Microstructure evolution in ultrafine-grained interstitial free steel processed by high pressure torsion

M Janeček¹, T Krajňák¹, J Stráská¹, J Čížek¹, D J Lee², H S Kim², J Gubicza³

¹Charles University in Prague, Czech Republic, Ke Karlovu 3, 121 16, Prague 2, Czech Republic
²Department of Materials Science and Engineering, POSTECH, Pohang 790-784 South Korea
³Department of Materials Physics, Eötvös Loránd University, Budapest, Hungary

E-mail: milos.janecek@met.mff.cuni.cz

Abstract. Commercial interstitial free steel was processed by high pressure torsion (HPT) at room temperature up to 5 revolutions. HPT resulted in strong grain refinement. The microstructure after HPT was inhomogeneous with refined grains mainly in regions near the specimen periphery, while coarse only slightly fragmented grains were observed in specimen centre. The microstructure inhomogeneity was continuously smeared out with increasing number of rotations by extending the fine grain region from specimen periphery towards its centre. However, even after 5 revolutions the microstructure remained inhomogeneous characterized by slightly coarser grains in central regions as compared to peripheral regions of the specimen. Positron annihilation spectroscopy (PAS) and X-ray line profile analysis (XLPA) were employed to characterize the structure inhomogeneity in individual specimens. Microstructure and dislocation density evolution were correlated with mechanical properties characterized by a detail microhardness measurement throughout the individual specimens.

1. Introduction

The grain size of polycrystalline materials influences many critical properties of the material including the strength and the resistance to plastic flow. Materials with small grain sizes have several advantages over their coarse-grained counterparts because they have higher strength and other favourable properties. Hence, tailoring microstructures of bulk ultrafine-grained (UFG) materials has attracted interest of scientific community over the past decades. Due to the grain size in the submicrometer or nanometre range UFG materials possess enhanced mechanical and other physical properties while retaining appreciable level of ductility [1-3]. UFG materials can be produced via severe plastic deformation (SPD) using a top-down approach which involves extremely large plastic strain to be imposed to the material while retaining the initial dimensions of the workpiece [4-6]. SPD top-down techniques are of particular interest owing to their ability to produce considerable grain refinement in fully dense, bulk-scale workpieces giving a promise for structural applications. Up to now a variety of techniques for the production of bulk UFG materials emerged, in particular equal channel angular pressing (ECAP) [7], high pressure torsion (HPT) [5], accumulative roll-bonding (ARB) [9], etc. Recently, inspired by the success of these "classical" techniques, which process samples with the simple shape like rods or disks, more exotic methods were developed enabling a higher throughput; for the list, see the recent review by Estrin [10].

Among "classical" techniques high pressure torsion, that involves a combination of high pressure (usually in GPa range) with torsional straining, became very popular among researchers as it allows the most efficient grain refinement of the material. Bulk solids with microstructures in submicrocrystalline range - pure metals, metallic alloys, intermetallics, composites, etc. - have been successfully produced using HPT [11-15]. The principles of modern HPT process have already been described extensively in the literature [16-17]. Two
important parameters in the HPT process are the magnitude of the imposed pressure (P) and the number of revolutions (N) applied to the disk sample [8].

Interstitial-free (IF) steels belong to an important class of steels having carbon content less than 0.01 wt%. These steels are widely used in automobile industry as sheet material due to their excellent deep drawability as a result of the low content of interstitial solutes and particular texture in cold rolled state [18-20]. Efforts have been made recently to improve the strength of this class of steels by grain refinement through SPD procedures, namely by ECAP [21]. Detailed reports of HPT processed materials are rather sparse.

The objective of this work is to investigate the microstructure evolution in IF steels processed by HPT and to correlate it with mechanical properties characterized by microhardness variations throughout the individual specimen. Light and electron microscopy as well as X-ray diffraction line profile analysis were employed to characterize the microstructure evolution. Complementary to a detailed microhardness measurement the PAS was employed to investigate the structure inhomogeneity.

2. Material and experimental
The material used in this investigation was an IF steel with a composition of 0.0026 wt.% C, 0.096 wt.% Mn, 0.045 wt.% Al, and 0.041 wt.% Ti. The material was procured from Pohang Steel Company (POSCO), Korea. The as-received material was homogenized at 973 K for 2 h and furnace cooled. A rod with the diameter of 10 mm was machined from the material. For HPT, disks with the thickness of 1 mm were cut from the rod using a diamond saw. A series of specimens with different number of turns N = ½, 1, and 5 was processed by HPT at room temperature using the pressure of 2.5 GPa. Additionally, a specimen which was only pressed with the same pressure of 2.5 GPa was also used in this study (marked N=0).

For metallographic examinations the samples were mounted in Epofix® resin and mechanically polished successively using 240-, 1200- and 2400-grit SiC papers. Subsequently, polishing by 3 μm and 1 μm diamond suspensions and a final etching in 2% Nital solution for 10 s were carried out. Light microscopy imaging using differential interference contrast (Nomarski contrast) was performed by an Olympus light microscope. Microstructure evolution in the HPT processed specimens was observed by high resolution scanning electron microscope FEI Quanta 200 FEG operated at 10 kV using backscattered electrons. Employing channelling contrast revealed the grain structure.

Automatic microhardness tester Qness Q10a was utilized for Vickers microhardness (HV) mapping (the applied load of 500 g and the holding time of 10 s were used). A regular network of indents consisting of concentric circles with the radius step of 0.25 mm and the distance of 0.25 mm along each circle was used for HV measurements, see Fig. 1. HV map on each specimen consisted of 1100 indents.
Mapping of the lateral distribution of defects in HPT-deformed samples was performed by means of spatially resolved Doppler-broadening (DB) spectroscopy [22]. A small drop (≈1 mm in diameter) of $^{22}$Na radioisotope with activity of ≈1 MBq deposited on 2 µm thick mylar foil was used as a positron source. Spatially resolved DB measurements were performed simply by positioning the positron source spot at various radial distances from the centre of the sample disk using a x-y moving stage. The uncertainty in position of the positron source was ≈0.1 mm. The DB of the annihilation photopeak was measured by a high-purity germanium (HPGe) detector with the energy resolution of 1.30 keV at 511 keV and was evaluated using S line shape parameter [23]. The central region for calculation of the $S$ parameter was chosen symmetrically around the 511 keV annihilation peak from 510.15 to 511.85 keV. All $S$ values presented in this work were normalized to the $S$ parameter $S_0 = 0.5026(5)$ measured in the well annealed pure α-Fe specimen.

The microstructure of the HPT-processed samples was also studied by X-ray line profile analysis (XLPA) along the radius of the disks. The X-ray line profiles were measured by a high-resolution diffractometer with CoKα radiation (wavelength: $\lambda = 0.1789$ nm). The size of the X-ray beam spot on the sample surface was about $2 \times 0.2$ mm$^2$, where the longer dimension of the rectangular spot was set to be perpendicular to the disk diameter. As the longer dimension of the beam spot (2 mm) was smaller than the radius of the disks (5 mm) only by a factor of 2.5, the microstructure could not be studied in the vicinity of the edge of the samples. Therefore, XLPA measurements were carried out only in the centre and the half-radius of the disks. Additionally, due to the 2 mm high beam spot the parameters obtained at the centre characterize an averaged microstructure between the centre and 20% of the disk radius. The line profiles were evaluated by Convolutional Multiple Whole Profile (CMWP) fitting method [24]. In this procedure, the diffraction pattern is fitted by the sum of a background spline and the convolution of the instrumental pattern and the theoretical line profiles related to the crystallite size and dislocations. The theoretical profile functions used in this fitting procedure are calculated on the basis of a model of the microstructure, where the crystallites have spherical shape and log-normal size distribution, and the microstrains are caused by dislocations.

3. Results and discussion

3.1. Microhardness

Fig. 2 shows the three-dimensional plots of Vickers microhardness HV0.5 for specimens subjected to different number of HPT turns. The variations of microhardness with position within the disk are clearly displayed by the colour code at these maps. Table 1 summarizes the average HV values at the centre, half-radius and edge of the HPT-processed disks. The microhardness increases with increasing the distance from the centre even in the specimen which was only pressed (N=0), which can be explained by the material outflow at the periphery of the HPT anvils and the consequent hardening under a semi-constrained quasi-hydrostatic pressing [5]. In HPT strained specimens (Fig. 2 (b)-(d)) two distinct regions are readily apparent – a central region with comparatively low HV and the peripheral one with enhanced HV. These 3D plots clearly demonstrate the microhardness behaviour found also in
many other materials [5, 10, 25], that HPT straining introduces an inhomogeneity in the material which is manifested by the minimum value of the hardness in the centre and much higher microhardness values in the specimen periphery. This inhomogeneous microhardness distribution is formed already in the specimen after 1/4 HPT turn where the pronounced drop of HV in the central region is clearly recognizable, see also Table 1. Note that this inhomogeneous character of HV distribution remains also in the specimen after 5 turns, even if the zone of enhanced HV is continuously extending from the edge towards the specimen centre.

3.2 Microstructure evolution

The microstructure of the specimen which was only pressed (N=0) is presented in Fig. 3. It consists of equiaxed grains with the average size of 80 µm. The pressing resulted in apparent grain reduction (by a factor of 3) without any preferred orientation, as reported in our previous work [21]. Owing to the inhomogeneous character of HPT straining with theoretically zero imposed strain in the specimen centre and linearly increasing strain values with distance from the centre one can reasonably anticipate the inhomogeneous character of the microstructure developed in specimens processed by different number of HPT turns. Fig. 4 displays the SEM micrographs obtained by channelling contrast for different zones in specimens after 1 and 5 HPT turns. The inhomogeneous character of the microstructure both with increasing distance from the centre and with increasing N in the same zone is clearly seen in Fig. 4. In the centre of the specimen N=1 the microstructure has a bimodal character consisting mostly of bands of elongated grains/subgrains, cf. Fig. 4(a). The average length and width of these grains is about 2 µm and 800 nm, respectively. This proves that the process of grain fragmentation has already started even in the central zones of the disks. However, original large grains from the initial as-pressed specimen (N=0) are also present in the microstructure. On the other hand, both in the zones near the middle of the radius and the edge the well-refined grain structure is clearly seen. The size of equiaxed grains in the half radius and the edge of the specimen processed by a single turn N=1 is 300-400 nm and 200-250 nm, respectively, cf. Figs. 4(b) and 4(c). Further straining by HPT up to 5 turns resulted...
**Fig. 2** Three-D plots of microhardness variations throughout the surface of individual specimens

**Table 1** Average values of HV in different zones of the disks

| Spec./HV | 0   | 1/4 | 1   | 5   |
|----------|-----|-----|-----|-----|
| Centre   | 117 | 135 | 153 | 214 |
| Half-radius | 141±8 | 200±8 | 255±7 | 337±3 |
| Edge     | 174±12 | 253±5 | 325±3 | 355±7 |

**Fig. 3** Microstructure of the specimen  
N=0

(a) \( N = 1 \) – centre (smaller magnification)  
(d) \( N = 5 \) – centre
Fig. 4 SEM micrographs displaying the microstructure in different part of specimens after N=1 and N=5 HPT turns (back scattered electrons – channelling contrast)

in grain refinement throughout the whole specimen. However, the laterally inhomogeneous grain structure still remains, with markedly larger grains in central zones (average size 400-500 nm), cf. Fig. 4(d) than in zones near disk half radius (average size 250-300 nm), cf. Fig. 4(e) or near the disk edge (average size 200-250 nm), cf. Fig.4(f). By comparison of Figs 4(c) and 4(f) one can see that in peripheral zones of the disk the grain size saturates already after one turn, while in zones near the half radius of the disk the grain refinement is not completed, cf. Figs 4(b) and 4(e).

3.3 PAS – DB mapping
Fig. 5(a) shows the dependence of the $S$ parameter on the radial distance $r$ from the centre of the specimen which was only pressed ($N = 0$) and the specimens subjected to various numbers $N$ of HPT revolutions. It is apparent that $S$ exhibits only moderate variations with $r$
and slightly increases from the centre of the sample towards the periphery. However, $S$ strongly rises with increasing level of HPT straining. This can be observed in Fig. 5(b) which shows the development of $S$ in the centre ($r = 0$ mm) and at the periphery ($r = 3$ and $4$ mm) with increasing number of HPT revolutions. During the first HPT turn $S$ strongly increases due to increasing density of defects and becomes saturated for $N > 1$. The $S$ parameter at the periphery is in most cases slightly higher than in the centre indicating slightly higher concentration of defects. However, the development of $S$ with increasing number of HPT revolutions in the centre and at the periphery is very similar.

Fig. 5 (a) Dependence of the $S$ parameter on the radial distance $r$ from the centre of the sample disk; (b) development of the $S$ parameter in the centre and at the periphery with increasing number of HPT revolutions $N$

3.4 XLPA

The area-weighted mean crystallite size ($<x>_{area}$) and the dislocation density ($\rho$) obtained from CMWP fitting procedure are listed in Table 2. The value of $<x>_{area}$ is calculated as $<x>_{area}=m\cdot\exp(2.5\sigma^2)$, where $m$ is the median and $\sigma$ is the square-root of the log-normal variance of the crystallite size distribution. The crystallite size was small (90-110 nm) while the dislocation density was high ($\sim 1.9 \times 10^{14}$ m$^{-2}$) already in the initial only as-pressed specimen ($N=0$). The dislocation density considerably increased while the crystallite size did not change significantly with the increasing number of revolutions during HPT. The dislocation density saturated earlier at the half-radius (after 1/4 turn) than in the centre (only after 1 revolution), since the strain evolution is faster at larger distance from the disk centre. The saturation values of the average crystallite size and the dislocation density in HPT-processing were about 80 nm and $8 \times 10^{14}$ m$^{-2}$, respectively. It should be noted that the crystallite size determined by XLPA for the HPT-processed samples is lower than the grain size observed by SEM, cf. Table 2. This phenomenon has been observed for other plastically deformed bulk metals [21] and can be attributed to the fact that the crystallite size determined from X-ray line profiles corresponds essentially to the mean size of cells or subgrains which is usually smaller than the conventional grain size measured in severely deformed metals by electron microscopy methods. Comparing the XLPA and SEM results (given in Table 2) it can be concluded that the grain refinement continued with the increasing number of HPT turns even after the saturation of the dislocation density. When the dislocation density reached its maximum the grains were further refined by the arrangement of these dislocation into grain boundaries and/or by recrystallization.

Table 2. The parameters of the microstructure obtained by X-ray line profile analysis: the area-weighted mean crystallite size ($<x>_{area}$) and the dislocation density ($\rho$). The average size
of equiaxed grains \((d)\) determined from SEM is also given in the table (the elongated grain size is not given in the table)

| Number of revolutions | \(\langle x \rangle_{\text{area}}\) [nm] | \(\rho\) \([10^{14} \text{ m}^{-2}]\) | \(d\) [nm] |
|-----------------------|-----------------|-----------------|---------|
| Centre                |                 |                 |         |
| 0                     | 110 ± 10        | 1.9 ± 0.2       | 80 x 10^3 |
| 1/4                   | 108 ± 11        | 3.6 ± 0.4       | 3 x 10^3 & 900^1 |
| 1                     | 114 ± 12        | 7.0 ± 0.7       | 2 x 10^3 & 800^1 |
| 5                     | 91 ± 9          | 8.4 ± 0.9       | 400-500  |
| Half-radius           |                 |                 |         |
| 0                     | 92 ± 9          | 1.9 ± 0.2       | 80 x 10^3 |
| 1/4                   | 82 ± 9          | 8.4 ± 0.9       | 650-750  |
| 1                     | 94 ± 9          | 8.3 ± 0.9       | 300-400  |
| 5                     | 84 ± 8          | 7.9 ± 0.9       | 250-300  |
| Edge                  |                 |                 |         |
| 0                     | n/a             | n/a             | 80 x 10^3 |
| 1/4                   | n/a             | n/a             | 300-400  |
| 1                     | n/a             | n/a             | 200-250  |
| 5                     | n/a             | n/a             | 200-250  |

* elongated grains (length & width)

### 3.5 Correlation between microhardness, dislocation density and microstructure evolution

From comparison of Figs. 2 and 5(a) one can conclude that the dependence of HV and \(S\) on the radial distance \(r\) from the centre of the sample disk exhibit different features. In the beginning of HPT straining \((N \leq 1)\) HV remarkably increases with \(r\), see Fig. 2(b,c). After one HPT revolution HV at the periphery of the sample becomes saturated \((HV \approx 340)\) and with further straining the periphery region with saturated HV gradually extends towards the centre. However, a well defined minimum of HV can be always observed in the centre even in the sample subjected to 5 HPT revolutions.

On the other hand, the \(S\) parameter exhibits only moderate increase with \(r\) in the beginning of HPT straining \((N < 1)\). When the first HPT revolution is completed \(S\) remains approximately constant across the whole sample, see Fig. 5(a). Different behaviour of HV and \(S\) can be understood considering that hardening is caused either by dislocations or by grain size reduction (the Hall-Petch relationship) while \(S\) parameter is influenced predominantly by the dislocation density. This confirms again the conclusion from our previous study on UFG Cu processed by HPT [22] that HV and DB mapping are not alternative, but rather complementary techniques providing information about different aspects of microstructure inhomogeneity in HPT deformed samples.

Fig. 4 shows that grains in the centre are substantially coarser than at the periphery even after 5 HPT revolutions, but the lateral distribution of dislocation density is likely much more uniform as confirmed by XLPA results, cf. Table 2.

### 4. Conclusions

The microstructure and dislocation density evolution in ultrafine-grained interstitial free steel polycrystals processed by high pressure torsion were correlated with microhardness variations throughout the surface of individual specimens subjected to various number of HPT turns. The following conclusions can be drawn from this experimental investigation:
- HPT straining results in strong grain refinement,
- HPT introduces lateral inhomogeneity of the microhardness which may be characterized by an apparent minimum in the specimen centre and much higher HV values near its periphery. With the increasing number of HPT turns the HV inhomogeneity is continuously smeared out by extending the zone of enhanced HV from the specimen periphery towards its centre. However, even after 5 HPT turns both the microhardness and the microstructure remain still inhomogeneous,
- with increasing strain the dislocation density saturates much quicker in zones further from the centre of the specimen than in the specimen centre. The dislocation density was found to saturate at lower strain than the grain size,
- HV variations throughout the specimen reflect both the dislocation density variations and grain size reduction while PAS DB mapping indicates the dislocation density variations only and it is quite insensitive to grain size changes.

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