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Damage-tolerant architectured materials inspired by crystal microstructure

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Summary: Architectured materials comprised of periodic arrangements of nodes and struts are lightweight materials that can exhibit combinations of properties which are inaccessible to conventional solids. However, with regards to their mechanical performance, they have an Achilles heel in that these materials can exhibit a catastrophic post-yielding collapse, causing substantial drops in strength and energy absorption during plastic deformation. This post-yielding collapse is the result of the activity of single shear bands, and originates from the single orientation of macro-lattices. We observe that this behaviour is analogous to deformation by slip in metallic single crystals. In this study we propose that, by mimicking the microstructure observed in crystalline materials, we may be able to employ hardening mechanisms found in crystalline materials to help us to develop robust and damage-tolerant architected materials. This study demonstrates that crystal-inspired meso-structures can play as an important role in the mechanical properties of architectured materials as do crystallographic microstructures in the case of metallic alloys. Consequently, designing meso-structures that mimic crystallographic microstructure in crystalline metals enables the fusion of metallurgy and architectured materials to transform the way of designing a new type of materials with desired properties.

A crystal is composed of atoms that are arranged in an ordered manner in space. The ordered arrangement of atoms in a crystal is called a crystal lattice which is, in turn, defined by its unit cell (Figure 1a) with the unit cell being the smallest group of lattice points that can describe the overall symmetry of a crystal. A single crystal consists of unit cells of the same type and same orientation, however, most crystalline materials are comprised of many domains with each having a lattice orientation different to that of its neighbours. Such domains are separated by boundaries and termed in metallurgy a crystal grain. Crystalline materials consisting of many crystal grains are called polycrystals. In addition to the crystal lattice, there are other important crystallographic microstructure features (namely dislocations, grains, phases and precipitates) that can be manipulated to control the plastic deformation of crystals under external load[1,2]. In particular, the slip of dislocations is the most common mechanism responsible for plastic deformation[1]. For single crystals, a dominant single slip mode occurs during plastic deformation (Figure 1b), causing strain to become localised and the stress required for further deformation to drop[3]. For poly-crystals, the orientation change across the grain boundary can impede or even stop dislocation movement from one grain to the next one (Figure 1c), resulting in the dependence of plastic deformation on the grain size[3]. This capacity for constraining the deformation and preventing the rapid propagation of slip minimises the stress drops[4] and strengthen poly-crystals[1]. The size of grains can significantly alter the mechanical strength, described by the wel-known Hall-Petch relationship[5-7] that the yield strength (σ_y) of polycrystals is inversely proportional to the square root of grain size (d),

$$\sigma_y = \sigma_0 + \frac{k}{\sqrt{d}} \quad (1)$$

where σ_0 is the friction stress which does not depend on the grain size, and k is a material constant

Similarly, precipitates and phases also control the slip in crystals thanks to the difference in lattice parameters, leading to other strengthening sources in crystalline alloys that are termed precipitation and phase hardening in metallurgy[1]. The capacity to control the strength and toughness of alloys via engineering the size, distribution and orientation of grains, precipitates and second phases forms the foundation of physical metallurgy.

Figure 1: Lattice structures and deformation behaviour. (a) Face-centred-cubic (FCC) crystal lattice, (b) Single slips in a single crystal (re-used from [8] with permission from Elsevier), (c) Slips at grain boundary in a polycrystalline steel (re-used from[9] with permission from Elsevier), (d) Achitectured FCC lattice, (e) Single slip in a single oriented lattice, (f) Unstable behaviour of architectured metallic lattices (reused from [10] under the Creative Commons Attribution 4.0 International License).

Similar to the mimicry of structures inside biological systems (or organic materials) to design bio-inspired materials [11-14], we can easily mimic the crystal structure of metals and alloys on a macroscopic scale by constructing a lattice unit cell comprised of an ordered arrangement of nodes (analogous to atoms) connected by struts (equivalent to atomic bonds) (e.g., Figure 1d). Consequently, architectured lattice materials are straightforwardly generated from such a macro-lattice unit in space using a suitable computer-aided design package and can be fabricated via additive manufacturing (also commonly termed 3D printing). Very similar to crystals, the properties of architectured materials are strongly affected by their lattice architecture[15]. The behaviour of architectured lattices can be modified through careful design of the lattice unit cells, giving access to unprecedented properties, e.g. lightweight[16,17] and negative Poisson ratio[18,19]. By further manufacturing our architected lattice designs using different materials we can develop architectured materials that are both lightweight and strong[20]. To date, however, work on these promising architectured materials has focused only on lattice materials with a single orientation[10,15,17,21] and, these materials have been observed to suffer precisely the same drastic collapse in mechanical strength when loaded beyond the yield point (i.e., post-yielding collapse) because of the occurrence of shear bands[10] (e.g. Figure 1f) with this deformation again becoming highly localised on specific planes with defined lattice directions[10,22], similar to the slip activity in single crystals (Figure 1e, f versus Figure 1b). Considering the similarity between single crystals and singly oriented lattice structures, we hypothesised that if it should be possible to develop damage-tolerant architectured materials by introducing hardening mechanisms analogous to those found in crystalline materials. This proposal is aligned to a recent approach that aims to bring the metallurgical microstructures closer to the component-size scale to better engineer the behaviour of components and structures [12,23]. However, if the hypothesis holds true, the approach proposed in this study is distintive on its own as it will enable a fusion of material specific hardening mechanisms and architectured materials. In this study, we present the methodology of designing meso-lattice features that mimic crystallographic microstructure to bring metallurgical hardening principles (grain size effect, precipitation and multiphase hardening) to the development of architectured materials. We demonstrate that crystal-inspired meso-structures can indeed strengthen the architectured materials, leading to the generation of highly damage-tolerant materials. The freedom in designing crystal-like meso-structures also offers alternative ways of studying complex metallurgical phenomena (e.g., slip transfer) in metallic alloys. We further show that the fabrication of lattices by polycrystalline materials leads to the generation of a new family of materials that contain atomic lattices within meso-lattices and polycrystal microstructure within polycrystal-like meso-structure; therefore, coined meta-crystals. The properties of such new materials can be easily to be tailored via numerous combinations of micro, meso and macro- lattices.

Grain boundary hardening

Tailoring the misorientation between two adjoining domains of macro-lattices creates a boundary between them in the same way as the boundary between two adjoining grains of crystal (Methods Section 1.1). Hereafter, each domain of the same lattice orientation is termed a meta-grain. Because atoms can bond with new neighbouring atoms while physical nodes cannot, there is the clear difference between polycrystal-inspired materials and polycrystals themselves. We chose, as an initial step, two well-understood phenomena in metallurgy to verify whether relationships observed in polycrystals are applicable to architectured materials containing polycrystal-like features: (*i*) symmetric slip in a twin-related bi-crystal (Figure 2a), and (*ii*) the aforementioned Hall-Petch relationship[5,6].

To investigate the phenomenon *(i)*, an architectured material containing two meta-grains separated by a twin boundary to mimic a twinned bi-crystal was designed and 3D printed (Methods 1.1, and Extended data - Figures E1, and E2a, b). The base material used to fabricate the twinned meta-grains was an elasto-plastic polymer (Methods, Materials and fabrication). Later we demonstrate that a polycrystal-inpsired architectured material fabricated by different base materials (including a stainless steel) exhibits the same behaviour in forming shear bands, showing a wide applicability of this polycrystal-inspired approach. Under compression with the loading parallel to the twin boundary, shear bands occurred and were parallel to the maximum shear stress planes (Figure 2b). In addition, we observe that the shear bands formed are symmetric about the twin boundary, confirming the shear band behaviour in meta-grain twins is similar to the slip activity in crystal twins (Figures 2a, b). To assist us in understanding this behaviour, finite element modelling (FEM) was used to understand the early stage of shear band formation (see Methods). The simulation predicts well the location and direction of shear bands in meta-grain twins as seen in the experiment (Figure 2c). By design, the (001) plane in each meta-grain is parallel to the maximum shear force in that meta-grain domain and FEM shows that as expected, deformation occurred via the buckling of <001> struts between two parallel (001) planes and along a <101> direction, leading to the localisation through the face-centred nodes parallel to the (001) plane of FCC (Extended data Video E1). The shear band system family <101>{001} is different to the <101>{111} slip system one in the FCC crystal, highlighting the difference between the FCC crystal lattice and architectured lattice. The symmetry of lattice orientation across the boundary causes the same localisation parallel to the (001) plane in the other half, resulting in the symmetric shear bands in the twinned meta-grains. The same twin boundary was used to further increase the number of meta-grains, in essence using the twin boundary to reduce the size of meta-grains (Methods 1.1 and Extended data - Figure E2) to study the phenomenon *(ii)* in the architectured materials. Fabricated polycrystal-like materials demonstrated a highly reproducible constitutive stress-strain behaviour (Extended data - Figure E7). The change in orientation across boundaries effectively controls the propagation of shear bands (Figure 2b, d and e). Most importantly, the yield strength of architectured materials substantially increases with the reduction in the meta-grain size, confirming that an effect similar to the Hall-Petch relationship exists in the architectured materials. Fitting against experimental data using Eqn. (1) gives $\sigma_0 = 2.58\,MPa$ and $k = 14.37\,MPa\sqrt{mm}$. The value of k falls well in the range for conventional polycrystalline metals[24]. The flow stress at a given strain is also observed to increase with reducing the size of meta-grains during plastic deformation (Extended data - Figure E8). In other words, the size of meta-grains strongly affects both the yield strength and the work-hardening during deformation of architectured materials. This observation of the size effect (together with the deflecting effect, Figure 2b, d and e) is significant as it confirms that the boundary strengthening is applicable to architectured materials. In particular, the grain size dependence in polycrystals is one of most widely used mechanisms in metallurgy to achieve outstanding combination between strength and ductility, e.g., development of a tough steel at low temperatures by the ultra-refinement of grains

[25]. However, it should be noted that that the grain size effect in metals is seen to be much more significant with the presence of incoherently high angle boundaries[26]. This means that the size reduction of high angle boundaries should substantially strengthen architectured materials. With the freedom of varying the size while keeping the same type of boundary, the current approach can provide clean data to obtain insights into the grain size effect of a specific type of boundary. Obtaining such clean data in metallurgy are not possible because there are no effective ways to reduce the grain size while maintaining the same single type of grain boundaries. This means that, while the rich knowledge in polycrystals provides important underlying science to engineering architectured materials, this polycrystal-inspired approach offers alternative ways to study complex phenomena in metallurgy.

Figure 2: Roles of lattice orientation in the deformation behaviour of crystals and architectured lattices. (a) Twin bi-crystal (re-used from [27] with permission from Elsevier), Shear bands in meta-grain twins observed in experiment (b) and predicted by FEM (c) (Note: Cut sections shows the deformation of internal struts. The sections were formed thanks to two cutting planes that were parallel to {001} planes of FCC lattice), (d) – (e): Shear bands were controlled by orientation of meta-grains: (d) 8 meta-grains and (e) 16 meta-grains (note the nominal strain was of 30%). (f) Yield strength versus the size of meta-grains, (g) Boundaries between meta-grains effectively stops cracks in brittle lattices, leading to a drastic increase in toughness of architectured materials.

Metallurgical studies suggest that microstructure (e.g. dislocation substructures and high angle boundaries) can even be able to stop the propagation of short cracks [28-30]. A series of samples from a different base materias were fabricated to study the effect of Incoherent High Angle Boundaries (IHABs) on crack propagation in architectured materials (Methods – Fabrication and Extended data Table 1). The base material used for these architectured materials is a brittle polymer to easily generate cracks under loading. For a single oriented lattice, cracks occurred and propagated rapidly throughout the macro-lattice, causing premature and fast fracture (dashed line, Figure 2g). In the case of the architectured material containing 8 IHAB meta-grains (Figure 2g - solid line Extended data – Figure E3), the orientation rotation of internal lattice led to the creation of incomplete unit cells on the free surface, causing a lower yield strength compared to the single oriented lattice. Nevertheless, cracks did stop at the IHABs, preventing fast brittle fracture (solid line, Figure 2g). It is well known in metallurgy that crystallographic microstructure is able to stop and deflect the propagation of cracks when the size of microstructure is comparable to the crack length [28,29]. The mimicry of microstructure on a macro-scale makes polycrystal-like structures comparable to macro-cracks. The change in lattice orientation across IHABs stops the propagation of cracks, and most importantly, thanks to the presence of IHABs, the architectured material retained its strength after yielding and was able to carry load up to large deformation, substantially improving the energy absorption of the material (Figure 2g): from ~194 kJ/m^3 by the singly oriented lattice to ~1309 kJ/m^3 absorbed by the designed polycrystal-like material. The drastic increase in the energy absorption associated with the presence of IHABs is in agreement with a widely accepted approach employed to strengthen alloys: the grain boundary engineering[25]. The same architectured material containing 8 IHAB meta-grains was also printed by two other base materials (elasto-plastic polymer and an austenitic stainless steel, Methods – Materials and fabrication). To study the influcence of the base material on the behaviour of polycrystal-like architectured materials, two other base materials were used to fabricate this 8 meta-grain architectured material. The selection of base material does affect the behaviour of macro-lattices: Compared to the same lattice fabricated by the brittle polymerisation resin, there were no brittle cracks in the lattices made by the elasto-plastic polymer and the steel because of the ductile behaviour of the two latter base materials whilst, in contrast, the localisation of strain in the lattice made of the two materials remains very similar (Extended data - Figure E9a, b). The localised deformation of struts led to the formation of shear bands in the 30°- and 60°-oriented meta-grains in both the ductile polymer and metallic lattices (Extended data - Figure E9a, b). The orientation change was able to alter the localisation in the architectured lattice fabricated by two different base materials, confirming this crystal-inspired approach is widely applicable to various materials. While the shear band activity is mainly governed by the lattice architecture, the constitutive stress-strain behaviour is strongly affected by the base material because different base materials have different yield strength and work-hardening behaviours. Most interestingly, the metallic lattice has substantial hardening during plastic deformation much more compared to that of the polymer lattice (Extended data - Figure E9c and d). This is because the base material (316 steel) itself hardens during deformation because of the change in its intrinsic microstructure (in particular increase in the dislocation density) [31]. This means that the application of the boundary hardening to metallic base materials can enable the synergistic strengthening associated with both crystallographic microstructure and designed meso-structures.

Precipitation hardening

Precipitation hardening is widely utilised in metallurgy to design and manufacture high performance alloys, e.g., Ni-based superalloys [32]. In precipitate-hardened alloys, precipitates act as obstacles to the movement of dislocations. The influence of precipitates on the strength of alloys depends on the size, shape, volume fraction and distribution of precipitates, and the coherency between precipitates and matrix [33]. Precipitation strengthening was incorporated into lattice design by introducing embedded lattice domains (termed *meta-precipitates*) that have different lattice parameters from those in the matrix. Lattice parameters (type, spacing and orientation) of meta-precipitates can be tailored to assign the different degrees of coherency between them and the matrix. In addition, the strut diameters of meta-precipitates can be varied to account for different "atomic" bond strengths, e.g., the Ni-Al and Al-Al bondings in the gamma' $L1_2$ in Ni superalloys [32].

Figure 3: Precipitation and multiphase hardening in architectured materials. (a) A single oriented without meta-precipitates. (b) Constitutive stress-strain responses of architectured materials without meta-precipitates (a) and with meta-precipitates (c, d). (e) Mechanical behaviour of single phase versus multiphase architectured materials. (f) Pseudo-superelasticity of Kresling lattice, (g) FEM simulation shows the strain localisation and local buckling of struts, and (h) Energy per unit volume of the first five cycles.

A single FCC architectured material containing meta-precipitates that were face-centred tetragonal and stronger than the matrix thanks to shorter lattice spacings was fabricated by 3D printing (Extended data - Figure E4). Fabricated FCC lattices containing meta-precipitates demonstrated a highly reproducible behaviour (Extended data - Figure E7d). It appears that the presence of the harder meta-precipitates had a governing effect on the shear band propagation (Figure 3a versus Figure3c-d): stabilising the overall lattice and substantially strengthening the overall architectured materials (Figure 3b). Shear bands were stopped at and bowed around the interfaces between meta-precipitates and matrix (Figure 3d). This governing effect has the same dependence as seen in the Orowan hardening effect in which the strength of crystals containing precipitates is proportional to the strength of precipitates and spacing between precipitates [33,34]. This remarkable parallel between architectured materials (containing meta-precipitates) and alloys (containing precipitates) means that the mechanical response of architectured materials can be readily manipulated by varying the lattice parameters, the volume fraction and distribution of meta-precipitates.

Multi-phase hardening

In multi-phase metals, such as martensitic stainless steels, hard phases offer high strength to enhance load-bearing capacity while soft phases accommodate the plastic deformation. As a crystal phase is defined by its lattice type, multiple phases can be imitated by assigning different lattice types to different macro-lattice domains. An architectured material comprising two phases (FCC in the top and bottom layers and BCC in the middle layer, Extended data - Figure E5) was designed. Because the unit cell of the FCC macro-lattice has a higher connectivity and higher density than the BCC, the FCC is stronger than the BCC one (solid grey versus dashed curve, Figure 3e). Similar to the behaviour of soft/hard multiphase crystals, the strength of the multi-phase architectured material mainly results from the strength of the hard FCC phase (black solid versus grey solid lines, Figure 3e). By contrast, we observe that plastic deformation was mainly accommodated by the soft BCC meta-grains, resulting in the confinement of shear bands mainly to the middle layer (top left inset, Figure 3e). Therefore, the mixture of hard and soft architectured phases offers additional means of tailoring properties and controlling shear bands along specific paths inside of an architectured materials.

Taking this one step further, some highly engineered alloys can also exhibit a solid state phase transformation whereby the crystal structure can transform by rearrangement of atoms in the unit cell under external loads. In some cases, reversible phase transformation lead to a shape memory effect and superelasticity [35]. To see whether such a transformation were a feasible option for crystal-inspired materials, a Kresling lattice[36] was designed to mimic a hexagonal close packed phase (Extended data - Figure E6). Under compression, the Kresling lattice twisted due to bending and local buckling of struts on prismatic and basal planes (Figure 3g and Videos E2 and 3), causing a helical movement of the nodes (Extended data - Figure E10), which altered the arrangement into a different one, enabling a “phase transformation”. It is worth noting that for accurate mimicry of a specific type of phase transformation in crystals, one simply needs to perform some additional design calculations of the helical movement of nodes to enable the change in the stacking sequence (e.g., ABABAB to ABCABC for HCP to FCC transformation as described Zhao *et al.* [37]). The transformed phase was not stable during unloading, but it will be straightforward to generate a lattice structure that is stable in both the original and transformed configuration by incorporating the design of multi-stable cellular structures[38,39]. Most interestingly, the transformation back to the original arrangment upon unloading leads to superelasticity (Figure 3f). The changes in elastic modulus observed (Figure 3f) are as a result of a change from overall elastic bending to local buckling of struts on the prismatic planes (Figure 3g). The increasing constraint in compression caused a gradual transition from the overall elastic bending to local buckling of struts near basal nodes. During unloading, the decreasing constraint led to a more abrupt change: struts unbuckled at defined strain (from 14 % to 12 % during unloading), leading to a change in bending moment. Such a change caused different friction on the interacting faces between the Kresling lattice and compressing plates, resulting in the energy dissipation during transformation (Figure 3h). This reversible alternation of node arrangements demonstrates that phase transformations can be mimicked in architectured phases, opening up the possibility of making lattices with high energy absorption: external work can be dissipated by a phase transformation, and the lattice then returns to its original shape via reverse transformation upon unloading.

Multiscale fractal crystal structures: *Å* to *mm* and beyond

Combining all sources of tailoring properties in architectured materials containing crystal-like meso-structures, it will be possible to design lightweight components in which meso-structures are designed in such a way that better responds to the external load, or to direct the accumulated damage (shear bands and cracks) along specific structural paths as desired (Figure 4a-d). An exciting avenue of this approach is when a crystalline material is used to fabricate crystal-like artchitectured materials (Extended data - Figure E9a, c), it leads to the generation of a new family of architectured materials comprising highly scalable fractal crystal structure consisting of crystallographic microstructure on *μm* and crystal-inspired mesostructures on *mm* and beyond (depending on the size of build volume of manufacturing methods), i.e., micro-crystals within macro-crystals. Such multi-scale fractal crystal structures are not available in nature, making the new architectured materials be of the meta-materials family, here coined *meta-crystals*. For example, (*i*) a *mm*-size FCC (Figure 4e) contains an atomic FCC lattice (Figure 4g), and (*ii*) a macro-polygrain structure on *cm* (Figure 4d) is made of a *μm*-size polygrain microstructure of an austenitic stainless steel 316L (Figure 4f). Similarly, a fractal γ/γ' super-lattice can be generated: micro-γ/γ' in Nickel superalloys (Figure 4h) can be scaled up by γ/γ'-like super-lattice (note: different diameters and lengths need to be used for specific struts in meta-precipitates to mimic the Ni-Al ordered atomic bonds in γ'-Ni_3Al phase). It is very easy to change crystal lattices on the atomic scale and crystal microstructure (e.g., by changing the base material) and architectured lattice meso-structures *by design*, making it possible to engineer these hierarchical lattices across multiple lengthscales. As discussed previously, the use of crystalline alloys enables the synergistic strengthening associated with both crystal microstructure and crystal-inspired meso/macro-structures. The interplay between microstructure and designed structures on different lengthscales might lead to exciting properties and potential applications for this new class of materials.

Figure 4: Lightweight and damage-tolerant architectured materials inspired by crystal microstructure. (a)-(d) Lightweight lattice component. (e)-(g) FCC fractal lattices from atomic up to cm scales, (h) γ/γ' scalable fractal super-lattices.

Conclusions

This current study presents a novel way of combining the hardening mechanisms in crystalline alloys and architectured materials to transform the way of designing materials with desired properties. A comprehensive study was carried out to substantiate: (1) the similarities between shear bands in crystal-inspired architectured materials and dislocation slip in crystals and (2) the applicability of strengthening mechanisms (grain boundary, precipitation and multiphase) to architectured materials. This study confirms that metallurgical concepts can be applied to enhance the mechanical behaviour of architectured materials. In return, this crystal-inspired approach provides alternative ways to study complex phenomena in metallurgy. This unique way of fusing physical metallurgy and architectured meta-materials opens new opportunities in (1) designing and engineering damage-tolerant architectured materials with desired strength and toughness and (2) enhancing the functionality and performance of architectured materials in response to external loads. Possibilities offered by this approach are not limited to those presented here. The application of this approach to crystalline metallic alloys opens an exciting frontier of research both experimentally and computationally to understand the possibilities that varying both intrinsic microstructure and designed meso-crystal structures of meta-crystals can afford us.

Data availability The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

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Contributions M.S.P. developed the idea and directed this research. C.L. carried out the computer-aided designs, fabrication and mechanical tests and post analyses. J.L. performed FEM simulations. I.T. discussed and contributed to the further development of the concept. All the authors participated in analysing and interpreting the data. The manuscript was written and approved by the authors.

Competing interest statement The authors declare the following competing interests: a patent developed on the basis of the approach proposed in this study was filed and managed by Imperial Innovations.

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Methods

1. Design

A unit cell of crystal lattice is defined by its lattice constants (which are the distances between atoms along three principal axes x, y, z: *a*, *b* and *c,* Extended data - Figure E1a) and the angles between them (Figure E1a). For cubic lattices, x, y and z axes are parallel to the three [100] orientations of lattice, $a=b=c$ and $\angle(a,b)=\angle(b,c)=\angle(c,a)=90°$. All macroscopic FCC unit cells in this study had the same node arrangement and connection as the one shown in Extended data - Figure E1a. Macroscopic cubes containing crystal-like mesostructures were designed and fabricated to study the roles of such meso-structures on the behaviour of architectured materials, e.g., Extended data - Figure E1b. A global (X,Y,Z)-coordinate attached to each cube was defined by the three orthogonal directions of the cube (Extended data - Figure E1b). The global dimension of macroscopic cubes in this study was 40mm × 40mm × 40mm. All lattice models presented in this study were designed by Solidworks and Element softwares.

1.1. Mimicry of polygrains

The global cubic matrix was fragmented into Voronoi domains. Each domain mimics a crystalline grain: It is infilled by macro-lattice whose orientation of lattice is different to those in adjoining domains. To create two twin meta-grains of FCC lattice (Figure 2b), a cube of single oriented FCC lattice was first created with its three <001>] orientations being parallel to the X, Y, Z axes (Extended data - Figure E1b and c). The FCC unit cell had lattice constants of 5mm × 5mm × 5mm and struts diameter of 1mm. Subsequently, a plane at the centre of the global cube and perpendicular to the X axis was defined as the boundary between two twin meta-grains. Lattices on each side of the boundary were rotated to the same angles but in opposite directions: lattices in the right side of the boundary (Extended data - Figure E2c) were first rotated counter-clockwise by 45° about X axis (Extended data - Figure E1d), and then 45° about the Y axis to generate a meta-grain of FCC lattice. Such a rotation sequence for the meta-grain is shown in Extended data - Figure E1c-e where x'', y'' and z'' axes were the final coordinates of lattices after the two counter-clockwise rotations. By contrast, lattices in the other side were rotated clockwise by 45° about the X, following by 45° about the Y axis to create the second twinned meta-grain. The two constructed meta-grains were shown in Extended data - Figure E2a with the one on the right-hand side highlighted. Because of the globally constrained dimension (40mm x 40mm x 40mm), the rotation of lattices caused incomplete unit cells on free surfaces (and at the boundaries between meta-grains) (Figure 2a), this weakened the architectured material containing multiple meta-grains with different orientations. A planar lattice frame with a 2D unit cell containing a centred node was introduced to help maintain the connectivity of struts (Extended data - Figure E2b). The dimension of the planar unit cell is 5mm x 5mm with struts diameter of 1mm. The size of meta-grains was reduced by further dividing the global cube into smaller meta-grains, i.e., increasing the number of meta-grains from 2 to 4, 8, 16, 18 and 27 (Extended data - Figure E2b-e).

A cube (Extended data - Figure E3) consisted of 8 meta-grains separated by incoherently high angle boundaries was designed to study the role of misorientation on crack propagation (Figure 2d) and the deformation of lattices in different base materials (Extended data - Figure E9). All meta-grains had the same size: 20mm × 20mm × 20mm (Extended data - Figure E3a). The unit cell was FCC with constants of 5mm × 5mm × 5mm and the strut diameter of 1mm. The cube consisted of two layers, each comprises 4 meta-grains. In the top layer, the top right meta-grain with the three [100] orientations coinciding with the global X, Y and Z axes (Extended data - Figure E3b) was chosen as the reference meta-grain. The unit cells of the three neighbouring meta-grains (moving anticlockwise) in the same layer were created by rotating the unit cell of the reference meta-grain by 30°, 45° and 60° about the Z axis (Extended data - Figure E3b). The reference point of rotation was the centre of the original unit cell. Meta-grains in the bottom layer were created in the similar way, but with a different sequence order (Extended data - Figure E3c) with an aim to increase the randomness of the misorientation distribution. The outer frame was not introduced to reveal the stopping effect of high angle boundaries on the crack propagation (Figure 2g).

1.2. Mimicry of precipitates

Precipitates were mimicked by embedded lattice domains (termed meta-precipitates) that have different lattice parameters from those in the matrix. Lattice parameters (type, spacing, orientation) of meta-precipitates can be tailored to design the different degrees of coherency between precipitates and the matrix. The diameters of strut lattices in meta-precipitates can be varied to account for different "atomic" bonds to mimic an ordered arrangement of atomic bondings, e.g., the Ni-Al in γ' in Nickel superalloys. Meta-precipitates in Figure 3c and d were designed by embedding twenty-five cubic meta-precipitates in a FCC matrix. Meta-precipitates near the free surfaces of the global cube and their locations in the matrix were shown in Extended data - Figures E4a and b, respectively. The [001]-orientations of the matrix unit cell were parallel to the global X, Y and Z axes (Extended data - Figure E4a). The unit cell of the matrix was FCC with the lattice constants of 5mm × 5mm × 5mm and the strut diameter of 1mm, Extended data - Figure E4c. Each meta-precipitate had the dimension of 7.5mm × 7.5mm × 7.5mm, and was made of a face-centred-tetragonal (FCT) unit cell with lattice constants of 3mm × 3mm × 4mm and the strut diameter of 1mm (Extended data - Figure E4d). The orientations of meta-precipitates were randomly rotated. Frames were introduced as the interfaces between the matrix and precipitates to increase the connectivity of struts across the interfaces.

1.3. Mimicry of phases

As a crystal phase is defined by its lattice type, multiple phases can be imitated by assigning different lattice types to different macro-lattice domains. Single FCC (or single BCC) macro-phase was generated by an FCC (or BCC) unit cell (Extended data - Figure E5). The global dimension of the multiple meta-phases was 40mm × 40mm × 40mm. The FCC and BCC unit cells had the same lattice constants (5mm × 5mm × 5mm) and the same strut diameter (1mm).

The mixture of the two FCC and BCC phases consisting fifty Voronoi meta-grains was designed by filling the 50 meta-grains with the FCC and BCC unit cells (Extended data - Figure E5c). The global (40mm × 40mm × 40mm) cube comprising the two meta-phases was divided into three layers: top, middle and bottom layers. Meta-grains in top and bottom were filled by the FCC phase while the middle layer meta-grains were made of the BCC phase: 11 FCC meta-grains for the top layer, 25 BCC meta-grains for the middle layer, and 14 FCC meta-grains for the bottom layer. The orientations of lattices in all the meta-grains were randomly rotated to increase the isotropy of the lattice cube.

1.4. Kresling lattice

A Kresling lattice cylinder was constructed to imitate a hexagonal close packed (HCP) phase (Extended data - Figure E6a) (Methods reference 1). Struts and nodes on the bottom (*ABCDEF*) and top (*A'B'C'D'E'F'*) surfaces of the Kresling unit cell formed two regular hexagons that are parallel and equal to the other. Similar to the terminology in the HCP crystal phase, the (*ABCDEF*) and (*A'B'C'D'E'F'*) were the two basal planes. The bottom hexagon was created with the side length of 8mm. Subsequently, the top hexagon *A'B'C'D'E'F'* was formed by translating the bottom hexagon along the z axis (i.e., c-axis), and then rotating the translated hexagon clockwise of 30° about the c-axis (Extended data - Figure E6a). The $\angle AOH$ (with H is the middle of AF) was 30°, making HA' perpendicular to AF. The c-parameter (i.e., the length of OO') of the Kresling HCP unit cell was 8mm. The unit cell was created by connecting *AA'*, *A'B*, *BB'*, *B'C*, *CC'*, *C'D*, *DD'*, *D'E*, *EE'*, *E'F*, *FF'* and *F'A*. The Kresling HCP-like phase was built by stacking three Kresling unit cells along the c-axis. The total height of the Kresling HCP-like phase was 24 mm with strut diameter of 3mm for *AA'*, *BB'*, *CC'*, *DD'*, *EE'* and *FF'*; 2mm for the diagonal struts and for the basal struts (Extended data - Figure E6b).

2. Materials and fabrication

Various base materials were used to fabricate designed materials to demonstrate the applicability of the polygrain-inspired approach. The choice of the base material was based on the phenomenon of interest. For example, while ductile polylactic Acid (PLA) polymer and stainless steel were for studying the shear band activity in architectured materials, a polymerisation resin that is brittle after curing and heat treatment was used to study the crack propagation in architectured materials.

Cubic (except for Figure 2g) and Kresling lattices were made of PLA filaments and a thermoplastic co-polyester Natural FLEX 45 filaments, respectively. The filaments were provided by the RS Limited. Lattices were fabricated by a fused deposition modelling (FDM) Ultimaker 2 printer. Designed models of meta-crystals were sliced into sequential layers by a pre-processing software Cura with the layer height of 0.1mm. The nozzle of the printer was heated (to 210°C for PLA and to 215°C for co-polyester) to melt the filament. The nozzle diameter is 0.4 mm. The molten material was extruded and deposited on a build plate layer upon layer according to the sliced sequences of designed lattices. The build platform was heated (to 60°C for the PLA and to 100°C for co-polyester) to increase the bonding between layers. The nozzle speed was 30 mm/s for PLA and 15 mm/s for co-polyester

To study the role of meta-grain boundaries in the propagation of macro-cracks, both single meta-grain and 8 meta-grains separated by incoincidently high angle boundaries (Extended data - Figure E3) were made of a photopolymerisation resin (Visijet M2 RWT) provided by 3D Systems Limited. The resin was brittle after curing and subsequently heat treatment. The meta-crystals were printed by a stereolithography (SLA) printer (ProJet MJP 2500) in which resin is cured by Ultraviolet (UV) light. Similar to FDM process, meta-crystal models were firstly sliced into layers with thickness of 0.1mm. The UV light cured resin according to the sliced sequence of designed models. A wax material was used to support built lattices, this helps improving the geometrical accuracy and quality of lattices. The wax was removed from built lattices by post-treatment in an oven with temperature of 60°C for a duration of 5 hours.

To study the influence of the base material on the behaviour of polycrystal-like lattices, the 8 meta-grains (Extended data - Figure E3) were fabricated by both the elasto-plastic PLA and a 316L austenitic stainless steel. While the PLA was printed by Ultimaker 2 with parameters given above, the steel was fabricated using a Renishaw AM250 printer in argon atmosphere. The dimensions of the 8 meta-grains were reduced by a factor of 2 for 316L, i.e. the global dimensions: 20mm x 20mm x 20mm, the size of each meta-grain: 10mm x 10mm x 10mm, lattice constants: 2.5mm x 2.5mm x 2.5mm and strut diameter: 0.5mm. A hatch pattern was used for inner area, with a power intensity of 180 W, a spacing of 150 μm, an exposure time of 110 μs and a point distance of 65 μm. The outer skin was made by two contours with a power intensity of 100 W, a spacing of 55 μm, an exposure time of 90 μs and a point distance of 40 μm.

The mechanical properties of the PLA and co-polyester filaments, brittle resin and 316L steel are given in Table E1.

3. Microstructure characterisation

Scanning electron microscopy (SEM) imaging and electron backscatter diffraction (EBSD) were carried out on an austenitic stainless steel 316L using a Zeiss Auriga SEM at a voltage of 20 kV for Figure 4e, f. γ/γ' microstructure in a Ni single crystal (with a composition similar to CMSX4) was revealed by chemical etching (2.5% phosphoric acid in methanol solution, applying 10 V for 30 s). Microscopic observation of γ/γ' microstructure (Figure 4h) was done using SEM Sigma300 at a voltage of 10kV.

4. Mechanical tests and analyses

Mechanical properties of all macro-lattices were studied by compression tests that were carried out by a 100kN Zwick machine using the displacement control at a strain rate of 10^{-3} 1/s at room temperature. To minimise the effect of friction, the top and bottom compression plates were lubricated with dry molybdenum disulphide. The loading direction was parallel to the build direction (i.e., the Z direction) of prints. Stresses were calculated by dividing the recorded forces by the nominal area, which enclosed lattices and was perpendicular to the Z direction. Engineering strains were derived by dividing the change in the length along the Z direction by the initial length: 40mm for the polymer lattice cubes or 24mm for the Kresling lattice. The compression tests of meta-crystals were recorded by a Nikon D7100 camera with 200mm Nikkor macro lenses for taking images for post-test analyses, and Canon SX210 for recording videos. Captured images were characterized by digital image correlation (DIC) via a commercial software DaVis. The resolution of analyzed image is $6000\, pixels \times 4000\, pixels$. The subset dimensions used in DIC analysis were $101\, pixels \times 101\, pixels$ with step size of $25\, pixels$. Matlab subroutines were written to analyse the stress-strain behaviour of tested lattices. Fitting by a linear function over different ranges of the most linear part (up to 2% strain) of a stress-strain curve was done to identify the elastic modulus. The yield strength was defined as the stress corresponds to the 0.2% strain offset of the fitting line.

5. Finite element analysis

Finite element method (FEM) was used to simulate the deformation of macroscopic lattices. Model of macro-lattices generated by nTopology Element software were imported to Abaqus for FEM simulations. The quasi-static compression of a macro-lattice was modelled using two displacement controlled rigid-body plates. The first rigid body plate was fixed underneath the macro-lattice with an encastre boundary condition. The second rigid body plate was used to compress the macro-lattice with a displacement boundary condition. The General Contact algorithm available in Abaqus was employed to simulate the interactions between all surfaces, with a penalty friction coefficient of 0.1 and ‘hard’ pressure-overclosure surface behaviour defined. Macro-lattices were meshed using 10-node quadratic tetrahedral elements (C3D10) with isotropic elastic-plastic material behaviour that was defined according to the mechanical behaviour data given in the Table E1. Because of the symmetry and periodicity of the arrangement of unit cells in the meta-grain twins, a reduced FEM model of the meta-grain twins was constructed. The FEM model consists the twin boundary and 3x3 unit cells in each meta-grain, and was constructed by 1,184,608 elements. The Kresling lattice was builted by 140,120 elements. The rigid body plates were meshed using 4-node bi-linear quadrilateral elements (R3D4).

Methods references

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Main figure captions

Figure 1: Lattice structures and deformation behaviour. (a) Face-centred-cubic (FCC) crystal lattice, (b) Single slips in a single crystal (re-used from [8] with permission from Elsevier), (c) Slips at grain boundary in a polycrystalline steel (re-used from[9] with permission from Elsevier), (d) Achitectured FCC lattice, (e) Single slip in a single oriented lattice, (f) Unstable behaviour of architectured metallic lattices (reused from [10] under the Creative Commons Attribution 4.0 International License).

Figure 2: Roles of lattice orientation in the deformation behaviour of crystals and architectured lattices. (a) Twin bi-crystal (re-used from [27] with permission from Elsevier), Shear bands in meta-grain twins observed in experiment (b) and predicted by FEM (c) (Note: Cut sections shows the deformation of internal struts. The sections were formed thanks to two cutting planes that were parallel to {001} planes of FCC lattice), (d) – (e): Shear bands were controlled by orientation of meta-grains: (d) 8 meta-grains and (e) 16 meta-grains (note the nominal strain was of 30%). (f) Yield strength versus the size of meta-grains, (g) Boundaries between meta-grains effectively stops cracks in brittle lattices, leading to a drastic increase in toughness of architectured materials.

Figure 3: Precipitation and multiphase hardening in architectured materials. (a) A single oriented without meta-precipitates. (b) Constitutive stress-strain responses of architectured materials without meta-precipitates (a) and with meta-precipitates (c, d). (e) Mechanical behaviour of single phase versus multiphase architectured materials. (f) Pseudo-superelasticity of Kresling lattice, (g) FEM simulation shows the strain localisation and local buckling of struts, and (h) Energy per unit volume of the first five cycles.

Figure 4: Lightweight and damage-tolerant architectured materials inspired by crystal microstructure. (a)-(d) Lightweight lattice component. (e)-(g) FCC fractal lattices from atomic up to cm scales, (h) γ/γ' scalable fractal super-lattices.

Extended data table captions

Table E1: Mechanical properties of printed polymers. * data provided by RS Limited and 3D Systems Limited. ** data obtained from 3 tensile tests of solid cylindrical samples (fabricated by powder-bed selective laser fusion) at room temperature and a strain rate of 10^{-3} 1/s).

Extended data figure captions

Figure E1: Mimicry of crystal lattice. (a) Unit cell of lattice, (b) A macro-lattice cube consisting of 8 x 8 x 8 macro-unit cells. (c) – (e) The rotation sequence to form a twin meta-grain of lattice.

Figure E2: A different number of meta-grains within the same global volume (40mm x 40mm x 40mm). (a) 1 meta-grain, (b)-(c) 2 twinned meta-grains: (b) with outer frame and (c) without the outer frame, (d) 4 meta-grains, (e) 8 meta-grains, (f) 16 meta-grains, (g) 18 meta-grains and (e) 27 meta-grains. The locations of boundaries were highlighted.

Figure E3: Mimicry of crystallie grains seperated by incoherent high angle boundaries. (a) Model of 8 meta-grains. The orientations of lattices in the four meta-grains in (b) the top layer and (c) the bottom layer

Figure E4: Mimicry of precipitates. (a) Meta-precipitate lattice. (b) Cubic morphology and locations of meta-precipitates inside the FCC meta-phase. (c) FCC unit cell of the matrix and (d) FCT unit cell of meta-precipitate.

Figure E5: Mimicry of multi-phases. (a) Single meta-grain of FCC meta-phase, (b) Single meta-grain of BCC meta-phase, (c) A cube of meta-polygrains consisting of two meta-phases: FCC (top and bottom layers) and BCC (middle layer).

Figure E6: Kresling lattice. (a) unit cell, (b) HCP meta-phase

Figure E7: The repeatibility of mechanical behaviour of architectured materials. (a), (b) and (c) materials consist of 2, 8 and 16 meta-grains, respectively; and (d) materials contain 25 meta-precipitates.

Figure E8: Effect of the size of meta-grains. (a) Stress-strain curves of architectured materials consisting a different number of Voronoi domains of lattices. (b) Flow stress of architectured materials containing meta-grains at a given nominal strain of 40% increases with reducing the size of meta-grains.

Figure E9: Deformation behaviours of an architectured material containing 8 meta-grains separated by incoherent high angle boundaries. (a) and (b) the macro-lattice was fabricated by 316L stainless steel and elasto-plastic polymer, respectively. (c) and (d) stress-strain constitutive behaviour of the macro-lattices fabricated by the steel and polymer.

Figure E10: Helical movement enables the change in the stack sequence of nodes. Red lines represent helical movements of basal nodes. Note: only the movement trajectory of basal nodes on the top plane were shown by the red curves.