Cu-Ag phase diagram. Evolution of different compositions is shown for the same alloy before and after HPT.
3.2. Diffusionless HPT-induced phase transformations

Contrary to diffusive phase transformations, non-diffusive ones require atomic shuffling for rather smaller, much less than interatomic, distances. Ti and Ti-based alloys possess a high pressure ω-phase that can be observed as metastable one at ambient conditions after release of applied pressure or after HPT treatment as well. The effect of alloying of titanium with iron (up to 10 wt % Fe) on the ω-phase formation during HPT was studied using XRD analysis and HREM [26-30]. During shear deformation under uniaxial pressure, a pronounced orientation relationship between adjacent grains of α- and ω-phases is accompanied by a strong local preferred orientation, which is recognized as a “basal” crystallographic texture in α-phase. It appears favorable for the realization of the α-to-ω transformation under HPT conditions. A defect-rich high-pressure ω-phase forms after HPT and persists in the samples also after the pressure release. The amount of retained ω-phase after HPT depends on the iron concentration. It increases from 40% in pure titanium, reaches maximum of 95% at 4 wt % Fe and then decreases again to 10% at 10 wt. % Fe. It is because the addition of iron influences the lattice parameters in β- and ω-phases in a different manner. The minimal lattice mismatch between β- and ω-phases is reached at 4 wt. % Fe (figure 4). A good conformity between the lattices of the β- and ω-phases enhances the probability of the martensitic (diffusionless) β-to-ω transformation.

4. Scaling up of high pressure torsion

Until recently, a major disadvantage of the HPT method has been the small dimensions of the samples (typically disks 10-20 mm in diameter and about 1 mm in thickness), which hinders its commercial use for structural applications. Recently, the research team at INT has proposed to scale-up the HPT method for industrial processing of advanced materials with at least three order of magnitude larger yield using a new technical approach [32]. The novel machine allows continuous processing of long rods instead of the thin plates used in conventional HPT. The basic idea of the method, which was called high pressure torsion extrusion – HPTE, is to use a continuous transmission of the shear deformation zone along the metallic rod that is pushed through the die (figure 5). The strain accumulated in a specimen after one pass can be controlled by adjusting the specimen diameters ratio (D1/D0) and the velocities of the translational (v) and rotational (ω) motions of the tools according to the equation:

\[ e = 2 \ln \frac{D_1}{D_0} + 2 \ln \frac{D_1}{D_2} + \frac{\omega \cdot R_1}{\sqrt{3} \cdot v} \]  \hspace{1cm} (1)

where R1=0.5D1 is the specimen radius.

The proposed method has an important advantage over other large scale SPD techniques, i.e., equal channel angular pressing, accumulative roll bonding, twist extrusion, etc., as it enables processing of the billet in one pass without intermediate steps and thus allows for continuous fabrication of nanocrystalline rods. Furthermore, HPTE provides exciting new opportunities for the purposely design of nano- and microstructures and creating of hybrid materials [33].
Using the HPTE method, copper billets with the diameter of 12 mm and length of 35 mm were processed [8]. As a result, a gradient structure with coarse microstructure in the central part and uniform UFG microstructure with equiaxed grains and mean grain size of 350 nm in the rest of the sample was formed (figure 6). Figure 7 shows the results of microhardness measurements for the specimens processed using HPTE at different translational velocities \( V \), i.e. different strain (equation (1)). It is clearly seen that the hardness distribution correlates well with the distribution of strain. In the central part of a specimen where the torsional deformation is negligible, the hardening occurs only due to the expansion-extrusion deformation. For the specimen processed at higher translational velocity, the microhardness gradually increases along the radius and reaches the value of about 116 HV at the sample’s periphery. In the case of specimen processed at lower velocity, the hardness saturates at about 120 HV already at the distance of 1 mm from the specimen axis. Therefore, using different processing parameters in HPTE one can easily adjust the volume fraction of the material with low or high hardness.

![Figure 6. Orientation maps (inverse pole figure maps) of a Cu specimen after annealing and processing by HPTE. Coloring corresponds to the projection of normal direction (ND) in the inverse pole figure.](image)

![Figure 7. Variation of the microhardness of copper samples along the specimen radius in initial annealed state and after HPTE with rotational velocity \( \omega = 1 \) rpm and different translational velocities.](image)

5. Conclusions and outlook

While the field of Severe Plastic Deformation (SPD) is already more than 20 years old, it is still vibrant and many groups worldwide continue to develop new processing routes based on severe plastic deformation, to invent new materials and material combinations and address scale up aspects towards technological applications. The field includes strong basic science aspects on the structure formation, the phase stability under severe plastic deformation and detailed characterization of the resulting structures, as well as modelling contributions.

It should be noted that the collaboration between the INT KIT and Valiev’s group at USATU resulted recently in another DFG-funded project entitled “Fundamental Parameters and Enhanced Properties of Amorphous Alloys Subjected to Severe Plastic Deformation (SPD)”. In this project, the deformation behavior of metallic glasses during SPD will be examined with a particular emphasis on the shear bands formation in metallic glasses and their structural characterization.
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