

Article

A new strategy to strength-toughen metals: Tailoring disorder

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ABSTRACT

Metals have been mankind's most essential materials for thousands of years. In recent years, however, innovation-driven development of major national security strategy and core areas of the national economy is highly impeded by a shortage of advanced higher-strength-toughness metals. One of the main reasons is that metals inherently exhibit the inverted-relationship of strength-toughness. The emergence of two types of disordered metals: amorphous alloys and high entropy alloys, provides a fully-fresh strategy for strength-toughening by tailoring the topological and/or chemical disorder. In this paper, we first briefly review the history of strength-toughening of metals, and summarize the development route-map. We then introduce amorphous alloys and high entropy alloys, as well as some case studies in tailoring disorder to successfully achieve coexisting high strength and high ductility/toughness. Relevant challenges that await further research are summarized in concluding remarks.

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1. Introduction

What types of materials are massively in service is an important sign of the progress of human civilization. For example, the development of steels is considered to be one of the driver forces to the first industrial revolution marked by the steam engine. From then on, various metallic materials have been widely developed and applied in high-tech fields such as transportation, manufacturing, defense, and aviation. Nowadays metals occupy a dominated niche in engineering structural materials, and they are irreplaceable in many fields [1]. The national strategy of energy saving, emission reduction and sustainable development urges stronger metals with higher toughness that can serve in demanding environments. In the past decades, a series of approaches to strength-toughen metals have been proposed based on concepts of hierarchical, multi-scale structure/phase construction [2–8]. Among them, amorphous alloys (AAs) [9–11] and high entropy alloys (HEAs) [12, 13] have attracted worldwide attention because of their record-high strength and ductility/toughness. Due to inherent topological/chemical disorder, the two types of disordered alloys contain trans-scale emergence and evolution of non-equilibrium disordered structures under external loading. Their plastic flow

therefore becomes spatio-temporally complicated and diverse, subverting the classical lattice-defect-mediated plastic mechanisms. Importantly, the research on plastic flow and fracture of disordered alloys provides a new way to crack the natural inverted-relationship between the strength and toughness [14]. Such fundamental researches will also greatly promote the applications of disordered alloys as a new generation materials for kinetic energy weapon, nuclear energy and spacecraft shielding [15–19].

2. Route-map for strength-toughening

Throughout the development history of metals for thousands of years, this type of materials is of two basic attributes. One is that their chemical compositions always contain one or two principal elements. The other is that their topological structures are packing mainly with ordered lattices. In this sense, traditional metals can be defined as ordered alloys dominated by chemical/topological order. Metallic materials are of two most important mechanical properties: strength and toughness. In this review, toughness is a loosely-defined property associated with cracking resistance, plastic deformation in compression, and uniform elongation (ductility) in tension. For ordered crystalline metals, the two properties depends not only on the types of "order", but also largely on the "disorders", i.e., lattice defects, in the "order" background.

In 1934, three scientists: Orowan [20], Polanyi [21] and Taylor [22], independently proposed the dislocation mechanism of crys-

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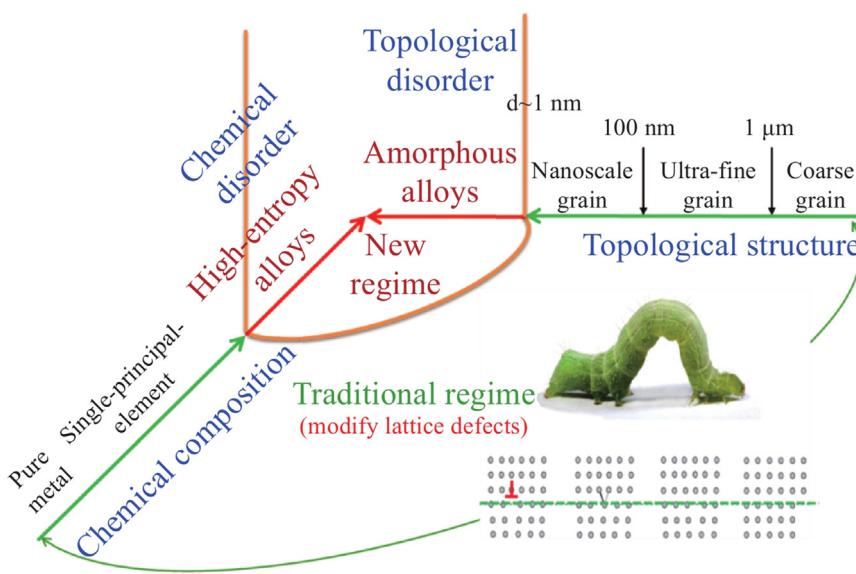


Fig. 1. Route-map for strengthening-toughening of metals.

tal plasticity. Dislocations are one type of line defects of lattice, and their activation and motion determine plastic yield and subsequent deformation. Traditionally, the core idea of strengthening metals is to introduce various forms of “disorder” through composition regulation and structural manipulation. These “disorder” will effectively slow-down or hinder the movement of dislocations, so that plastic deformation has to surmount higher barrier, thus increasing strength significantly. As illustrated in Fig. 1, there are two main routes to achieve such strengthening. The first route is to add some alloying elements to base metals. For example, high-strength steel or stainless steel can be obtained by adding minor C, Cr elements to pure Fe. Such alloying technique can give rise to solid solution strengthening, precipitation strengthening, phase transition strengthening, etc. The second route is to reduce the grain sizes of bulk metals down into ultra-fine ($\leq 1 \mu\text{m}$) or even nanostructured ($\leq 100 \text{ nm}$) regime. One of the motivations for such push for the very small grain sizes stems from the well-known Hall-Petch relationship that predicts a continuous rise of strength with decreasing grain size [23, 24]. In the ultra-fine or nanostructured metals, dislocations are limited within small grains and their movements are highly impeded by plenty of grain boundaries, thus contributing to unprecedented mechanical strength.

Every coin has two sides. Existence of immovable dislocations will inevitably weaken the ability of materials to deform plastically, ultimately sacrificing ductility or toughness and particularly uniform elongation in tension. Therefore, the strength of metals is usually irreconcilable with their toughness or ductility, an effect referred to as the strength-toughness trade-off [14], which seems an insurmountable hurdle. However, if there are multiple or excess plastic mechanisms in addition to dislocations that can be stimulated and act synergistically during the external loading process, it is possible to achieve a certain optimal balance between strength and toughness. Based on this idea, a few highly-strong-tough metals have been developed in the past decades [2–8]. These metals are usually constructed by hierarchical, multi-scale structures or phases, so that multiple plastic mechanisms such as slips, twinning, phase transition, and grain boundary diffusion could be activated synergistically upon loading. Nevertheless, these heterogeneous metals still belong to chemically or topologically ordered systems, and their strength-toughening (S-Ting) level is gradually reaching the upper boundary. A question naturally arises. Is it possible to break through such traditional routes, and to realize a new

strategy for S-Ting which is dominated and regulated by the topological and/or chemical disorder?

3. A new strategy: case studies

In recent years, some new types of high-strength-toughness metals represented by AAs and HEAs continue to emerge [25–32], signifying the possibility of this new strategy. AAs and HEAs subvert traditional metals that have existed in an orderly form for thousands of years. They inherently are of topological and/or chemical disorder, and therefore belong to the disordered alloys. AAs are formed upon cooling high-temperature melts fast enough to avoid crystal nucleation, but by deep supercooling to the glass transition temperature where the liquid structure is suddenly “frozen” to a solid. This new type of glassy solid resides in thermodynamically metastable states, and the atomic packing in AAs is only of short-range-order [33–35], but lacking in long-range order and transitional symmetry [36–38]. Such unique features endow AAs with inherent structural heterogeneities on nanoscale. In a sense, AAs can be regarded as the topological disorder limit of nano-structuring of metals, see the “topological structure” axis of Fig. 1. HEAs break through the traditional concept of alloy design using one or two dominant metal elements. Instead, HEAs are multi-principal-element metallic systems, which are formed based on the idea of entropy-stabilized random solid solution [12, 13]. They contain high concentration (20%–25%) of multiple elements with different crystal structures, but can crystallize as a single phase. Recently, the phase composition of HEAs tends to be multiphase or eutectic [19, 28, 39–41], which refreshes the precipitated-phase strengthening, the second-phase strengthening or other strengthening mechanisms. Equiatomic or near-equiatomic HEAs are the chemical disorder limit of element-devising of metals, see the “chemical composition” axis of Fig. 1. For HEAs, there are four core effects [42, 43]: (i) high-entropy effect in thermodynamics; (ii) sluggish effect in kinetics; (iii) severe lattice distortion in structures; and (iv) cocktail effect in properties. Next, we present some case studies about the new S-Ting strategies of the two types of disordered alloys: both AAs and HEAs.

Case 1: Shear-banding-mediated S-Ting.

In 2004, Schroes and Johnson [25] reported a monolithic Pt_{57.5}Cu_{14.7}Ni_{5.3}P_{22.5} bulk AA with both 1.4 GPa strength and pro-

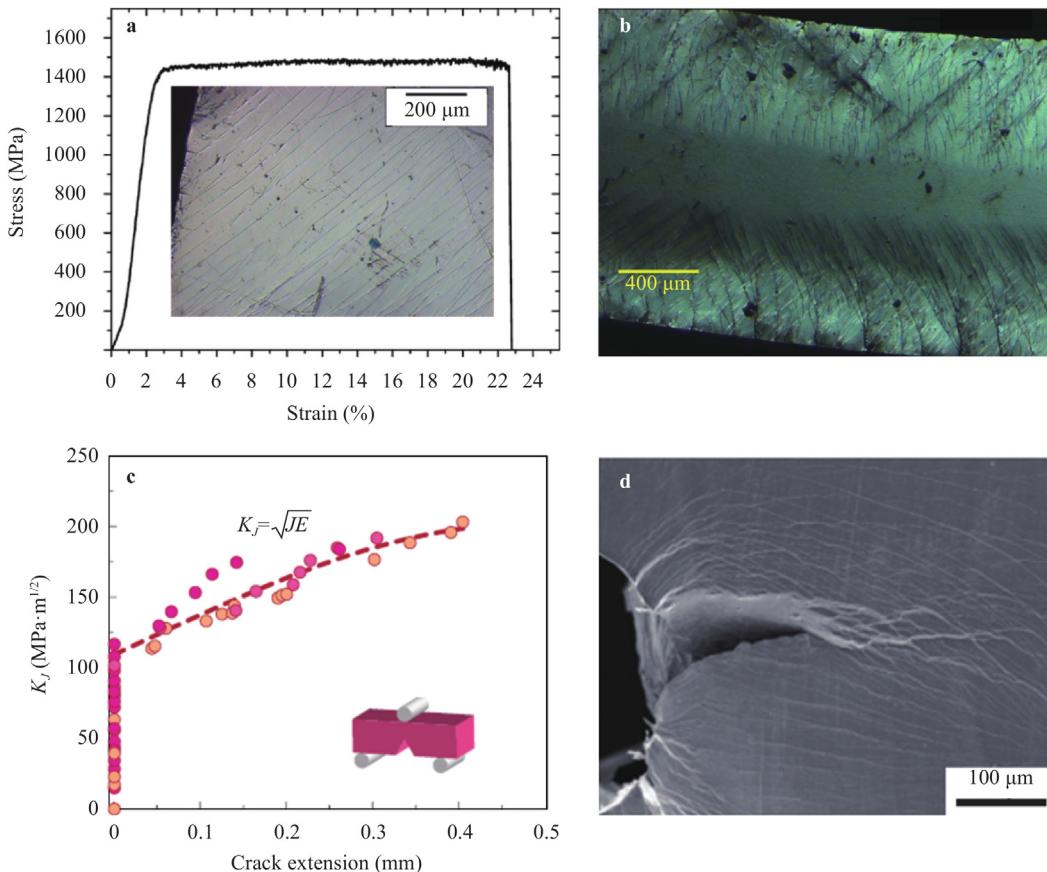


Fig. 2. (a) Compressive stress-strain curve of a $\text{Pt}_{57.5}\text{Cu}_{14.7}\text{Ni}_{5.3}\text{P}_{22.5}$ bulk AA, with multiple shear bands after 15% strain. (b) Multiplication of shear bands in the $\text{Pt}_{57.5}\text{Cu}_{14.7}\text{Ni}_{5.3}\text{P}_{22.5}$ bulk AA after bending to $\sim 7\%$ strain. (c) Fracture resistance curve of a $\text{Pd}_{79}\text{Ag}_{3.5}\text{P}_6\text{Si}_{9.5}\text{Ge}_2$ bulk AA with (d) multiple shear bands at crack tip [25, 26].

nounced global plasticity. Under quasistatic compression, this high-strength AA shows a very large plastic strain of 20%, which is ascribed to multiplication of shear bands during loading, as shown in Fig. 2a. Mediated by multiple shear bands; see Fig. 2b, this Pt-based alloy also shows permanent deformation and a strain exceeding 3% before failure during bending. In 2011, Demetriou et al. [26] reported a $\text{Pd}_{79}\text{Ag}_{3.5}\text{P}_6\text{Si}_{9.5}\text{Ge}_2$ bulk AA with the yield strength close to 1.5 GPa in tension. This Pd-rich metal-metalloid alloy has a very high fracture toughness in terms of a stress intensity, K_I , of ~ 200 MPa·m^{1/2}, measured by the fracture resistance curve in Fig. 2c. Again, such remarkable damage-tolerance results from multiplication of shear bands at the tip of an opening crack, and the extensive shear-band sliding promotes significant crack-tip blunting, as shown in Fig. 2d.

Shear-banding in AAs results from stress-driven structural disordering via an avalanche of shear-transformations (STs) [44–46]. Homogenous nucleation and stable propagation of shear bands could effectively balance strength and plasticity of alloys. Usually, a shear band will rapidly evolve into an opening crack, particularly in disordered systems. However, according to a recent study [47], introducing chemical inhomogeneity/disorder will lead to more difficult cracking nucleation. Obviously, the shear-banding-mediated S-Ting of the two AAs mentioned above is closely associated with unique topological and chemical disorder in these materials.

Case 2: Rejuvenation-induced S-Ting.

Metastable AAs tend to physical ageing, which is detrimental to the plastic deformation ability, known as ageing-induced brittle-

ness [48]. Recent studies [27, 49–52] have shown that either aged or as-cast AAs can be effectively rejuvenated into more disordered high-enthalpy states. The glass rejuvenation is not spontaneous, but requires external energy injection to ‘shake up’ the frozen-in disordered structure [53]. Without loss of high strength, the rejuvenated AAs can show enhanced deformability [27, 31, 54–57]. In 2015, Ketov et al. [27] reported that, after repeated cryogenic cycling between 338 K and 77 K, rejuvenated Zr-based AA rods with ~ 1.68 GPa strength show an increase in plastic strain under uniaxial compression, as shown in Fig. 3. Further studies [58–60] have revealed that such rejuvenation-induced S-Ting results from thermally activated disordering of topological structures, pointing towards heterogeneities in the fictive temperature.

In addition to thermal activation, glass rejuvenation can be achieved by mechanical energy working to directly introduce non-affine strain in disordered structures [49, 50, 54, 61–63]. Pan et al. [49] have performed uniaxial compression on a Zr-based AA rod with a circumferential notch, and they found that the notch region can experience an extreme rejuvenation without any shear banding. Furthermore, Pan et al. [31] cut a rod from the rejuvenated notch region, as shown in Fig. 4a. Upon uniaxial compression, this rejuvenated AA rod shows extensive strain-hardening (indicated as an increase of yield strength with successive loading-unloading) and homogeneous flow, as shown in Fig. 4b. It is revealed that these intriguing properties result from a gradual decrease of topological disorder (i.e., free volume) induced by compressive plastic strain applied. Such ordering or relaxation mechanism leads to strain-hardening and suppression of shear banding, which is consistent with the theoretical analyses by Jiang et al [64].

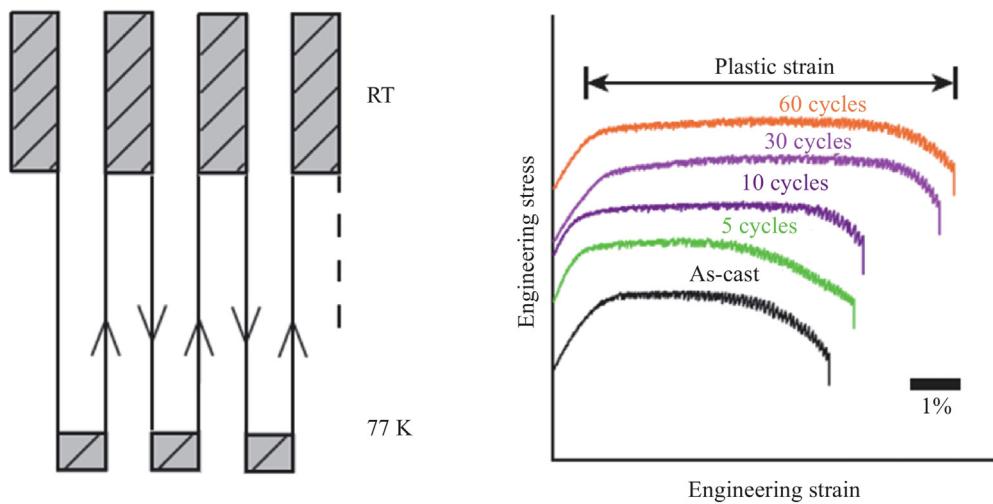


Fig. 3. After cryogenic cycling between 338 K and 77 K, a Zr₆₂Cu₂₄Fe₅Al₉ AA shows an increase in plastic strain under uniaxial compression [27].

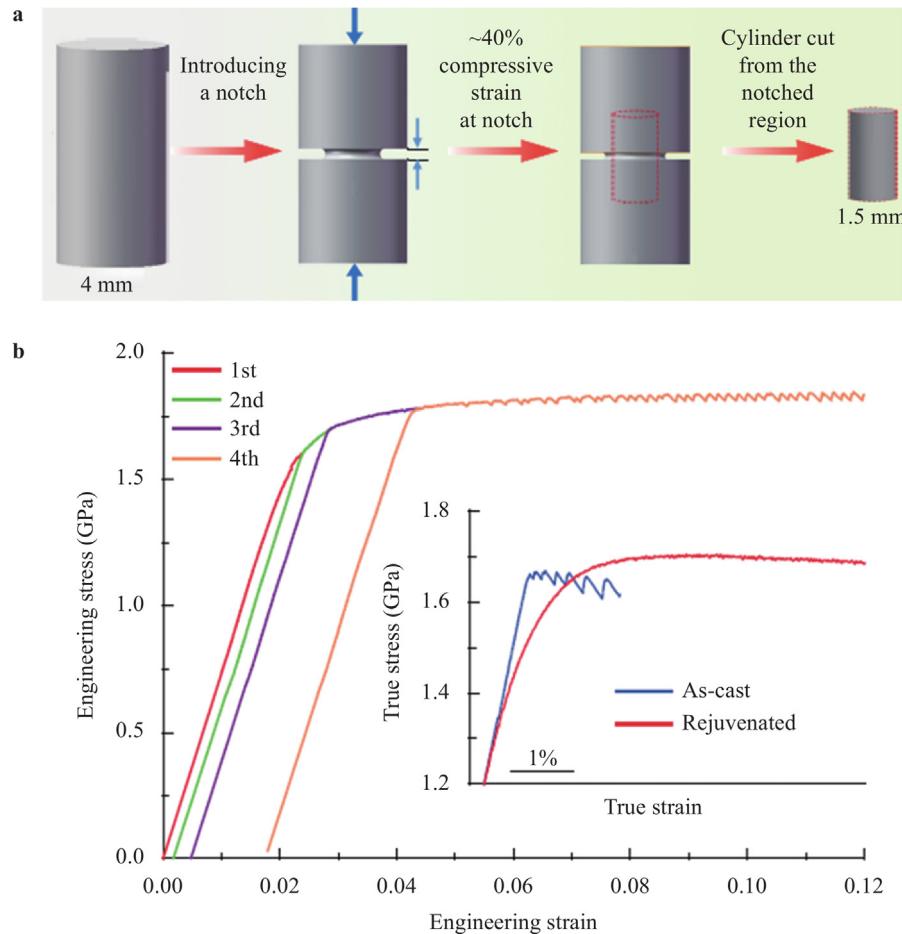


Fig. 4. (a) A Zr_{64.13}Cu_{15.75}Ni_{10.12}Al₁₀ AA rod is machined to have a circumferential notch. Uniaxial compression produces a highly rejuvenated region within the notched rod, from which a cylinder is machined. (b) Compressive stress-strain curves of one sample rejuvenated as in subfig (a) under three unloading-reloading cycles. The inset compares the true stress-true strain curves of a rejuvenated sample and an as-cast one of the same composition [31].

Case 3: S-Ting via metastability engineering.

The single-f.c.c.-phase Fe₂₀Mn₂₀Ni₂₀Co₂₀Cr₂₀, i.e., the so-called Cantor alloy [13], is a case in point, and this alloy is the most successful HEA so far. In 2014, Gludovatz et al. [65] reported a grain-refined Cantor alloy with 6 μm grains. When the temperature decreases from 293 K to 77 K, this alloy shows a syn-

chronous increase in both strength and ductility, as shown in Fig. 5a. But its fracture toughness ($\sim 220 \text{ MPa}\cdot\text{m}^{1/2}$) remains unchanged, which is ascribed to a synergy of deformation mechanisms. In 2016, Li et al. [28] modified the equiatomic Cantor alloy to the non-equiatomic Fe₅₀Mn₃₀Co₁₀Cr₁₀. The latter consists of the f.c.c. matrix (of $\sim 45 \mu\text{m}$ grain size) and the h.c.p. phase laminate layers (ranging from several nanometers to 10 μm in thick-

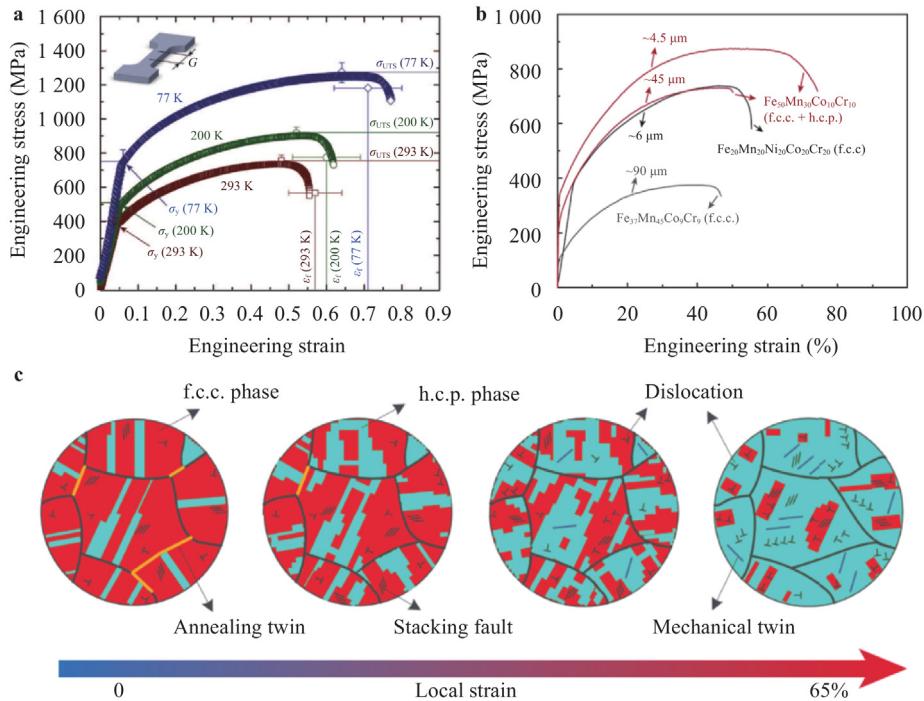


Fig. 5. (a) Tensile stress-strain curves of the $\text{Fe}_{20}\text{Mn}_{20}\text{Ni}_{20}\text{Co}_{20}\text{Cr}_{20}$ HEA with decreasing temperature from 293 K to 77 K; (b) Comparison of tensile properties between metastable dual-phase $\text{Fe}_{50}\text{Mn}_{30}\text{Co}_{10}\text{Cr}_{10}$ and two other single-f.c.c.-phase HEAs; (c) Schematic sketches a synergy of deformation mechanisms in the dual-phase HEA [28, 65].

ness). Despite of distinct topological structures, the two phases are of the same composition distribution even at phase boundaries. Such metastable, dual-phase HEAs exhibit a mechanical behavior almost identical to that of grain-refined Cantor alloy [65], as shown in Fig. 5b. More importantly, on grain refinement to $\sim 4.5 \mu\text{m}$, both strength and ductility of this alloy will further increase remarkably. The excellent combination of strength and ductility stems from the existence of metastable f.c.c. phase where a large number of stacking faults are present. When subjected to external loading, these stacking faults act as the nuclei of martensitic transformation (f.c.c. \rightarrow h.c.p.). This metastable phase transformation can significantly accommodate plastic deformation, and at the same time, effectively activate dislocation slip and mechanical twinning due to increased phase boundary density, as illustrated in Fig. 5c. Recently, Huang et al. [29] have confirmed that such metastability engineering still works well in brittle b.c.c. HEAs. The metastability engineering combines the chemical compositions adjustment with spontaneously changes of the topological disorder upon loading. This strategy suggests that the near-infinite compositional space of HEAs offers a broader room for S-Ting via topological/chemical disorder regulation.

Case 4: S-Ting via chemical short-range order.

In the beginning, researchers take it for granted that the constituent elements of HEAs occupy the lattice sites completely randomly, although their packing structures still keep solid-solution crystals. However, recent studies by simulations and experiments have shown that chemical short-range order (CSRO) inherently exists in HEAs or medium-entropy alloys (MEAs) [66–70]. In other words, some elements due to enthalpic interactions will preferentially form locally clustered domains, usually on nanoscale. These local CSRO domains could roughen dislocation pathways and thus affect their selection in slip, faulting and twinning [67, 69]. This provides a new avenue for S-Ting of MEAs and HEAs.

Recently, such CSRO-induced S-Ting has been successfully achieved in a TiZrHfNb HEA doped with 2 at% oxygen [30]. It is

surprising to find that addition of 2.0 at% oxygen to the TiZrHfNb HEA simultaneously improves strength and ductility, as shown in Fig. 6a. However, doping this HEA with 2 at% nitrogen still presents the usual strength-ductility trade-off. Structural characterizations reveal that the S-Ting of the oxygen-doped HEA is due to the existence of ordered oxygen complexes (OOCs) that are promoted by CSRO. More specifically, oxygen tends to occupy interstitial positions adjacent to Zr and/or Ti-enriched lattice sites in an orderly manner, as shown in the inset of Fig. 6a. These nanoscale oxygen-containing complexes severely distort the local lattice, leading to a large strain field around them. On the other hand, during deformation, these OOCs interact with dislocations, via pinning and promotion of dislocation double cross-slip, as shown in Fig. 6b. Ordered oxygen interstitial complexes change the dislocation shear mode from planar slip to wavy slip, ultimately leading to dislocation multiplication. This is a new S-Ting mechanism, different from either conventional interstitial strengthening or nano-precipitate strengthening [8].

4. Concluding remarks

From the four case studies about S-Ting of AAs or HEAs, we suggest that topological/chemical disordering of alloys provides near-infinite space where the disorder-property relationship can be effectively tailored. These strategies are beyond the simple modification of lattice defects and conventional microalloying, which is thus expected to break the long-standing strength-ductility trade-off in metals. It seems to be exciting and intriguing, but some fundamental mechanics problems should be further clarified to pave the path. We summarize these problems as follows.

(i) **Plastic carry.** In AAs, the basic carriers of plastic deformation are dynamic STs [64, 71–73], i.e., local irreversible rearrangements occurring within zones a few to hundreds of atoms. In the background of long-range topological disorder, the structural origin of STs and their tempo-spatial evolution re-

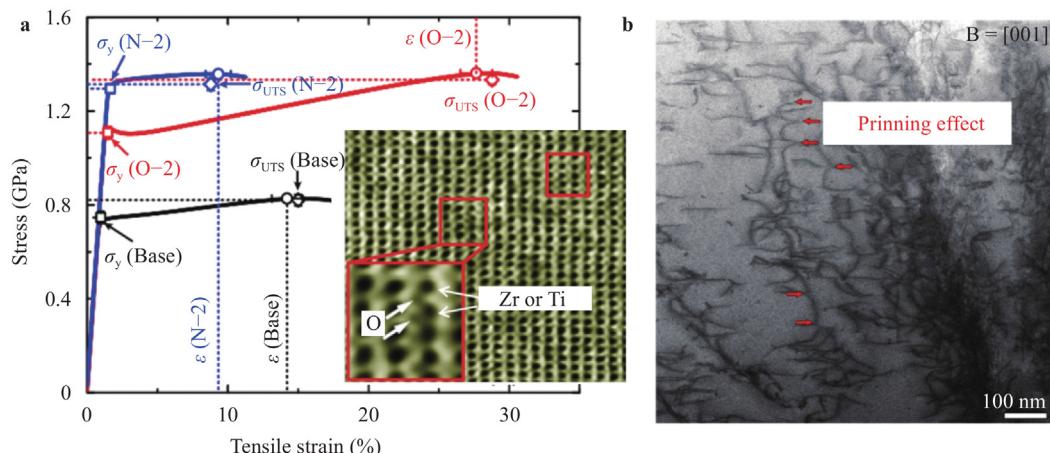


Fig. 6. (a) Room-temperature tensile stress-strain curves of the as-cast TiZrHfNb, (TiZrHfNb)₉₈O₂ and (TiZrHfNb)₉₈N₂ HEAs; The inset shows the existence of ordered oxygen complexes. (b) The dislocation pinning effect in the (TiZrHfNb)₉₈O₂ HEA after being pre-strained to 8% [30].

main mysterious [74–76]. Although lattice defects (e.g., dislocations) can be defined in HEAs, their characterizations face a big challenge. This is because that everything becomes fluctuated, from lattice parameters, Burgers vector, to stacking-fault energies [77–79]. In HEAs, solvent and solute atoms are indistinguishable, which also greatly challenges the conventional solid-solution concept.

- (ii) **Constitutive theory.** To develop constitutive theory for AAs, the first challenge is the intrinsic interaction between vibration and activation, or elasticity and plasticity. Such an interaction is highly nonlinear and occurs in mesoscale [74]. Therefore, we must introduce proper state-variables to describe the two (fast and slow) processes [80, 81]. Moreover, the inherent Reynolds dilatancy [73, 82] of amorphous deformation must be taken into account. For HEAs, their constitutive theory in principle can be developed from the conventional theories of crystalline plasticity [83, 84]. However, the challenge is to consider the structural fluctuation and the resulting multiple plastic mechanisms. In fact, it is necessary to consider the entropy (configurational and vibrational) contribution to deformation in the development of constitutive theory for both AAs and HEAs.
- (iii) **Flow instability.** In either AAs or HEAs, the plastic flow can become tempo-spatially unstable in the form of shear-banding [17, 44], cavitation or microvoid [85–87], necking [32, 88] and flow serration [89, 90]. These flow instabilities, as precursors of cracking, are closely associated with internal transport processes and external loading conditions. In addition to usual momentum and energy transports, the intrinsic transport from disorder should be carefully considered and described. More importantly, these flow instabilities will compete with each other, in which some effects from nonlocal, rate, and inertia should play key roles.

- (iv) **Fracture mechanism.** In which way AAs and HEAs fracture directly determines their S-Ting levels. For crystalline alloys, the crack propagates either by a certain blunt mechanism such as dislocations, or with a sharp tip to cleavage along a proper crystallographic plane. This picture, however, cannot be applied to AAs where any concept based ordered lattices is not well defined. Instead, the crack becomes blunt via ST-mediated shear banding, or advances in a brittle manner by a series of nano-cavitation [85, 91, 92]. The two mechanisms' competition highly depends on the degree of locally topological or chemical disorder at the crack tip. In HEAs, the crack propagation mechanism is also complicated, due to the uncertain selection of crack-tip dislocations via slip, faulting and twinning.

(v) **S-Ting strategy.** For AAs, the S-Ting strategies include multiplication or suppression of shear bands at crack tips. The shear-banding behavior depends closely on the tempo-spatial feature of STs. But the challenging is to determine the relationship between STs and topological/chemical disorder. For HEAs, in addition to CSRO-induced S-Ting mentioned above, some conventional strategies such as second-phase [40], fine-grain [41], solution [19] and nanoprecipitate [93] still work well. But near-infinite compositional space of HEAs provides a huge tailoring room where the disorder-property relationship holds the key of S-Ting.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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