Dual-gradient structure leads to optimized combination of high fracture resistance and strength-ductility synergy with minimized final catastrophic failure

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ABSTRACT

Nature-inspired gradients can be implemented in metallic materials to achieve a synergy of strength and ductility. However, due to the small (often microscale) size of the gradient structured samples, their fracture properties have remained relatively unexplored. By fabricating centimeter-sized gradient-structured pure nickel samples using direct-current electroplating technique, we demonstrate that a dual-gradient architecture in pure nickel, comprising grain-size transitions from coarse grains to nano grains and then back to coarse grains (CG→NG→CG), achieves an optimized combination of strength-ductility synergy and exceptional fracture resistance – a crack-initiation toughness exceeding 300 MPa m½ – while minimizing the problem of final unstable catastrophic failure. Significantly, this dual-gradient CG→NG→CG structure can effectively arrest any brittle fracture in the nano grains by inducing a stable rising R-curve with an enhanced crack-growth toughness exceeding 350 MPa m½. We believe that this dual-gradient CG→NG→CG structure provides a promising prototype for designing multi-layer graded structures with exceptional combinations of mechanical properties which can be readily tuned to meet the advanced requirements of safety-critical applications.

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1. Introduction

Technological advancement in engineering systems has imposed unprecedented demands for superior mechanical performance. This has motivated material scientists to seek for ways to improve mechanical properties of structural materials. A unique strategy that Nature devises to enhance mechanical properties is through the generation of gradients [1–6]. For example, the density of vascular bundles in the
bamboo stems decreases from the exterior inwards to the center, creating a gradient structure that enhances strength and stiffness simultaneously [7–9]. Recent attempts have utilized the concepts of gradients in metallic materials to achieve a favorable synergy of strength and ductility [10–17] as well as other material performance, such as fatigue and wear resistance [18–22]. However, these replicated gradients have invariably been generated in small-sized samples or within a relatively shallow sub-surface depth, invariably sized in the scale of micrometers; accordingly, the specific effects of such gradients on the fracture toughness have rarely been able to be explored. As the toughness represents a vital service performance of structural materials, its study and the resulting effect on safety is of considerable significance to the engineering application of such graded materials.

We have fabricated centimeter-sized samples of pure nickel with gradients in grain size from the nano-to-microscale utilizing a direct-current (DC) electroplating technique [16], and, for the first time, gradient effects on the variation in fracture toughness have been systematically investigated [23]. Interestingly, the fracture resistance of gradient structures is a marked function of the crack-growth direction. The crack-initiation toughness is far higher in the gradient direction of coarse-grains (CG) to nano-grains (NG), i.e., in CG→NG gradient structure. However, unstable crack propagation can occur in these CG→NG structures as the crack encounters the NG zone. In contrast, the NG→CG gradient structure, which has a significantly lower initiation toughness, is less vulnerable to outright fracture. Accordingly, neither of these single gradient structures is ideal for structural applications where safety is a major concern. This poses an intriguing question - whether one can design a gradient architecture that possesses a high crack-initiation toughness but with a rising crack-growth resistance R-curve to inhibit unstable brittle fracture, all without compromising the good combination of strength and ductility?

To tackle this, we have developed a simple dual-gradient architecture in pure nickel in bulk (centimeter-sized) samples which encompasses gradients transitioning from coarse grains (~4 μm) to nano-grains (~30 nm), and back to coarse grains (~4 μm), i.e., a dual-gradient CG→NG→CG structure. We show that this CG→NG→CG structure achieves an optimized combination of strength-ductility synergy and exceptional fracture resistance with a high crack-initiation toughness coupled with a rising R-curve to suppress catastrophic fracture. This dual-gradient structure represents sound microscale architecture for structural materials displaying superior crack resistance for crack growing in either direction without compromise in global strength and ductility.

### 2. Experimental methods

#### 2.1. Materials

Single-gradient structured and dual-gradient structured Ni plates with a thickness of 1.4 mm were synthesized by a DC electrodeposition technique, as shown in Fig. 1a and in more detail in Fig. S1 in the Supplementary Materials. By increasing the current density from 10 to 100 mA cm⁻² and the additive concentration (sodium saccharin) from 1 to 6 g/L, the grain size can be continuously refined from ~4 μm to ~30 nm (Fig. S1). In order to fabricate centimeter-sized single edge-notched bend SE(B) specimens for fracture toughness testing, 1.6 mm-thick layers of monolithic nano-grained Ni were further coated to the single- and dual-gradient structured Ni plates on both ends, forming a sandwich plate with a final dimension of 60 × 30 × 4.6 mm³ (Fig. 1a). For comparison, pure nano-grained (NG) and pure coarse-grained (CG) SE(B) specimens were prepared by replacing the gradient structured (GS) layer of the sandwiched plate with uniform-grained NG and CG Ni having grain sizes of ~30 nm and ~4 μm (Fig. 1b). All the sandwich-structured plates were annealed at 393 K for 12 h to relax the residual stress induced by electrodeposition before wire electrical-discharge machining the SE(B) specimens.

#### 2.2. Mechanical characterization

To determine the tensile properties of the monolithic and gradient structures, dog-bone plate specimens with a gauge length of 5 mm and a gauge cross-section of 2 × 0.5 mm² were machined from the monolithic and gradient Ni plates. The loading direction was aligned perpendicular to the deposition direction. Uniaxial tensile tests were performed at room temperature by Instron 5848 Micro-Tester system (Instron Corporation, USA) at a strain rate of 5.0 × 10⁻⁴ s⁻¹. A contactless laser extensometer (MTS LX300) with displacement resolution of 1 μm was used to measure the engineering strain within the gauge section during the tensile test. At least three specimens were tested for each gradient and monolithic structure.

To evaluate the fracture properties of the monolithic and gradient structures, SE(B) specimens, with thickness of B = 2 mm, width of W = 4 mm and total length of 24 mm, were extracted from the gradient/monolithic sandwich plates (Fig. 1a and b). Notches, 1.4 mm in length with notch root radii of ~100 μm, were cut with their orientation aligned perpendicular to the gradient or monolithic ligament. The cracking direction in the CG→NG specimens translates from the coarse-grained (~4 μm) zone to the nano-grained (~30 nm) zone, and vice versa for the NG→CG specimens. For the CG→NG→CG specimens, the crack was propagated from coarse grains to nano grains and then back to coarse grains. All specimen surfaces were mechanically polished down to 1-μm mirror finish.

Fracture toughness tests were performed in accordance with ASTM Standard E1820 [24]. Fatigue pre-cracking was carried out using a 3 kN MTS fatigue testing machine (MTS Corporation, USA), operated under a stress intensity range of ΔK = Kmax - Kmin of 8–11 MPa m¹/² at a constant frequency of 10 Hz with a load ratio R = Kmin/Kmax = 0.1. The initial crack size, a0, i.e., the notch length plus the fatigue pre-crack length, was produced in the range of 0.45 ≤ a0/W ≤ 0.55. To improve the constraint conditions at the crack tip, all the specimens were side-grooved using diamond wire cutting to depths of 0.2 mm, which resulted in a net specimen thickness of B0 of 1.6 mm. This thickness reduction did not exceed 0.25B, as mandated by ASTM Standard E1820 [24]. Nonlinear elastic-fracture mechanics involving R-curve measurements, characterized in terms of the J-integral as a function of crack extension Δa, were used to evaluate the fracture toughness of the gradient-structured Ni. Details of the
methods used for these R-curve measurements are described in the Supplementary Materials.

Because of the ductility of Ni, several of the measured fracture toughness values were strictly invalid in terms of ASTM Standard E1820 [24] as described in the footnote to Table 1. Although we believe that our measured $J_{IC}$ and $K_{IC}$ toughness values still reflect the ranking of the fracture resistance of the various uniform and gradient samples tested, we additionally measured the widely-used work of fracture (i.e., plastic work density, $\omega_f$) from the uniaxial tensile tests, using $\omega_f = \int_0^{\epsilon_f} \sigma \, d\epsilon_p$, from the area under the true stress-plastic strain curve, where $\epsilon_{pf}$ is the plastic strain corresponding to the elongation to failure, $\epsilon_f$. This provides an alternative, albeit approximate, measure of the toughness.

2.3. Material characterization

Prior to mechanical testing, the monolithic and the gradient microstructures in the 1.4-mm wide ligament of the SE(B) specimens were characterized using a Zeiss Supra 55 scanning electron microscope (SEM) operating at a voltage of 20 kV in the backscatter electron (BSE) imaging mode. To obtain the grain-size distributions along the gradient/monolithic ligaments, we characterized the microhardness profile along the deposition direction and calculated the grain size, $d_g$, by means of the Hall–Petch relationship ($HV = 1.9346 + 16.79 \, d_g^{(1/2)}$), using the method described in [23]. The microhardness $HV$ profile was measured by a Qnes Q10 A+ microhardness tester using a Vickers indenter with a peak load of 50 g and a dwell time of 10 s. The determination of the grain-size distribution from the microhardness profile was verified by direct X-ray diffraction (XRD) measurements (for grain sizes below ~3 μm) and SEM imaging (for grain sizes above ~3 μm) (Fig. S2) [23]. Further verification was conducted for grain sizes below ~1 μm by transmission electron microscopy (TEM) characterization (Fig. S3c).

To discern the deformation mechanisms in the vicinity of the crack tip and crack wake under plane-strain conditions, the crack–path profile was examined along the mid-thickness section of the test specimens. Certain SE(B) specimens were unloaded from different loading levels and sliced through the thickness into two halves. The interior surface of one half was progressively polished to a 0.05-μm surface finish, followed by a final vibration polish using 0.05-μm colloidal silica and an
electrical polish using electrolyte consisting of perchloric acid, glacial acetic acid, and alcohol with a volume ratio of 1:3:4, respectively. The microstructure along the crack wake and the crack-tip region were imaged using backscattered electrons in a Zeiss Supra 55 SEM operating at 20 kV.

3. Results

3.1. Microstructure

Centimeter-sized SE(B) specimens for fracture toughness testing were fabricated from monolithic- and gradient-structured sandwich Ni plates synthesized by a DC electrodeposition technique (Fig. 1a). Back-scattered electron (BSE) images taken from the graded ligaments (Fig. 1c) clearly revealed that the gradients in grain size transition smoothly from the coarse grains to nano-grains, nano-grains to coarse grains, and coarse grains via nano-grains to coarse grains, respectively for the CG/NG, NG/CG and CG/NG/CG materials. Uniform NG and CG structures were also characterized by BSE imaging (Fig. 1c). The uniform coarse-grained structure comprised columnar crystal grains with a size of ~30 μm in the crack-propagation direction (i.e. the electrodeposition direction) and ~4 μm in the transverse direction (Figs. S3b and d).

Figure 1d presents the grain-size distribution profiles characterized along the crack-propagation ligaments of the monolithic and gradient SE(B) specimens. The measured grain size can be seen to continuously decrease from ~4 μm to ~30 nm in the CG→NG specimen, whereas it increases from ~30 nm to ~4 μm in the NG→CG specimen. The grain size in the CG→NG→CG specimen decreases from ~4 μm to ~30 nm and then increases to ~4 μm again. Specifically, the grain sizes within the initial and final 400-μm regions of these gradient

| Table 1 – Tensile and fracture properties of the monolithic (pure NG and pure CG) and single- and dual-gradient structured Ni at room temperature. |
|-------------------------------------------------|
| Yield strength, σy (MPa) | NG | CG | NG→CG | CG→NG | CG→NG→CG |
|--------------------------|-----|----|------|------|----------|
| 1095 ± 66                | 383 ± 3 | 687 ± 59 | 687 ± 59 | 499 ± 21 |
| Tensile strength, σuts (MPa) | 1437 ± 50 | 592 ± 4 | 1094 ± 38 | 1094 ± 38 | 738 ± 20 |
| Elongation to failure, εf | 6.6 ± 0.1% | 14.2 ± 0.6% | 11.0 ± 0.4% | 11.0 ± 0.4% | 16.9 ± 1.7% |
| Plastic work density (or work of fracture), ωf (MJ.m⁻³) | 79.0 ± 2.1 | 72.4 ± 4.4 | 103.4 ± 7.4 | 103.4 ± 7.4 | 113.5 ± 9.9 |
| Provisional J-integral at crack initiation, J₀ (kJ.m⁻²)* | 13.2 | 396.0 | 33.4 | 338.6 | 459.2 |
| Provisional fracture toughness at crack initiation, KIC (MPa.m¹/²) | 54.1 | 296.2 | 86.0 | 273.9 | 319.0 |
| Crack growth J-integral at Δa = ~1 mm, Jₐ (kJ.m⁻²) | 66.3 | 442.5 | 231.9 | 436.4 | 618.2 |
| Crack growth toughness at Δa = ~1 mm, Kₐ (MPa.m¹/²) | 121.2 | 313.1 | 226.7 | 310.8 | 370.1 |
| ASTM “valid” * | | | | | |
| ASTM “invalid” * | | | | | |

*According to ASTM Standard 1820 [24], for the provisional toughness J₀ to be considered as a size-independent fracture toughness (JIC), the validity requirements for the J-field dominance and plane-strain conditions shall be respectively met, i.e., that b₀; B > 10J₀/σ₀, where the b₀ and B are the initial ligament length and the specimen thickness, respectively. The flow, or effective yield, stress, which is the mean of the yield and tensile strength: σ₀ = ½ (σy + σuts), is 1266 MPa, 488 MPa, 891 MPa, and 619 MPa for the NG, CG, single-gradient and double-gradient samples, respectively. The calculated 10(J₀/σ₀) values for NG, CG, NG→CG, CG→NG, and CG→NG→CG samples are 0.1 mm, 8.11 mm, 0.37 mm, 3.8 mm, and 7.42 mm, respectively. The J₀ and KIC of NG and NG→CG samples satisfy the specimen size requirements, b₀; B > 10J₀/σ₀, and thus they are regarded as ASTM valid J₀ and KIC values. CG, CG→NG, and CG→NG→CG samples are close, but do not strictly meet the ASTM validity requirements.
profiles remain constant, indicating that there is a uniform grain size at the beginning and end of each grain-size gradient. The CG→NG and NG→CG specimens exhibit completely identical grain-size distribution but in opposite directions. The grain-size distribution of the CG→NG→CG specimen can be regarded as a combination of those from the CG→NG and NG→CG specimens but with a steeper gradient slope.

3.2. Tensile properties

Representative engineering stress–strain curves obtained from uniaxial tensile tests on the monolithic (pure NG and pure CG) and single- and dual-gradient structures are presented in Fig. 2a. The tensile properties are summarized in Table 1. Results show that the tensile strength ($\sigma_{uts}$) of the single-gradient structure at ~1094 MPa is higher, by 85%, than that of pure CG structure (where $\sigma_{uts} = 592$ MPa), but lower, by 24%, than that of pure NG structure (where $\sigma_{uts} = 1437$ MPa). Compared to single gradient structure, the tensile strength of the dual-gradient structure at ~738 MPa is 33% lower, but still higher, by 25%, than that of uniform CG structure. In contrast to the tensile strength, the total elongation ($\epsilon_f$) of the single-gradient structure at ~11% is 23% smaller than that of the CG structure (where $\epsilon_f \sim 14.2\%$), but 67% larger than that of the uniform NG material (where $\epsilon_f \sim 6.6\%$). Most significantly, the dual-gradient structure exhibits the highest total elongation at ~16.9%, which is respectively 156%, 19% and 54% higher than those of pure NG, pure CG, and single-gradient structures.

The trade-off between strength and ductility is clearly shown for the uniform NG and CG structures, where the increase of strength with a finer grain size is attained at a cost of reduced ductility. However, consistent with results for a range of gradient- and heterogeneous-structured materials [10–17], the combination of strength and ductility is enhanced in our single-gradient structure, yet is more significantly enhanced with a dual gradient. The optimized combination of strength and ductility can be clearly seen in the increase in the plastic work density (or the work of fracture), i.e. the area under the true stress–plastic strain curve, which is increased from ~79 MJ m$^{-3}$ and ~72 MJ m$^{-3}$ respectively in uniform CG and NG specimens to 103 MJ m$^{-3}$ and 114 MJ m$^{-3}$ respectively in the single- and dual-gradient structures. This result strongly indicates that in general gradient structures can give rise to a favorable synergy of strength and damage tolerance.

3.3. Crack-resistance curves and fracture toughness

To evaluate the fracture resistance of the gradient- and monolithic-structured Ni, we applied nonlinear-elastic fracture mechanics analysis to measure the J-integral based R-curves, i.e., $J$ as a function of the stable crack extension, $\Delta a$. The $J$-$\Delta a$ curves for the uniform CG and NG Ni and the single-gradient (CG→NG and NG→CG) and dual-gradient (CG→NG→CG) Ni are summarized in Fig. 2b. Rising R-curve behavior is seen in all the microstructures, although the extent of toughening varies significantly in the five different structures.

The uniform CG and NG structures exhibit typical “ductile” and “brittle” R-curve behavior (see comment in the Supplementary Materials), where the CG structure shows a high initiation toughness almost 4.5 times higher than that of the NG structure. The crack-driving force $J$ to sustain a crack extension of $\Delta a \sim 1$ mm is ~443 kJ m$^{-2}$ in the CG structure; this is almost six times higher than the $J_{\text{req}}$ ~66 kJ m$^{-2}$ that is required for simple crack extension in the NG structure.

The R-curve behavior of the single-gradient structures are quite different. Compared to the NG structure, where the R-curve slope, which is a measure of the crack-growth toughness, remains almost constant but is extremely shallow, the NG→CG structure displays a higher fracture resistance as crack extends. Moreover, as shown in Fig. 2b for this single-gradient structure, the slope of the R-curve gradually increases once crack enters the gradient zone.
Unlike the substantial enhancement in fracture resistance of the NG→CG structure compared to the NG structure, the gradient CG→NG structure shows only marginally improved fracture resistance compared to the uniform CG structure (Fig. 2b); the advantage of gradient structure in this case is its much higher strength properties (Fig. 2a). The initial R-curves of the CG and CG→NG structures, for crack extensions Δa < 0.2 mm, are essentially identical, as would be expected; both structures exhibit high crack-initiation toughness. Beyond Δa ~0.2 mm, a slight rise in the R-curve of the CG→NG structure can be observed; however, this is terminated abruptly at Δa ~0.33 mm due to a rapid unstable cracking through the fine-grained region at the end of the gradient out to Δa ~1 mm. Although the J-integral toughnesses at Δa ~1 mm of the CG→NG and CG structures are similar, respectively 436 and 443 kJ m⁻², the catastrophic final fracture behavior of the CG→NG structure is highly undesirable.

As listed in Table 1, both single-gradient structures provide a far improved balance of strength and toughness to either of the uniform grained-sized structures. Nevertheless, the toughness of the NG→CG structure is still relatively low, whereas the CG→NG structure has a high crack-initiation toughness but is susceptible to eventual unstable fracture in the finer grains. Accordingly, our objective here was to develop a structure with high strength, ductility and toughness but which was not prone to eventual “brittle” fracture, i.e., a structure more appropriate for safety-critical applications. This combination of properties, however, can be achieved with the dual-gradient CG→NG→CG structure. In addition to combining sound values of strength and ductility (Fig. 2a), the CG→NG→CG structure exhibits a crack-initiation toughness superior to all the other structures (uniformed grained and single-gradient) (Table 1), with a sharply-rising initial R-curve for stable crack extensions out to Δa ~0.4 mm (Fig. 2b). Subsequent crack extension, for -0.4 mm < Δa < -0.8 mm, involves a short period of brittle propagation within the central region of nano-grains — this is indicated by the dashed purple arrow in Fig. 2b — before further crack extension into the second region of coarse grains results in a rising R-curve again with no final catastrophic failure. By assessing the crack-growth toughness in terms of the J-based crack-driving force at Δa ~1 mm, the measured J of ~618 kJ m⁻² for this dual-gradient structure is higher than those of the CG→NG, CG, NG→CG, and NG structures, respectively by ~42%, ~40%, ~166%, and ~836%.

Stress-intensity based fracture toughness values for all structures were estimated using the mode-I J-K equivalence from the critical value of J at crack initiation, i.e., at the intersection of the R-curve with the 0.2-offset line. As summarized in Table 1, the CG→NG→CG structure displays the highest JKI toughness of ~319 MPa m⁵ of all tested structures. This dual-gradient toughness was, respectively, ~16% and ~8% higher than that estimated for the CG→NG and CG structures, and almost 2.7 and 5 times higher than the corresponding values for the NG→CG and NG structures.

3.4. Plane-strain crack-path profiles

To understand why the fracture resistance of CG→NG→CG structure is superior to all the other monolithic/gradient structures, we examined differences in the failure mechanisms across the various structures. For this purpose, it is necessary to discern the development of the deformation and failure modes during crack propagation under plane-strain conditions, which we achieved for specific SE(B) specimens using the procedure described in Section 2.3. Resulting crack-path profiles and the deformed microstructure near the crack tip and in the plastic-wake region, imaged using the BSE in the SEM, are shown for the single-gradient CG→NG and NG→CG and dual-gradient CG→NG→CG structures in Fig. 3.

Figure 3a shows the plane-strain crack profiles of the CG→NG specimens unloaded from two load levels on the load–displacement curve, i.e., at 676 N, which is just past the maximum load, and at 335 N, where significant load drop is followed. At a load of 676 N, a pronounced blunted crack tip is clearly formed in the initial coarse-grained zone. As revealed by in situ observation on the surface [23], the excessive crack-tip blunting is accommodated by extensive [111] dislocation slip operating in favorably orientated coarse grains within the large-scale plastic zone surrounding the crack tip. Such crack-tip blunting is clearly responsible for the high initiation toughness of the CG→NG structure (Fig. 2b). Further loading of the CG→NG specimen leads to a fast load drop. The corresponding crack profile at 335 N shows a major blunted crack connected with a second brittle crack. The tip of the blunted crack resides in the initial coarser-grained region of the gradient zone. The second brittle crack initiates at a location away from the blunted crack-tip near the nano-grained end of the gradient zone. Two branches of the brittle crack in opposite directions are visualized. One branch, aligns almost with the horizontal line, extends into the NG zone, while the other branch, slightly inclined to the horizon line, propagates towards the coarser grains in the gradient zone and links to the blunted major crack tip. The formation of a brittle crack ahead of the blunted crack tip is likely ascribed to the highest triaxial stresses located ahead of the blunted major crack, which is analogous to the formation of stress-controlled brittle cracks in steels [25,26]. This burst of brittle cracking (Fig. 3a) is consistent with the R-curve behavior (Fig. 2b) where unstable crack propagation occurs when crack extends ~130 μm into the gradient zone.

For the NG→CG structure, the crack profile unloaded from 334 N, which is just prior to the peak load, shows a typical mode I brittle crack with its tip near the nano-grained end of the gradient zone (Fig. 3b). The propagation of this macroscopically brittle crack in the nano-grains is driven by the conjoining of micro-voids and microcracks resulting from the restricted plastic deformation within the nano-grains ahead of the main crack [23]. After the peak load is exceeded, the load drops quickly at first, followed by decrease at a gradually lowered rate. A crack-path profile taken at 142 N (Fig. 3b) shows a pronounced blunted crack with its tip arrested in the coarse-grained zone. This toughening effect, again resulting from crack-tip blunting in the coarser grains, is consistent with the R-curve behavior (Fig. 2b) where crack resistance is progressively increased as the crack propagates through the NG→CG gradient zone.

In contrast to these single-gradient structures, the load–displacement response for the CG→NG→CG structure
Fig. 3 – Load-displacement responses and plane-strain crack–path profiles characterized on the mid-thickness section of specimens unloaded from different load levels. (a) CG → NG structure: Blunted ductile crack arrested in the initial part of the CG → NG gradient zone, followed by a brittle crack initiated away from the blunted crack front. (b) NG → CG structure: Brittle crack initiated from the NG zone which arrested in the gradient NG → CG zone and the CG zone due to substantial crack-tip blunting in the CG region. (c) CG → NG → CG structure: Blunted ductile crack in the initial coarser grains, followed by brittle crack through the central nano-grains which arrested in the latter coarser grains in the gradient zone.

4. Discussion

Nature is especially adept in creating gradients in biological materials to confer functions and benefits to organisms. These gradients can involve variations in the local chemistry and/or the structural characteristics associated with the arrangement, distribution, dimension and orientation of the structural constituents. The result of such graded materials is not only the generation of impressive properties but more importantly the creation of unusual combinations of properties, such as those described for bamboo stems in the Introduction.

We have demonstrated that a simple gradient in grain size in centimeter-sized samples of pure nickel can generate an excellent combination of the often mutually exclusive properties of strength and tensile ductility, which in turn results in a significantly improved fracture toughness, compared to that of the corresponding uniform grain-sized structures [23]. However, as described above, by far the toughest (single) gradient structure, involving a gradient from coarse to fine grains to promote a markedly rising R-curve, suffers from final (unstable) brittle fracture in the fine-grained region. We have shown here that the use of a simple dual-gradient structure, involving a transition from coarse to fine grains and then back to coarse grains, can effectively solve this problem.

Figure 4 shows plots of the measured toughness as a function of strength for all the uniform-grained, single-gradient and dual-gradient structures tested. Here the strength is plotted as the yield (tensile) strength, and three measures of toughness are shown: the plastic work density (work of fracture), the crack-initiation toughness represented by $K_{IC}$, and the crack-growth toughness represented by $K_{SS}$, i.e., the stress intensity required to generate stable crack extension of $\Delta a \sim 1$ mm. Whichever measure of strength and toughness is considered, it is clear that the dual-gradient CG-NG-CG structure (depicted in purple) exhibits an optimal
combination of these properties – not necessarily with the highest strength but certainly with the highest toughness without too much compromise in strength.

Mechanistically, the nano-grained regions of all these gradients can confer the highest strength but cracking in these regions is unstable and brittle in nature and therefore significantly lowers the fracture resistance for both the initiation and propagation of cracks. The coarse-grained regions conversely are of much lower strength; this results in greater ductility, which then is translated into a higher toughness. In terms of mechanisms, the main function of these coarser-grained regions is to induce excessive crack blunting which markedly elevates the crack-initiation and -growth toughness. The advantages of the dual-gradient structure are that these combinations of properties are retained, and often enhanced, but with coarse grains at each end of the gradient, the structure is simply not prone to final unstable fracture, which would likely be a mandatory characteristic for most safety-critical applications. Indeed, we believe that such a dual-gradient prototype could serve as a “unit cell” for designing multi-layered gradient materials with unparalleled levels of damage-tolerance.

A final thought here is that solving the strength vs. toughness “conflict” [27] to produce superior damage-tolerant structural materials has been in vogue of late through the development of multiple principal element, or high-entropy, alloys which can display unprecedented strength and toughness properties (e.g., ref. [28]). However, as pointed out by Li and Lu [29,30], because of the many alloying elements involved, this can represent an expensive option. They advocate instead a “compositional planification” approach with simpler materials where these properties are achieved, as in many natural materials, through the creation of gradient structures. Although few bulk materials have been developed with this concept due to difficulties in macro-scale processing, the current work shows that excellent damage-tolerance can be induced in a single element bulk material through gradients in grain size, which are specifically designed to optimize strength and toughness without making the material susceptible to final unstable catastrophic failure due to the directional aspects of the fracture resistance.

5. Conclusions

Taking a lesson from Nature by leveraging the strength and weakness of single-gradient (NG→CG and CG→NG) structures, we have designed a dual-gradient CG→NG→CG structure in pure nickel where the grain size transitions from coarse grains (CG) of ~4 µm to nano-grains (NG) of ~30 nm, and then back to ~4 µm again with the intent of developing a structure with optimized combination of high strength, tensile ductility and toughness, which will not fail by final catastrophic fracture. Our major findings include:

1. As compared to the uniform-grained and single gradient structures, the dual-gradient structure achieves the highest fracture toughness for crack initiation and crack growth, the latter via the generation of a stable rising crack–resistance curve while maintaining a good balance of strength and ductility.
2. The high crack-initiation toughness of the dual-gradient CG→NG→CG structure originates from the toughening due to the excessive crack-tip blunting in the initial coarser grains. Further loading results in rapid crack extension through the central nano-grained region; however, such brittle cracks are readily arrested once they reach the latter coarser-grained region.

3. In simple terms, the dual-gradient structure combines the tensile strength of the nano-grains with the ductility and toughness of the coarse grains, but without compromising the structure by making it susceptible to final catastrophic failure.

Data availability

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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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Appendix A. Supplementary data

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