

The conflicts between strength and toughness

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The attainment of both strength and toughness is a vital requirement for most structural materials; unfortunately these properties are generally mutually exclusive. Although the quest continues for stronger and harder materials, these have little to no use as bulk structural materials without appropriate fracture resistance. It is the lower-strength, and hence higher-toughness, materials that find use for most safety-critical applications where premature or, worse still, catastrophic fracture is unacceptable. For these reasons, the development of strong and tough (damage-tolerant) materials has traditionally been an exercise in compromise between hardness versus ductility. Drawing examples from metallic glasses, natural and biological materials, and structural and biomimetic ceramics, we examine some of the newer strategies in dealing with this conflict. Specifically, we focus on the interplay between the mechanisms that individually contribute to strength and toughness, noting that these phenomena can originate from very different lengthscales in a material's structural architecture. We show how these new and natural materials can defeat the conflict of strength versus toughness and achieve unprecedented levels of damage tolerance within their respective material classes.

A requirement for almost all engineering structural materials is that they are both strong and tough (damage tolerant) yet invariably, in most materials, the properties of strength and toughness are mutually exclusive. This is not always widely appreciated, as in the common vernacular these terms are often taken to mean the same thing. Whereas strength (or hardness) is invariably a stress representing a material's resistance to non-recoverable (for example, plastic) deformation (at least in ductile materials), toughness is a material's resistance to fracture and, as such, is measured as the energy needed to cause fracture. Toughness can also be measured using fracture-mechanics methods, which evaluate the critical value of a crack-driving force, for example, the stress intensity K , strain-energy release rate G , or nonlinear elastic J -integral, required to initiate and/or propagate a pre-existing crack.

The ability of a material to undergo limited deformation is a critical aspect of conferring toughness, as this feature enables the local dissipation of high stresses that would otherwise cause the material to fracture; this is the reason that hard materials tend to be brittle and lower strength materials, which can deform more readily, tend to be tougher (Fig. 1a). These deformation mechanisms can be widely diverse; although plasticity associated with dislocation motion in crystalline materials is documented in any materials textbook, inelastic deformation can also occur through such processes as *in situ* phase transformations in certain metals and ceramics¹, the sliding of mineralized collagen fibrils in tooth dentine and bone², the frictional motion between mineral platelets in seashells³, and even by mechanisms that can also lead to fracture such as micro-cracking in geological materials and bone⁴, and shear band propagation in metallic glasses⁵. The operative word here is often 'limited', as with too much deformability the material can lose its toughness. The property of toughness is thus a compromise (indeed, it is a series of compromises); traditionally it is considered to represent the combination of strength and deformability (ductility) — two mechanical properties that also tend to be mutually exclusive.

With the trend today of seeking ever stronger materials, does this mean that such materials can never be tough? Well, not exactly — the development of toughness, or lack thereof, is not that simple; indeed, toughness can even be generated without ductility.

A prime example of this is in brittle materials such as ceramics, which invariably cannot be toughened by promoting plasticity⁶. To provide a context for this, it is useful to consider the process of fracture as another conflict — in fracture mechanics terms, as a mutual competition between what can be termed intrinsic damage processes that operate ahead of the tip of a crack to promote its propagation, and extrinsic crack-tip-shielding mechanisms that act mostly behind the crack tip to inhibit this (Fig. 1b)^{7,8}. Damage mechanisms depend on the nature of the nano/microstructure and involve such processes as the cracking or debonding of a second phase within the process zone ahead of a crack tip. Intrinsic toughening is associated with making these processes more difficult and is primarily related to plasticity, that is, enlarging the plastic zone; as such it is effective against both the initiation and propagation of cracks. With extrinsic toughening, conversely, the inherent fracture resistance of the material is unchanged; instead, mechanisms such as crack bridging act principally on the wake of the crack to reduce (shield) the local stresses and strains actually experienced at the crack tip — stresses and strains that would otherwise have been used to extend the crack. (Extrinsic mechanisms can be quite diverse, involving such processes as crack bridging by unbroken fibres or a ductile phase in composites, the frictional interlocking of grains during intergranular fracture in monolithic ceramics, and the presence of collagen fibrils spanning microcracks in bone^{6–8}.) By operating principally in the crack wake, extrinsic mechanisms (unlike intrinsic mechanisms) are only effective in resisting crack growth — as there has to be a crack for them to operate, they can have no effect on crack initiation. Moreover, their effect is dependent on crack length (or size). A major consequence of this is so-called rising crack-resistance curve (R-curve) toughness behaviour⁸, where the fracture toughness in terms of the required crack-driving force must be increased to maintain the subcritical (non-catastrophic) extension of the crack due to the enhancement in extrinsic toughening generated in the crack wake.

Intrinsic toughening is the primary source of fracture resistance in ductile materials. Most metallic materials are toughened in this manner, and are therefore subject to the initial conflict between strength and toughness referred to above — the stronger

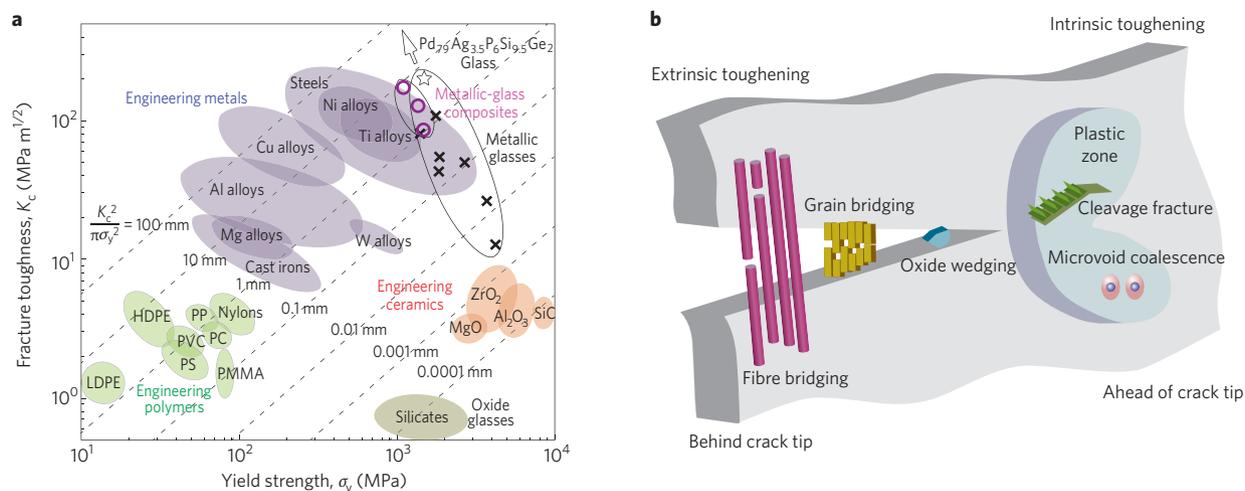


Figure 1 | Conflicts of strength versus toughness. **a**, Ashby plot showing strength–toughness relationships for engineering materials¹¹. Diagonal lines show the plastic-zone size, $K_c^2/\pi\sigma_y^2$, where K_c is the fracture toughness and σ_y the yield strength. The white star and purple circles refer, respectively, to the Pd-glass and metallic-glass composites, as compared with monolithic glasses (black crosses), which are discussed below. We believe that through composition changes, further toughness enhancement is possible (white arrow). Figure reproduced from ref. 11, © 2011 NPG. **b**, Schematic illustration showing how strength and fracture behaviour can be considered in terms of intrinsic (plasticity) versus extrinsic (shielding) toughening mechanisms associated with crack extension. Illustration shows mutual competition between intrinsic damage mechanisms, which act ahead of the crack tip to promote crack advance and extrinsic crack-tip-shielding mechanisms, which act primarily behind the tip to impede crack advance. Intrinsic toughening results essentially from plasticity and enhances a material’s inherent damage resistance; as such it increases both the crack-initiation and crack-growth toughnesses. Extrinsic toughening acts to lower the local stress and strain fields at the crack tip; as it depends on the presence of a crack, it affects only the crack-growth toughness, specifically through the generation of a rising R-curve⁷.

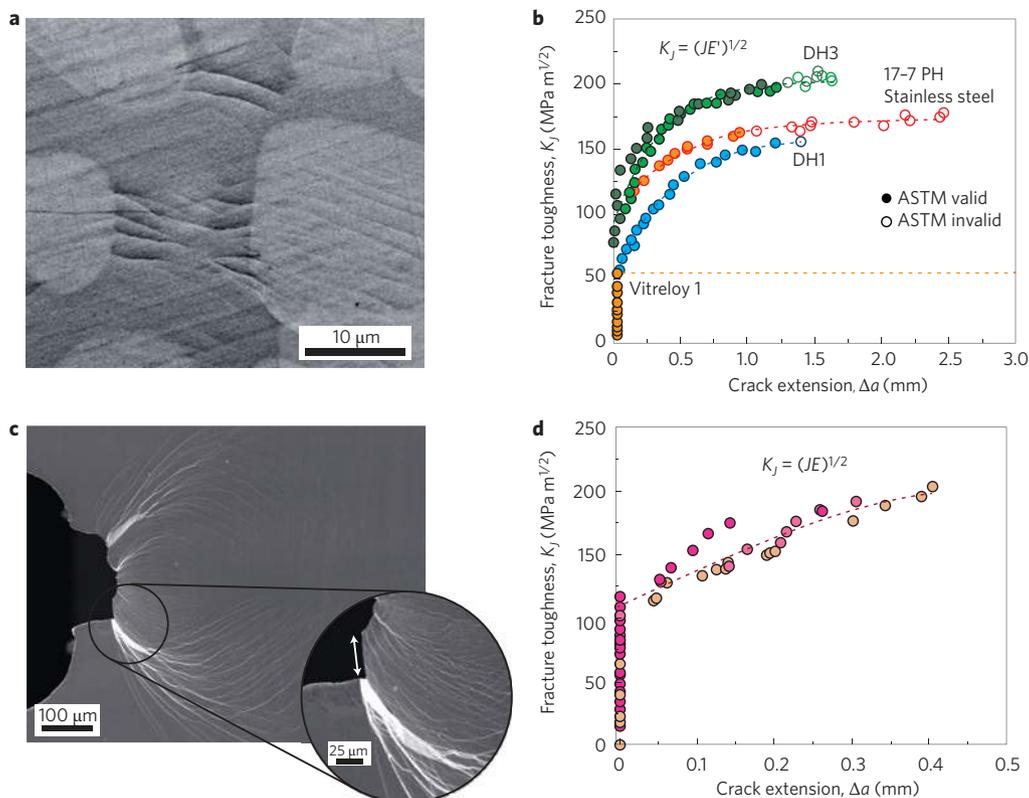


Figure 2 | Strength and toughness strategies for BMG alloys. Achieving high toughness and strength in BMGs involves preventing single shear-band formation, which transverses the material to cause failure at near-zero strains. **a,b**, One approach is to add a second phase to arrest shear bands, here crystalline dendrites in a Zr–Ti–Nb–Cu–Be glass matrix, where the interdendritic spacing is smaller than the crack size for failure (**a**); compared with the monolithic matrix alloy (Vitreloy 1), this leads to a three- to fourfold toughness increase to ~150 MPa m^{1/2} in two alloys DH1 and DH3 (refs 9,10) (stainless steel is shown for comparison) (**b**). **c,d**, Another approach involves achieving a high bulk-to-shear-modulus ratio; this makes shear-band formation easier but cavitation leading to fracture within bands more difficult. The result, for a monolithic Pd–Ag–P–Si–Ge glass, is multiple shear-band formation without unstable fracture, exceptional strength (~1.5 GPa), large crack-opening displacements (white arrow) (**c**) and toughnesses of ~200 MPa m^{1/2} (ref. 11) (**d**). K_j is the toughness measured using the J -integral; E is Young’s modulus. Figures reproduced with permission from: **b**, ref. 10, © 2009 AIP; **c,d**, ref. 11, © 2011 NPG.

the material, the less plasticity is available for (intrinsic) toughening. For this reason, most safety-critical structural applications, from steel pressure vessels and pipelines to aluminium alloy airframes, are manufactured from the comparatively lower-strength versions of these alloys to avoid problems from premature failure.

There are instances, though, where the conflict of attaining strength and toughness can be overcome. A seemingly unexpected example of this is in certain new bulk metallic glass materials (BMGs)^{9–11}. Being 100% amorphous, BMGs tend to be strong with tensile strength that can easily be in the range of 1–2 GPa. In the absence of dislocations, they deform by the initiation and propagation of shear bands, but this can be the source of extreme brittleness; in tension, a single shear band can form and traverse the entire sample, causing failure at vanishingly small strains. Locally arresting shear bands before they can cavitate and become cracks is thus the key issue to making these strong materials tough, as this encourages multiple shear band formation, which can relieve locally high stresses^{9–11}. This can be achieved by making BMG-matrix composites where a crystalline second phase — dendrites — is added implicitly to arrest shear bands (Fig. 2a,b); however, the spacing of the dendrites must be small enough to stop a shear band/crack before it can cause catastrophic failure, that is, the characteristic microstructural lengthscale of the interdendritic spacing must be matched to the characteristic mechanical lengthscale of the critical crack size for failure^{9,10}. Indeed, Zr–Ti–Nb–Cu–Be glasses containing 42–67 vol.% dendrites have shown fracture toughnesses of 100–160 MPa m^{1/2} at tensile yield strengths of 1.1–1.5 GPa, representing one of the best combinations of strength and toughness in a structural material so far (Fig. 2b)¹⁰. Amazingly, even better strength and toughness properties have been reported more recently in a monolithic (100% amorphous) Pd–Ag–P–Si–Ge glass alloy (Fig. 2c,d)¹¹. Here a very different approach was used in that the alloy composition was chosen to achieve a high bulk modulus but a low shear modulus. The latter is related to the ease

of shear band formation, the former to the difficulty of cavitation within these bands. On loading, these alloys generate a proliferation of shear bands mimicking large-scale plasticity (Fig. 2c), but the bands themselves are restrained from becoming actual cracks. The strength and toughness properties of these alloys are truly astounding, with fracture toughnesses of ~200 MPa m^{1/2} (Fig. 2d), and tensile strengths greater than 1.5 GPa; in fact, they seem to be the most damage-tolerant materials ever made. Unfortunately they are very expensive and currently can only be processed in very small section sizes of ~6 mm (ref. 11).

As noted above, extrinsic toughening is the primary source, and in many cases the only source, of toughening in brittle materials. In monolithic ceramics, such as silicon carbide, silicon nitride and alumina for example, intrinsic toughening would require changing the bond strength — not a feasible undertaking — yet these materials can readily be toughened extrinsically by promoting crack deflection and grain bridging (Fig. 3)⁶. Intergranular fracture is generally an essential requirement here for the operation of these mechanisms. The fracture toughness of SiC, for example, is ~2–3 MPa m^{1/2} when it fractures transgranularly (Fig. 3a,c), whereas it can approach 10 MPa m^{1/2} for intergranular fracture (Fig. 3a,b)¹², the key microstructural feature being the presence of brittle nanoscale glassy films along the grain boundaries, which promote boundary cracking¹³, grain bridging and hence the higher toughness.

Some materials rely on both intrinsic and extrinsic toughening; natural materials are a good example here. Indeed, one only has to look at seashells to realize that nature has been far more successful than us in making damage-tolerant (hard and tough) materials^{3,14}. One reason for this is the hierarchical architecture of most biological and natural materials, which have characteristic structural features on multiple lengthscales from molecular to near-macroscopic dimensions^{15–17}. The origin of the intrinsic (plastic deformation) mechanisms tends to be on the smaller, submicrometre lengthscales, akin to the nanometre scale of dislocation Burgers'

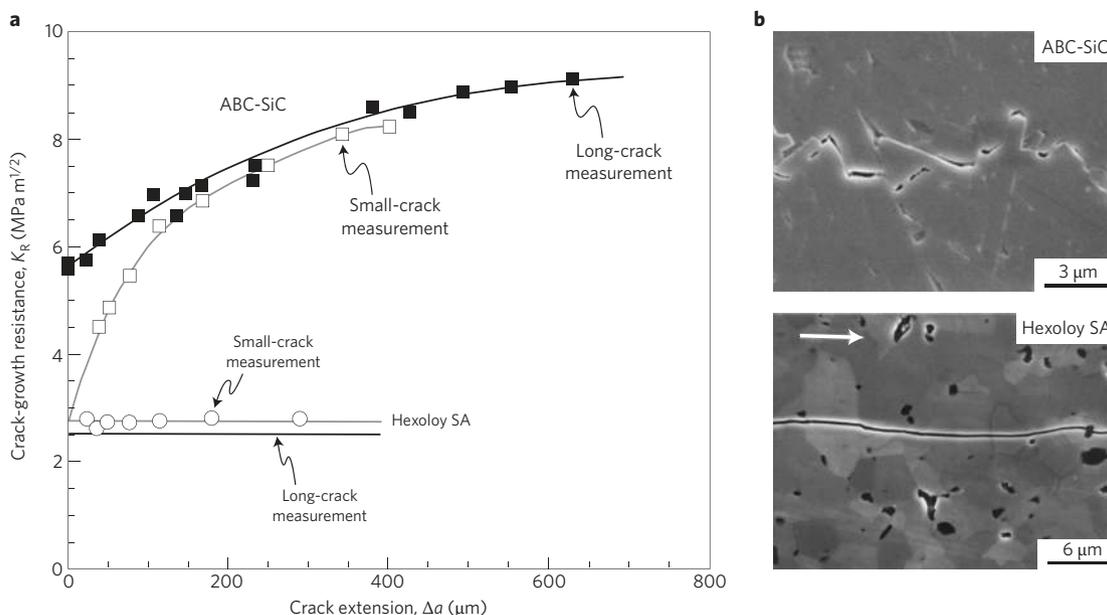


Figure 3 | Extrinsic toughening in monolithic ceramics. The fracture toughness of monolithic ceramics depends largely on their fracture mode. Silicon carbide, processed with dopants of aluminium, silicon and boron (termed ABC-SiC)¹², develops nanoscale glassy films along the grain boundaries (so called wet boundaries). **a**, ABC-SiC correspondingly shows significant rising R-curve toughening. **b**, This is because it fails intergranularly along the brittle grain-boundary films, resulting in extrinsic toughening through grain bridging (that is, frictional interference in the boundaries between interlocking grains as the material fails intergranularly acts to oppose crack opening, as shown schematically in Fig. 1). **c**, The commercial SiC Hexoloy, conversely, has so-called dry boundaries with no glassy films; it fails transgranularly with no extrinsic toughening and hence no rising R-curves. The crack-initiation toughness of both materials is essentially identical, whereas the crack-growth toughness is three times higher in the grain-bridging material¹². The horizontal arrow represents the general direction of crack propagation. Figures reproduced with permission from ref. 12, © 1997 Wiley.

vectors in metals, whereas the processes of extrinsic toughening and fracture occur on much coarser lengthscales typically well into the micrometre range¹⁸. Human cortical bone provides an excellent example of this (Fig. 4)¹⁸. Intrinsic toughening, that is, plasticity, derives from a fibrillar sliding mechanism on the scale of tens to hundreds of nanometres, the lengthscales associated with the mineralized collagen fibrils. With aging, irradiation and certain diseases, however, the bone can embrittle due to increased cross-linking of the collagen¹⁹, which inhibits this mechanism; the intrinsic toughening then switches to higher lengthscales and is accommodated by inelasticity associated with microcracking. However, the principal source of toughness in bone is extrinsic and arises from crack bridging and crack deflection as a growing crack encounters the more-mineralized interfaces of the osteonal structures^{20,21}; as the size and spacing of the osteons are in the range of tens to hundreds of micrometres, the characteristic lengthscales for these events approach millimetre dimensions.

Mollusc shells are also another fine example of nature's design of damage-tolerant materials. Dating back as a species to some 545 million years ago, these materials, such as nacre (red abalone shell), have a 'brick-and-mortar' structure; the 'bricks' are ~0.5 μm thick, 5–10 μm wide, platelets of the mineral aragonite (a polymorph of calcium carbonate) that comprise some 95 vol.% of the structure, separated by an organic biopolymer 'mortar' in-between (Fig. 5a)^{22,23}. The ceramic mineral accounts for the high strength; however, as the mineral is inherently brittle, if the aragonite platelets were rigidly locked together, the resulting toughness would be very low as there would be no means of relieving any locally high stresses. This is the role of the organic 'mortar', which acts like a lubricant by allowing some movement between the platelets; this constitutes the intrinsic toughening plasticity mechanism³. This sliding between the mineral platelets must be limited to ~1 μm though, or the material would lose its strength; nature achieves this by roughening the surface of the mineral platelets to provide frictional stops^{3,24} and by having

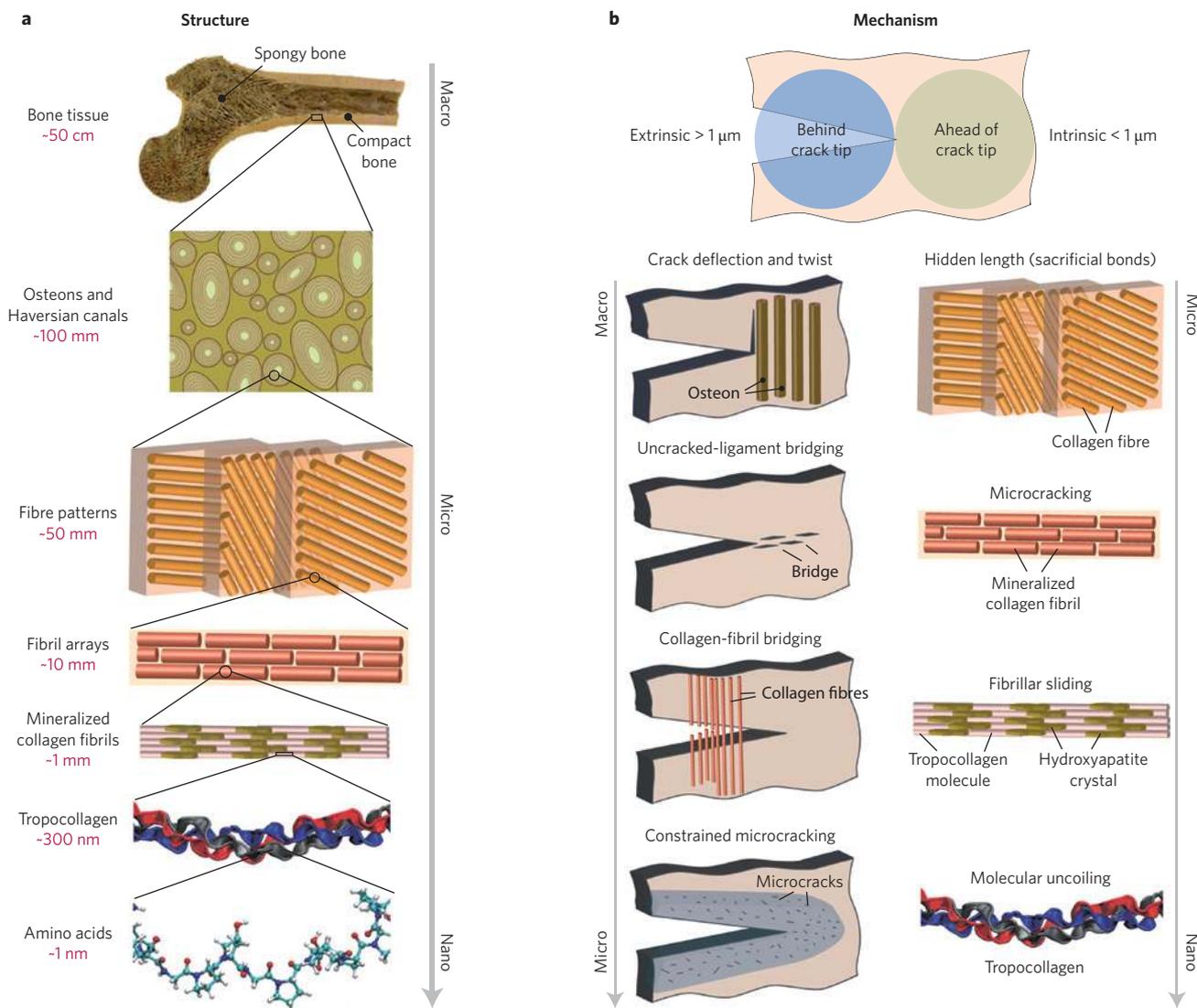


Figure 4 | The structure of bone showing the seven levels of hierarchy with the prevailing toughening mechanisms. **a**, The seven levels of hierarchy¹⁵. **b**, The prevailing toughening mechanisms¹⁸. At the smallest level, on the scale of the tropocollagen molecules and mineralized collagen fibrils, (intrinsic) toughening, that is, plasticity, is achieved through the mechanisms of molecular uncoiling and intermolecular sliding of molecules. At coarser levels, on the scale of the fibril arrays, microcracking and fibrillar sliding act as plasticity mechanisms and contribute to the intrinsic toughness. At micrometre dimensions, the breaking of sacrificial bonds at the interfaces of fibril arrays contributes to increased energy dissipation, together with crack bridging by collagen fibrils. On the largest lengthscales, in the range of tens to hundreds of micrometres, the primary sources of toughening are extrinsic and result from extensive crack deflection and crack bridging by uncracked ligaments, both mechanisms that are motivated by the occurrence of microcracking. Figures reproduced with permission from ref. 18, © 2010 Annual Reviews.

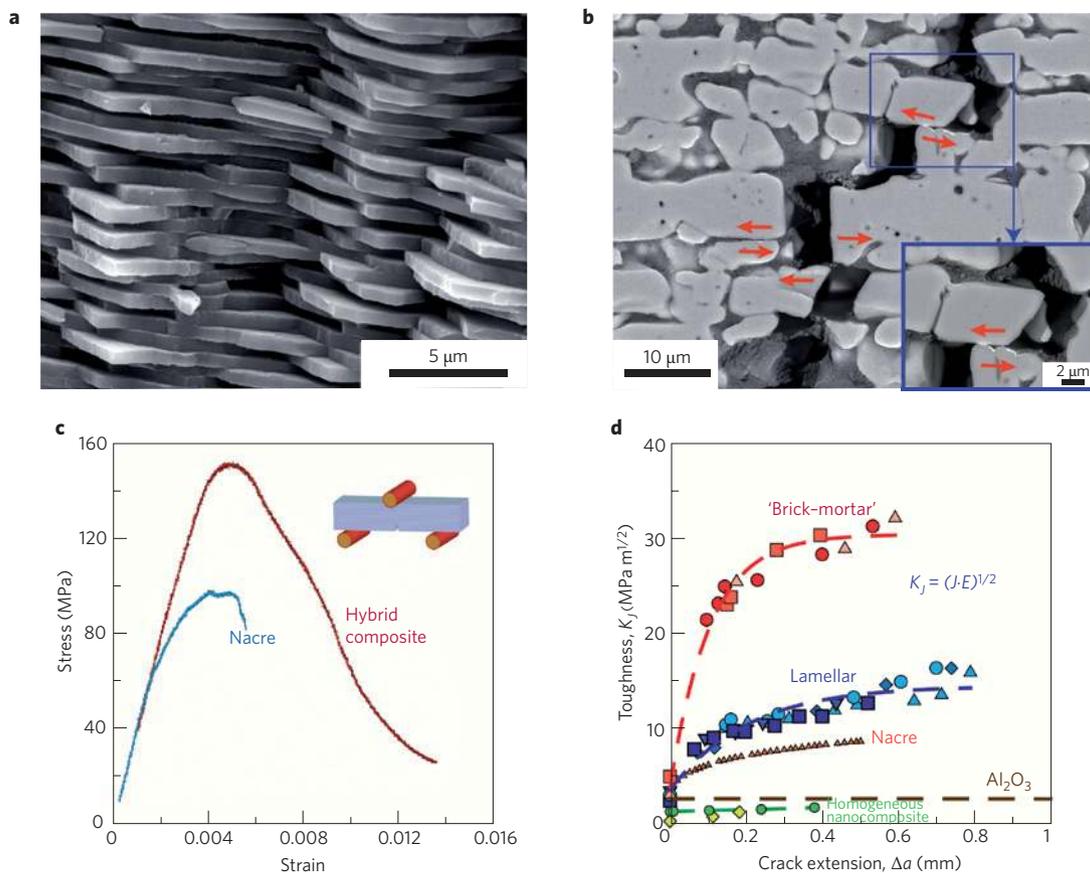


Figure 5 | Toughening in mollusc shells (nacre) and in corresponding biomimetic ceramics. **a**, Nacre has a natural structure of $\sim 0.5\text{-}\mu\text{m}$ thick aragonite mineral 'bricks' separated by a biopolymeric 'mortar'. **b**, Bioinspired synthetic alumina-PMMA 'brick-and-mortar' structures²⁵ made by freeze-casting²⁶ in the image of nacre. **c,d**, Although both natural and synthetic materials comprise brittle ceramics (CaCO_3 and Al_2O_3), they display significant toughness and tensile ductility (**c**), and marked R-curve behaviour (**d**). Toughening is associated in part with 'brick' pull-out (inset in **b**) and frictional sliding along mineral interfaces in the lubricant polymeric layer (red arrows in **b**). This nacre-like alumina-PMMA ceramic shows exceptional toughness exceeding $30\text{ MPa m}^{1/2}$, which is twice as tough as lamellar structures and far tougher than corresponding alumina-PMMA nanocomposites²⁵. Moreover, characteristic of many natural materials, it is an order of magnitude or more tougher than its constituent phases. Toughness is measured using the J -integral. Figure parts **b** and **c** reproduced with permission from ref. 25, © 2008 AAAS.

small mineral 'bridges' linking the layers¹⁴. The tortuous crack paths and pull-out of the mineral platelets further provides a major contribution from extrinsic toughening. The result is a hybrid material with toughness of at least an order of magnitude higher than either of its constituent phases.

Recently, synthetic, yet bioinspired, bulk ceramic materials have been made in the image of the nacre structure²⁵. Using alumina ceramic powders mixed with water and frozen using a freeze-casting (ice-templating) technique^{25,26}, ceramic scaffolds can be processed with layer thicknesses (of $\sim 1\text{--}100\text{ }\mu\text{m}$) controlled by the rate of cooling, and interlayer roughnesses controlled in part by the addition of dopants (sugar, salt or alcohol). After cold pressing and infiltrating with a polymeric lubricant phase (poly(methyl methacrylate), PMMA), 'brick-and-mortar' 85 vol.% alumina ceramic-PMMA hybrid materials have been made in bulk form in the image of nacre (Fig. 5b). The resulting mechanical properties of these bioinspired ceramics are nothing short of remarkable, with strengths comparable to pure alumina but fracture toughnesses an order of magnitude larger (Fig. 5c). Indeed, toughness values can exceed $30\text{ MPa m}^{1/2}$ (Fig. 5d) making these materials the highest toughness ceramics on record²⁵.

These examples hopefully convey that although the conflicts between the generally mutually exclusive properties of strength and toughness are very real, there are ways to attain both properties in a single material through the presence of multiple plasticity

and toughening mechanisms acting on differing lengthscales. Indeed, in this quest for such damage tolerance, a common feature of many of these materials is the notion of a nano/microstructure comprising a hard phase to provide for strength but incorporating: (1) mechanisms for alleviating locally high stresses through limited inelastic deformation to provide for intrinsic toughness, be it dislocation plasticity in metals, multiple shear-band formation in metallic glasses, fibrillar sliding and microcracking in bone, or mineral-platelet sliding in seashells; and (2) further extrinsic mechanisms, such as crack deflection and bridging, that can provide a very potent source of toughening on much larger lengthscales. Unlike prototypical strong yet brittle solids where cracks, once initiated, immediately propagate in an unstable (often catastrophic) fashion, these mechanisms allow for the stable (sub-critical) growth of cracks. It is the attainment of this crack-growth toughness from extrinsic mechanisms, which is characterized not by parameters such as the critical value of K at crack initiation, K_{Ic} , but by the existence of R-curve toughening behaviour, together with the intrinsic (plasticity) contributions to the crack-initiation toughness, that are the essential features of these strong and tough damage-tolerant materials.

Finally, one might sensibly ask where these new damage-tolerant materials may find application. With such cutting-edge properties, one could hope for important, safety-critical, applications such as in the aerospace, transportation or power-generation industries,

although if the past is any judge, the timespan for the adoption of new structural materials is often measured in decades rather than years! There are many confounding issues here, one being density. The bioinspired ceramics, actually ceramics in general, display excellent specific strengths, comparable to metals. Indeed, the nacre-like alumina-based ceramics described in Fig. 5 have specific strength and toughness properties similar to metallic aluminium alloys, and as such could find use as lightweight armour. Many metallic glasses may well not possess such low densities. The palladium-based alloy described in Fig. 2, despite having exceptional damage tolerance, has a density some 20% higher than nickel-based alloys and a factor of two higher than titanium, which would undoubtedly be a major consideration in its potential use. However, most structural materials including carbon-reinforced plastics invariably find application initially in consumer goods. Metallic glasses are no exception here; they will probably be used as casings for mobile phones and laptops long before any aerospace applications are contemplated, owing more to their ease of fabrication than their specific mechanical properties.

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Additional information

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