Extreme grain refinement by severe plastic deformation: A wealth of challenging science

Y. Estrin, A. Vinogradov

Abstract

This article presents our take on the area of bulk ultrafine-grained materials produced by severe plastic deformation (SPD). Over the last decades, research activities in this area have grown enormously and have produced interesting results, which we summarise in this concise review. This paper is intended as an introduction to the field for the “uninitiated”, while at the same time highlighting some polemic issues that may be of interest to those specialising in bulk nanomaterials produced by SPD. A brief overview of the available SPD technologies is given, along with a summary of unusual mechanical, physical and other properties achievable by SPD processing. The challenges this research is facing—some of them generic and some specific to the nanoSPD area—are identified and discussed.

Keywords: Severe plastic deformation; Ultrafine-grained materials; Modelling; Properties

1. Historical overview

Grain size can be regarded as a key microstructural factor affecting nearly all aspects of the physical and mechanical behaviour of polycrystalline metals as well as their chemical and biochemical response to the surrounding media. Hence, control over grain size has long been recognized as a way to design materials with desired properties. Most of the mentioned properties benefit greatly from grain size reduction. As the race for better materials performance is never ending, attempts to develop viable techniques for microstructure refinement continue. A possible avenue for microstructure refinement of metals is the use of severe plastic deformation (SPD)—a principle that is as old as metalworking itself. Recent essays [1–4] tell a fascinating story of the art of ancient swordmaking through SPD. The modern-day history of SPD technology has its beginnings in the seminal work by P.W. Bridgman who developed the scientific grounds and techniques for materials processing through a combination of high hydrostatic pressure and shear deformation [5,6], which today are at the core of SPD methods. Bridgman effectively introduced the defining characteristics of SPD processing in the early 1950s. In a strict sense generally accepted in the materials engineering community, an SPD process is currently defined as “any method of metal forming under an extensive hydrostatic pressure that may be used to impose a very high strain on a bulk solid without the introduction of any significant change in the overall dimensions of the sample and having the ability to produce exceptional grain refinement” [7]. In this Diamond Jubilee issue of Acta Materialia it is appropriate to mention that many of the modern ideas of theromechanical processing involved in virtually all SPD schemes were already addressed in the first volume of Acta Metallurgica in 1953. Carreker and Hibbard [8] pointed out that the yield strength of high-purity copper benefits substantially from grain refinement and this effect is more pronounced at low temperatures. They also noticed that the effect of the initial grain size vanishes at strains larger than 0.1 and for that reason the grain size has little or no influence on the strength under monotonic loading.
similar effect is well known in fatigue where the grain size of wavy-slip materials has no bearing on the fatigue limit. These observations can be associated with the vital role of the dislocation substructure, which forms during deformation (be it monotonic or cyclic), and it is the size of the substructure which determines the strength characteristics of metallic materials. Gow and Cahn [9] emphasized the significance of crystallographic texture for the deformation and recrystallization behaviour of metals and the effect of evolving texture on the resultant properties. Bell and Cahn [10] outlined many fine features of mechanical twinning, which play an important part in plastic deformation when accommodation by dislocation slip is hindered. Beck [11] highlighted the possibility of relieving the work-hardening effects by post-processing recovery. As will be seen in the following sections, these ideas have had a great impact on the development of the SPD processing and are pivotal to the modern concepts underlying these techniques. Nowadays, the subject of SPD processing is represented very prominently on the pages of Acta, as illustrated by the analysis in Ref. [4]. Its revival is due to the work of Segal et al. [12] in the Soviet Union in the mid-1970s. These authors developed the method of equal-channel angular pressing (ECAP), which later evolved into what is now the most popular SPD technique. It should be mentioned, however, that in the time between the publication of Bridgman’s studies and the reintroduction of this subject in the metal science literature, exploration of the possibilities of changing the properties of materials through combined high pressure and shear deformation went on both in the Soviet Union and in the West. This less known work has been reviewed in Ref. [13]. In particular, credit should be given to the work of N.S. Enikolopian conducted mainly on polymers.

A real appreciation for the new possibilities for improving the properties of metallic materials provided by SPD techniques came with the work of the group of Valiev [14,15], which demonstrated the relation between the enhanced strength and the extreme grain refinement imparted by SPD processing to a range of metals and alloys. The seminal work of this group, emphasizing the great potential of SPD processing with regard to property improvement through grain structure modification, has heralded what has been described as the “microstructural age” of SPD research [4]. Over the last decade, the nano-SPD community (www.nanospd.org) has grown to an impressive group of researchers, and thousands of publications on ultrafine-grained (UFG) and nanostructured materials produced by SPD have been published. It is probably not surprising that in the year of the Diamond Jubilee of this journal, the Acta Materialia Gold Medal goes to Professor Terry Langdon—one of the world leaders in the area of nanoSPD materials. A representative collection of the relevant articles on the subject can be found in the proceedings of symposia on UFG materials [16,18] and nanoSPD conferences [19,20]—the most recent ones in a series of five such forums. Further useful sources include the reviews [21,22] and special issues of Advanced Engineering Materials [23], Materials Science and Engineering A [24] and Materials Transactions [25,26].

What makes SPD processing techniques so popular is the possibility of using them to enhance the strength characteristics of conventional metallic materials in a quite spectacular way: by a factor of up to eight for pure metals such as copper and by some 30–50% for alloys [7,27]. Despite the impressive property improvement achievable with SPD techniques, their uptake by industry has been rather sluggish. However, things are now starting to change, and there is a common feeling in the nanoSPD community that major breakthroughs in terms of industry-scale applications of SPD-based technologies are imminent. We have been working in this area for more than a decade and have followed its developments closely. In this article we present our views on what has been achieved, what is possibly achievable, and what future trends are to be expected from SPD processing technologies. This article does not represent a full review of the SPD area (one could almost say the discipline of SPD, considering the firm place this group of material processing techniques has taken in literature). Rather, it is our personal take on the SPD area and an attempt to foretell its future development. Emphasis is placed on the scientifically challenging aspects of SPD, and not so much on technological issues, although some insights into the promises and limitations of SPD technologies will also be given.

2. SPD methods

Among the procedures devised for grain refinement, SPD techniques are of particular interest and are the focus of the present review. These techniques enjoy great popularity owing to their ability to produce considerable grain refinement in fully dense, bulk-scale work-pieces, thus giving promise for structural applications. The achievable grain sizes lie within the submicrometer (100–1000 nm) and nanometer (<100 nm) ranges. SPD-processed materials with such grain sizes are generally referred to as nanoSPD materials [7], although only the latter ones can be regarded as being nanostructured according to the conventional definition. Several comprehensive reviews have focused on various nanoSPD processing techniques [22,28–33]. We refer the reader to the original works for specific details and only briefly outline the general SPD methodology underlining the common features and the most important differences between the nanoSPD processes. By no means do we claim that our list of currently available manufacturing schemes is exhaustive.

After the landmark work by Bridgman mentioned above [6,34], Langford and Cohen [35] and Rack and Cohen [36] demonstrated in the 1960s that the microstructure of Fe–0.003% C subjected to high strains by wire drawing was refined to subgrain sizes in the 200–500 nm range. These microstructures could not be regarded as UFG proper in
Table 1
Schematic illustration of some basic and modern SPD techniques.

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<tr>
<th>Process</th>
<th>Schematic illustration</th>
<th>Equivalent strain</th>
<th>Ref.</th>
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<tr>
<td><strong>Basic processes</strong></td>
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<tr>
<td>(a) Equal-channel angular pressing (ECAP)</td>
<td><img src="image" alt="ECAP schematic" /></td>
<td>$e_{\text{eff}} = N \frac{1}{\sqrt{3}} \cot(\phi)$</td>
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<td></td>
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<td>$N$, the number of ECAP passes</td>
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<td>(b) High-pressure torsion (HPT)</td>
<td><img src="image" alt="HPT schematic" /></td>
<td>$e_{\text{eff}} = N \frac{1}{\sqrt{3}} \frac{r}{t}$</td>
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<td>$r$, the distance from the axis, $t$, the thickness of the sample, $N$, the number of revolutions</td>
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<td>(c) Accumulative roll bonding (ARB)</td>
<td><img src="image" alt="ARB schematic" /></td>
<td>$e_{\text{eff}} = N \frac{1}{\sqrt{3}} \ln \left( \frac{t_0}{t} \right)$</td>
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<td>$t_0$, the initial thickness of the sample, $t$, the thickness of the sample after rolling, $N$, the number of passes</td>
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<td>(d) Multi-axial forging</td>
<td><img src="image" alt="Multi-axial forging" /></td>
<td>$e_{\text{eff}} = N \frac{1}{\sqrt{3}} \ln \left( \frac{t_0}{t} \right)$</td>
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<td>Strain is non-uniform, $N$, the number of processing steps</td>
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<td>(e) Twist extrusion (TE)</td>
<td><img src="image" alt="TE schematic" /></td>
<td>$e_{\text{eff}} \approx 0.4 + 0.1 \tan \gamma$, $e_{\text{max}} \approx N \frac{1}{\sqrt{3}} \tan \gamma$</td>
<td>[61]</td>
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<td>$\gamma$ is the twist line slope, $N$ is the number of passes. Deformation is non-uniform</td>
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<td><strong>Derivative processes</strong></td>
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<td>(f) Repetitive side extrusion</td>
<td><img src="image" alt="Side extrusion" /></td>
<td>ECAP equivalent</td>
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<th>Process</th>
<th>Schematic illustration</th>
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<td>(g) Rotary-die ECAP</td>
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<td>ECAP equivalent</td>
<td>[66]</td>
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<td>(h) Cyclic extrusion–compression (CEC)</td>
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<td>$\varepsilon_{\text{eff}} = N 4 \ln \left( \frac{D}{d} \right)$</td>
<td>[75]</td>
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<td>(i) Cyclic close-die forging (CCDF)</td>
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<td>$\varepsilon_{\text{eff}} = N \frac{1}{2} \ln \left( \frac{H}{W} \right)$</td>
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<td>(k) Repetitive corrugation and straightening (RCS)</td>
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<td>$\varepsilon_{\text{eff}} = N \frac{4}{3} \ln \left( \frac{r + t}{r + 0.5 t} \right)$</td>
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<td><strong>Integrated processes</strong></td>
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<td>(l) Integrated extrusion + ECAP</td>
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<td>(m) Parallel channel ECAP (PC-ECAP)</td>
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<td>ECAP equivalent</td>
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<td><strong>Continuous processes</strong></td>
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<td>(n) ECAP- Conform</td>
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the sense of the commonly accepted definitions [7], because most of the sub-boundaries were low angle. Indeed, it is the prevalence of high-angle grain boundaries that is commonly believed to be a signature of UFG materials manufactured by SPD. This constitutes a clear demarcation line between nanoSPD materials and more conventional materials with subgrain structures produced by cold rolling or other common metal forming techniques. This distinction notwithstanding, these works opened the gates for microstructure refinement by deformation to gigantic strains.
Imparting large plastic strains to a work-piece is a challenging and technically formidable task. It requires a considerable investment in tool design, which on one hand should be durable enough to sustain repetitive high loads during material forming, and on the other hand be suitable for materials processing without causing damage to the work-piece. A distinctive feature of SPD processing, which meets these requirements, is that the high strain is imposed without any significant change in the overall dimensions of the work-piece. This is achieved due to special tool geometries that prevent free flow of the material and thereby produce a significant hydrostatic pressure. The presence of this hydrostatic pressure is a clue for achieving the high strains required for exceptional grain refinement. Many crystalline materials, including those which are brittle under normal conditions (e.g. tungsten oxide, B_2O_3 glasses and amorphous materials), gain substantial ductility under high hydrostatic pressure and can be deformed to large strains without failure. Nowadays many variants of SPD techniques, which expressly or tacitly employ this generic feature of high hydrostatic pressure, are readily available for fabrication of a great variety of UFG materials.

2.1. Principal processing schemes

**Equal-channel angular pressing (ECAP)**, less appropriately referred to as equal-channel angular extrusion (ECAE) in some publications, is at present the most highly developed SPD processing technique (Table 1a). A simple shear strain is introduced when the billet passes through the plane where the two channels meet. The cross-sectional dimensions of the billet remain unchanged, thereby permitting repetitive pressing, leading to accumulation of very large strains. For example, the equivalent (von Mises) strain, \( \varepsilon_{eq} \), introduced per pass in ECAP with a 90° angle between the channels amounts to 1.15 [37,38]. Different ECAP variants involving rotations of the billet about the pressing axis between the passes are possible, and they generally lead to different results in terms of the microstructure and texture produced. The definitions of these ECAP routes to which we refer below can be found in Refs. [14,15].

Against the backdrop of a flood of publications on ECAP processing, it is easy to forget where it all started. It is therefore timely to recall that the key advantages and fundamentals of ECAP, including the mechanics of extrusion, the derivation of the optimal process conditions involving a balance between friction, tool geometry, strain path and its efficiency for grain refinement, were formulated by V. Segal in a series of early publications [12,38–42]. He defined ECAP as “a deformation technique to impart intensive, uniform and oriented simple shear for materials processing”. He also demonstrated that ECAP is effective if (i) friction between the billet and the die walls is kept at a minimum; (ii) the angle between the channels is close to 90°; and (iii) the sharp outer corner is fully filled ensuring that the shear zone is as narrow as possible. The first requirement stimulated development of dies with reduced friction by implementing surface hardening of the channel walls, mobile walls [37,43], etc., as well as the introduction of new effective lubricants [44]. The third requirement led to the understanding of the significance of back-pressure for processing of billets with uniform microstructure and improved mechanical properties [43,45,46]. Segal showed that the performance of a die may be compromised by perceived simplicity of the die design [38]. In particular, he warned against the use of dies with a corner arc which leads to the occurrence of a widely spread fan-shaped plastic zone. This is equivalent to artificially increased friction that spreads shear and gives rise to significant heterogeneity of strain. Unfortunately, this important warning was disregarded in many later studies, which utilized a “simplified” die design with a rounded outer corner and had to pay a high price in form of substantial heterogeneity of the deformed structure. By contrast, by following Segal’s philosophy, samples with uniform microstructure throughout the billet could be fabricated [47,48]. While standard laboratory-scale ECAP rigs can handle billets with cross-sectional dimensions in the range of 10–20 mm, Segal’s ideas enabled development of industry-scale ECAP facilities for processing of billets as large as 50 × 50 mm² in cross-section and 500 mm in length [43].

**High pressure torsion (HPT)** refers to processing that evolved from Bridgman’s anvils [6], cf. Table 1b, and involves a combination of high (GPa range) pressure with torsional straining. Today this technique is appreciated by many researchers as the one that allows the most efficient grain refinement. A handicap of the method is that only small coin-shaped samples, typically 10–15 mm in diameter and 1 mm in thickness, can be processed. The readers are referred to a comprehensive review on the subject [30] for details. Because of size restrictions, the samples manufactured by HPT are used primarily for research purposes. An important issue for many SPD processing schemes, including HPT, is the non-uniformity of deformation. For instance, during HPT straining, Table 1b, the shear strain at the rotation axis should be zero, increasing linearly in the radial direction if the geometry of the work-piece does not change. This means that the material near the rotation axis of the sample should stay undeformed. This is not supported by numerous microstructural observations and microhardness measurements showing a reasonably uniform distribution of grain dimensions and microhardness, provided the compressive pressure and the number of revolutions of the anvil are sufficiently large (as in Fig. 1) [49–51]. Vorhauer and Pippan [52] explained this discrepancy by the fact that it is virtually impossible to realize an ideal HPT deformation due to the misalignment of the axes of the anvils. Alternatively, the development of a reasonably uniform strain (Fig. 2) and homogeneous microstructure was explained in terms of gradient plasticity theory coupled with the microstructurally based constitutive modelling [53]. This model will be addressed in the
following section. Axial inhomogeneities observed in an HPT-processed Zr$_3$Al intermetallic [54] were associated with softening effects related to nanostructuring.

**Accumulative roll-bonding (ARB),** Table 1c, was introduced by Saito et al. [55] as a process which was supposed to overcome major limitations of the ECAP and HPT, namely the low productivity of the former and the small work-piece size of the latter. The greatest technological advantage of ARB is that it makes use of a conventional rolling facility. A metal sheet is rolled to 50% thickness reduction. Then, the rolled sheet is cut in two and both halves are stacked together, thus restoring the original thickness of the sheet. The contact faces are degreased and wire-brushed before stacking of the sheets, which are then rolled together to half the thickness. This sequence of rolling, cutting, brushing and stacking operations is repeated so that ultimately a large strain is accumulated in the sheet. ARB was successfully applied to a wide range of materials, including commercial-purity (CP) Al, the Al–Mg alloy AA5083 and interstitial-free steel, and was also used to process Al- and Mg-based laminated structures and composites [56]. In addition, ARB can be applied for the production of metal–matrix composites by sheathing mixed powders and subjecting them to a roll-bonding process [57]. Similarly to conventional rolling, the UFG structure formed in the course of repetitive rolling is not equiaxed but rather exhibits pancake-like grains, irrespective of the type of metal or alloy processed.

**Multi-directional forging (MDF),** Table 1d, was proposed as a technique for structure refinement in the first half of the 1990s [58–60]. Multiple free-forging operations include repeated setting in three orthogonal directions. Since MDF is commonly performed in the temperature interval of 0.1–0.5T$_m$, where T$_m$ is the melting temperature, grain refinement during MDF is usually associated with dynamic recrystallization. The homogeneity of the strain produced by MDF is lower than for ECAP or HPT. However, the method can be used for microstructure refinement in rather brittle materials owing to elevated temperatures and the low specific loads on tooling involved. Furthermore, MDF was demonstrated to be a potent technique for manufacturing large-size billets with microcrystalline (UFG) structures and was successfully applied to a wide range of materials [61].

**Twist extrusion (TE),** Table 1e, is another variant of a simple shear deformation process that was introduced by Beygelzimer et al. some ten years ago [62–64]. Under TE processing, a prismatic billet is extruded through a “twist die”. While the advantage of the process is its high upscaling capacity, it suffers from essentially the same generic problem as the HPT: deformation is non-uniform, being smallest near the extrusion axis. Investigation by Orlov et al. [65] showed that this technique is slightly less effective in producing UFG structure than ECAP or HPT.

### 2.2. ‘Derivative’ SPD processes

Inspired by the success of the above “classical” SPD techniques, more “exotic” methods were developed...
recently with the aim of processing samples other than simple rod or disk stock and/or enabling a higher throughput. Some of them are illustrated in Table 1. A list of these techniques (which is admittedly not exhaustive) includes:

- repetitive side extrusion [66];
- rotary die ECAP [67];
- parallel channel ECAP [68];
- hydrostatic extrusion [69–71] and hydrostatic extrusion combined with torsion [72];
- repetitive corrugating and straightening (RCS) for processing of sheets or plates [73–75];
- constrained groove pressing [76];
- cyclic extrusion–compression (CEC) [77];
- cyclic closed-die forging (CCDF) [78];
- cone–cone method (CCM) [79];
- cryogenic rolling [80,81];
- asymmetric rolling (ASR) [82];
- continuous frictional angular extrusion (CFAE) [83,84];
- friction stir processing (FSP) [85,86];
- super short interval multi-pass rolling (SSMR) [87,88];
- severe torsion straining (STS) [89,90];
- torsion extrusion [91];
- ECAP with rotation tooling in which the conventional fixed die is replaced by rotating tools [92];
- reversed shear spinning [92];
- transverse rolling [92];
- non-equal channel angular pressing (NECAP) for plate-shaped billets [93];
- tube channel pressing [94];
-KOBO forming [95];
-high-pressure tube twisting (HPTT) for thin-walled tubes [96];
-cyclic expansion–extrusion CEE—a modified CEC process [97];
-simple shear extrusion [98,99];
vortex extrusion [100];
-helical rolling [101];
-high-pressure sliding [102].

From this list alone one can see that there are really no bounds to the imagination and resourcefulness of SPD process designers, and more and more new SPD techniques have been emerging recently. From a purist’s viewpoint, not all of them would qualify to be termed “nanoSPD” processing according to the definition in Ref. [6], but most of these techniques are cognate with the principal processes—ECAP, HPT, TE or ARB—they derive from and bear some semblance with. Most of these processes use shear deformation in conjunction with hydrostatic pressure to produce large strains. The potential benefits of these “derivative” techniques include simplified tool design, lower loads, reduced material loss, the possibility to process larger work-pieces, automated handling and/or potential continuous operation.

It is broadly recognized that strength and ductility may greatly benefit from a combination of ECAP with interme-
diate annealing and/or post-ECAP processing by conventional rolling, drawing or extrusion. The advantageous effect of post-processing was confirmed by many researchers who combined different post-ECAP techniques to further enhance strength [103–105], modify texture [106] or improve ductility by subsequent annealing [107–109]. Finally, new integrated processing schemes, which adopt features of different processes and combine them in a single-step integrated processing workflow [110–112], have recently been developed, cf. Table 11. The use of the integrated semicontinuous processing techniques may be a promising way of overcoming obstacles to uptake of SPD techniques by industry.

Among the recent developments of SPD methods one can recognize a trend to target thin products, particularly thin-walled tubes, and produce grain refinement by friction-induced shear. One of the work-piece dimensions used in such processes, namely the thickness, is much smaller than the other two dimensions. The cone–cone method [79,113] and high-pressure tube twisting [96] are in that category, as is a modified tube-twisting technique suggested in Ref. [114]. Depending on the wall thickness, grain refinement can be achieved throughout the tube wall thickness or only within near-surface regions of the wall. This method was also applied for producing bimetallic Al–Cu tubes with ultrafine grain size (as small as about 140 nm near the interface of the two metals) [115].

In a similar vein, Umemoto [116–118] made a point that conventional metal processing techniques, such as shot peening, drilling and wear [119], can be used as an effective way to create UFG structure and concomitant strengthening in near-surface regions of metals and alloys.

2.3. Continuous SPD techniques and post-processing

Many of the SPD methods presented above involve a large number of discrete steps and are not labour and cost efficient. Furthermore, they suffer from the inability to deliver sufficiently large work-pieces as required for industry-scale applications. A number of approaches to SPD processing seek to alleviate these disadvantages. In what follows, we touch upon them briefly.

Continuous forming (CONFORM), Table 1m, is a well-known process, the principles of which were first formulated by Etherington [120] with the aim of improving the efficacy of materials recycling. They were later adapted by Segal and co-workers to continuous ECAP of bulk materials [37]. These principles were implemented by Raab et al. in a rig for production of Al and Ti rods [121]. In this process, the rod is placed in a groove within a rotating shaft and is driven forward by frictional forces and then extruded through an outlet cannel of the die similarly to ECAP. A modification of this process was proposed by Saito et al. [122] as continuous shearing, Table 1o, for processing of sheets or strips.

Continuous confined strip shearing (C2S2), sometimes referred to as ECA-rolling process, Table 1p, is a
modification of the CONFORM method for processing of sheets or strips [123,124].

Continuous ECAP for sheet manufacturing was discussed by Lapovok et al. [125,126]. For the Al alloys tested it was established that just a single ECAP pass was sufficient to obtain a significant reduction of normal and in-plane anisotropy. A variant of the process is continuous equal-channel angular drawing [125,126].

Repetitive corrugating and straightening (RCS) has an obvious advantage of providing a simple modification of rolling to enable continuous SPD processing, as illustrated graphically in Table 1q [74,75].

Incremental ECAP (I-ECAP). Rosochowski and co-authors extended general knowledge of incremental metal forming operations, such as rolling, swaging or rotary forging, and adapted it to ECAP by modifying it for processing of long billets. This process was dubbed incremental ECAP (I-ECAP) [127]. The basic version of I-ECAP is shown in Table 1r. The deformation mode is simple shear, and it is uniform within the marked zone similarly to the shear deformation in “classical” ECAP. Separation of the feeding and deformation stages reduces or eliminates friction during feeding; this substantially reduces the feeding force and enables processing of very long or continuous billets.

Continuous manufacturing of bolts by ECAP. The group of Prof. Y.-T. Im at KAIST in Korea developed a method [128,129] which overcomes the discrete character of ECAP by integrating what they call “spring-loaded ECAP” in a continuous bolt manufacturing process (Table 1t). AA6016 bolts produced using this technology were shown to be superior to those manufactured in a conventional way in terms of their tensile strength and fatigue strength.

Continuous high-pressure torsion. An advanced version of the HPT technique was proposed by Edalati and Horita [130], who demonstrated its viability as a method to produce sheets 0.6-mm thick and 3 mm wide, which possess UFG structure, in a continuous fashion, cf. Table 1s. While for most structural applications upscaling of the SPD technologies is required, there may be niche applications where downscaling would be desirable. The feasibility of such downscaling was demonstrated for the ECAP process [131]. Miniaturized dies with channel diameters in the millimeter range were used to deform Al specimens and achieve grain refinement in a single pass.

An SPD-like process of an entirely different type was proposed by Estrin et al. [132]. In this “solid-state infiltration” method, solid aluminium was forced to fill a porous steel preform under high pressure in much the same way via are filled with metal in fabrication of metallic interconnects by the force-fill process in microelectronics [133]. The random paths taken by the plastically flowing Al are pretty tortuous and involve numerous kinks. Some of them may be similar to those seen in ECAP channels and induce ECAP-like localized shear zones and ensuing grain refinement. Penetration of Al into the porous steel preform is illustrated in Fig. 3.

Concluding this section we would like to note that a great variety of SPD techniques are now available. Their common features are the high hydrostatic pressure and the tool geometry permitting multipass operation to achieve ultrahigh strains. Differences are mainly related to the deformation mode, the work-piece shape, the efficacy with respect to the strain imposed per pass and the load involved. All these factors affect, to a varying extent, the resultant microstructure, the properties of the product and the upscaling capacity of the technique used. These aspects of SPD processing will be addressed in the next sections.

A great advantage of the SPD techniques is that they are based on a “top-down” approach involving grain refinement through “breaking down” the microstructure of the bulk to the submicron scale. SPD processing is thus free from problems of excessive residual porosity and contamination, which are common in nanostructured materials manufactured in a “bottom-up” fashion, e.g. by consolidation of nanopowders. Furthermore, no health hazards potentially associated with handling of nanopowders are involved in SPD processing.

As will be seen below, perhaps the most important disadvantage of SPD is that the efficiency of grain refinement drops with strain [134]. A way to overcome the problem by suppressing dynamic recovery, e.g. by using SPD processing at cryogenic temperatures, was suggested in Ref. [81]. However, the microstructures obtained in this way retain a large volume fraction of low-angle boundaries, giving rise to considerable thermal instability and coarsening of the ultrafine microstructure produced.

Given the rapid progress in the field, we are confident that new processes with higher throughput, upscaling capacity and greater cost-effectiveness will emerge, meeting
the demand for advanced high-performance structural materials in modern industries.

3. Mechanisms of grain refinement by SPD

The main aim of SPD processing, its ultimate raison d’être, is extreme grain refinement and the ensuing strengthening of the processed material. There is no longer any doubt that this is achievable with most malleable and even with many hard-to-deform materials, and innumerable experimental results documented in review articles and conference proceedings (e.g. [16–20]) are a convincing testimony to that. Despite this body of experimental evidence, the mechanisms of grain refinement, which are pivotal in designing the routes to property improvement, are far from being understood. In particular, there is no generally accepted scenario of grain fragmentation by subdivision of grains, and the underlying processes have remained a riddle for researchers to the present day.

3.1. Disclination models of grain fragmentation

Early attempts at unravelling this riddle go back to the work of Honeycombe [135], Rybin [136–137], Vladimirov and Kusov [138], Mughrabi et al. [139], Indenbom and Orlov [140] and others. This research has prepared the ground for models introducing grain fragmentation by dislocation wall formation, notably in form of rows of dislocation dipoles. These concepts have led to a description of the grain fragmentation process in terms of disclinations [141,142]. The process of grain subdivision is represented by the nucleation and propagation of incomplete disclinations, producing misorientation between the adjacent grain fragments. Models based on coupled disclination–dislocation dynamics were shown to provide a reasonably good description of the microstructures formed at large strains [142]. A recent review of disclination–dislocation dynamics simulations were being devoted to this problem. The emergence of a dislocation cell structure is one of the composite principle and detailing the evolution of the dislocation densities in the cell walls and cell interiors, including interactions between the two “phases” of the composite. While Zehetbauer’s model postulated constancy of the volume fraction of cell walls, Estrin et al. emphasized that this volume fraction must decrease during stage IV of hardening in order to account for the nearly constant hardening coefficient commonly found experimentally in stage V.

The two-dislocation density models [153,154] have become a useful platform for modelling SPD processes (cf. [155,157,158]), and we shall present them in a condensed form here. The models apply to dislocation cell-forming materials and address sufficiently large strains when a cell structure is already formed. The “primordial” stage of the deformation in which this happens is not considered. Understanding the self-organization processes leading to the emergence of a dislocation cell structure is one of the greatest unsolved problems of the dislocation theory, and numerous modelling efforts, including discrete dislocation dynamics simulations [159], are being devoted to this problem. An assumption made in the models [153,154] is that the average dislocation cell size, \( d \), scales inversely with the square root of the total dislocation density, \( \rho \):

\[
d = K / \sqrt{\rho},
\]

with the intrinsic length scale \( L \) corresponding to a characteristic length scale of the cell structure, e.g. the cell size, that determines the dislocation mean free path. Here \( k_0 \) is a constant or a slowly varying quantity and \( k_2 \) is a mechanism-dependent phenomenological parameter sensitive to strain rate and temperature. The Kocks–Mecking model has been extremely successful in providing a description of stages II and III of strain hardening. However, an adequate description of stages IV and V of strain hardening, which are predominant at large strains [146], requires a more detailed representation of the dislocation population. It involves treating dislocations in the dislocation cell walls and cell interiors as separate entities, thus introducing two distinct evolving dislocation densities. This was done by Prinz and Argon [147] and later by Nix et al. [148] who adopted the approach proposed by Mughrabi [149,150], in which the cell walls and cell interiors are treated as two distinct phases of a ‘composite’. These models were able to account for stage IV and stage V hardening. More complex models with three internal variables [151] also provided an adequate description of mechanical response at large strains and were successfully applied for metal forming simulations [152].

Estrin et al. [153,154] and Zehetbauer et al. [146,155,156] proposed constitutive models based on Mughrabi’s composite principle and detailing the evolution of the dislocation densities in the cell walls and cell interiors, including interactions between the two “phases” of the composite. While Zehetbauer’s model postulated constancy of the volume fraction of cell walls, Estrin et al. emphasized that this volume fraction must decrease during stage IV of hardening in order to account for the nearly constant hardening coefficient commonly found experimentally in stage IV.

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\[
d = K / \sqrt{\rho},
\]

where \( K \) is a proportionality constant. Eq. (2) goes back to an early work of Holt [160] in which patterning in a dislocation population was first considered. While a substantial body of evidence supports the validity of Eq. (2) for steady-state deformation, extending it to the dynamic
case of evolving dislocation structure is a major assumption, which definitely needs further substantiation.

The total dislocation density is considered to be given by the weighted sum of the dislocation densities in the cell walls (subscript “w”) and cell interiors (subscript “c”)

$$\rho = f \rho_w + (1 - f) \rho_c,$$

where $f$ denotes the volume fraction of the cell walls. Obviously, the latter quantity is linked to the cell size $d$ and the wall thickness $w$ through a relation that should account for cell morphology. As this relation is not very sensitive to the particular shape of the cells, a simple expression

$$f = \frac{d^3 - (d - w)^3}{d^3},$$

corresponding to the idealized case of cube-shaped cells is a reasonably good one. In modelling SPD processes [161,162], it was assumed that the volume fraction decreases from an initial value $f_c$ to a saturation value $f_\infty$. This approach of saturation is governed by a phenomenological equation:

$$f = f_\infty + (f_c - f_\infty) \exp(-\gamma/\gamma),$$

which is supported by experiments on Cu. The parameter $\gamma$ represents the inverse of the rate of this variation with the plastic shear strain $\gamma$. The growth of the total dislocation density $\rho$ and the concomitant decrease of the average cell size $d$ with strain suggests that a decrease of the cell volume fraction $f$, which is implied by Eq. (5) for $f_\infty < f_c$, is only possible if the cell wall thickness decreases fast enough with strain. This assumption was implicit in the original model [153,154]. “Thinning” of cell walls with the progress of straining is a reasonable notion, as “geometrically unnecessary” dislocations not contributing to misorientation between neighbouring cells will recover with strain. However, the phenomenological ansatz made in Eq. (5) can be replaced by a more physical assumption of the variation of the cell wall thickness. For instance, $w$ can be defined in terms of a characteristic length controlling the decay of the stress field of the cell wall dislocations, which should scale with $1/\sqrt{\rho_w}$. In this way, both the average cell size and the cell wall thickness would be expressed in terms of the dislocation densities.

The evolution of the dislocation ensemble is captured in a set of coupled differential equations for the dislocation densities in the cell interior and cell walls under the “Taylor”-type assumption that the shear strain is the same in both phases:

$$\frac{d\rho_w}{d\gamma} = 6b' \left(1 - f\right)^{2/3} + \sqrt{\frac{3}{b}} \frac{b' \left(1 - f\right) \sqrt{\rho_w}}{fb} - k_0 \left(\frac{\gamma}{\gamma_0}\right)^{-1/n} \rho_w,$$

$$\frac{d\rho_c}{d\gamma} = \alpha' \frac{1}{\sqrt{3}} \frac{\sqrt{\rho_w}}{b} - b' \left(1 - f\right)^{1/3} - k_0 \left(\frac{\gamma}{\gamma_0}\right)^{-1/n} \rho_c.$$

Here $b$ is the magnitude of the Burgers vector and the coefficients $\alpha'$ and $b'$ are numerical constants. The various terms in the evolution equations represent possible dislocation reactions involved. Their physical origin was explained in Refs. [153,154]. For example, the loss of cell interior dislocations to the walls where they get entrapped is represented by the first term of Eq. (6) and the second term of Eq. (7). The last terms in both equations account for the dynamic recovery. The quantity $\gamma_0$ denotes a reference shear rate. In the low-temperature regime typical for SPD processing, i.e. below half the melting temperature, the exponent $n$ in both equations can be taken to be inversely proportional to the absolute temperature $T$, while the coefficient $k_0$ can be considered constant. This corresponds to dynamic recovery being governed by dislocation cross-slip.

In Zehetbauer’s model [146,155,156], it was assumed that dynamic recovery in cell walls is controlled by dislocation climb—a non-conservative process involving vacancy diffusion. This process is characterized by $\gamma_0$, which is given by an Arrhenius equation with the activation energy representing that for self-diffusion, the exponent $n$ being a constant.

As vacancy formation is influenced by hydrostatic pressure, this approach introduces a direct effect of hydrostatic pressure on the dislocation recovery kinetics [163]. An appropriate modification of the dynamic recovery term in Eq. (7) to include the hydrostatic pressure made it possible to account for the effect of back-pressure on the ECAP processing of an Al alloy [164].

The type of model described above proved to be a useful descriptive and predictive tool for computer simulations of ECAP processing of copper [157], aluminium [161], Al alloy 6016 [164], interstitial-free steel [165], CP titanium [158] and HPT processing of copper [53,166]. The focus of these simulations was the calculation of the strength characteristics of the ECAP-processed materials. However, the model was also used for calculating the texture of copper developing as a result of ECAP processing, albeit in a very simplified way [167]. We shall return to the issue of texture below.

The model of grain fragmentation presented above is based on the idea that fine granularity is attained through dislocation cell formation with (tacitly assumed) accumulation of misorientations across the dislocation cell boundaries. In other words, gradual transformation of dislocation cell walls, or at least a large proportion of them, to high-angle grain boundaries is implied. Pantleon [168] and Estrin et al. [169] considered the increase of dislocation cell misorientations with progressing straining in terms of a probabilistic approach. In both models, preferred storage of dislocations with a certain sign of the Burgers vector, which occurs locally within a cell wall, gives rise to misorientation between the cells separated by the wall. This occurs stochastically, leading to a continual increase in the misorientation angle, which eventually saturates. The results are consistent with the observed levels of misorientation angles (several degrees) associated with the so-called “incidental dislocation boundaries” (in the terminology of the Hansen school [170–172]), but cannot account for the
occurrence of a very significant fraction of high-angle grain boundaries usually found experimentally in SPD-processed materials, which are referred to as “geometrically necessary boundaries” [170,171]. Malygin, who also considered the evolution kinetics of cell size and misorientation angle [173], associates the occurrence of the geometrically necessary boundaries with elastic bending of the crystal caused by non-uniformity of plastic deformation and the attendant local distortions in the crystal bulk or the outer shape of the specimen. Obviously, such boundaries are not accounted for by the models discussed.

Although Eq. (2), which describes cell/grain size evolution with the accumulation of the dislocation density, was introduced in a heuristic way, there are strong reasons to believe that it is a robust relation which can be used in current and future models. This can be expected based on both dimensional considerations and rudimentary modelling of dislocation pattern formation [171,174]. Nevertheless, developing a model which would provide an adequate description of the evolution of dislocation cell size and misorientation angle distributions is as much a challenge as is the development of a full theory explaining the formation of a dislocation cell pattern in the first place.

Obviously, according to Eq. (2), the cell/grain size $d$ has to saturate, asymptotically reaching a steady-state value prescribed by the steady-state values of the dislocation densities governed by Eqs. (6) and (7). (We note that the volume fraction of cell walls, $f$, also tends to saturation.) Observations tell us that even when $d$ approaches saturation, e.g. with the number of ECAP passes or the rotation angle in HPT, the misorientation angle still evolves visibly (cf. [175]). Data for a Ni single crystal processed by HPT (Fig. 4 [176]) show that the various characteristics of the microstructure, such as the mean length of small- and high-angle boundaries, saturate at rates different from that for the average cell/grain size. This is at variance with the thesis [159] that the dislocation cell size and the misorientation angle tend to saturation with the same rate and that their product is independent of strain.

It is pretty obvious that the mechanism of grain fragmentation considered cannot lead to an arbitrarily small grain size. Indeed, an estimate of the dislocation density in Eq. (2) in terms of the applied stress $\sigma = M\tau$ (where $M$ is the Taylor factor, which accounts for texture) leads to the following estimate for the smallest achievable grain size $d_s$:

$$\frac{d_s}{b} \approx K M \sigma \frac{G}{\sigma_m}.$$  (8)

Here $\sigma_m$ is the highest possible stress, $G$ is the shear modulus and $\alpha$ is a numerical constant, typically of the order of 0.5. With $K$ of the order of 10 and $M$ around 3 it is easily seen that even for $\sigma_m$ close to the theoretical strength the final grain size cannot be smaller than $d_s \sim 100b$. That is to say, the average grain size cannot attain values in the “nano range”, i.e. below 100 nm, if the assumed mechanism of grain subdivision via dislocation cell formation controls grain refinement.

There is also a further limitation on the smallest grain size achievable by this mechanism. As suggested in Ref. [177], below a certain critical value of the grain size, $d_c$, diffusive accommodation of dislocations in the walls is predominant and their storage is negligible. In this regime, no strain hardening will occur. The critical grain size is given by:

$$d_c = \left(\frac{D_{GB} b}{\gamma}\right)^{1/3},$$  (9)

where $D_{GB}$ is the grain boundary diffusivity. The critical grain size $d_c$ estimated for room temperature SPD of copper is about 250 nm. This is, indeed, consistent with the average grain size found in copper processed by ECAP at

Fig. 4. Tendency to saturation with equivalent strain for various characteristics of the microstructure of monocrystalline Ni fragmented into a dislocation cell/grain structure. The data in the right-hand side figure represent the mean lengths of cell walls (misorientation angle below 15°) and high-angle grain boundaries (misorientation angle above 15°), as well as the fraction of the high-angle grain boundaries. After Ref. [176] (reprinted with permission).
room temperature [161, 178]. As seen from Eq. (9), $d_s$ is a decreasing function of the strain rate and, through the grain boundary diffusivity, an increasing function of temperature. Clearly, during SPD-induced grain fragmentation the grain size should saturate at the level of the larger of the two critical quantities, $d_s$ and $d_c$.

In deciding what mechanisms control the saturation of the microstructure evolution, one can be guided by the above semi-intuitive arguments. However, a strict theory based on a detailed analysis of the dislocation–grain boundary interactions that could shed light on these mechanisms is still lacking. For instance, it is not quite understood at what grain size scale the grain boundaries lose their ability to effectively hinder dislocation motion and to act as storage sites where dislocation pile-ups form—a mechanism commonly accepted as the cause of the Hall–Petch effect. As dislocation-mediated plasticity is at play even in truly nanostructured materials [179], a systematic study of the relative roles of diffusion- and dislocation-controlled processes using the entire available arsenal of modelling techniques is the order of the day.

A further, relatively new, modelling approach to grain fragmentation was proposed in Ref. [180]. Grain subdivision was treated there as a result of the development of lattice curvature within an individual grain due to constraints imposed by the neighbouring grains. Specifically, it was assumed that the rotation of the crystallographic planes in a grain due to dislocation slip is impeded near the grain boundaries. Due to this retardation the lattice rotation at the periphery of a grain is smaller than in the middle part. The geometrically necessary dislocations associated with the lattice curvature thus produced were considered as the main cause of grain subdivision. Used in conjunction with the dislocation-density model [153, 154], the grain subdivision rule defined in [180] enabled the prediction of the strain-hardening behaviour of copper, along with a simulation of the evolution of the grain size distribution and texture. The model predictions were found to be in excellent agreement with experiment. This consideration of grain subdivision in texture simulations is a feature that was not implemented in earlier work, which was reviewed in Ref. [181]. With the grain subdivision model [180], the agreement between the calculated and the experimental textures obtained from ECAP processing of copper was shown to be improved. The method was further advanced in a very recent work on SPD processing of Al [182].

In this relatively short overview, we cannot address all possible mechanisms relevant for SPD and discuss all models proposed to account for them. Some complementary material can be found in earlier reviews [183, 184]. What we would like to mention, however, is a radically different pathway to grain refinement, based on the work by the group of Ke Lu at the Institute of Metal Research in Shenyang, which was summarized in Ref. [185]. It is based on dynamic plastic deformation (DPD), which involves high strain rates, often in combination with low deformation temperature. Both strain-rate and temperature effects are captured in the magnitude of the Zener–Hollomon parameter $Z$ [186]. For the case of copper, the authors demonstrated that deformation-induced grain refinement is favoured by large values of $Z$. Thus, the mean transverse grain size was shown to decrease from 320 to 66 nm when $\ln Z$ was raised from 22 to 66. The smallest grain size values obtained are substantially smaller than $d_c$—the lower bound of the grain size for the case of gradual transformation of the dislocation cell structure to a new, ultrafine grain structure. This implies that an entirely different mechanism of grain refinement operates at large $Z$-values. It was concluded in Ref. [185] that this mechanism involves formation of nanotwin bundles, which transform to nanograins by fragmentation of twin/matrix lamellae due to interaction of twin boundaries with dislocations or shear banding. Nanotwinning was also shown to be a determining factor in the inner structure enabling a favourable combination of strength and ductility of copper processed by low-temperature ECAP followed by cryodrawing and cryorolling [187] and SUS316L austenitic stainless steel with low stacking fault energy [188, 189].

Grain subdivision associated with nanotwinning, producing a “dynamic Hall–Petch effect” under high $Z$-value conditions possible for materials with a pronounced propensity for twinning, is not universal. However, it can be conjectured that quite generally more “violent” SPD deformation characterized by a large $Z$-value would promote formation of a finer grain structure and a high strength of the processed material. Indeed, suppression of dynamic recovery for increased $Z$-values [190] would promote the formation of finer granularity according to the scenario of a “peaceful” transformation of dislocation cell structure to the grain structure. In extreme cases, for most energetic DPD conditions with very high $Z$-values, one probably cannot rule out situations when severe local distortions (or even amorphization) of the crystal lattice may induce nanocrystallization. The possibility of deformation-driven amorphization was recently discussed in a stimulating and thought-provoking paper by Raabe et al. [191]. We realize, of course, that at this stage the amorphization/ nanocrystallization scenario of nanostructuring by high-$Z$ SPD is somewhat speculative. It would take both modelling and experimentation to verify or disprove its feasibility—another formidable task to work towards. It is interesting to note that amorphization of the intermetallic compound $\text{Zr}_3\text{Al}$ by repeated cold rolling with subsequent nanocrystallization was reported recently [192].

4. Properties of UFG materials produced by SPD

4.1. Strength and ductility and their impact on other mechanical properties

Investigations of the behaviour of UFG materials have been greatly motivated by the expectations that they may possess unique properties as well as by the desire to understand the fundamental mechanisms underlying the specific
<table>
<thead>
<tr>
<th>Material Description</th>
<th>Processing</th>
<th>$\sigma_{0.2}$ (MPa)</th>
<th>$\sigma_{UTS}$ (MPa)</th>
<th>$\delta$ (%)</th>
<th>$\sigma_g$ (MPa)</th>
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<td>H18</td>
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<td>25</td>
<td>350***</td>
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<td>[394] ECAP 4Bc50–400°C, F400–300°C, D,</td>
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<td></td>
<td>AA350°C 6 h</td>
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(continued on next page)
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<tr>
<th>Material</th>
<th>Processing</th>
<th>$\sigma_{0.2}$ (MPa)</th>
<th>$\sigma_{UTS}$ (MPa)</th>
<th>$\delta$ (%)</th>
<th>$\sigma_{0.6}$ (MPa)</th>
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<tr>
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<td>ECAP, A 480 °C 72 h</td>
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<td>733</td>
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<td>RSE 10</td>
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<tr>
<td>SUS 316L stainless steel</td>
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<td>560</td>
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<td>210</td>
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<tr>
<td></td>
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<td>900</td>
<td>30</td>
<td>360</td>
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<tr>
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<td>420</td>
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<tr>
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<td>ECAP 3Bc 150 °C</td>
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<td>1340</td>
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<td>ECAP 4Bc 150 °C</td>
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<td>1560</td>
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</table>
properties associated with extreme grain refinement. In the present section we review the state-of-the-art of both aspects of research.

Strength and ductility are primary grain-size-dependent characteristics of a material, which determine virtually all facets of its mechanical response. We shall, therefore, first address these properties, especially as they have been repeatedly claimed in literature as the ones that benefit most from grain refinement. Fatigue resistance and the toughness are the most important “secondary” properties, which are directly governed by strength and ductility. The ideal structural material should combine high strength with sufficient ductility, along with high fracture toughness. However, high strength and good ductility are often mutually exclusive, and improving both at the same time is a very challenging task. A strategy for strength improvement follows straightforwardly from the Hall–Petch relation between the yield stress $\sigma_y$ and the grain size $d$:

$$\sigma_y = \sigma_0 + K_H P d^{-1/2}. \quad (10)$$

Here $\sigma_0$ is the so-called friction stress and $K_{HP}$ is a constant for a given material. Grain refinement has long been used as an efficient tool to improve both strength and toughness of steels simultaneously. (This was the basis of the ancient art of swordsmaking mentioned in Section 1.) It should be noted that due to the additive nature by which the various mechanisms contribute to strength, pure materials and dilute solutions, which are most responsive to grain refinement, have a great potential for replacing some conventional alloys. Replacing titanium alloys with leaner CP Ti is one example that will be discussed below. Starting from the early studies in SPD [21,37,193–196], a substantial increase in strength (say, by a factor of 3–8) over that of the well-annealed or conventionally manufactured samples was reported repeatedly. This is seen in Table 2, which displays a list of currently available experimental data representa-

go of different groups of pure metals, as well as of some model and commercial alloys.

Despite the broad diversity of structural states and processing schedules used in SPD, cf. Table 1, the common trends seem to be clear: a spectacular enhancement of strength upon SPD processing concurs with a loss of ductility. This is vividly illustrated in Fig. 5, where the improvement in strength with the number of ECAP passes is shown. The main reason for this loss of ductility is a combination of high flow stress and low strain-hardening capability of SPD-processed materials. The Considère criterion signifying the onset of necking at the point where the strain-hardening coefficient drops below the value of the flow stress (see Eq. (11) below) is therefore more readily fulfilled in materials modified by SPD. Nevertheless, we note that the tensile ductility of materials processed by SPD is actually higher than that of the nanostructured materials produced, for example, by cryomilling [197]. In some cases the ductility of the SPD-processed materials may even exceed that of their conventionally strain-hardened analogues. Thus, enhancement of ductility was demonstrated for ECAP-processed CP Al, as well as for ARB-processed UFG Al and AA6016 [198,199]. In reviewing the data available for Al alloys, Markushev and Vinogradov [200] came to the conclusion that no improvement in ductility was found in non-age-hardenable Al–Mg alloys, such as AA5056 [201]. The situation is quite different with the age-hardenable Al alloys, which were found to be most responsive to SPD in terms of structure refinement, strength enhancement and fatigue life and ductility improvement [27,201–205]. For example, Roven et al. [206] reported a considerable (25–35%) increase in the ultimate tensile strength, $\sigma_{UTS}$, in various Al alloys of the 6xxx series as compared with their conventional T6-heat treated analogues. Not only was this strength increase achieved without compromising ductility, but, in fact, tensile

---

Table 2 (continued)

<table>
<thead>
<tr>
<th>Material</th>
<th>Processing</th>
<th>$\sigma_{0.2}$ (MPa)</th>
<th>$\sigma_{UTS}$ (MPa)</th>
<th>$\delta$ (MPa)</th>
<th>$\sigma_{0}$ (MPa)</th>
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<td>0Kh18N10T stainless steel</td>
<td>HR, Q1050 °C</td>
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<td>623</td>
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<td>990</td>
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<td>1121</td>
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</table>

$d$, Grain size; $\sigma_{0.2}$, conventional yield stress; $\sigma_{UTS}$, ultimate tensile strength; $\delta$, elongation at break; $\sigma_{0}$, endurance limit (on the basis of $10^7$ symmetric push–pull cycles, and stress ratio $R = -1$, if not otherwise specified); processing temperature is shown where it differs from ambient; O, as received condition, no SPD processing; CR, cold rolling; HR, hot rolling; F, forging; D, drawing; MF, multistep forging; S, solution treatment; Q, quenching; A, annealing; AG, ageing; NA, natural ageing; BP, back pressure.

*** Data obtained from the total strain amplitude control test at about $10^6$ cycles to failure.

$^*$ $R = 0.05$.

$^*$ $R = 0$.

$^*$ Rotation-bending test.

† Based on $5 \times 10^5$ cycles to failure.

†† Data obtained from the total strain control test at about $2 \times 10^5$ cycles to failure.
elongation to rupture increased by a factor of 1.5–2. Similarly, it was reported that the influence of SPD on the room temperature ductility of Al alloys may be ambivalent [200].

It is evident that the uniform elongation does not commonly improve as a result of SPD processing; however, the material’s resistance to localized plastic flow in the post-necking regime can increase remarkably. This is exemplified by numerous observations, e.g. on Al alloy 6061 [207], Ti [208] and Fe–36Ni Invar [209], cf. also Figs. 5 and 6 for SUS 316L steel and copper, respectively.

Enhancement of both strength and ductility is, however, tricky and is not common. Some successful examples, which are exceptions rather than the rule, have, however, been reported. These include the results on Ti [210], Cu and Cu–Al alloy [187,211,212], Cu–Zn [213], Al–Mg–Sc [214] and Al–Mg–Si [215]. In all these examples, success was based on a savvy choice of chemical composition and the specific processing schedules combining the above principles. For instance, Zhao et al. [213] developed a multistep processing schedule involving ECAP followed by cryodrawing and cryorolling. They demonstrated a tremendous improvement of strength coupled with enhanced ductility (with uniform tensile elongation for Cu amounting to 9–10%, Table 2). However, the authors of this impressive report used sub-size specimens of 0.1 mm thickness. They suggested that thicker samples would exhibit even higher ductility, but we are not aware of any published result confirming the ductility being the same or higher for conventional size specimens. The problem of small-size specimens, which are commonly prepared after HPT or other kinds of SPD processing, has been addressed to some extent by Zhao et al. [216,217], who demonstrated that tests without appropriate strain measurements across the gauge length conducted on sub-size specimens return considerably overestimated elongations to failure.

Another strategy for achieving a simultaneous increase in ductility and strength can be borrowed from the classical “coarse-grain” world: it has been recognized in conventional metallurgy that improved ductility can be associated with “delayed necking”. This can be achieved by activating mechanisms of deformation other than dislocation-based ones, such as phase transformations or twinning, when
dislocations are blocked and cannot accommodate imposed plastic strains effectively. These well-known mechanisms, widely utilized in steels, are referred to as transformation-induced plasticity (TRIP) [218] and twinning induced plasticity (TWIP) [219]. The formation of a tensile neck is accompanied by a concentration of stress and an increase in stress triaxiality at the neck [220]. As a result of the increased triaxiality and raised stress, the probability of martensite nucleation increases in austenitic TRIP steels [221]. A local phase transformation at sites with high stress concentrations leads to intensive local hardening and blunting of the crack tip. Therefore enhanced uniform elongation is achieved because of retardation of local necking. So far the efficacy of the TRIP effect in metastable elongation is achieved because of retardation of local necking. The authors 

Of course, grain refinement alone is not sufficient for achieving enhanced strength and improved fatigue performance, particularly for hcp-Mg-based alloys where the texture produced by ECAP, or a “derivative” thereof, is not favourable for strength [226,227]. Indeed, under ECAP with the commonly used 90° angle between the entrance and the exit channels the texture is such that the basal planes in the majority of the grains are predominantly oriented at 45° to the pressing direction [226]. In subsequent tensile tests with the tensile direction aligned with or close to the pressing direction, basal slip is activated relatively easily. The associated texture-related reduction in the yield strength acts against the Hall–Petch-type strength enhancement due to grain refinement, while ductility is improved, as was demonstrated for Mg alloy AZ31 [234].

An impressive ductility enhancement was observed for Mg alloys processed by ECAP [226–228], which is believed to be due to the specific texture that develops under simple shear in hexagonal close-packed (hcp) materials. Record tensile elongations were reached in ECAP-processed Mg alloys ZK60 [229–232] in the superplastic deformation regime. An exceptionally good combination of tensile strength and ductility was also achieved with titanium processed by ECAP [233].
estimated the recovery times for the grain interior and the grain boundaries, which are substantially different. They concluded that in UFG steel, unlike in coarse-grained ones, the grain interior dislocations reach the grain boundaries where they recover much faster than within the grains.

The ensuing reduction of the dislocation density inside the grains in the UFG material reduces strain hardening in comparison with the case of large grain size.

The second concept exploits the mechanistic approach on the basis of the Considère criterion:

\[
\frac{d\sigma}{d\varepsilon} \leq \sigma, \tag{11}
\]

for the onset of macroscopic instability associated with necking in terms of the true stress \(\sigma\) and true strain \(\varepsilon\). As the reduction of the grain size down to the sub-micron or nanoscale range leads to a pronounced increase in the flow stress with a simultaneous drop in the capacity of the material to strain harden, the necking criterion given by Eq. (11) is met at small tensile strains, which means low uniform elongation. It is easy to recognize through a simple exercise that both concepts are essentially the same. Eq. (1) is integrated analytically with the initial condition \(\rho(0) = \rho_0\) and yields the dislocation density \(\rho(\varepsilon)\) as a function of strain. The flow stress \(\sigma\) is linear in the square root of \(\rho\), as prescribed by the Taylor relation, which holds almost regardless of the particular arrangement of dislocations for a wide range of dislocation mechanisms:

\[
\sigma(\varepsilon) = \rho_0 + 6\pi G b \sqrt{\rho(\varepsilon)}. \tag{12}
\]

Solving Eq. (1) (for simplicity, for a constant \(L\)) one gets:

\[
\sigma(\varepsilon) = \sigma_0 + 6\pi G b \sqrt{k_0 k_2 L (1 - e^{-k_2 \varepsilon}) + \rho_0 e^{-k_2 \varepsilon}}. \tag{13}
\]

Using the necking criterion expressed by Eq. (11), the critical strain \(\varepsilon_c\) at which the strain hardening rate has dropped to the level of the applied stress is obtained as:

\[
\varepsilon_c = \frac{1}{k_2} \ln \left[ k_2 b L \left( \frac{k_0}{k_0} - k_2 \rho_0 \right) \left( 1 + \frac{2}{k_0} \right) / 2k_0 \right]. \tag{14}
\]

It is obvious from this equation that \(\varepsilon_c\) is reduced when the dynamic recovery rate controlled by the coefficient \(k_2\) increases. The parameter \(k_2\) is strain rate and temperature dependent. In the formulation by Essmann and Mughrabi [149,236] this phenomenological coefficient is associated with the dislocation annihilation length. In the Kocks–Mecking model it is related to dislocation cross-slip or climb probability [144,145]. This very simple reasoning can be extended in various ways, particularly by replacing the Considère criterion (11) with Hart’s criterion, which includes the effect of the strain-rate sensitivity of the flow stress [237,238]. In essence, however, Eq. (14) provides a formal footing for the popular speculative argument about the vital role of recovery and dislocation annihilation in reduced ductility of UFG metals and alloys. Besides, in the low strain limit \(k_2 \varepsilon < 1\) when steep hardening is observed, cf. Fig. 5a (stainless steel) and Fig. 6b (copper), Eq. (13) can be simplified further. Routine manipulation of Eqs. (13) and (14) leads to a simple functional relation between the proof stress \(\sigma_{0.2}\) (for \(\varepsilon = 0.002\)) and the uniform elongation \(\varepsilon_c\) in the form of \(\sigma_{0.2} \propto \frac{k_0}{k_2} \exp \left( \frac{-k_2}{k_0} \right)\), which is in general agreement with the results presented in Ref. [239]. It is also in accord with Fig. 7, showing an exponential decrease of \(\sigma_{0.2}\) with \(\varepsilon_c\) for Cu specimens manufactured in different ways. The above derivation was based on the assumption that dislocation-mediated mechanisms of plasticity remain predominant in the UFG range. Thus, most of the salient features of strength and plasticity of UFG materials, including their response to cyclic loading, see below, are captured well by the dislocation-based model.

Indeed, one can see that specimens deformed by one ECAP pass or rolled to the same equivalent strain exhibit similar hardening behaviour and nearly the same critical strain for necking, \(\varepsilon_c\), with just slightly different \(k_2\) values, close to 90 ± 6. The second path by route A gives rise to extension and thinning of grains, leading to a reduction in the dislocation mean free path. The ensuing rapid hardening raises the \(k_2\) value to 280 ± 20. A drop in uniform elongation is recorded. When further straining to four passes reduces the \(k_2\) value effectively to about 120, the uniform elongation rises again, as predicted by Eq. (14).

Based on the outcomes of the Considère analysis discussed above, Höppel et al. [240,241], and then Wang et al. [239] suggested that a bimodal grain size distribution, with micrometer-sized grains embedded in a sea of nanocrystalline and ultrafine (≤300 nm) grains, is conducive for good ductility. In a somewhat simplistic fashion it was claimed that the nanoscale fraction of the grain population was responsible for high strength, while the coarse grains contributed to ductility by promoting the strain hardening necessary to stabilize the tensile deformation against strain localizations. Improved tensile ductility (up

Fig. 7. Experimental data illustrating the relation between the yield strength and the uniform elongation in coarse grained (CG) and ultrafine-grained (UFG) copper including the results for the samples with a bimodal structure produced by annealing for 1, 3 and 10 min at 200 °C after ECAP (four B shear passes). Data are compiled from Refs. [178,239,248–251].
to 65% elongation to failure, and 30% uniform elongation) in copper produced by cold rolling at liquid nitrogen temperature, followed by an annealing treatment at a moderate temperature, which again led to a bimodal grain size distribution, was reported [239], cf. Fig. 7. A positive effect of the bimodal structure on tensile ductility was found in several investigations, although in most cases the high strength was sacrificed to a degree. It is also backed by results of numerical simulations of the deformation behaviour of elastic–viscoplastic materials [242] (see also Ref. [243]). The numerical results show the impact the variance of the grain size distribution has on the mechanical response of a material with a bimodal grain size distribution. Not surprisingly, the importance of the coarsest grains within the microstructure due to their lowest yield strength is clearly seen. This provides general guidelines for developing fine-grained materials with bimodal grain size distributions, suggesting that the two “modes” should be relatively close to each other. This result is in line with the recommendations of Wang et al. [239]. After their publication, many researchers advocated the advantages of using UFG materials with a bimodal grain size distribution for obtaining strong and ductility. Although some improvement of ductility cannot be denied [244], we believe that the magnitude of the effect does not meet the high expectations raised, and controversy still exists. For example, Mughribi et al. [107,245] showed that only a marginal improvement of the low-cycle fatigue (LCF) performance was achieved in UFG α-brass with a bimodal grain size distribution, despite the systematic way in which the parameters of the annealing treatment were varied. The original data reported in Ref. [239], as well as a compilation of new data, are presented in Fig. 7. This indicates that, contrary to the results of Wang et al. [236], the samples with bimodal structures still follow the common trend and this kind of structure does not necessarily result in a substantial increase in ductility. While bimodal structures were found in Mg alloys whose superplastic properties were boosted by ECAP [229,232], the role of bimodality in superplasticity was questioned in Refs. [230,246]. The idea of structural bimodality may even be counterproductive for high-cycle fatigue (HCF) properties, which are known to benefit strongly from a uniform grain size distribution [247]. It can be concluded that the bimodality recipe cannot generally be regarded as a panacea for the low-ductility malaise of SPD-processed materials.

In some investigations the high ductility was attributed to pre-existing growth twins in the nanostructured Cu [252]. The positive effect of twinned structure on ductility and toughness is well documented [253–255]. It may be associated with the shorter mean free path of dislocations in such structures, giving rise to a higher strain-hardening capacity of the material. If twinning occurs in situ, during the deformation, a “dynamic Hall–Petch” effect, as mentioned above, can lead to a continual decrease in the dislocation mean free path and the attendant delay of necking.

### 4.2. Fatigue

For prospective engineering applications of UFG materials, their cyclic properties need to be considered along with the strength and ductility under monotonic loading. Improvement of fatigue properties is both very important and challenging. Whereas strength under monotonic loading obeys the Hall–Petch relation extended to sub-micron grain sizes, the 150-year history of fatigue studies has taught us that the cyclic stress–strain behaviour of face-centred cubic and body-centred cubic metals with conventional grain size does not exhibit a strong grain-size dependence (see a review by Mughribi [256]). These properties were extensively studied in relation to UFG metals and several comprehensive reviews cover this topic specifically [257–259]. So far, most of the experimental results related to fatigue of UFG metals were obtained on specimens produced by ECAP in combination with other thermomechanical treatments. Analysis of the currently available body of experimental data shows that the fatigue behaviour of UFG metals follows common trends determined by the fatigue life approach based on the interplay between the strength and ductility. It can be conjectured that without loss of generality similar principles can be expected to apply for other processing schedules. The results depend on how they manage strength and ductility, as well as the key microstructural factor, uniformity of strain, they produce.

The LCF and HCF regimes are conventionally distinguished on the basis of the applied strain amplitude [257,260,261]. HCF testing corresponds to probing a material’s resistance to crack initiation, whereas LCF testing corresponds to assessing the material’s tolerance to defects in a regime when fatigue life is controlled by crack propagation. Combining these two regimes, the total strain range \( \Delta \epsilon_t \) is considered as a sum of two additive components: the elastic, \( \Delta \epsilon_{\text{el}} \) and the plastic, \( \Delta \epsilon_{\text{pl}} \): \( \Delta \epsilon_t = \Delta \epsilon_{\text{el}} + \Delta \epsilon_{\text{pl}} \). An empirical formula relates the total fatigue life represented by the number of cycles to failure, \( N_f \), to the strain amplitude, \( \Delta \epsilon_t \), as follows:

\[
\frac{\Delta \epsilon_t}{2} = \frac{\sigma'_{\text{f}}}{E} (2N_f)^{b'} + \epsilon'_{\text{f}} (2N_f)^{c'}. \tag{15}
\]

Here the first and the second terms on the right-hand side correspond to the elastic and the plastic components of the total strain, respectively; \( E \) is Young’s modulus, \( \sigma'_{\text{f}} \) is the fatigue strength (which is believed to be related to the yield stress or the ultimate tensile strength of the material), \( b' \) is the Basquin exponent, \( \epsilon'_{\text{f}} \) is the fatigue ductility coefficient and \( c' \) is the fatigue ductility exponent, also known as the Coffin–Manson exponent. Hence, the fatigue life under a given total strain amplitude is expressed in terms of four material parameters: \( c' \), \( \sigma'_{\text{f}} \), \( \epsilon'_{\text{f}} \) and \( b' \). Although direct equivalence of the parameters \( \epsilon'_{\text{f}} \) and \( \sigma'_{\text{f}} \) to the tensile ductility and the ultimate tensile strength, \( \sigma_{\text{UTS}} \), respectively, is rarely observed in experiment, a correlation between these pairs of quantities does often exist. At large strain
amplitudes corresponding to short fatigue lives, the plastic strain component is prevalent in the total applied strain and the fatigue life is determined primarily by ductility. At long fatigue lives, the elastic strain amplitude is more significant than the plastic one, and fatigue life is dictated by the fracture strength, so that the endurance limit increases with tensile strength [209,247,262]. This trend is clearly seen in Fig. 7. As already discussed, in most cases the highest strength levels are achieved at the expense of ductility (cf. Table 2). Process design aimed at enhancing fatigue properties is hindered by the fact that no commonly accepted physically based models capable of explaining the physical origin and predicting the values of the four parameters, \(c^0\), \(\sigma^0\), \(\varepsilon^0\), and \(b^0\), in Eq. (15) are available to date even for structurally simple pure polycrystals.

Based on the foregoing consideration, a traditional fatigue improvement strategy relies on the following correlation: the higher the strength under monotonic loading, the higher is the endurance (fatigue) limit. This is certainly not a firm “law”, and the effectiveness of the strategy for improving fatigue properties by raising the ultimate tensile strength depends on the material and the grain refinement process used. The most successful example of the use of this strategy in combination with SPD techniques was demonstrated by the group of R.Z. Valiev for Ti[208,210,263–267]. Starting off from ordinary ECAP processing of CP Ti, this group designed increasingly sophisticated strain-hardening schedules involving cold rolling, forging and/or drawing to “convert” the residual ductility after ECAP into strength via further grain refinement and/or dislocation hardening. While a material’s performance under monotonic loading is improved considerably with the grain size reduction by SPD, for many metals and alloys the HCF properties, particularly the endurance limit, are not enhanced to the same extent [109,201–203] (cf. Table 2).

More troublesome are the results for LCF: regardless of the SPD technique used to produce UFG and nanostructured metals, they are consistently inferior in their ability to sustain cyclic loads at fairly large imposed plastic strains. In line with the fatigue life approach based on Eq. (15), this is rationalized in terms of lower ductility, susceptibility to strain localization in shear bands and greater availability of grain boundaries in orientations favourable for intergranular crack propagation in a material with a finer grain structure.

In some cases, however, it turns out to be possible to improve both HCF and LCF performance. For example, it was demonstrated [224,225] that by combining ECAP of several dilute Cu–Cr–Zr alloys with subsequent optimized ageing it is possible to fabricate UFG materials with superior multifunctional properties, including excellent strength and very high endurance limit exceeding by far those for pure copper. Even more strikingly, the endurance limit was higher than that of a representative group of highly alloyed Cu (Fig. 8). The significant improvement of tensile strength and HCF strength observed was achieved without sacrificing ductility and LCF properties. Similarly, an improvement of monotonic and fatigue strength was reported by Xu et al. [268] for Cu–0.69Cr–0.05Fe–0.02Ni, albeit with a markedly lower fatigue limit.

Since the LCF performance relies on the resistance to flaws either induced during processing or created during testing, the crack growth resistance was found to follow the common trends for materials exhibiting increased strength and reduced ductility. The fatigue threshold is usually lower and the fatigue crack growth rate is considerably higher in UFG materials as compared to their coarse-grained counterparts (Fig. 9) [201,204,257,262,269–271].

Many discussions in the literature are concerned with the particular role of grain boundaries in the properties
of UFG materials [21]. Obviously, the interfacial energy and higher diffusivity of grain boundaries (see the next section) cannot be disregarded for many phenomena. In fatigue of SPD metals, for instance, grain boundaries play a significant role [251,272]. On one hand, the fine-grained structure usually gives rise to longer fatigue life—at least under stress-controlled cycling—than the coarse-grained one. On the other hand, the grain boundaries appear to contribute to the relatively low stability of the UFG structure and the tendency toward recovery and grain coarsening during cycling [241] and fatigue crack growth (cf. Fig. 9) [269]. Furthermore, grain boundaries are believed to play a role in the frequently observed shear banding as well as crack initiation and propagation [251,257,262,269,273–276]. Therefore grain boundaries arguably represent the most critical structural element.

Despite the complexity of the nanocrystalline structures, the fatigue behaviour of UFG metals can be more easily described than that of ordinary polycrystals and single crystals. The main reason for this simplification is the lack of dislocation patterning within the UFG structures since the characteristic dimensions of the major structural elements—grains or cells—are smaller than the characteristic length scale of dislocation structures that self-organize during cyclic loading would induce. Vinogradov et al. [109,208] suggested that in light of this argument it is sensible to describe the shape of a stable hysteresis loop and the cyclic stress–strain curve in terms of the one-internal variable approach and apply Eq. (1). It was assumed that the mobile dislocations generated at a grain boundary pass through the grain and disappear at the opposite grain boundary, i.e. the grain boundaries act as effective sources and sinks for dislocations and the grain (cell) size determines the dislocation mean free path scale. Because TEM observations do not reveal any substantial differences between the initial and the post-fatigue structures, at least in some UFG metals, it is plausible that in these cases dislocations are not accumulated inside the fine grains during cycling. The model was tested on UFG Al–Mg alloy AA5056 [109,208] and CP grade 2 titanium [109,208] after ECAP, and it showed a surprisingly good agreement with experiment. Later the same model was successfully applied by Klemm [277] for UFG nickel after different processing schedules (see also Ref. [258]). Both groups of researchers arrived at essentially the same conclusions and obtained very reasonable values of the slip distance \( L \) (of the order of the grain size) entering Eq. (1) explicitly, and the annihilation distance \( y \) which is implicitly hidden in \( k_2 \). As an example, a typical ascending part of the stable hysteresis loop of CP (grade 4) titanium in three different structural states created under different SPD processing schedules is plotted in Fig. 10a. A good agreement between the model fit and the experimental data is seen (cf. [109,208]).

Furthermore, the model applies reasonably well to severely deformed copper single crystal of \{110\} initial crystallographic orientation which was pressed through a 90° ECAP die [48] (cf. [278]) and then subjected to push-pull cyclic loading. It was demonstrated that no high-angle grain boundaries formed during a single ECAP pass; rather, a well-developed cell structure was observed [279,280] in line with ideas of microstructure formation discussed in Section 3.2 and with early experimental observations of fragmented structures formed during plastic deformation [136,137]. A very large strengthening effect comparable with that in polycrystals of the same purity deformed under the same conditions was achieved entirely due to dislocation storage in cell walls that gave rise to low misorientation angles (“incidental” boundaries). Although “old” grain boundaries had no effect on the monotonic and cyclic strength and ductility, the \( \sigma_{0.2}, \sigma_{UTS} \) and \( \sigma_f \) values were quite high: 260, 275 and 105 MPa, respectively. Such dislocation ensembles are not stable thermally or mechanically and tend to recover during cyclic loading. This recovery is a root cause of a well-known cyclic softening, i.e. the reduction of the cyclic stress amplitude with the number of cycles (Fig. 10b). The model [109,208] correctly captures this feature as well.

4.3. Creep behaviour

Reports on the creep behaviour of UFG materials manufactured by SPD are still relatively scarce. Sklenička et al. [282–284] evaluated the roles of different factors affecting the creep performance of pure aluminium, pure copper and the binary Al–0.2 wt.% Sc alloy processed by ECAP. It was found that the creep behaviour of ECAP materials depends strongly on the number of passes, a decrease in the creep resistance being associated with each successive pass. This may be attributed to a number of factors, including microstructural changes, homogenization of the
microstructure and the microtexture and nanoporosity induced by ECAP.

High-purity aluminium processed by ECAP was tested under creep conditions at 200 °C in Ref. [285]. The results showed conventional power-law creep with a stress exponent of $n = 5$, which is consistent with intragranular dislocation processes involving glide and climb of dislocations. The results suggested that diffusion creep was not important in these tests because the ultrafine grains produced by ECAP were not stable at the test temperature. (In the opposite case of a material whose extremely fine grain structure persists under service conditions and creep is controlled by diffusion, e.g. by the Nabarro–Herring or Coble mechanism, its use is obviously contraindicated.) There are reported cases when SPD processing leads to improved creep resistance. Thus, binary Cu–0.02Zr alloy after ECAP exhibited a significantly better creep resistance than its coarse-grained counterpart in the homogenized or the cold-rolled condition. By contrast, the formation of meso-

Fig. 10. Typical ascending part of the stable hysteresis loop of (a) grade 4 titanium in two different structural states: ECAP followed by hot rolling (ECAP + HR), and ECAP followed by hot rolling and annealing for 30 min at 500 °C (ECAP + HT + A500 C 30 min) [281]; and (b) copper single crystal after one ECAP pass—the three curves correspond to the cycle numbers 40, 70 and 100, showing reduction in the stress amplitude due to continual cyclic softening; curve fits by Eq. (13) are shown by thin lines.

microstructure and the microtexture and nanoporosity induced by ECAP.

4.4. Properties other than mechanical

4.4.1. Thermal stability

Improving several properties of a material at the same time to provide it with the often-desired multifunctionality is a very challenging task materials science is facing quite generally. Strengthening often involves considerable trade-offs, as other material properties may be compromised. Since strengthening is commonly a main goal and the usual outcome of SPD-induced grain refinement, it can be expected that this occurs at the expense of some other properties. For instance, beside tensile ductility and fatigue behaviour discussed above, thermal stability, electrical conductivity and corrosion resistance are among the most important properties that may be at risk of being sacrificed. Depending on the material and the targeted application, a full portrait of the properties after SPD processing needs to be obtained. Unfortunately, this has rarely been done [286]. As is typical for cold-worked materials in general, thermal stability is an Achilles heel of many SPD-treated materials. For example, SPD-manufactured pure oxygen-free copper shows relatively poor thermal stability [287,288] (even compared to copper cold-rolled to the same strain [289]). It tends to recover during storage even at room temperature due to annihilation of excess dislocations accumulated during severe straining [250] (Fig. 11a). It is seen that the greater the number of ECAP passes, the faster is the rate of recovery. No significant change of microstructure is typically observed in SPD-manufactured copper up to 120–150 °C, while in the temperature range between 150 and 250 °C recovery followed by recrystallization and abnormal grain growth takes place, as seen in the micrographs in the insets in Fig. 11b. One can follow the transformation of UFG structure into a bimodal one after annealing at 200 °C for 10 min, which then evolves to a fully recrystallized coarse-grained structure at higher temperatures. Of course, the temperature signifying loss of stability depends on the purity of copper and the amount of the imposed strain.

Strategies towards microstructure stabilization and enhancement of multifunctional properties of SPD materials align well with the general philosophy of materials design based on a balance between the basic strengthening
mechanisms—grain refinement, strain hardening, solid-solution hardening and precipitation hardening—and stress relief or recovery. As applied to UFG metals, the following measures have proven to be effective for this purpose.

(a) Post-process annealing below recrystallization temperature to relieve internal stresses increases the work-hardening capacity, thereby improving the overall ductility of cold-worked materials [107,109,235].

(b) Avoiding materials that exhibit wavy slip. Thus, titanium with its hcp crystal lattice shows reasonably high thermal and microstructural stability under cyclic loading, retaining its UFG microstructure up to annealing temperatures of 450 °C [290] and exhibiting no cyclic softening during LCF [208]; see also Ref. [291] for ECAP-processed iron.

(c) Stabilization by solutes preventing grain coarsening by pinning of grain boundaries [47,292].

(d) Particle-induced stabilization [47,203,213].

(e) Grain boundary engineering—a concept proposed by Watanabe [293] as a means of designing high-temperature materials, which exploits the idea of higher stability of special grain boundaries with low energy. Specifically, Σ3 (twin) boundaries should be particularly beneficial for enhanced thermal stability (see also Section 4.3.4 for further discussion). This is clearly illustrated in Fig. 10c [294] for the SUS 316L austenitic steel nanostructured by mechanical twinning (cf. Fig. 5), which remains stable at temperatures up to 500 °C.

A judicious choice of materials in conjunction with optimized thermomechanical treatment helps to overcome microstructural instability, which was identified as a most serious drawback of SPD processing. For instance, alloying of pure copper with Zr in small concentrations helps to stabilize the UFG microstructure against thermal [244] or mechanical (both monotonic and cyclic) influences [48]. Similarly, alloying with Sc provides aluminium with stability of its SPD-induced UFG microstructure [203,295–300]. The excellent fatigue properties of the ECAP-processed Cu–Cr–Zr alloys [224–225] mentioned in the previous section may be related to their remarkable thermal stability. Transmission electron microscopy observations showed that their ECAP-induced structure remained fine, the grain size not exceeding 250 nm even after heating to temperatures up to 600 °C. Grain growth was obviously suppressed due to nanoscale CuZr precipitates at the grain boundaries [224,225]. Grain coarsening set in only at temperatures as high as 650–700 °C. A similar result was reported for a Cu–Zr alloy in Ref. [301].

4.4.2. Diffusion properties

It is intuitively obvious that grain refinement by SPD should lead to enhanced atomic transport. Indeed, SPD promotes fast short-circuit diffusion through an increased volume fraction of grain boundaries and a higher dislocation density in the processed material. It is not obvious a priori, however, whether the effect of grain refinement is restricted just to this increased availability of fast diffusion pathways, or the diffusivity for the short-circuit paths itself

![Figure 11. Thermal stability of ECAPed (eight B, passes) copper (99.96%) (a and b) and SUS 316L stainless steel (c). (a) Reduction of Vickers microhardness during natural ageing of UFG Cu sample at room temperature for 1 year; (b) the effect of annealing on the grain size; (c) Vickers hardness of the SUS316L steel after one ECAP pass and annealing for 1 h.](image-url)
changes upon SPD processing. There is some controversy in literature about this issue, as both nearly unaltered [302] and enhanced [303,304] grain boundary diffusivities were found in SPD-processed materials. The complexity of singling out the contribution of grain boundaries from the overall diffusivity is compounded by the fact that old, “equilibrium” grain boundaries coexist with “non-equilibrium” ones [305–307], freshly produced by SPD. Such non-equilibrium grain boundaries possess a higher energy and a larger excess free volume, which is believed to lead to higher diffusivities than those of equilibrium grain boundaries in well-annealed coarse-grained materials.

The research group of Wilde and Divinski in Münster did a lot to shed light on the specifics of diffusion in SPD-processed materials. Based on their thorough radio-tracer measurements [308–312], a hierarchy of short-circuit diffusion paths in UFG materials produced by SPD was experimentally established. It was suggested that alongside relaxed (“equilibrium”) high-angle grain boundaries, there exist interfaces with significantly higher diffusivities. The enhanced diffusion rates for solute and self-diffusion in UFG materials prepared by SPD were attributed to the “non-equilibrium” state of the grain boundaries [267]. However, the most recent study [313] showed that this is not universally true. Rather, the effect of SPD processing was shown to be selective: grain boundary diffusivity of Co in ECAP-modified Ti was slower than in coarse-grained Ti, in contrast to accelerated grain boundary diffusivity of Ag atoms in Ti. This is an indication that for interstitially diffusing Co atoms trapping or scattering due to high concentration of lattice defects within non-equilibrium grain boundaries occurs, which retards their diffusion. By contrast, for Ag atoms, which migrate by substitutional diffusion, SPD-induced grain boundary defects boost the diffusion along non-equilibrium grain boundaries.

The studies by the same group revealed furthermore that, in addition to the more conventional, grain-boundary related, fast diffusion paths, some new, “ultra-fast” short-circuit paths specific for UFG materials produced by SPD may also exist. It was initially assumed that the ultra-fast diffusion channels were associated exclusively with the non-equilibrium grain boundaries [308,312]. This idea was later disproved [311,312,314], as evidence had emerged that a long-range network of interconnected nano- and micropores created by SPD and residing predominantly at non-equilibrium grain boundaries and triple junctions gives rise to ultra-fast diffusion [310,311]. An example of such pore structure in ECAP-deformed Cu is shown in Fig. 12. It should be noted that no such percolating porosity was found in Ni deformed by room-temperature ECAP and the ultra-fast diffusion observed is believed to be governed by a specific non-equilibrium state of the SPD-induced grain boundaries [315].

The occurrence of nano- and micropores as a consequence of SPD processing may appear worrisome, but the levels of porosity in question are extremely low, about 2 ppm [312]. The mechanical properties of SPD-treated materials do not seem to suffer from it anyway. It should be mentioned that nanopores dispersed in the bulk are very efficient obstacles to dislocation motion, so that their presence may be adding to strength.

Fig. 12. SEM micrographs of ECAP-processed copper obtained by field ion beam milling at different locations around a triple junction. The locations are indicated by the coloured lines; the corresponding micrographs are provided within a frame of the same colour. The grain boundaries (assumed to be SPD-induced, non-equilibrium ones) are visible through thermal grooves formed on them. Also seen are micro- and nanopores at grain boundaries. Note that the 1 μm scale bars have different lengths in the micrographs (a), (b) and (c). (Adapted from Ref. [309].) (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
With the mentioned exception of interstitial diffusion being slowed down, SPD can be generally considered to enhance diffusion of solutes as well as self-diffusion. It can thus be expected that kinetic properties of metals and alloys that involve diffusion will be accelerated as a result of SPD. Confirmations of this general expectation can be seen in the acceleration of such processes as plasma nitriding of steels [316] and hydrogen sorption in magnesium alloys [317]. The latter effect will be presented below.

4.4.3. Corrosion resistance

Another property of great interest for prospective engineering applications is corrosion resistance. Corrosion in single-phase polycrystalline aggregates is greatly influenced by grain boundaries and their area, which is determined by the grain size. Normally one would expect that any strengthening mechanism involved in SPD processing should deteriorate the corrosion behaviour of a defect-free single-phase metal. Indeed, at first glance both the SPD-induced structural inhomogeneities leading to local differences in the surface potential [318] and the higher reactivity of grain boundaries intersecting the surface of an UFG material could promote its degradation in a corrosive medium.

The nature of environmental degradation of metals and alloys is a multidisciplinary subject, which should be studied with respect to many aspects, including the three major ones: corrosion (chemical, electrochemical, pitting, etc.), stress corrosion cracking (SCC) and corrosion fatigue. To the best of our knowledge, the only material which has been investigated with regard to all these aspects is ECAP-processed copper [319–323]. A general encouraging conclusion obtained in these first investigations is that SPD, while enhancing mechanical characteristics, does not compromise the overall corrosion resistance and improves the SCC and corrosion fatigue resistance. Moreover, the homogeneity of corrosion damage of UFG Cu, which contrasts the localized intergranular corrosion in coarse-grained Cu polycrystals (cf. Fig. 13a and b), is seen as an advantageous property of ECAPed copper that makes it attractive for engineering applications. This finding may appear surprising in view of the general concern that grain boundaries are regions with high excess energy that act as preferred sites for corrosion attack.

The abundance of grain boundaries as preferential sites of attack by corrosive media increases the propensity of an UFG material to undergo intergranular cracking in the low-strain-rate SCC tests. This is compounded by a higher dislocation density in SPD-processed materials. Despite these negative factors, the corrosion damage on the surface of UFG copper turns out to be rather uniform at the macroscale, whereas obvious attack at grain boundaries and selective corrosion of some grain interiors were observed in coarse-grained copper.

These first observations were later confirmed by different researchers who found enhanced (or at least not reduced) corrosion resistance of UFG Cu [324–325], Al and a variety of Al-alloys [318,326–328], titanium [329], interstitial-free steel [330], austenitic stainless steels 316L [331] and 304 [332], FeCr [333], Mg [334] and Mg-based alloy ZK60 [335]. The improvement in corrosion resistance of ZK60 by the integrated extrusion + ECAP process (Table 1l), was interpreted in terms of two factors: grain refinement and the redistribution of Zn and Zr solutes within the microstructure. These microstructural changes impacted on the anodic and cathodic reaction kinetics of corrosion in a favourable manner, through both structural and chemical variations. A detailed understanding of the enhanced corrosion resistance observed for several SPD processed materials is, however, lacking, and this area definitely requires the attention of corrosion scientists.

In contrast to coarse-grained copper specimens showing a transgranular fracture when immersed in an aggressive liquid during SCC or fatigue deformation, the fracture of UFG specimens occurred intergranularly under the same conditions. This shows the dominant role grain boundaries played in the latter case. While it is true also for this case that increasing aggressiveness of a corrosive environment reduces the fatigue resistance, the observations in Ref. [321] demonstrated that the bulk microstructure, which is reflected in the surface properties, is a major factor
governing the damaging effect of the environment and the degree of degradation under load.

As mentioned above, the grain boundaries act as sites of preferential corrosion attack because of their imperfect atomic arrangement giving rise to increased local energy, higher excess volume, and stresses. Furthermore, the corrosion resistance is very sensitive to the grain boundary type. Thus, grain boundaries having low energy, such as special boundaries with low reciprocal coincident site lattice ratio $\Sigma$ ($\Sigma 3, \Sigma 7$, etc.), are particularly resistant to corrosion [336]. Following the above-mentioned concept of grain boundary engineering [286], micro- and nanostructures with the majority of energetically stable $\Sigma 3$ boundaries were artificially produced by mechanical twinning during “smooth” [189] or “violent” [337] processing, and these structures are considered to be beneficial for sustained resistance against corrosion.

4.4.4. Biocorrosion properties

As one of the prospective uses of SPD-processed UFG materials is in biomedical implants (see below), their corrosion behaviour in bodily fluids has become a vigorously growing research area. For brevity, we can only offer some sketchy glimpses of this research.

The biocorrosion properties of severely deformed CP titanium were shown to be similar to [338] or better than [339,340] those of its coarse-grained counterpart. Of course, it should not be forgotten that this occurs against the background of the hugely enhanced mechanical strength of the material.

Whereas improved corrosion resistance of ECAP-modified copper was reported by several authors for salty or acidic media [323,341], Xu et al. observed an increase in corrosion current in a simulated body fluid [342]. They suggested that this property of ECAP-modified copper could be used in intrauterine contraceptive devices.

Making a virtue of necessity and putting the high reactivity of magnesium to service, numerous researchers are working on the development of biodegradable Mg alloys [343]. A critical review of current methodologies and their limitations can be found in Ref. [344]. Low corrosion resistance is, of course, essential in bioresorbable Mg-based implants (e.g. bone implants or vascular stents), but the excessively high rate of degradation, which is accompanied by hydrogen evolution, is a hindrance to the use of uncoated Mg implants. Fortunately, SPD dampens the degradation rate of Mg and its alloys, especially in the early stages of exposure to body fluids, while improving their mechanical performance [345].

4.4.5. Biocompatibility of SPD-processed materials

Contemporary development of metallic implant materials is driven by the need for improved mechanical performance of biomedical implants, while fulfilling demanding biocompatibility requirements. Different paradigms govern this development for permanent and temporary (biodegradable) implants. While materials for permanent implants, e.g. for bone or tooth replacement, obviously need to be as inert in bodily fluids as possible, those for temporary implants must degrade at a rate suitable for the targeted application. Two archetypal alloy systems—those based on titanium and magnesium—are the favourites in these two respective categories.

In the case of permanent implants, titanium alloys, particularly Ti-6Al-4V, have dominated the market for decades. Titanium-based implants owe their great popularity to a very favourable combination of properties. These include a high corrosion resistance due to the protective surface layer of titania they form, reasonable bioinertness, high strength-to-weight ratio and very good fatigue resistance. An obvious choice for bioresorbable metallic implant material is magnesium, which is highly biodegradable. There is a huge literature on the medical implant applications of these materials, which we are not going to review here. Our goal is much narrower: a discourse on the suitability of SPD processing for improving the properties of Ti and Mg relevant to medical implant applications.

Current research focuses on improving the biocompatibility and the mechanical performance of these systems through variations in alloy composition, microstructure and surface treatment. In the case of titanium, a target is to improve the strength characteristics of CP grades, which could replace currently used alloys, thus avoiding potential biotoxicity of the alloying elements. If CP titanium is ever to replace Ti alloys, the deficit of strength in the unalloyed material will have to be compensated for. As discussed above, this can be readily achieved by SPD. ECAP [346], twist extrusion [63], helical rolling [101] and other SPD techniques are well suited for this purpose. Microstructure modification of CP titanium by ECAP was shown to be very efficient [347–349], with average grain sizes of 250–300 nm being achieved, and tensile strengths and HCF endurance limits (depending on the Ti grade used) approaching the levels for conventional Ti-6Al-4V. A surprising “by-product” of ECAP is that the response of the living cells to the surface of CP titanium after extreme grain refinement in the bulk is affected positively, resulting in increased adhesion and growth. This was found by in vitro assays with fibroblast cells [347,348], pre-osteoblast cells [349,350] and human bone marrow-derived mesenchymal stem cells [351]. On the negative side, it was found that bacterial growth on the surfaces of ECAP-modified CP Ti is also enhanced [352,353]. Evidence that the surface morphology, including nanoroughness, may be a major governing factor in cell attachment and proliferation is firming up [351,354]. However, many other factors may also be responsible for these phenomena, including hydrophilicity, surface chemistry, texture [355], etc., and the mechanisms of enhanced bioactivity of the ECAPed CP Ti have yet to be unravelled. In addition, machining and subsequent surface treatment involved in implant manufacturing may change these factors dramatically, and it is not known whether the benefits of bulk grain refinement for
cellular response will survive such surface-modifying processes.

The potential for using Mg alloys in bioresorbable vascular stents or bone implants is currently attracting huge interest [356,357]. A major challenge is the excessively high rate of degradation of these materials, which is problematic both in terms of the durability of the implant and the high rate of hydrogen evolution during corrosion, e.g. in vascular stent applications. As discussed above, the biodegradation of Mg and Mg alloys can be manipulated by SPD. What can be stated safely at this stage is that enhancement of mechanical performance (particularly, the fatigue characteristics) can be achieved with Mg alloys by such processes as ECAP without a loss (or even with some increase) in their resistance to biocorrosion [358]. However, bulk grain refinement does not seem to be sufficient to reduce the biocorrosion rate to the levels required by clinical needs. A tractable way to contain corrosion of Mg and achieve controllable corrosion rates will most probably be by surface modification, particularly through smart coating design. Possible interactions between the surface treatment and the fine granularity of the SPD-processed Mg alloys should be an interesting subject for research.

Development of metallic materials for bioimplants and medical devices is currently a buoyant area of research with the promise of technological, commercial and societal benefits. Bulk and surface modification by SPD is a scientifically challenging and potentially rewarding avenue to achieving better manufacturability and improved properties of the targeted products.

4.4.6. Radiation tolerance

A further benefit of SPD-processed UFG materials is the greater radiation tolerance they may offer. This can be expected, as the area of grain boundaries, which act as sinks for radiation-induced point defects, is significantly increased by grain refinement. Interplay between rapid healing of radiation effects due to an abundance of closely spaced sinks for point defects and segregation of solutes to grain boundaries is quite complex, however. Atom probe evidence for grain boundary segregation of Si and Ni induced by ion irradiation of 316L austenitic stainless steel nanostructured by HPT was reported by Pareige et al. [356] and Etienne et al. [359,360]. The possibility to manipulate the kinetics of irradiation damage and potentially suppress such unwanted phenomena as swelling, radiation creep, etc., in nuclear materials by controlled grain refinement by SPD opens up exciting new opportunities in nuclear power plant development. Compelling evidence that radiation tolerance of UFG materials is higher than that of the coarse-grained ones has begun to crop up [361–363]. More work in this area is required in order to develop efficient SPD processing routes leading to controlled improvement of radiation damage tolerance of structural materials.

4.4.7. Hydrogen sorption kinetics

Magnesium and its alloys are attractive materials for hydrogen storage due to their high storage capacity and the reversibility of hydrogen absorption/desorption. Unfortunately, the kinetics of hydrogen absorption and desorption are too slow, which hinders practical application. A way to change both the kinetics and the thermodynamics of hydrogen storage by SPD was first proposed by Skripnyuk et al. [317] who observed a pronounced enhancement of hydrogen desorption rate in Mg alloy ZK60 upon ECAP (Fig. 14). A recent publication reported an even greater acceleration of hydrogen desorption in ZK60 after ECAP [115]. The success of this approach prompted further studies [129,131,352,364].

The group of Horita investigated the effect of HPT on hydrogen storage in pure Mg and also found that the hydrogenation rate can be increased substantially [365,366]. Further studies employing HPT [367–370] confirmed the beneficial effects of grain refinement by SPD on hydrogen storage capacity or the sorption kinetics, or both. HPT processing also proved to be effective in enhancing the sorption of MgH2 powders [368].

With more and more experimental data on the effect of SPD processing on the thermodynamics and kinetics of hydrogen sorption in Mg and Mg alloys becoming available, the time has come to develop a better understanding of the physical mechanisms underlying these effects.

4.4.8. Further functional properties

There is hardly a physical property of a solid that is not affected by extreme grain refinement down to the submicron range. Despite the long history of SPD processing, the many possible effects are still largely unexplored, and there may be some hidden treasures to be uncovered out there. We briefly mention some effects that have recently been discovered.
There is evidence that magnetic properties are affected by SPD. Examples of coercivity measurements in HPT-processed Fe, Ni, Fe–17Co, Fe–3Si and Fe–6.5Si [113] show that the decrease in coercivity with decreasing grain size in the nanorange, which should follow the $d^{-2}$-law, is actually much slower—most probably as a result of the severe deformation the materials have undergone.

A marked influence of SPD processing on the properties of shape memory alloys was observed by a number of researchers (e.g. [114,233,371,372]). A summary of these effects is given in Ref. [113]. As was found in a recent study [373], ECAP of NiTi shape memory alloy can improve its strength without affecting the transformation temperature. A big increase (by &sim;80%) in the actuation stress was demonstrated, which is of significance for applications in miniaturized devices where high actuation stresses are at a premium.

An ultrafine grain structure and a high density of dislocations and point defects generated by SPD inevitably diminish the thermal conductivity [374] as well as the electrical conductivity [375]. This is bad news for Cu, Cu alloys and other conductors, as the intention behind SPD processing is to raise their mechanical strength with only moderate loss of conductivity. Post-processing, including annealing, helped to restore a large part of the lost conductivity up to acceptable values (75% of International Copper Annealed Standard – IACS) [225,375], but this was based on a rather empirical approach. A lot more needs to be done to provide guidance for rational process optimization. As an aid in evaluating the magnitude of the possible effect of the SPD on thermal conductivity, we refer to a simple model [374], which relates the thermal conductivity $\kappa$ of the processed UFG material to that of the unperturbed bulk material, $\kappa_{\text{bulk}}$:

$$\kappa = \kappa_{\text{bulk}} \left(1 + \frac{4\lambda}{3x d(1-r)}\right)^{-1}. \quad (16)$$

Here $\lambda$ is the electron mean free path and $r$ is the electron reflectivity, which accounts for the fact that the electrons reflected from a grain boundary do not contribute to heat transfer. Finally, $x$ (which is smaller than unity) is a shape factor accounting for the grain topology. This formula was shown to describe the grain size dependence of the thermal conductivity of ECAPed copper very well.

SPD processing of thermoelectric materials gives promise as a potentially viable strategy for raising the “figure of merit” that characterizes the efficiency of a thermoelectric material and its suitability for applications in thermoelectric generators. As these materials are commonly very brittle, HPT appears to be the only SPD technique suitable for processing them. The figure of merit is proportional to the square of the thermopower and inversely proportional to the electrical resistivity and thermal conductivity. Potentially, HPT can increase the thermopower and reduce the thermal conductivity. This needs to be done in such a smart way that these beneficial effects are not outstripped by a simultaneous increase in the electrical resistivity. The case of HPT-deformed thermoelectric compounds with skutterudite structure studied by the Vienna group [376] is a demonstration that this strategy may work. In this first exercise the increased electrical resistivity due to microcrack formation did not, however, produce an overall increase in the figure of merit. The most recent publication of the same group [377] reports further improvements in the figure of merit for skutterudite Sr$_{0.07}$Ba$_{0.07}$Yb$_{0.07}$Co$_6$Sb$_{12}$, mainly due to substantially reduced thermal conductivity. The problem with HPT-induced microcracks however, still remains. The challenge now is to find suitable processing conditions that would suppress the occurrence of microcracks.

5. Discussion and outlook

In the foregoing sections we presented a brief history of SPD techniques as a means of producing UFG materials, gave a summary of the existing methods and looked into the mechanism-based models that describe the material behaviour during SPD. The intention was to give the reader an overview of the subject and to offer insights in the mechanisms underlying SPD as well as the unusual material properties that can be achieved by SPD processing. We hope that this expose will serve as an introduction and a reference for the “uninitiated”, while also presenting our views on this area to those specializing in “nanoSPD”. We also hope that through this exercise we were able to show that the microcosm of the specific scientific challenges this area is facing reflects the fundamental problems of the physical theory of strength and plasticity at large. We tried to highlight these challenges throughout the manuscript, within the context of the particular subjects discussed, and are not going to list them here again. However, the most salient ones will be recapitulated below. An attempt at predicting where the nanoSPD research may be heading in the future will also be made.

First and foremost, in our view the question of how dislocations organize themselves in characteristic patterns, such as dislocation cell structures, and how these structures evolve in the process of straining, is largely unresolved. In particular, a comprehensive theory of the transformation of the dislocation cell structure into a new grain structure with a large proportion of high-angle grain boundaries needs to be developed. Such a theory should provide insights in the mechanisms and governing factors that determine the smallest achievable grain size. Ideally, this will be a probabilistic theory able to predict grain size and misorientation angle distributions and their variation with strain for different deformation paths. The models
to be developed will have to possess the predictive power to adequately represent the strain hardening and texture evolution during SPD processing, while also providing a reliable basis for simulations of the post-SPD performance of the processed material under service conditions. Understanding the nature of dislocation–grain boundary interactions for small grain size systems and the role of grain boundaries as sinks and sources of dislocations is a particularly interesting goal of research.

It would be naïve, of course, to believe that an all-embracing model of SPD based on dislocation dynamics is feasible. As indicated above, a distinction needs to be made between the case when a dislocation cell structure formed early on transforms smoothly to the final, smaller-scale grain structure and the situations when a violent (high-Z) deformation process breaks down the grain structure to a finer one. It is yet to be explored whether amorphization of the material on the way to a final structure is a necessary prerequisite for the formation of nanostructure. Experimentation with such demanding deformation paths supported by modelling and atomistic simulations may reveal interesting possibilities for nanostructuring.

There are a number of concepts which have established a firm place in the literature on nanoSPD materials, but for which a thorough justification is missing. This refers, in particular, to the widespread belief that grain boundary sliding may become a predominant mechanism of deformation of SPD materials. Indeed, the increased strain-rate sensitivity of flow stress often found in severely deformed materials suggests that grain boundary sliding or other diffusion-controlled mechanisms may contribute to their plasticity. However, there is no solid evidence that such deformation mechanisms are prevalent. Another concept that warrants scrutiny is that of bimodality of the grain structure being responsible for a good balance between strength and ductility. There are indications that this may be so, but a strict relationship between improved strength–ductility balance and the occurrence of a bimodal grain structure cannot be considered as proven.

An exciting area of research is the possible link between surface and bulk properties of SPD-treated materials. The enhancement of corrosion resistance and cell attachment and proliferation discussed in the previous section are in the category of surface phenomena affected by this link. It is fair to say that the mechanisms for these phenomena are not understood, which calls for more research on the subject. Of particular interest is the interaction of bulk nanostructured metals with the living matter—an area that may be full of surprises and exciting opportunities.

Further unusual effects may be expected from the application of SPD techniques to modify various physical or physicochemical properties of functional materials. The exemplary studies mentioned in this article (including the effects of SPD treatment on shape memory effect, hydrogen sorption, magnetic and thermoelectric properties, etc.) have possibly just scratched the surface of a hidden goldmine of opportunities in terms of improving properties. Understanding the mechanisms through which functional properties can be affected by SPD processing and identifying promising applications in this space pose interesting scientific problems and challenges.

The greatest expectations are being placed in achieving superior mechanical properties of structural materials. Despite a steady growth of the SPD research area and the promise it offers, an overly cautious attitude towards this group of processing techniques has until recently prevailed in the metal-forming industry. There are various reasons for this, one of them being the degree of overpromise of the benefits of SPD processing in the past two decades. Now that this research has matured, the situation is changing, and there are real and tangible applications of SPD processing ready to be picked up by industry. Manufacturing of biomedical implants, where commercial products have emerged recently, is a market-ready area par excellence [378]. Nevertheless, the challenge of providing compelling and persuasive arguments to promote SPD technologies still remains.

In addition to such psychological barriers to taking SPD processing from the laboratory to the shop floor, there are, of course, real technological challenges, such as the need for upscaling the processes and making them continuous or semicontinuous. Some examples of processing routes of this kind, which promise successful transfer to industrial-scale manufacturing [111,121,128,129,379], were given in this review.

SPD methods that were initially developed for processing of bulk materials as an extension of conventional metalworking techniques can also be used for other purposes, such as efficient compaction of powders [380], particularly for producing alloys from blended elemental powders [381], and swarf [112,382]. We also expect that SPD techniques targeting improvement of surface, rather than bulk, properties will experience a period of growth. In particular, in the spirit of an eighteenth-century aphorism by George Christoph Lichtenberg that “the most important things are done through tubes”, we see great opportunities in nanostructuring of tubes by SPD methods [96,114,115].

An attractive new application of SPD processing, which should give new life to mature SPD methods, was suggested in Ref. [383]. It was proposed to produce simultaneous architecturing and nanostructuring of hybrid materials by using established SPD techniques. In particular, twist extrusion, HPT and some newer methods appear suitable for producing a range of spiral architectures beneficial for strength and ductility. This field is an excellent playground for creative materials and process design, and we predict an exciting future for it.

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