

Hierarchical crack buffering triples ductility in eutectic herringbone high-entropy alloys

Authors: Peijian Shi¹, Runguang Li², Yi Li¹, Yuebo Wen¹, Yunbo Zhong^{1*}, Weili Ren¹, Zhe Shen¹, Tianxiang Zheng¹, Jianchao Peng³, Xue Liang³, Pengfei Hu³, Na Min³, Yong Zhang², Yang Ren⁴, Peter K. Liaw⁵, Dierk Raabe^{6*} & Yan-Dong Wang^{2,7*}

Affiliations:

¹State Key Laboratory of Advanced Special Steel, Shanghai Key Laboratory of Advanced Ferrometallurgy, School of Materials Science and Engineering, Shanghai University, Shanghai, China.

²Beijing Advanced Innovation Center for Materials Genome Engineering, State Key Laboratory for Advanced Metals and Materials, University of Science and Technology Beijing, Beijing, China.

³Laboratory for Microstructures, Shanghai University, Shanghai, China.

⁴X-Ray Science Division, Advanced Photon Source, Argonne National Laboratory, Argonne, IL, USA.

⁵Department of Materials Science and Engineering, The University of Tennessee, Knoxville, TN, USA.

⁶Department Microstructure Physics and Alloy Design, Max-Planck-Institut für Eisenforschung, Düsseldorf, Germany.

⁷Key Laboratory for Anisotropy and Texture of Materials (Ministry of Education), School of Material Science and Engineering, Northeastern University, Shenyang 110819, China.

*Corresponding author. Email: yunboz@staff.shu.edu.cn; d.raabe@mpie.de; ydawang@mail.neu.edu.cn

ABSTRACT

Tolerating cracks in human-made malleable materials is counterintuitive as micro-damage usually limits material lifetime rather than enhancing it. Some composites, such as bone, have hierarchical microstructures that feature crack tolerance, but cannot withstand high elongation. We successfully reconcile crack tolerance and high elongation, demonstrated with a directionally-solidified eutectic high-entropy alloy (EHEA). The solidified alloy has a hierarchically-organized herringbone structure that equips a bionic-inspired hierarchical crack buffering. This effect guides stable, persistent, crystallographic nucleation and growth of multiple micro-cracks in abundant poor-deformability microstructures. The structure helps the cracks avoid catastrophic growth and percolation, due to a hierarchical buffering by adjacent dynamic strain-hardened features. Our self-buffering herringbone material yields an ultrahigh uniform tensile elongation (~50%), three times that of conventional non-buffering EHEAs, without sacrificing strength.

40 Cracks occur in materials if loads cannot be fully dissipated by elastic-plastic work,
41 exposing human lives to risk of failure and integrity loss of safety-critical components
42 (1–6). Some hierarchical composites, such as high-toughness bone (3), feature excellent
43 crack tolerance, but they usually cannot withstand high elongations due to the lack of
44 conventional lattice defects to bear tensile deformation (1–3). By contrast, tolerating
45 cracks in human-engineered formable materials is counterintuitive as extensive cracks
46 tend to trigger the premature failure (6). More specifically, these cracks are generally
47 initiated from the localized severe plastic deformation, and their propagation cannot be
48 effectively buffered and arrested (5–8). This situation is due to the fact that the locally-
49 deformed microstructures are usually not characterized by sufficiently sustainable
50 strain-hardening capability used to relieve locally high stresses at the propagating tips
51 of cracks (1–4). So even though some ductile metallic composites exhibit crack
52 tolerance, only limited additional tensile ductility can be achieved (5–11). Overall, in
53 human-made materials the total elongation in conjunction with crack tolerance is
54 usually not as high as we would expect (5–11). We show that the conflict between
55 extensive crack generation and high uniform elongation can be broken in eutectic high-
56 entropy alloys (EHEAs). EHEAs are a family of recently developed multi-principal-
57 element lamellar composites (12–15). We demonstrate that in EHEAs a herringbone-
58 like hierarchical eutectic microstructure design tends to generate a high density of cracks
59 upon tensile deformation. However, the hierarchical crack buffering prevents them from
60 growing and percolating catastrophically across a huge straining range of ~25%.
61 Consequently, such a high density of cracks is not detrimental to the elongation, but

62 instead can serve as an effective strategy to compensate for the limited tensile ductility
63 of the poor-deformability lamellae. This renders ultrahigh isostrain forming conditions
64 among adjacent lamellae with different deformability, thus achieving a surprisingly high
65 uniform elongation of ~50%. This value is three times that of conventional EHEAs
66 without this type of crack tolerance.

67 We studied two as-cast $\text{Al}_{19}\text{Fe}_{20}\text{Co}_{20}\text{Ni}_{41}$ (at.%) EHEAs (15) fabricated by
68 conventional casting and directional solidification (DS), respectively. The
69 conventionally-cast EHEA served as the reference material and exhibited a typical
70 lamellar microstructure (Fig. 1A) formed during a eutectic transformation (14). The
71 structure is comprised of L_{12} (soft ordered face-centered-cubic) and B2 (hard ordered
72 body-centered-cubic) dual-phase lamellae with varying growth directions in different
73 near-equiaxed grains (Fig. 1B, C). The directionally-solidified EHEA displays a
74 directionally-grown microstructure (Fig. 1D). It consists of columnar grains aligned
75 along the DS direction (Fig. 1E). They contain aligned (grain center) and branched (rims
76 of the grains) eutectic colonies, both of which comprise soft L_{12} and hard B2 lamellae
77 with nano-indentation hardness of ~4.2 GPa and ~5.6 GPa, respectively (Fig. 1E).
78 Lamellae consisting of aligned eutectic colonies (AEC, accounting for ~33 vol.%)
79 generally grow along the DS direction, whereas lamellae comprising branched eutectic
80 colonies (BEC, 67 vol.%) are inclined at 30~60 degrees to the DS direction, and have a
81 more branched morphology. With these features the directionally-solidified EHEA
82 assumes a new type of hierarchically-arranged herringbone microstructure (Fig. 1F).
83 We show that this bone-like structure is formed by the directional growth of cellular

84 solid-liquid interfaces along the DS direction (16–21). The lamellae of both colonies
85 grow perpendicular to the cellular interfaces (Fig. 1I and fig. S1). We did not detect any
86 precipitates or other phases in the dual-phase lamellae by selected-area-electron
87 diffraction patterns and high-resolution high-angle annular dark-field scanning
88 transmission-electron microscopy (HAADF-STEM, Fig. 1G and fig. S1). We confirmed
89 this observation with synchrotron high-energy X-ray diffraction (SHE-XRD, Fig. 1H).
90 Our HAADF-STEM energy-dispersive spectroscopy analysis shows that the DS process
91 has a negligible effect on the chemical composition distribution of the eutectic lamellae
92 (fig. S2). We also obtained three-dimensional stereographic microstructure images of
93 the two as-cast EHEAs (fig. S3). The average width of the columnar grains and its L1₂-
94 phase content are ~54 μm and ~59 vol.% in the directionally-solidified EHEA,
95 respectively. Both values are slightly larger than in the conventionally-cast reference
96 EHEA (grain size ~48 μm and L1₂-phase content ~55 vol.%). These two discrepancies
97 are due to the slower cooling rate of the molten EHEA during DS compared to the
98 condition that during transient solidification when preparing the reference EHEA. The
99 eutectic lamellar spacing of the reference EHEA is ~2.1 μm, which is smaller than that
100 of the AEC (~2.8 μm) and the BEC (mainly varying in the range 3~8 μm). Variable
101 lamellar spacing in the BEC results in significant hardness fluctuations (192~247 HV)
102 compared to the AEC (263 ± 10 HV).

103 We observed a remarkable ductility improvement, quantified from engineering stress-
104 strain curves (Fig. 2A), of the directionally-solidified compared to the conventionally-
105 cast reference material. The uniform elongation tripled, increasing from ~16% for the

reference EHEA to ~50% for the directionally-solidified EHEA. Additionally, the directionally-solidified EHEA has a ~150-MPa higher yield strength than the conventionally-cast EHEA. Although the directionally-solidified EHEA has a high content of hard, low-ductility B2 phase (~41 vol.%) (15, 22, 23), it nonetheless exhibits a large uniform elongation of ~50%. Its elongation is comparable to that of widely-studied, high-ductility, and fully homogenized face-centered-cubic high-entropy alloys (HEAs) (24–27). The resulting strength–ductility combination, and especially the uniform ductility, in the directionally-solidified EHEA outperforms that of any other as-cast eutectic and near-eutectic HEAs (14, 15, 19, 20, 22, 23, 28–33) (Fig. 2B).

We attribute the gain in ductility of the directionally-solidified material to its hierarchical herringbone microstructure and the effect that this structure has on crack buffering, as we revealed with detailed characterizations of the deformation and fracture mechanisms (Fig. 3). The post-fractured specimen surface of the EHEA, which we characterized by scanning-electron microscopy (SEM), shows a large number of micro-cracks (Fig. 3H). The micro-crack density is as large as $\sim 8 \times 10^4 \text{ mm}^{-2}$ in the AEC, and the micro-crack spacing is as small as $\sim 0.83 \text{ }\mu\text{m}$ in some B2 lamellae of the AEC. We do not find the otherwise typical large secondary cracks that are observed in conventional as-cast materials (5) (fig. S5). The micro-cracks we observed are mainly distributed in the hard B2 lamellae of the AEC (Fig. 3H), yet they seem to have ultrahigh stability. The cracks remain strictly confined in the individual B2 phase where they formed, and no crack percolation into neighboring B2 lamellae occurs. This applies even in B2 regions with multiple micro-cracks that are separated by only a single L1₂ lamella

(Fig. 3H). Interestingly, we detected high micro-crack populations already at early and medium strains of up to $\sim 25\%$ (Fig. 3J). Considering the $\sim 50\%$ total ductility, the material features a capability to withstand $\sim 25\%$ more straining after the onset of the first massive crack initiation at modest strains. In this deformation regime, no crack percolation or catastrophic failure event occur irrespective of the extreme abundance of micro-cracks (Fig. 3E–H). This observation substantiates our claim that this alloy has extreme crack tolerance and a crack-mediated ductility reserve of $\sim 25\%$.

To further verify the exceptional crack tolerance, we evaluated the fracture resistance of the directionally-solidified herringbone EHEA by measuring J -integral-based R -curves— J as a function of the stable crack extension, Δa —using single-edge bend specimens in accordance with the ASTM Standard E1820 (34). The crack-resistance (R -curve) behavior (fig. S6A) displays a surprisingly high crack-initiation fracture toughness J_{Ic} , determined essentially at $\Delta a \rightarrow 0$, of $\sim 318 \text{ kJ/m}^2$, and a crack-growth toughness J_{ss} of $\sim 430 \text{ kJ/m}^2$ at a valid crack extension Δa of $\sim 1 \text{ mm}$. These toughness values are over 2 times higher than those in the conventionally-cast EHEA (fig. S6A). These fracture properties are comparable with those of high-toughness gradient-structured materials reported recently (35). Thus, these trends demonstrate extreme crack resistance and damage tolerance of the herringbone EHEA.

To illuminate the underlying mechanisms responsible for the extraordinary crack tolerance, we characterized the dynamic micro-crack evolution in the directionally-solidified herringbone structure (Fig. 3E–H). We revealed an in-situ developing

149 hierarchical interplay between crystallographic micro-crack guidance and crack
150 blunting. We first observed the evolution of dense slip lines on the pre-polished surface
151 of the hard B2 lamellae (Fig. 3E). These slip lines mark regions of strong linear strain
152 localization, stemming from prevalent activation of the primary slip system with highest
153 Schmid factor (35–38). This promotes formation of micro-cracks in the B2 lamellae and
154 their linear crystallographic propagation along these slip lines (Fig. 3E, F). Quantitative
155 analysis of these micro-cracks reveals a rapid increase in crack density in this strain
156 stage (Stage I in Fig. 3J), which subsequently increases modestly towards near-
157 saturation. This behavior differs from the gradually-increasing crack density observed
158 in most common materials (10). Surprisingly, this rapid increase in micro-crack
159 population occurs in a relatively-small intermediate strain range of 25~30%. The
160 resulting micro-crack density is up to $\sim 5.5 \times 10^4 \text{ mm}^{-2}$, which is over twice the density
161 increase found in the subsequent large strain range of 30~50% (Fig. 3J). Interestingly,
162 these multiple cracks do not cause specimen rupture, thus revealing an impressive
163 tolerance of the plastically-deformed material against percolative crack expansion and
164 catastrophic failure.

165 We observed that the micro-cracks emerging in the B2 phase got arrested at the
166 interfaces to the alternatingly adjacent L1₂ lamellae of the AEC (Fig. 3F). These softer
167 L1₂ lamellae serve as soft crack buffers, that act on the tips of micro-cracks, blunt them,
168 and shield the associated high local stresses (10, 35, 39). Without the presence of such
169 alternating soft buffer layers, the cracks would have percolated forward. When the
170 micro-cracks have penetrated an entire B2-lamellar cross section, the neighboring soft

171 L1₂ lamellae blunt the crack tips, as evidenced by their rounded shapes (10) (Fig. 3G).
172 Also, for triggering sample fracture, micro-cracks would not only have to cut through
173 the L1₂ lamellae and thus cut the whole AEC, but also penetrate into the adjacent BEC
174 zone. This means that the herringbone microstructure has two hierarchical levels that
175 feature crack arresting properties, namely, the alternating hard-soft (B2-L1₂) phase
176 layers and the changing alignment of the eutectic colonies. Further three-point bending
177 experiments reveal that this hierarchical effect effectively blunts the crack tip and
178 buffers the crack propagation (fig. S8) (21), thus rendering the directionally-solidified
179 herringbone EHEA with concurrently excellent fracture toughness (fig. S6A) (39).

180 Another important effect is that during tension new micro-cracks are frequently
181 generated in the free intact material portions between these fully-extended cracks within
182 every single B2 lamella (Fig. 3G, H). This crack pattern refinement releases stress
183 concentrations around phase interfaces and weakens the stress intensity at the tips of
184 larger micro-cracks (1–6).

185 Thus, further growth of these fully-extended micro-cracks requires increased
186 mechanical loads for developing them into unstable and critical larger cracks that can
187 cause failure (1–3). However, the overall directional topology of the phases and
188 interfaces means that these micro-cracks can only grow with an alignment towards the
189 tensile (and not transverse) direction in the B2 lamellae of the AEC. Numerous micro-
190 cracks develop in such a fashion assuming stable parallelogram-like shapes (Fig. 3H).
191 We confirmed these experimental observations with high-resolution focused-ion-beam

192 imaging (fig. S5C) and quantitative micro-crack studies (Stage III in Fig. 3J). This stage
193 features a very slow increase in micro-crack density and length. But these micro-cracks
194 in the low-ductility B2 lamellae of the AEC exhibit a surprisingly high capacity to
195 accommodate their shapes and thus carry strains, as revealed by their parallelogram-like
196 morphology (Fig. 3H). Even in this high load regime no interface delamination cracks
197 are found in the herringbone structure. This effect is attributed to their semi-coherent
198 interface structure that can bear high shear stresses (17) (fig. S9).

199 By contrast, in the absence of crack tolerance, early failure occurs in the
200 conventionally-cast EHEA. In the directionally-solidified material, the strain tolerated
201 by the hierarchical herringbone structure, with its high density of cracks in the 25~50%
202 tensile strain regime, exceeds the overall elongation (~16%, Fig. 2A) of the
203 conventionally-cast EHEA. In general, the cumulative crack damage of the material's
204 cross section will gradually reduce its effective load-bearing capability per unit area,
205 thus decreasing the apparent nominal tensile stress (6–11). However, this trend does not
206 appear in the directionally-solidified material (Fig. 2A), despite its high crack density.

207 We investigated the microstructure in detail at the nanoscale to better understand the
208 crack-tolerance mechanisms. Aberration-corrected transmission-electron microscopy
209 (TEM, Fig. 4) shows that the directional movement of massive dislocations in the crystal
210 interior produces the pronounced crystallographic slip-line structures (35–38), also
211 visible on the pre-polished sample surfaces (Fig. 3). The deformation of the B2 lamellae
212 is dominated by planar slip of screw dislocations on $\{110\}<111>$ slip systems, and the

213 $L1_2$ lamellae show planar-dislocation slip on $\{111\}\langle 011\rangle$ (Fig. 4A, B and fig. S10). As
 214 deformation proceeds, the B2 lamellae undergo increasing planar-dislocation shear
 215 (characterized by dramatically-reduced dislocation spacing at tensile strains of 10~25%,
 216 fig. S11H). This leads to a gradual exhaustion of deformability (fig. S11E–H). The
 217 exhausted node corresponds roughly to ~25% tensile strain, thus triggering extensive
 218 micro-crack initiation at the low strain range of 25~30% (Fig. 3E, J) and their
 219 crystallographic propagation along these slip lines inside the B2 lamellae (Fig. 3F, G).
 220 In the $L1_2$ lamellae, however, we found a dynamic substructure refinement governed by
 221 sequentially-activated multi-slip dislocation shear and microband formation (Fig. 4). At
 222 a tensile strain of ~5%, the deformation of the $L1_2$ lamellae is at first dominated by
 223 planar-dislocation slip evolving into a banded shear morphology (Fig. 4A). In these slip
 224 bands, the spacing among adjacent dislocations gradually decreases from the lamellar
 225 interior towards the lamellar interfaces (fig. S11A), thereby establishing a long-range
 226 strain gradient (40, 41). These dislocations, piled up against the interfaces, are known
 227 as geometrically-necessary dislocations (GNDs), creating back-stress hardening (40).
 228 At ~15% strain, these pronounced planar dislocations evolve into well-developed slip
 229 bands (42) (Fig. 4D). Subsequently, we observed deformation-driven refinement of slip
 230 bands. Fresh slip bands are constantly generated in the free space between initially-
 231 existing ones (Fig. 4E). The increasing slip-band density supports substantial dislocation
 232 storage, accompanied by back-stress hardening. By conducting loading-unloading-
 233 reloading experiments, we detected a back stress-dominated high kinematic
 234 strengthening effect (holding about two-thirds of the applied stress, fig. S12) (40, 43).

235 This quantitatively confirms the leading role of back-stress hardening in the $L1_2$
236 lamellae at this stage. Upon further straining we observed dislocations in wavy-slip
237 patterns between slip bands (Fig. 4E). This suggests that dislocation cross-slip is
238 activated, inducing forest dislocation hardening (43) (fig. S12). At higher strains of
239 35~42%, we observed deformation-induced microbands that subdivide the $L1_2$ lamellae
240 into numerous plate-like misoriented domains (43–45) (Fig. 4C, F). Thus, these
241 microbands, analogous to low-angle grain boundaries (not twins, evidenced by Fig. 4F,
242 inset), are associated with further lamellar subdivision and refinement. This mechanism
243 promotes a microband-induced hardening effect, as demonstrated by the surprisingly
244 high strain-hardening rate (Fig. 2A, inset).

245 As we further discovered using low-angle annular dark-field STEM (fig. S13), this
246 dynamic substructure refinement favors substantial local strain hardening in the vicinity
247 of the incoming crack tips (3), thus turning the $L1_2$ lamellae into very efficient crack
248 buffer regions. This phenomenon endows the herringbone structure with excellent crack
249 tolerance (Fig. 3K). Besides the plastic buffering and crack blunting effect, strong strain
250 hardening of the $L1_2$ lamellae (Fig. 4C–F) contributes to the high load-bearing capability
251 of the material. This counteracts the potential softening effect caused by micro-cracks
252 in the B2 phase, as revealed by the fact that the material does not lose but gain tensile
253 strength (Fig. 2A). To provide quantitative insights into the exceptional load-bearing
254 capability, we performed in-situ synchrotron experiments. We observed a real-time
255 stress partitioning effect (46–48) between the B2 and $L1_2$ phases, based on the SHE-
256 XRD results (Fig. 4G). As we expected, the yielding of the $L1_2$ phase leads to stress

257 relaxation and stress transfer to the hard B2 phase. This trend is known from other dual-
258 phase composites (46–48). Stress partitioning to hard phases usually increases
259 continuously until fracture (46). However, in the herringbone structure the B2 phase
260 bears decreasing stress after reaching a tensile strain of ~25%, whereas the L1₂ phase
261 exhibits an opposite trend with remarkable strain-hardening behavior (Fig. 4G). This
262 marks a gradual transfer of the load from the hard but brittle B2 phase to the initially
263 soft but gradually strain-hardened L1₂ phase. This means that the SHE-XRD probing
264 elucidates that the load-bearing capacity of the L1₂ phase increases substantially due to
265 dynamic substructure refinement, while that of the B2 phase decreases gradually caused
266 by increasing internal micro-cracking. To compensate the limited TEM-sampling area,
267 we also conducted 2D SHE-XRD diffraction investigations (48), covering the entire
268 360°-azimuthal range recorded for different planes near the point of fracture (Fig. 4H).
269 No new diffraction lines are detected, suggesting that the high load-bearing capability
270 of the herringbone structure is not caused by conventional phase transformation.
271 However, the diffraction lines of the B2 phase (e.g., marked by red lines) deviate
272 severely from their initial angle. This is caused by the huge elastic strain of the B2 phase
273 (up to ~5.5%, fig. S14). This observation is also revealed by the conventionally-cast
274 EHEA, which shows delayed dislocation shear in the B2 lamellae (fig. S15).

275 When further exploring the hierarchical herringbone structure and its microstructural
276 evolution during tensile deformation, we identified a slip-line-mediated sequential
277 deformation transfer from the soft BEC to the hard AEC (Fig. 3A–D) (21). This finding
278 reveals that the BEC features high deformability that allows compatible deformation of

adjacent columnar grains (Fig. 3D). At the early deformation stage, dense dislocation
pile-up arrays against phase interfaces were observed in the L1₂ lamellae of the BEC
(Figs. 3A, 4A). The associated pile-up stresses are accommodated by the elastic
deformation of the plastically-less compliant B2 lamellae (Fig. 3B), thereby shielding
high stress concentrations (48), a mechanism which supports compatible co-
deformation of adjacent phases and colonies (49). The large local misorientation shown
in the kernel average misorientation (KAM) map (Fig. 3I) confirms that the plastic strain
incompatibility can be well accommodated in the BEC. As the deformation progresses,
stable micro-cracks, as observed in the AEC, can also be generated in the low-ductility
B2 lamellae of the BEC, assisting its compliance (fig. S16). These mechanisms of
mechanical energy release in the adjacent phases and colonies reduce the chance of
grain-boundary decohesion or cracking (40, 49) (fig. S5B), unlike accessible in
conventionally cast EHEA. The statistically-distributed lamellar arrangements in the
differently-oriented grains cannot sustain a compatible co-deformation among them
(21). Consequently, grain-boundary cracking concomitant with mechanical instability
triggers premature failure (i.e., deteriorating tensile ductility) (fig. S17–19) and also
limits the fracture toughness level of the conventionally-cast material (40, 49) (figs. S6,
7).

Natural materials with high toughness often also comprise hard and soft components
in hierarchical layered architectures (50–52). The lamellar cortical bone, a prime
example, consists of mineralized collagen fibrils and a non-fibrillar organic matrix,
which acts as a ‘glue’ that holds the mineralized fibrils together (50–52). Healthy

301 lamellar bone resists fracture through complementary intrinsic and extrinsic
302 contributions throughout its hierarchical structure (50, 52). The glue-mediated fibrillar
303 sliding mechanism, analogous to the dislocation-assisted inelastic deformation in our
304 herringbone EHEA, is essential to promote high plasticity (50). In both materials,
305 plasticity and the resultant ductility provide a major contribution to the intrinsic
306 toughness by dissipating energy and forming plastic zones surrounding incipient cracks,
307 which further serves to blunt crack tips, thereby reducing the driving force for cracking
308 (3, 50). The extrinsic mechanisms, such as collagen-fibre bridging and crack deflection,
309 act principally on the wake of cracks to reduce (shield) the local stresses/strains
310 experienced at crack tips and inhibit their propagation (50). These effects exhibit a
311 marked similarity to what we identified in our herringbone material (see Fig. 3E–H and
312 fig. S8). Of course, a salient characteristic of bone is its ability to remodel itself to heal
313 and repair damage—a trait that is difficult to replicate in our synthetic materials (50,
314 51).

315 In summary, we presented a hierarchical microstructure design approach, realized in
316 a directionally-solidified bulk EHEA, that allows reconciliation of crack tolerance and
317 high uniform elongation, features that are usually mutually exclusive in both, human-
318 made and biological materials. The crack tolerance is maintained over a huge range of
319 ~25% tensile elongation and enables to triple the ductility of the material by a factor of
320 3 relative to conventionally solidified material, without sacrificing strength. These
321 proposed mechanisms exhibit practical merits in guiding a broader group of eutectic-
322 type cast HEA and traditional alloy development. The microstructure approach can be

323 potentially realized also in other bulk materials consisting of hard and soft phases that
324 can be rendered into hierarchically organized herringbone microstructures, enabling the
325 design of crack-tolerant yet high-deformability materials not by avoiding cracks but by
326 guiding and buffering them. Furthermore, this hierarchical herringbone microstructure
327 design approach and the salient effect it has on crack buffering show a promising
328 guidance in designing not only new hierarchically-structured alloys with high
329 elongation but also new bone substituting biomaterials with excellent fracture toughness.

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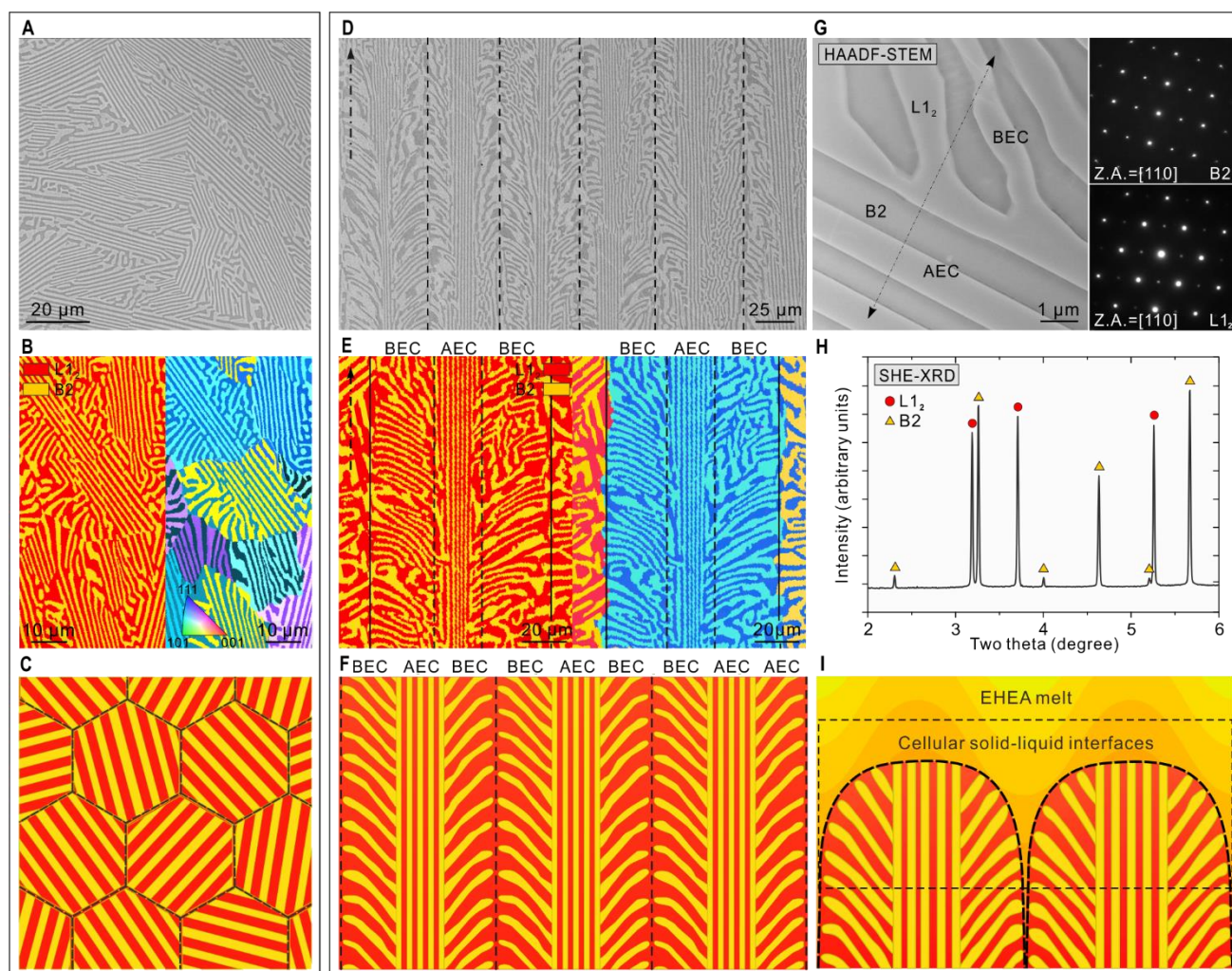


Fig. 1. Hierarchically-arranged herringbone microstructure. (A to C) Conventionally-cast EHEA serving here as reference material. (A) Scanning-electron microscopy (SEM) backscattered electron image. (B) Electron back-scattering diffraction (EBSD) phase map (left) and inverse-pole-figure (IPF) map (right). (C) Schematic diagram. (D to I) The directionally-solidified EHEA with a hierarchical herringbone microstructure. The black arrows in (D) and (E) indicate the DS direction, and also the tensile loading direction in Fig. 2A. (D) SEM backscatter electron image showing that the microstructure is composed of columnar grains. Grain boundaries are marked by black dotted lines. (E) Enlarged EBSD phase and IPF maps showing the

492 columnar grain consisting of AEC and BEC. Black solid and dotted lines mark grain
493 and colony boundaries, respectively. (F and I) Schematic diagram of herringbone
494 structure and its formation principle, respectively. (G) HAADF-STEM image and
495 related selected-area electron diffraction patterns of B2 and L1₂ phases. The HAADF-
496 STEM image shows clean dual-phase lamellae without evidence of nanoprecipitates or
497 other phases, which is also indicated in (F). (F) SHE-XRD of B2 and L1₂ phases.

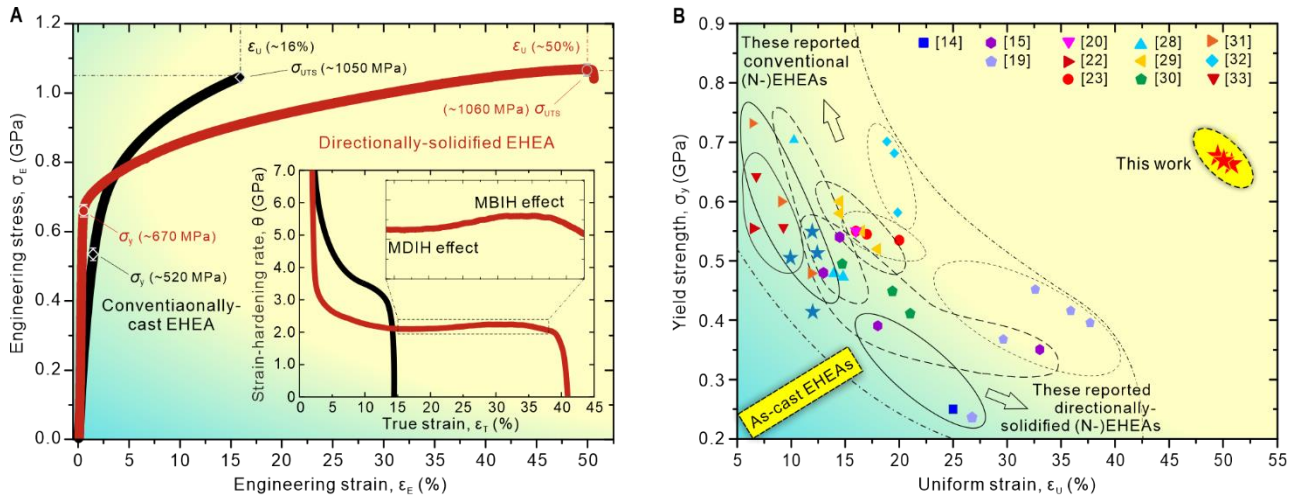


Fig. 2. Tensile response at ambient temperature. (A) Engineering stress–strain curves of the directionally-solidified EHEA compared with the conventionally-cast EHEA, displaying a substantial increase in uniform tensile ductility without any strength reduction. The directionally-solidified EHEA shows no post-uniform ductility, which is also confirmed by the absence of macroscopic necking at the fracture end in the inset of fig. S5A. Tensile loading was performed along the DS direction. Inset, the corresponding strain-hardening curves. MDIH and MBIH refer to multi-slip dislocation-induced hardening and microband-induced hardening, respectively. (ϵ_U) uniform strain. (σ_y) yield strength. (σ_{UTS}) ultimate tensile strength. (B) Yield strength versus uniform strain of the directionally-solidified EHEAs compared with those of previously reported as-cast eutectic and near-eutectic HEAs (14, 15, 19, 20, 22, 23, 28–33). These blue pentagrams indicate some of our recently-designed eutectic and near-eutectic HEAs (unpublished). (N-)EHEAs refers to eutectic and near-eutectic HEAs. The conventional (N-)EHEAs include the directly-cast and the arc-melting eutectic and near-eutectic HEAs.

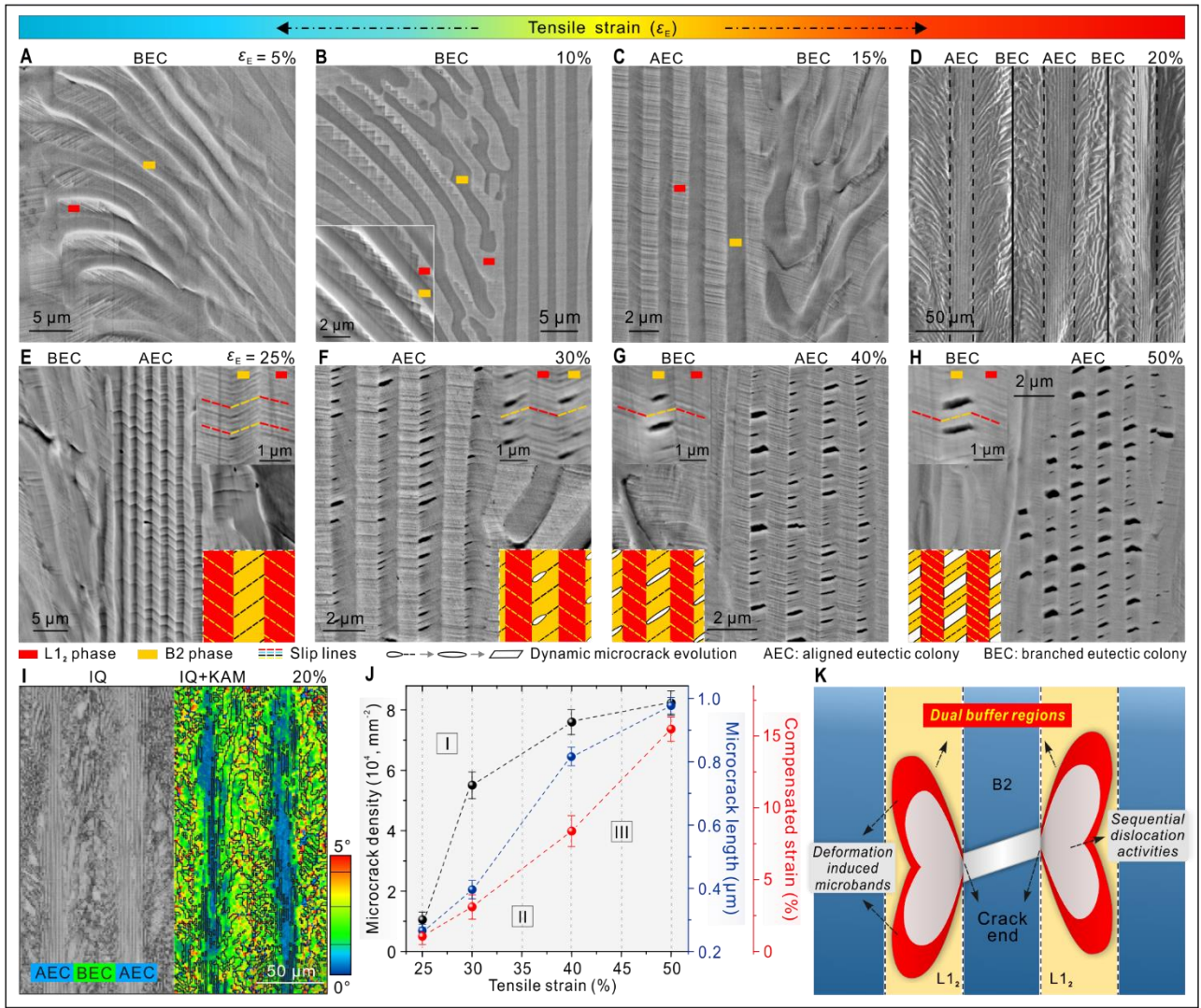


Fig. 3. Hierarchical crack buffering. (A to C) SEM backscattered electron images showing sequentially-activated slip lines from soft BEC to strong AEC. The inset in (B) shows enlarged cross-slip lines. (D) SEM image exhibiting the compatible deformation between adjacent columnar grains and no grain-boundary cracks. Black solid and dotted lines mark grain and colony boundaries, respectively. (E to H) Well-controlled micro-crack evolution in the AEC. These (upper) enlarged and (lower) colored insets illustrate dynamic micro-crack evolution. (I) EBSD-based image quality (IQ) and IQ with kernel average misorientation (KAM) maps. The KAM is calculated up to the fifth neighbour shell with a maximum misorientation angle of 5° , which is indicative of the deformation

524 degree. **(J)** The evolution of micro-crack length, micro-crack density, and compensated
525 strain in the AEC. Stages I–III correspond to tensile strains of 25–30%, 30%–40%, and
526 40%–50%. The error bars are standard deviations of the mean. **(K)** Schematic diagram.

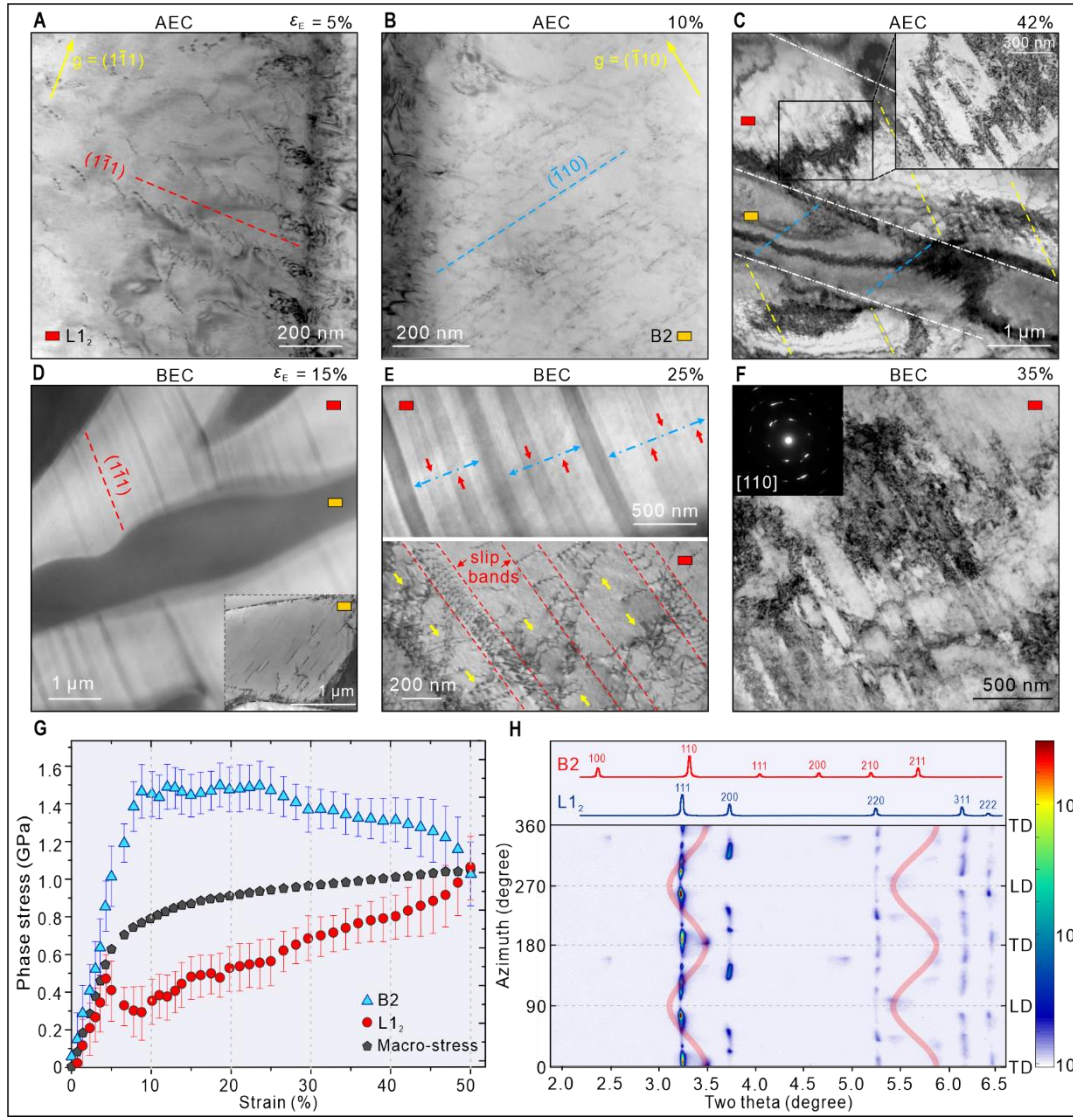


Fig. 4. Microstructural and micromechanical observations of load bearing response for L1₂ lamellae. (A and B) Planar-slip dislocations in L1₂ and B2 lamellae, respectively. (C) Deformation-induced microbands in L1₂ lamellae. The inset exhibits clearer microband structure. (D and E) Dynamic slip band refinement (D and Upper E shown by HAADF-STEM) and fresh slip bands and cross-slip dislocations marked by red lines and yellow arrows, respectively (Under E). (F) Deformation-induced microbands. The ring-like selected-area electron diffraction pattern (inset) suggests that these microbands are similar to low-angle grain boundaries (rather than mechanical twins). The beam directions are $[011]$ in (A, D and E) and $[001]$ in (B). g indicates the

537 direction of the diffraction vector. (ϵ_E) tensile strain. **(G)** Real-time stress partitioning
538 of B2 and L1₂ phases (i.e., σ_{B2} and σ_{L12}) during tensile loading (21). **(H)** Selected 2D X-
539 ray diffraction images along the full azimuthal angle η (0°–360°) at tensile strain of
540 ~48%. Note that 90° and 180° correspond to the loading direction (LD) and transverse
541 direction (TD), respectively.