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# Twenty-five years of ultrafine-grained materials: Achieving exceptional properties through grain refinement ☆

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**Abstract**

Twenty-five years ago, in 1988, there appeared a classic description of the application of severe plastic deformation (SPD) to bulk solids in order to achieve exceptional grain refinement to the submicrometer level. This report and later publications initiated considerable interest in materials science laboratories around the world and many experiments were subsequently performed to evaluate the principles and practice of SPD processing. The present report provides an overview of the more recent developments in this field, with special emphasis on the opportunities for achieving homogeneity in the as-processed materials and on the general characteristics of the mechanical properties achieved after SPD processing. For simplicity, special emphasis is placed on the two techniques of equal-channel angular pressing and high-pressure torsion as these are currently the most popular procedures for applying SPD processing.

*Keywords:* Equal-channel angular pressing; High-pressure torsion; Homogeneity; Severe plastic deformation; Ultrafine grains

## 1. Introduction

It is well known that the flow behavior of polycrystalline materials is readily divisible into two distinct regimes, depending primarily upon the operating temperature. These regimes are depicted most clearly using deformation mechanism maps [1,2], where there is a sharp division, occurring at $\sim 0.5T_m$ where $T_m$ is the absolute melting temperature, between low-temperature behavior controlled by the conservative motion of dislocation glide and high-temperature behavior in which diffusion-controlled flow is dominant through processes such as dislocation climb and diffusional creep.

Inspection shows that, for both these flow regimes, the grain size is the most important, and indeed the dominant, structural parameter in polycrystalline materials. Thus, in the low-temperature regime, the yield stress $\sigma_y$ varies with the grain size $d$ through the Hall–Petch relationship, which is given by [3,4]

$$\sigma_y = \sigma_o + k_y d^{-1/2} \qquad (1)$$

where $\sigma_o$ is the lattice friction stress, and $k_y$ is a constant of yielding. Conversely, in the high-temperature regime, the creep rate under steady-state conditions $\dot{\varepsilon}$ is expressed by a relationship of the form [5–7]

$$\dot{\varepsilon} = \frac{ADG\mathbf{b}}{kT}\left(\frac{\mathbf{b}}{d}\right)^p\left(\frac{\sigma}{G}\right)^n \qquad (2)$$

where $D$ is the appropriate diffusion constant ($=D_o\exp(-Q/RT)$, where $D_o$ is the frequency factor, $Q$ is the activation energy for the flow process, $R$ is the gas constant, and $T$ is the absolute temperature), $G$ is the shear modulus, $\mathbf{b}$ is the Burgers vector, $k$ is Boltzmann's

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constant, $\sigma$ is the flow stress, $n$ and $p$ are the exponents of the stress and the inverse grain size, respectively, and $A$ is a dimensionless constant.

It follows from inspection of Eq. (1) that a small grain size is advantageous because it leads to a significantly higher strength. Additionally, Eq. (2) shows that a small grain size leads to faster strain rates, and this provides the possibility of achieving a superplastic forming capability at rapid rates that may be readily employed in industrial forming operations. Thus, grain refinement is an important processing tool for achieving optimum properties in metallic materials. The advantage of grain refinement was recognized many years ago and led to the development of thermo-mechanical processing operations wherein materials were subjected to annealing treatments and mechanical straining in order to reduce the grain size to values that are typically within the range ~3–10 μm.

Twenty-five years ago, in 1988, a landmark report was published demonstrating the potential for achieving even smaller grain sizes within the submicrometer range through the application of severe plastic deformation (SPD) to bulk coarse-grained solids [8]: this approach is now generally termed SPD processing. The publication of this report attracted much attention, and it led to the initiation and development of many research activities around the world devoted to processing and measuring the characteristics of materials with exceptionally small grain sizes. It is interesting to note that the research activity in this field has both continued to develop and very much expanded up to the present day.

It is often not recognized that the general concept of SPD processing has a long history which may be traced back for more than two thousand years. Thus, the pioneering concept of SPD processing, introduced 25 years ago, lay not specifically with the processing method per se but rather with the availability of new advanced analytical and microscopic tools which provided, for the first time, direct evidence that the improved mechanical properties of these processed metals was due primarily to the introduction of exceptional grain refinement.

As described elsewhere [9], the historical background of SPD processing divides readily into three separate periods spanning more than two millennia. These periods are outlined briefly in the following section, and the subsequent sections provide detailed descriptions of some of the more recent developments in this important research field.

## 2. The historical background of SPD processing

### 2.1. The ancient age

A comprehensive review of the history of SPD processing shows that the general concept dates back at least to the Han dynasty of ancient China in ~200 BC [10]. At that time, the local artisans developed a new technique for the processing of steel for use in swords, where the metal was repetitively forged and folded to form the famous Bai-Lian steels. Many archeological artifacts are now available from this area and they often have inscriptions providing a historical record of the processing operation. Thus, a high-strength 50-Lian steel sword was prepared using 50 separate forging and folding operations. Subsequently, the principles of this technology spread to Japan where it was used for the processing of samurai swords, to India where it led to the development of the wootz ultrahigh carbon steel [11], and then to the Middle East where it produced the famous Damascus steel [12]. It is important to note that, although these developments spread readily across Asia, the fundamental principles of the processing technique lacked scientific rigor and, ultimately, in about the middle of the 18th century, the principles of this technique were lost.

### 2.2. The scientific age

The first attempt to introduce scientific principles into the procedures now known as SPD processing lies unambiguously in the classic work of Professor P.W. Bridgman at Harvard University. Beginning in the 1930s, Bridgman conducted a remarkably comprehensive series of experiments on the application of high pressures to bulk solids [13,14] and, in 1952, the results from these many experiments were succinctly summarized in a book [15]. It is important to note that Bridgman received the Nobel Prize in Physics in 1946 with the citation reading “for the invention of an apparatus to produce extremely high pressures, and for the discoveries he made therewith in the field of high pressure physics”. A review of this work shows that Bridgman was the first to propose the processing of metals through a combination of compression and torsional straining. This approach was later further developed in the former Soviet Union [16] and ultimately evolved into the procedure now known as high-pressure torsion (HPT).

A second major influence on SPD processing may be traced to the classic work of Dr. V.M. Segal and his colleagues, conducted in the 1980s in Minsk in the former Soviet Union (now the capital of Belarus). Segal and coworkers were the first to develop the process now known as equal-channel angular pressing (ECAP) (or equal-channel angular extrusion) [17] and this process has now become the most important and the most used of all SPD processing techniques.

Nevertheless, a deficiency of these earlier investigations was the absence of any detailed microstructural analysis. In practice, the detailed examination of microstructure became possible only with the later development of sophisticated analytical tools, including high-resolution transmission electron microscopy (HRTEM), electron back-scatter diffraction (EBSD), orientation imaging microscopy (OIM) and modern X-ray techniques.

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