

Ultra-severe plastic deformation for room-temperature superplasticity and superfunctionality

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Keywords: Severe Plastic Deformation (SPD), High-Pressure Torsions (HPT), Superplasticity, Strain Rate Sensitivity, Functional Properties

Abstract. Ultra-severe plastic deformation (ultra-SPD) is a terminology used for the introduction of extremely large shear strains (over 1000) to material so that the thickness of sheared phases geometrically reaches the subnanometer level. Under such extreme shearing conditions, new nanostructured phases with unique properties are formed even from the immiscible systems. Various metallic alloys and ceramics were developed by this concept for different applications such as room-temperature superplasticity, room-temperature hydrogen storage, photocatalytic hydrogen production, photocatalytic carbon dioxide conversion, etc. This article reviews recent advances regarding ultra-SPD with a focus on low-temperature superplasticity, which was reported for the first time at room temperature in aluminum and magnesium alloys.

Introduction

Severe plastic deformation (SPD) is a popular technology to introduce large plastic strain in a workpiece [1,2]. There are various SPD methods such as high-pressure torsion (HPT), equal-channel angular pressing (ECAP) and accumulative roll-bonding (ARB) [3,4]. The main target to employ SPD is the generation of ultrafine grains (UFG) [5,6]. Formation of such UFG structures leads to strengthening and enhanced functional properties [7,8]. A review paper written by 49 experts in SPD referred to these SPD-processed UFG materials as superfunctional materials due to their excellent functional properties [9]. SPD has been used since ancient times, but its scientific principles were documented by Bridgman in the 1930s [10,11], Segal et al. in the 1970s [12], and Valiev et al. in the 1980s [13]. For the latest progress in the SPD field, the readers are referred to review papers published in a special issue of Materials Transactions [14].

Ultra-SPD is one of the new directions which is getting attention to synthesize new materials with promising properties [15]. This manuscript reviews recent advances in the application of ultra-SPD with a focus on room-temperature superplasticity, which could be realized for the first time in aluminum and magnesium alloys by ultra-SPD [16,17].

Ultra-Severe Plastic Deformation

The concept of ultra-SPD was suggested in 2017 as an attempt to find a strategy to synthesize new materials [18]. Ultra-SPD is defined as SPD processing in which the shear strain is drastically increased so that the thickness of sheared phases reaches the subnanometer level, as schematically shown in Fig. 1a [15]. The necessary shear strain depends on the size of phases and their co-deformation, but the applied shear strain is at least 1000 [15]. Besides the objective of material synthesis, ultra-SPD has also raised questions regarding the behavior of metallic materials in the steady state, where the strength is supposed to remain constant with increasing shear strain [19].

High-Pressure Torsion for Ultra-Severe Plastic Deformation

Ultra-SPD studies use HPT, a process schematically shown in Fig. 1b. The method, in which a disc [20] or ring [21] is strained under pressure by torsion, has some merits for ultra-SPD. First,

shear strain in HPT ($\gamma = 2\pi rN/h$; γ : shear strain, r : radial distance from the rotation center, N : rotation numbers, h : thickness of disc- or ring-shaped specimen) can be increased with almost no limits by increasing the rotation numbers [9-11]. Second, pressure in HPT is high, and thus it is applicable to hard metals (Hf [22], Mo [23], W [24]), glasses [25-27], semiconductors (Si [28,29], Ge [30,31]), oxides (TiO₂ [32], ZrO₂ [33], BiTaO₃ [34]), and multi-component alloys (intermetallics [35-37], single-phase high-entropy alloys [38-40], dual-phase high-entropy alloys [41-43]). Third, it is applicable for cold consolidation of powders [44-46], because of high pressure/strain and minor temperature rise [47,48]. Fourth, the method uses the advantages of high pressure/strain to control phase transformations [49-51]. Fifth, the method continuously applies strain, resulting in a high concentration of defects [52-54] and ultra-fast dynamic interdiffusion [55-57].

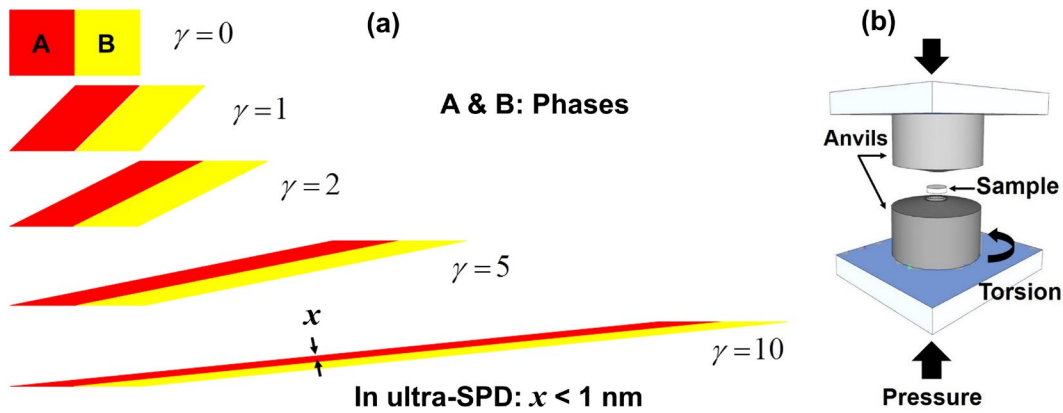


Fig. 1. (a) Schematic illustration of (a) ultra-SPD, and (b) HPT.

Main Findings by Ultra-Severe Plastic Deformation

Ultra-SPD is promising to synthesize a variety of materials including conventional binary/ternary alloys [15], intermetallic compounds [18], high-entropy alloys [18], hydrides [58], and high-entropy ceramics [59]. Such materials show promising properties like high strength, room-temperature superplasticity, hydrogen storage, biocompatibility, superconductivity, thermal stability, and photocatalysis, as reviewed in [60]. A summary of materials processed by ultra-SPD and their characteristics are given in Table 1 [61-88]. These properties are briefly summarized below, but room-temperature superplasticity is discussed in the next section.

SPD-processed materials usually show low thermal stability due to a large fraction of defects [89-93], but the materials processed by ultra-SPD can have high thermal stability. For example, the application of ultra-SPD to produce supersaturated Al-Ca [69], Al-Fe [70], Al-Zr [75], and Al-La-Ce [76] alloys, leads not only to high thermal stability but also to age hardening.

Ultra-SPD is effective in the stabilization of phases that do not exist in the phase diagram such as BCC, HCP and FCC phases in the immiscible Mg-Ti [62], Mg-Zr [63] and Mg-Hf [64] systems.

Table 1. Summary of the application of ultra-SPD to various materials.

Alloy	Shear Strain	Characteristics	Ref.
Mg-Li	7800	Superplasticity at room temperature	[16]
Mg ₂ X (X: 21 elements)	5500	Storing hydrogen	[61]
Mg-Ti	5500	Storing hydrogen	[62]
Mg-Zr	55000	Storing hydrogen in new phases	[63]
Mg-Hf	3900	Biocompatible new phases	[64]
Mg-V-Cr	50000	Storing hydrogen	[65]
Mg ₄ NiPd	59000	Storing hydrogen at room temperature	[66]
MgTiVCrFe	12000	Storing hydrogen	[67]
MgTiH ₄	17000	Storing hydrogen	[68]

Al–Ca	39000	Thermal stability	[69]
Al–Fe	39000	Thermal stability	[70]
AlNi	4700	Enhanced microhardness	[71]
Al ₃ Ni	4700	Enhanced microhardness	[72,73]
Al–Cu	3900	Quick interdiffusion	[74]
Al–Zn	7800	Superplasticity at room temperature	[17]
Al–Zr	39000	Thermal stability and age hardening	[75]
Al–La–Ce	39000	Thermal stability and age hardening	[76]
TiAl	2000	Enhanced strength/plasticity	[77]
TiV	5500	Storing hydrogen with no activation treatment	[78]
Ti–Nb	5900	Biocompatibility with good strength/elasticity	[79]
TiZrHfNbTa	2000	Biocompatibility with good strength/elasticity	[80]
TiZrHfNbTaO ₁₁	7800	Photocatalytic H ₂ evolution and CO ₂ conversion	[81,82]
TiZrHfNbTaO ₆ N ₃	3900	Photocatalytic H ₂ evolution and CO ₂ conversion	[83,84]
TiZrNbTaWO ₁₂	3900	Photocatalytic O ₂ evolution	[85]
FeNi	3900	Quick phase transition	[86]
Ni ₂ AlTi	4700	Enhanced microhardness	[87]
Nb–Ti	3900	Superconductivity	[88]

Intermetallic compounds with nanograin sizes are of interest, but these materials should be produced by two-step procedures like high-temperature sintering of nanopowders [93-96]. However, ultra-SPD is a single-step process to produce bulk nano-intermetallics like Mg₂X (X: other elements) for hydrogen storage [61], AlNi [71] and Al₃Ni [72,73] with high hardness, TiAl with high strength and plasticity [77], L1₀-FeNi [86] and Ni₂AlTi [87].

Orthopedic materials should have biocompatibility, high strength and low elastic modulus. Ti-based biomaterials suffer from low strength and high modulus [97], but SPD can improve their strength [98-101]. Ultra-SPD was successfully used to synthesize alloys with a good combination of high strength and low elastic modulus from the Ti–Zr–Hf–Nb–Ta biocompatible system [79,80].

Nanostructured Nb–Ti superconductors are commercially fabricated by several repetitions of wire drawing and long-time annealing [102-105]. Ultra-SPD followed by short annealing is a fast method to produce such superconductors with properties comparable with commercial ones [88].

The design and synthesis of magnesium alloys that can reversibly store hydrogen at room temperature is a long-term challenge [106,107]. Ultra-SPD not only contributed to the synthesis of various hydrogen-storing compounds such as Mg₂X intermetallics (X: other elements) [61], immiscible Mg–Ti [62,68], Mg–Zr [63] and Mg–Hf [66] alloys, ternary Mg–V–Cr alloy [65] and high-entropy MgVTiCrFe alloy [67], but also it introduced Mg₄Ni₁pd as the first Mg-based material with reversible hydrogen storage performance at room temperature [66].

Ultra-SPD introduced the first high-entropy photocatalysts for hydrogen and oxygen production from water splitting as well as for CO₂ conversion. The high-entropy oxides TiZrHfNbTaO₁₁ [81,82] and TiZrNbTaWO₁₂ [85] and the high-entropy oxynitride TiZrHfNbTaO₆N₃ [83,84] are the first high-entropy photocatalysts with high activity.

Room-Temperature Superplasticity

Superplasticity is defined as the capability of a material for large tensile deformations over 400%. Such elongations can be achieved at homologous temperatures over 0.5, where grain-boundary sliding is dominant. Superplasticity is quantified by the creep equation with a strain rate sensitivity of ~0.5 [108]. The equation suggests two solutions to achieve low-temperature superplasticity: (i) grain size reduction which was used to attain low-temperature superplasticity by SPD processing [109-111]; and (ii) acceleration of grain boundary diffusion by grain boundary engineering, which was used to attain room-temperature superplasticity in Mg–Li and Al–Zn by ultra-SPD [16,17].

The application of ultra-SPD (200 HPT turns) to a two-phase Mg–8wt.%Li alloy (Mg-rich α phase + Li-rich β) resulted in 440% elongation at room temperature (Fig. 2a) with a strain-rate sensitivity of 0.37 (Fig. 2b) [16]. However, the extrusion and SPD process for 5 HPT turns did not

lead to such an elongation in agreement with earlier studies [112]. Superplastic deformation after ultra-SPD is promising because room temperature corresponds to a homologous temperature of 0.35 for the alloy. Room-temperature superplasticity occurs due to fast grain-boundary diffusion by increasing the fraction of α/β interphase boundaries and the segregation of lithium along the α/α grain boundaries, as shown in Fig. 2d using high-angle annular dark-field (HAADF) image and in Figs. 2d and 2e using energy-filtered transmission electron microscopy (EFTEM).

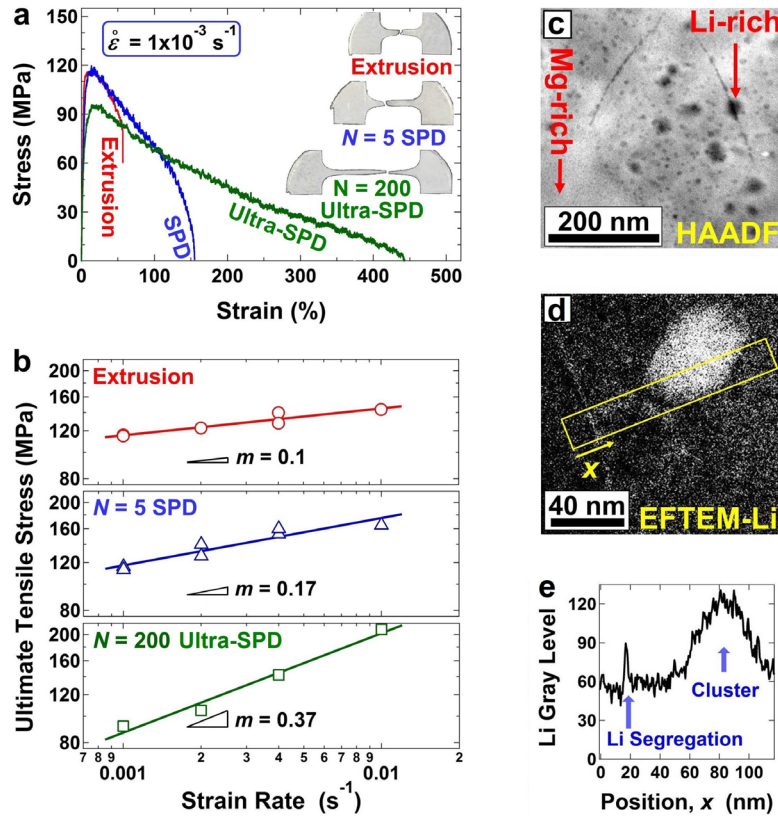


Fig. 2. (a) Stress-strain curves, (b) ultimate tensile strength versus strain rate for Mg-Li alloy processed by extrusion, SPD with 5 HPT turns and ultra-SPD with 200 HPT turns (m : strain-rate sensitivity). (c) HAADF image, (d) EFTEM-Li image and (e) gray level versus position shown in (b) for Mg-Li after ultra-SPD [16].

Ultra-SPD (200 HPT turns) was applied to an Al-30at%Zn alloy with two phases of Al-rich α and Zn-rich η [17]. The tensile behavior of the material was examined after ultra-SPD and after 100 days of aging at room temperature. As shown in Fig. 3a, the homogenized sample shows limited plasticity, but elongation is 480% after ultra-SPD and 330% after aging. It should be noted that this alloy did not show room-temperature superplasticity after SPD using 20 HPT turns [113]. The strain-rate sensitivity for ultra-SPD-processed samples reached a high value of 0.41, as shown in Fig. 3b. Superplasticity after ultra-SPD at room temperature (i.e. the homologous temperature of 0.36) is due to fast grain-boundary diffusion by increasing the fraction of α/η interphase boundaries and the segregation of zinc along the α/α grain boundaries, as shown in Fig. 3c and 3d using HAADF image and in Figs. 3e and 3f using energy-dispersive X-ray spectroscopy (EDS).

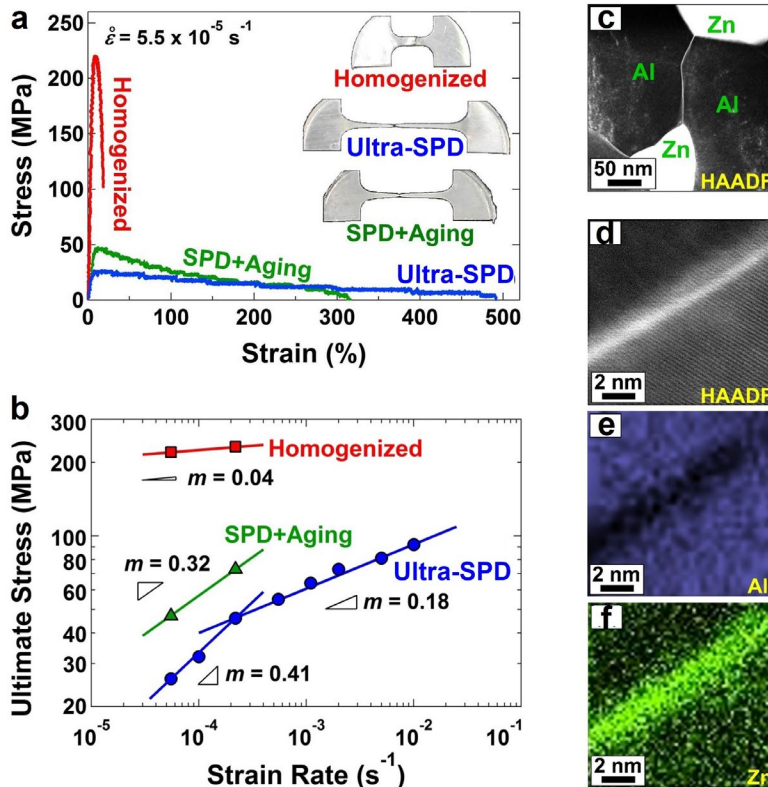


Fig. 3. (a) Stress-strain curves, (b) ultimate tensile strength versus strain rate for Mg-Li alloy processed by homogenization, ultra-SPD with 200 HPT turns and ultra-SPD followed by 100 day aging (m : strain-rate sensitivity). (c, d) HAADF images, and (e, f) EDS elemental mappings of region in (a).

Summary

The concept of ultra-SPD is effective to develop various superfunctional materials for energy, electrical and biomedical applications. One of the interesting results achieved by ultra-SPD is the development of first room-temperature-superplastic aluminum and magnesium alloys.

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