ACOUSTIC EMISSION IN ULTRA-FINE GRAINED COPPER

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1. Introduction

During the past decade, the ultra-fine grained (UFG) materials (both nanostructured and submicro-crystalline) have attracted much of attention largely due to their superior mechanical properties such as a high strength with significant plasticity (or even superplasticity) at relatively low temperatures [1]. Despite considerable efforts, the mechanisms of plastic deformation of these advanced materials remain unclear [2–5]. Strain (stress) localization is an important factor which controls degradation of mechanical properties in a wide range of materials. Modern high strength materials such as ceramics are rather brittle and sensitive to the stress concentration. Hence, the investigation of strain (stress) localization in UFG materials is of utmost importance for characterization of their mechanical stability. An acoustic emission (AE) technique provides real-time information on the structural rearrangements in very small volumes of solids. For this reason, in the present work we employ AE to explore possible plastic instabilities in UFG copper subjected to monotonic tensile loading.

AE at the onset of plastic deformation of metallic materials is usually associated with the motion of dislocations. AE parameters are usually connected to dislocation velocity, mean free pass, etc. A strong effect of grain size on AE has long been recognized [6–15]. The AE experimental results which are available to date reflect a pretty complicated role of grain boundaries in plastic deformation. Comprehensive reviews concerning the grain size and grain boundary effect on AE are given in refs. [11–13]. To our knowledge, no reports are available on dislocation AE detected in pure polycrystalline metals with the grain size smaller than 10 μm.

From the aforementioned data on AE in the crystalline solids it is hard to expect measurable AE during plastic deformation of pure nanocrystals. Furthermore, adopting the criterion for AE detectability proposed by Wadley and Mehrabian [12] \( naV \geq 0.035 \text{ m}^2 \text{ s}^{-1} \) (\( a \) is the radius to which a dislocation loop can expand before arrest at a pinning point, \( V \) is the radial velocity and \( n \) is a number of dislocations involved into a co-operative motion) we obtain that for \( a = 10^{-7} \text{ m} \) \( nV \) should be greater than \( 3.5 \times 10^5 \text{ m/s} \). This is unreasonable value to expect, particularly in view of the estimates made in ref. [4] where it was shown that only one dislocation passes in average through each grain of 200 nm per second upon loading with a strain rate of \( 10^{-3} \text{ s}^{-1} \). Although these estimates are rather rough, it seems that there is neither experimental nor theoretical basis for dislocation AE in nanocrystals. However, there are at least two strong points in a favour of possible AE in UFG polycrystals. (1) The idea of AE measurements in these materials was stimulated by former observations of shear bands and fine slip lines during cyclic and tensile experiments [4, 16, 17]. It has been shown that plastic flow in UFG Cu is not limited to the intergranular deformation and extends over a large number of grains. Localization of plastic deformation in the form of shear bands implies that the co-operative rearrangements in the ensembles of defects occurs, facilitating AE detection. (2) Several authors have argued...
both experimentally [4, 16] and theoretically [5, 18] that the unusual mechanical behaviour of sub-microcrystalline materials may be due to a large volume fraction of the specific grain boundary phase. This glassy-like phase is believed to be responsible for the similarity in the mechanical behaviour of ultra-fine grained and amorphous metals: 1) a very high yield stress [4, 19] in both materials; 2) shear banding in the direction of the maximum shear stress with no significant strain hardening upon monotonic [20] and cyclic testing [16, 17]; 3) temperature dependence of Young modulus [19, 20]; 4) enhanced diffusivity and excess free volume [21], etc. Although there are no “traditional” crystallographic dislocations in amorphous solids, AE has been detected in strained metallic glasses and analysed in detail [22, 23].

The present paper is the first report on AE generated during plastic deformation of pure UFG materials. Therefore, we focus ourselves on the general characterization of AE appearance and on the comparison of the AE observed with known results obtained for both conventional polycrystals and metallic glasses.

2. Experimental Procedure

2.1 Sample Preparation

UFG copper of 99.98% purity with the mean grain size of 200 nm was produced by severe plastic deformation, using an equi-channel angular pressing (ECAP) technique [4, 21]. UFG materials are thermally unstable. Their structure and properties change drastically upon annealing at relatively low temperatures [19]. To explore the effect of preliminary heat treatment on the plastic behaviour and AE of UFG Cu we used three kinds of sample: (1) the samples in the as-received state (specimen A); (2) as-annealed at 473 K for 3 min (specimen B); (3) as-annealed above the recrystallization temperature at 773 K for 2 h (specimen C).

The specimens of nearly square cross-section of $2 \times 2 \text{ mm}^2$ and 6 mm gauge length for mechanical testing were cut by spark erosion. They were then mechanically and electrolytically polished to remove a damaged layer and to obtain a mirror like surface suitable for optical microscopic observations during testing. Bright field transmission electron microscopy (JEOL-2000EX) was used for structural characterization of the specimens before mechanical testing.

2.2. Tensile Testing and AE Apparatus

Tensile tests were done under a constant nominal strain rate from a range of $1.4 \times 10^{-5}$ to $1.4 \times 10^{-2}$ s$^{-1}$ on a screw-driven testing machine at room temperature. The broad band (100–1000 kHz) AE-sensor AE-900M (NF Electronics Instruments) was securely mounted on the face surface of 5 mm apart from the gauge part. The measuring system and the algorithms of data processing have been described in ref. [24]. The output of the 60 dB preamplifier is connected with a 12 bits analog-to-digital converter having a sampling frequency of 4 MHz. The digitized image of the AE signal is stored in the 16 ksamples buffer memory before transferring to a computer for further saving and processing. A total gain was of 90 dB. The ADC threshold was set slightly above the peak noise level. The electrical noise signals coming from various parts of laboratory equipment were recorded just before testing for further identification of the “false” events to be excluded from AE analysis.
3. Results and Discussion

3.1. Structural Characterization of the Specimens

Microstructure, texture and grain boundary distributions in UFG materials produced by ECAP were formerly characterized by TEM both in the as-received state after fabrication and after annealing at various temperatures [4, 20, 25, 26]. A typical TEM photograph of the fine-grain structure of as-received UFG copper is shown in Fig.1a (specimen A). This structure does not differ notably from that described in the papers mentioned above. The grains in the samples prepared by severe plastic deformation are usually separated by high-angle grain boundaries with a random spectrum of misorientations [25]. These boundaries are often specified as “non-equilibrium” because of their long range stress fields. A nature of high internal stresses in UFG materials has been discussed, for example, in terms of random extrinsic grain boundary dislocations [18], junction disclinations [27] and dislocation pile-ups [28]. Some smaller grains are almost free of dislocations while chaotically distributed dislocations are clearly visible in larger grains. The average dislocation density is seen to be very high. Its value for the same UFG ECAP Cu was obtained about $5 \times 10^{14}$ m$^{-2}$ (that is, however, a very rough estimate) [4].

Figure 1. TEM micrographs of the as-received (a) and as-annealed for 3 min at 473 K (b) ultra-fine grained copper produced by ECAP.
The grain growth occurs in UFG ECAP copper during annealing at 423–473 K for sufficiently long time [19, 26]. However, as emphasized in ref. [4], the short-time annealing at 473 K may not result in the significant grain growth but reduces internal stresses due to recovery processes in non-equilibrium grain boundaries and inside the grains. This, in turn, results in the changes of mechanical properties (the yield stress decreases, for instance). Figure 1b gives a bright field TEM image of microstructure after annealing of the as-received specimen at 473 K for 3 min (specimen B). No significant growth in the mean grain size can be noticed. Thus, the specimens A and B have approximately the same grain size. Nevertheless, the state of grain boundaries in these specimens is supposed to be different due to recovery caused by a short-term heat treatment. The samples C have the grain size of 50–70 μm typical for polycrystalline copper used in former AE studies.

3.2. AE in Coarse-Grained and Fine-Grained Copper

Figure 2 demonstrates a typical example of AE observed in a relatively coarse-grained copper (specimen C) obtained by annealing of the as-received UFG specimen above the recrystallization temperature. AE appears in a time domain as a noise-like continuous signal formed by a flux of overlapped pulses of low and moderate amplitudes. The AE-level attains its maximum near the yield point and decreases gradually as the deformation proceeds. Such AE behaviour in pure fcc metals has been well established [6–10]. The AE decay during plastic deformation is often associated with the increase in lateral constraints for dislocation movement. Thus, the refinement of structure and substructure restricts dislocation mobility, resulting in the total disappearance of AE at a certain fairly small grain size [13]. For the same reason AE is negligible in ordinary pure cold-worked materials so that we do not detect any acoustic emissions in rolled copper, for instance. However, we observe some clearly pronounced AE signals in the UFG specimens in the as-received state (specimen A), Fig.3. One can see that acoustic emissions appear randomly in time near the yield point. A total number of AE events is
rather small, ranging from a few signals to a few hundreds in dependence on the strain rate. Fracture occurs in a ductile manner and is not accompanied by notable AE.

Figure 3 represents a typical record of AE during tensile deformation of the UFG sample A. The stress-strain curves do not reveal any hardening. The noticeable stress decrease after yielding should be attributed to the change in the sample crosssection due to necking. The UFG specimen in the as-received state (sample A) shows burst type AE (the AE pulse is clearly visible in Fig.3) in contrast to that in coarse grain polycrystals. This striking difference reflects an apparent dissimilarity in AE sources in UFG and coarse grained copper.

AE during plastic deformation of metals is irreversible. This property of AE is usually addressed to the so-called Keiser effect [29] which states that once a material has been loaded to a certain stress (strain) and then unloaded, no new AE is produced in the second loading until the previous stress (strain) is exceeded. We do not detect any AE during unloading and subsequent loading of UFG Cu.

The slip markings on the surface of specimen A strained to $\varepsilon = 0.06$ are shown in Fig.4. They appear just after yielding and extend over a fairly long distance compared to a grain size. The slip lines are oriented at about 45 degrees with respect to the tensile axis, that corresponds to the maximum shear stress. Presumably one can suppose that AE is caused by this slip or, at least, by the coarsest slip events. However, the nature of AE in UFG Cu is still ambiguous and is a subject of further investigations.

3.3. Strain Rate Dependence of AE in Ultra-Fine Grained Copper

As shown in Fig.5, AE in UFG copper is strain rate dependent similarly to that in conventional polycrystals: the AE activity increases with increasing strain rate (in conventional polycrystals, the AE maximum power is usually proportional to the strain rate [30]). When the strain rate is low enough AE is not detected at all, Fig.5c. For burst-type AE of a low activity in UFG copper one cannot say that a certain AE parameter is simply proportional to the strain rate. However, a certain correlation between
the AE activity and the strain rate apparently exist. The similar correlation was found in metallic glasses subjected to tensile deformation [22].

3.4. Amplitude and Duration Distributions

The AE signals in UFG specimens are clearly separated. They can be characterized by a rather high average amplitude $<U_p>$ of 84 $\mu$V (the peak-to-peak noise level taken from the sensor output is of 7 $\mu$V) and duration $<\tau>$ of 64 $\mu$s. The sample standard deviations for these parameters are of $\sigma_u = 8.0$ $\mu$V and of $\sigma_\tau = 6.5$ $\mu$s correspondingly. Empirical frequency histograms are shown in Fig.6a and b for the amplitude $U_p$ and duration $\tau$ respectively. A distinct feature of these AE parameters in UFG copper is that they are nearly normally distributed (as proved by the Kolmogorov-Smirnov test of normality), while in ordinary pure polycrystals we usually observe truncated Pareto distributions [31]. Gaussian distributions corresponding to the calculated mean values and variances for both $U_p$ and $\tau$ are also plotted in Fig.6 by smooth solid lines.

3.5. Effect of the Heat Treatment

Several investigations have shown that the properties of UFG materials strongly depend on the structure and properties of grain boundaries [1, 5]. In particular, it has been suggested that the mechanical properties of ECAP fine-grained materials are influenced by the non-equilibrium grain boundary structure formed during severe plastic deformation [4, 18, 27]. Compare the mechanical behaviour of the specimen A and B which have approximately the same grain size but differed by their internal stresses and the state of grain boundaries. Their stress-strain curves are plotted in Fig.3. The decrease in the yield stress after heat treatment agrees with the results reported in ref. [19]. We do not observe any other significant differences in the tensile behaviour of these samples. However, no AE is detected upon loading of the specimen B. Figure 3 shows the AE activity for the specimen A only. Thus, the recovery which usually commences from grain boundaries has apparently resulted in de-localization of plastic flow due to reduction of internal stresses. This makes it evident that the grain size is not a unique factor which determines the mechanical behaviour of the ECAP materials.

3.6. On the Mechanism of AE in UFG Materials

Let us make a short note concerning the possible mechanism of AE discussed. ECAP produces extremely hardened metals so that no stress/strain hardening is usually observed during mechanical
testing. The load-carrying ability of individual grains attains its maximum possible value due to hardening imposed upon fabrication (the yield stress in the UFG state is about 20 times greater than that in the annealed state and is near the ultimate tensile stress). Hence, it seems plausible to suppose that the enhanced plastic properties of ECAP materials should be attributed to the mechanisms of strain accommodation, which are related to grain boundaries. These mechanisms might be similar to those taking place during superplastic deformation (plastic rotation, grain boundary sliding, etc).

It has been noticed that in plastically deformed UFG materials grain sliding as well as grain boundary migration may serve as effective channels of stress relaxation by facilitating the absorption of lattice dislocations in the grain boundaries [4]. However, according to the results discussed in the literature, these mechanisms of stress relaxation either do not contribute to AE [32] or their contribution is negligible [33] in fine grained polycrystals. A certain resemblance between the AE appearance in UFG copper and in metallic glasses makes it possible to assume that the AE sources in these different materials are also similar. In our opinion, a highly disordered non-equilibrium grain boundary phase in

Figure 5. Stress-strain curves and AE activity in the as-received UFG copper (sample A) tested at different strain rates: (a) \( \dot{\varepsilon} = 1.4 \times 10^{-2} \) s\(^{-1}\); (b) \( \dot{\varepsilon} = 1.4 \times 10^{-3} \) s\(^{-1}\); (c) \( \dot{\varepsilon} = 1.4 \times 10^{-4} \) s\(^{-1}\).
the UFG ECAP materials may deform in a glass-like manner, showing many features typical for inhomogeneous plastic flow of metallic glasses: AE is one of clear examples.

4. Summary

Acoustic emission (AE) is observed during tensile testing of ultra-fine grained (UFG) copper produced by severe plastic deformation. The AE appearance is investigated over a wide range (about three orders of magnitude) strain rates. Experimental findings can be summarized as follows:

- AE is of a burst type;
- AE signals appear randomly in time near the yield point;
- AE in UFG Cu is irreversible;
- AE in UFG Cu is strain-rate dependent similarly to that of coarse-grained polycrystals and of metallic glasses, i.e. the AE activity increases with strain rate;
- AE in UFG Cu is largely dependent on the specific state of grain boundaries.

The experimental AE results allow to conclude that strongly localized plastic deformation of the mesoscopic scale can occur in UFG materials upon loading. The mechanism of this mode of deformation is not clear yet. We believe that precise investigations of individual AE pulses may help to further understanding of the mechanisms of strain localization in UFG materials. Detailed statistical and
spectral analysis of AE signals in both time and frequency domains is currently in progress and will be published elsewhere.

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