Highly Mineralized 2D Natural Composites Structure Opens Pathways to Bioinspired MXene-Based Composites

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# Abstract

MXene is a new class of two-dimensional (2D) materials with an excellent combination of mechanical, chemical, and electrical properties. These materials can be designed for specialized applications using a plethora of element combinations and surface termination layers, making them an attractive material for highly optimized multifunctional composites. However, multiple critical engineering applications demand that such composites balance specialized functions with mechanical demands. The current knowledge of the mechanical performance and optimized traits necessary for MXene-based composite design is severely limited. Here we review structure-function connections for highly mineralized 2D natural composites such as nacre and exoskeletal of windowpane oysters to extract underlying design principles relevant for MXene-based engineered systems. Finally, we discuss future perspectives and opportunities for MXene-based composites in areas of multiscale modeling, nanomechanical characterization, and potential machine-learning design approaches to present a path forward.

# Keywords

MXene, Bioinspired, 2D materials, nacre, multiscale modeling, machine learning

# Statements and Declarations

## Competing interests

None

## Conflict of Interests

The authors declare that they have no conflict of interest

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## Authors' Contributions

AP prepared the main write-up. VV and DN contributed equally during the manuscript write-up and preparation of figures. GJF contributed towards the manuscript design and revisions. MR performed literature review for material property acquisition and prepared a MATLAB program for plotting several figures. All authors read and approved the final manuscript.

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# Introduction

As traditional engineering materials are quickly achieving their performance limits, there is a pressing need of next-generation materials for a wide range of applications in biomedical, robotics, aerospace, and defense. New demands include stretchable materials for soft robotic and electronics [1,2], ultrathin flexible materials for electromagnetic interference shielding [3], high density and mechanically robust energy storage devices [4,5], biocompatible and mechanically optimized biosensors [6,7], and damage-tolerant flexible body armors [8,9], to name a few. These examples highlight the multifunctionality requirements of next-generational materials. The critical functional focus, such as biosensing, electromagnetic interference (EMI) shielding, or damage-tolerance, needs to be balanced with structural requirements of toughness and flexibility. Balancing functional needs and structural requirements necessitates the discovery of new materials and design traits.

Recently several classes of two-dimensional (2D) materials (both elemental and hetero-nuclear) have been synthesized, including graphene, borophene, transition metal dichalcogenides or TMDs, and MXenes, each of which has a unique combination of mechanical, chemical, and electrical properties [10–15]. Significant effort has focused on their processing and on identifying new applications [15,16]. Among these, MXenes are the most recent entry with a general representation of (M = early transition metals, X = carbon or nitrogen, and T = functional surface terminations). MXenes can exist in several forms through element permutations and surface terminations and thus, offer an exciting opportunity for a wide range of applications from body armor, EMI shielding, eye shields, and sensors [10,16–19]. MXenes have a sizeable in-plane area and high in-plane stiffness. However, MXenes based processed films are brittle and are relatively weak in shear and have low out-of-plane stiffness [20]. These characteristics can affect the design and selection of suitable MXene-based composites for various applications. To date, while there has been an immense early focus on synthesizing various MXene structures, developing MXene-based composite designs with control over geometry has received far less attention.

While MXenes have similar geometrical and property constraints as 2D structures like graphene, it is notably different from the single-layered and single element-based (carbon) structure of graphene. MXenes can have multiple element combinations and can exist as several atomic layers thick, represented by the "n” in the MXene formula. For example, and are monolayer and bilayer MXenes with n=1 and 2, respectively. Due to their greater thickness, MXenes have higher bending stiffness than graphene. Through elemental combinations and design of surface termination layers, MXenes can have many different functional interactions between layers. The mechanical design requirements for MXene-based composites can also significantly vary across applications. For example, in EMI shielding applications for miniature mobile devices, the demand for mechanical flexibility and environmental stability should not compromise its primary function of providing an ultrathin layer with high electrical conductivity [3]. Similarly, in next-generation body armor, the demand for increasing flexibility and decreasing weight to improve wearability should not compromise its primary goal of higher toughness. Hence, MXene-based design for specific applications requires a careful selection of optimized design features and a suitable choice of composite phase to balance the different performance demands.

Natural composites provide an excellent template of tightly balanced design traits that have been carefully optimized over several millennia through evolutionary forces [21,22]. Specifically, layered architecture with carefully designed interfaces is the hallmark of many natural composites allowing them to achieve strength and functionality far exceeding their constituents [21,22]. For example, despite being made from more than 95% mineral, nacre achieves three orders of higher fracture resistance than its mineral phase through its unique brick-and-mortar organization and careful design of organic-mineral interfaces [23,24]. The exoskeletal forewings of the diabolical ironclad beetle (*Phloeodes diabolicus*) despite having 88 mineral content, achieve extreme toughness through a combination of design features including laminated microstructure, stiffness gradient, and ellipsoidal geometry [25]. The ultrathin coating (≈ 70 ) on the dactyl club of mantis shrimps has 88 densely packed hydroxyapatite nanoparticles, but it achieves high toughness and impact resistance through an interspersed organic matrix within its nanoparticles [26]. The geomaterial sheet silicates montmorillonite (MMT) or nanoclay is another example of a 2D composite that has been extensively used for membrane separation and flame retardant due to its increased thermal stability, hydration swelling, and water dispersion properties [27–30].

Exploring the structure-function relationship of these highly mineralized 2D natural composites provides an excellent template for designing MXene-based engineered composites. With that aim, this paper first presents examples of recently developed 2D MXenes and MXene-based composites to illustrate examples of layered architecture typically achieved in MXenes, draw similarities of the architecture to mineralized 2D layered natural materials, and identify the relevance of certain features to EMI shielding applications. This is followed by a review of highly mineralized 2D layered natural composites to identify unique structural and design traits governing their mechanical stiffness and toughness response. Following the above, we connect back to MXene and MXene-based composites to discuss the relevance of these bioinspired design features to their design and present the current state of MXene-composite processing with examples from applications beyond EMI, and identify key challenges for their design. The paper concludes with a discussion of some exciting opportunities in MXene-based nanocomposite design for future work.

# Two-dimensional layered MXene composites

MXenes are versatile materials with applications in many different fields [10,16]. The first example of MXene-based layered composite film developed showed high capacitance, demonstrating its applications for flexible and wearable energy storage devices [31]. In another early example, layered MXene-based composite film showed high EMI shielding capability comparable to metals of the same thickness [32]. In general, layered MXene-based structures find applications such as supercapacitance, EMI shielding, energy storage, wastewater treatment, air filtration, gas separators, body armors, and wearable electronics [33]. The desired structural traits of MXene-based composites will vary across these different applications, making the design requirements challenging to cover its vast field. Here, we limit our discussion to heavily filled layered MXene-polymer composite (MPC) for EMI shielding due to its immense demand. Multiple recent reviews have discussed MPCs and their applications [33,34], while others have focused on EMI shielding application needs and materials systems [35–38]. Hence below is not a review of the field of EMI shielding and materials but instead summarizes structural and materials parameters affecting shielding effectiveness and presents some examples of free-standing MXenes and MPCs developed for EMI shielding application.

The effectiveness of EMI shielding is quantified by the measure of its shielding efficiency (). Fig. 1a shows a schematic of a layered material subjected to EM wave and shielding contributions. The measured in is given by equation 1 and includes contributions from external reflection (), absorbance (), and internal surface reflection (). These variables in turn depend on the frequency of the incident wave (), material properties such as electrical conductivity (), relative permeability (), and relative permittivity (), and structural parameters such as porosity, layering, and depth () [35,36,39,40]. Furthermore, many EMI shielding applications such as those in miniaturized mobile devices require ultrathin, mechanically robust, and environmentally stable shields. These demands need to be balanced with the primary function of high conductivity, wavelength-tunable absorption, and internal reflections to improve .

equation [1]

MXenes and MPC fulfill these multifunctional demands. Single pristine MXene sheets are only a few atoms thick with high electrical conductivity and high in-plane stiffness. Typically, several of such sheets are layered together during processing and are called flakes, one example is shown in Fig. 1b. Using these MXene sheets and flakes as building blocks, several ultrathin free-standing and MPC shields have also been developed in the foam, aerogel, and more coearannepalmmon layered film form (example in Fig. 1c). A few examples of MXene and layered MPC films primarily directed for EMI applications are discussed below.

Various pristine MXene films have been produced by the common technique of acid etching followed by post-processing as necessary for enhancing performance. A hydrofluoric etching was followed by hydrazine-induced foaming to form hydrophobic porous thin flexible films with 70 [41]**.** Another study used hydrofluoric acid etching to form ultrathin mono and bilayer of free-standing MXene () 1.6 thick and in-plane dimension of 10 , and demonstrated high mechanical performance with Young’s modulus of 330 GPa [42]. Pristine multilayer MXene flakes () with up to 60 and environmental stability were produced first by acid etching, followed up by the removal of external intercalants between layers by using acid colloidal treatment [43]. In another work, MXene () inks were prepared using a high viscosity solvent (DMSO or dimethylsulfoxide) and synthetic binder molecules [44]. The prepared ink was inkjet printed on multiple substrates, resulting in highly conductive and humidity-sensitive flexible films with up to 50 . Other approaches for enhancing the and strength of free-standing films included heat treatment [45] and surface functionalization [46]. MXenes films with 16 different combinations of MXenes were prepared to show EMI applications beyond the family [47]. A significantly faster MXene-film formation was demonstrated by combining vacuum filtration with ion-induced dispersion of MXenes () into microgels [48]. The modified sheets showed 49 to 94 depending on film thickness and stacking.

MPC for EMI involves MXene film preparation and their organization in layers in the presence of a polymer material. A hydrofluoric etched MXene () film was stacked with nacre-like interlayer binders using sodium alginate, which resulted in structurally stable and high internally reflecting films of 8 and of 57 [32]. Another biomimetic nacre-like organization used etched MXene layers ultrasonically mixed with conductive polymers poly(3,4-ethylenedioxythiophene)–poly(styrenesulfonate) (PEDOT:PSS) to develop multilayered and flexible / PEDOT:PSS composite with high tensile strength and 42 in a film thickness of 11 [49]. Similarly, cellulose nanofibers (CNF) have been used in producing layered MPC with enhanced mechanical properties and of 40 [50,51]. Other approaches include the use of polyvinyl alcohol (PVA) [52] and multiphase materials such as MXene/CNF/carbon nanotubes (CNT) [53] and MXene/CNF/Ag nanoparticles [54] to improve shielding performance. Examples also include MXene/Ag nanowire/ poly(vinylidene fluoride (/PVDF) composite thick films prepared by solution casting with of up to 42 for 600 film thickness [55]. In other recent examples, a multilayered MXene/ polyurethane films film was prepared by layer-by-layer spraying [56]. The film showed of 51 with 28.6 MXene and film thickness of 52 . MXene sheets () anchored with nanoparticles between layers (/)were assembled using electrostatic self-assembling and freeze-drying technique was organized in a matrix of paraffin wax [57]. With only 3% of /, the composite design showed of 42 , attributed to the presence of which prevented closed self-stacking of MXene layers.

The above demonstrates multiple examples and significant progress to date in processing pristine and layered MXene structures using natural and synthetic polymers as adhesives for interlayer stability for EMI applications. However, a fundamental understanding of the mechanics of free-standing MXenes and MPCs and the correlation between structural design and electromechanical performance is limited. Simultaneously, while polymer additives are used in MPCs for structural stability and property enhancement, understanding the MXene-polymer interfaces and interactions is needed to select suitable MXene-polymer systems for EMI and other targeted applications. With the above two focuses, we look into some highly mineralized and layered 2D natural composites and their mineral-organic phase interactions to extract design rules for heavily filled MPCs.

**Graphical user interface

Description automatically generated with medium confidence**

**Fig 1**: Schematic of EMI shielding and examples of ultrathin layered MXenes and MPC films. (a) Layered material subjected to incident EM wave () with shielding via reflection (), absorbance (), and internal reflections (b) Free-standing MXene flake () post annealing at 350 0C showing significant internal porosity [45]. (c,d) MPC composed of alternating layers of and CNF for improved mechanical performance and [51]. Images b to d taken from cited literatures.

# Two-dimensional layered natural composites

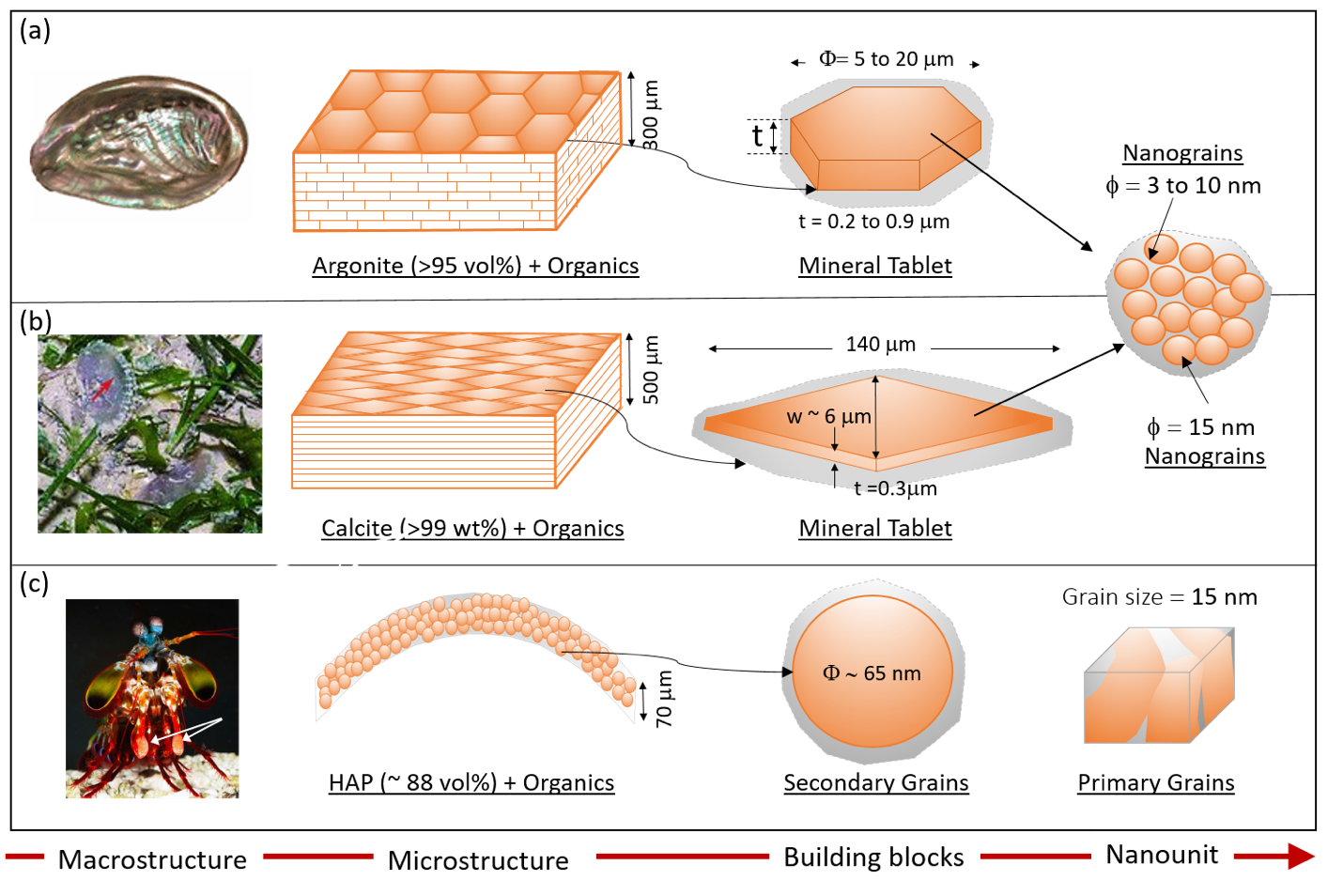
Classic examples of 2D natural composites with high mineralization (> 80 ) include nacreous shells, the exoskeleton of windowpane oyster (WO)*,* and the thin outer coating on the dactyl club of mantis shrimp (DCMS). Due to their superior mechanical properties, specifically high toughness and low compressive to tensile strength ratios, the structural organization and underlying mechanisms driving their behavior have been the focus of intense investigations [21,22,24,58,59,59–62]. While the nacre and exoskeleton of WO have layered organization and thus have direct similarity with the layered MPCs illustrated above, the DCMS structure is nanograined. However, DCMS do possess overlapping features of relevance to MPCs such as high mineralization and mineral-polymer interactions which are responsible for their superior toughness under impact loading. Furthermore, as we will discuss later, nanogranularity is another overlapping feature that is present not only in DCMS but also within the tablets of nacre and WO. Hence DCMS is also included in the discussion below on natural systems of relevance to MXenes and MPCs. Here, some key aspects of the structural organization are briefly summarized to identify and compare their design features for driving further discussions. Fig. 2 shows the structural organization of the three natural composites.

## Structural organization of 2D layered natural composites

Nacre is a classic example of a 2D layered structure and is present in the inner surface of the exoskeletal of certain molluscan shells. It is primarily made from stiff mineralized tablets (≈ 95 aragonite or ) interspersed with soft organic polymer phase (polysaccharide and hydrated protein) in a brick-and-mortar architecture [63,64]. Fig. 2a shows the hierarchical organization of nacre derived from molluscan shells. The “brick” is the aragonite present as a continuous lamellar sheet of plate-like polygonal tablets 5 to 20 diameter and 0.2 to 0.9 thick, with an aspect ratio of 8 to 14 [24,65–67]. Each tablet is “glued” to the adjacent tablets via a thick intra-tablet organic “mortar” layer 20 to 50 thick [23,68]. The tablets are not single-crystal but instead are made from clusters of “mesocrystal” comprised of polygonal aragonite nanograins 3 to 10 in size, again glued using the interspersed polymer phase between the nanograins [69–72]. Multiple parallel tablet-polymer zones are arranged together in a staggered brick-mortar organization to achieve a total thickness of 300 [23]. This tiled nacreous zone is sandwiched between mesolayers 20 thick, which is also made of mineral phase with interspersed organic phase [23,73,74]. The mesolayers are also called the growth band since it is believed to originate during the growth phase due to the variation in the feeding and temperature patterns, resulting in interruptions in the brick-mortar layering pattern.

The exoskeleton of WO (*Placuna placenta*) is another example of a highly mineralized structure (≈ 99 calcite or ), which can simultaneously achieve high optical transparency, stiffness, toughness, and high strain impact resistance [75,76]. Like nacre, it has a lamellar organization made from mineral tablets and interspersed organics. The tablets are elongated diamond-shaped calcite tiles (length 140 , width 6 tip angle 10°, and thickness 0.3 ) glued together by an ultrathin (≈ 2 ) organic phase (Fig. 2b). The entire shell comprises 2000 such laminar layers, leading to a total thickness of approximately 500 . Many pairs of screw-like connection centers are present within each layer to provide growth pathways for biomineralization [77–79]. These connection centers also play a significant role in energy dissipation via interlocking of tablets and localizing damage [75], as discussed in later sections.

The outer coating of DCMS is another example of mineralized material (≈ 88 Hydroxyapatite or HAP) designed for impact resistance [26,80,81]. Its structural architecture is different from the tablet-like laminated organization of nacre and WO but shares with them the nanogranularity feature which is present within the tablets of layered systems and here at two different length scales (primary and secondary grains). The ultrathin (≈ 70 ) outer coating is made from dense packing of crystalline HAP nanoparticles or secondary grains (≈ 65 ) embedded in an organic matrix (polysaccharide and protein). The nanoparticles are not single crystals [81] but instead consist of highly aligned primary grains (≈ 15 ) of preferred orientation, separated from adjacent primary grains with low-angle grain boundaries [26]. The hydrated organic matrix interpenetrates the primary and the secondary grains, similar to the organic material present in inter- and intra-table regions of nacre and WO. The grain boundaries and the organic phase provide pathways for energy dissipation and crack localization, leading to extreme damage resistance under high impact [26,82], as discussed in later sections.



**Fig. 2**: Structural organization of thin mineralized composite (a) nacre, (b) exoskeleton of windowpane oyster (WO), and (c) outer coating on dactyl club of mantis shrimp (DCMS) showing the mineral-organic organization at the microscale, underlying building block, and the nanounit within the building block (macrostructure courtesy: nacre [83], windowpane oyster [76], and mantis shrimp [26]).

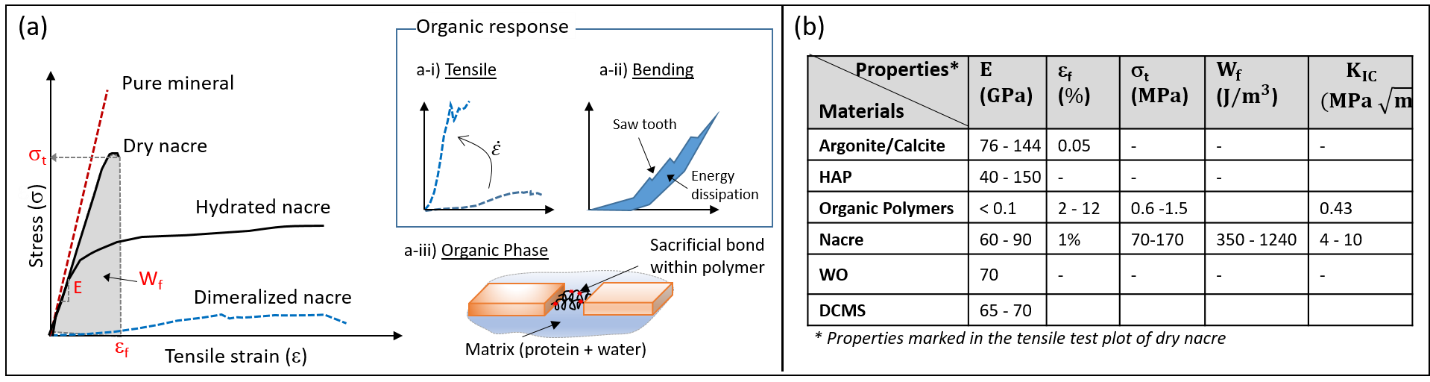
## Mechanical response of 2D layered natural composites

The structural organization of the nacre, the exoskeleton of WO, and the outer coating of DCMS show examples of highly mineralized composite made of nanogranulated minerals organized either in laminar tablets or in crystalline grain-like structure, interspersed with soft organic materials. The above structural organization leads to their unique mechanical response. Fig. 3a shows a schematic of the typical stress-strain response of nacre and its constituents. Fig. 3b shows values of some of the common mechanical properties of these natural composites and their constituents.

The minerals aragonite and calcite are brittle and orthotropic with elastic modulus between 76 to 144 , and a low strain to failure of 0.05% [63,84–86]. Similarly, HAP is brittle with a modulus of 40 to 150 , the higher of the values reported for synthetic crystals [87,88]. The organic polymers (polysaccharide and hydrated protein) have a significantly lower modulus of 1 to 136 depending on the strain rate, low strength of 0.6 to 1.5 , and fracture toughness of 0.43 [63,89–91]. However, it shows significant strain to failure (2 to 12%), deformation strengthening, and viscoelasticity [89–91]. In contrast to their constitute phases, all of the natural composites offer a remarkable combination of properties. Nacre has been the most widely studied material among the three systems. It has a tensile elastic modulus of 60 to 90 comparable to its mineral phase and shows strain hardening post-yield with large strain to failure (≈ 1 to 8%) [24,69,92–94]. The tensile strength varies from 70 to 170 with a work of fracture between 350 to 1240 [24,92,94]. Its fracture toughness is reported between 4 to 10 [94–96]. These properties of nacre are in stark contrast to its primary constituent aragonite, for which the is 3000 times lower and and is almost 20 to 30 times smaller [61,96].

Though limited experimental data is available for WO [75,76] and DCMS [80,97], they show a similar trend as nacre, unique from their primary mineral constituent. Indentation tests on freshly cleaved WO reveal a high elastic modulus of 70 that is comparable to the modulus of a single calcite crystal, but a substantially increased plasticity with 55% higher hardness (3.8 vs. 2.5 for calcite) and localized damaged response [75,76]. A theoretical calculation of interfacial energy dissipation in WO revealed interfacial fracture toughness of 100 , which is two orders of magnitude higher than its constituent mineral crystal [75]. Unlike nacre and exoskeleton of WO that fail under high strain rate impact [22], DCMS coating can resist high-velocity impact [80,97]. It has a similar modulus of 65 to 70 as nacre and exoskeleton of WO [80,97]. These data reveal that the structure is highly optimized for extreme toughness and fracture resistance, far higher than its constituent materials.

The properties reported above have high sensitivity to hydration, as revealed through testing of nacre. Fig. 3a shows schematically the typical response under dry and hydrated conditions, revealing the hygromechanical sensitive response. Lower modulus and higher failure strain correspond to a hydrated condition of testing [66,94,98]. Hydrated nacre also shows higher plasticity and viscoelasticity, observed through the hysteresis loops in tension or material pile-up in indentation [93,99,100]. The property modification of nacre in the presence of water can be attributed to multiple factors, such as the plasticizing effect of bulk water presence in inter and intra-tablet mineral gaps and pores, a reduction of surface energies through chemical absorption of water on mineral surfaces, and the stabilization via hydrogen bonding of the organic phase [94,101,102]. These enable the gradual transition of stress from the organic phase to the tablet interfaces during sliding, resulting in higher toughness [102]. Consequently, properties of nacre specifically its toughness and high strain rate sensitivity are highly influenced by its organic phase. The insets (a-i) and (a-ii) of Fig. 3a show the typical response of the organic phase. The tensile curve shows high strain-rate sensitivity. The bending curve shows variable stiffness under loading with saw-tooth-like patterns and energy dissipation upon unloading. Many of the features of the response of hydrated nacre and other natural composites can be attributed to the mechanical influence of the hydrated matrix explained above, though other features such as its helical structure and sacrificial bonds also play an important role and are discussed later.



**Fig. 3**: (a) Schematic views of stress-strain response of nacre under dry and hydrated conditions based on properties reported in the literature, and (b) summary of typical values reported for constituent phases and corresponding natural composites (E = Elastic modulus, = strain to failure, =tensile strength, = work of fracture measured as the area under the stress-strain curve, and = mode-I fracture toughness). The inset in “a” shows the response of the organic phase under tensile (a-i) and bending (a-ii) loading, with some unique characteristics revealed including strain-rate sensitivity, saw-tooth pattern, and energy dissipation. These characteristics can be explained through the presence of a hydrated matrix comprising of organic polymer with helical polymer structure and sacrificial bonds (Fig a-iii). Data is taken from multiple sources: aragonite and calcite [63,84–86], HAP [87,88], organic phase [63,89–91], nacre [69,92–94,96], WO [75,76], and DCMS [80,97].

# Design features of mineralized composites for stiffness and toughness

Several design features lead to the extraordinary mechanical response of these highly mineralized composites, which can be assigned to three broad categories, namely (a) nanogranular organization, (b) mechanical interlocking of tablet interfaces, and (c) interphase zone of organic materials. These key design traits along with additional mechanisms such as deformation twinning and aspect ratios of tablets which also influence the mechanical response are discussed next.

## Nanograin architecture drives flaw resistance.

The nanogranular arrangement within the tablets is a crucial design feature of biomineralized composites. In nacre, nanograins 3 to 10 in size are bonded together to form the polygonal tablet [69,72]. The size of the granules can vary within a tablet, thereby controlling the density distribution of organic interphases and consequently inhomogeneity in elastic modulus, leading to improved fracture response [72,103,104]. In the exoskeleton of WO, the elongated diamond-shaped calcite tiles are also comprised of nanograins ≈ 50 in size [76]. The coating of DCMS has a more apparent nanogranular organization than within the tablets of nacre and WO. The nanogranules are present both as secondary grains ≈ 60 in size and as aligned primary grains ≈ 15 in size, which organizes within the secondary grains.

Nanoscale organization of natural materials has been explained through crack tolerance design [105]. Using Griffith criteria and an idealized laminar architecture consisting of staggered mineral tablets in a protein matrix, equations for the fracture response of the mineral tablet were derived, as given by equations 1 and 2. The fracture strength of the tablet at failure () is given by equation 2, and the critical thickness () for its fracture strength to be identical to the strength of the perfect crystal is given by equation 2. The value of is calculated as 30 for values , , and assuming [105].

While the above method is a simplified analysis for an idealized tablet without accounting for geometrical constraints or strength contribution of the organic phase, it explains the nanogranular organization as a means for improving crack tolerance. These relationships also provide a method for comparing crack tolerance responses across natural composites. For example, the nanograins in nacre (3 to 10 ) are below the h\* values, which indicates its tablet design emphasizes toughness over maximizing strength. The larger granules within the tablet of WO (≈ 50 ) compared to a finer organization of nacre (3 to 10 ) and DCMS coating (15 ) predicts that tablets of WO are the least resistant to tablet cracking of these three material systems.

equation [1]

equation [2]

where = thickness of mineral platelet, = critical thickness of mineral platelet, = fracture strength of mineral platelet, = theoretical strength of perfect mineral crystal, = surface energy of mineral (), = elastic modulus of mineral, and for half-cracked platelet.

## Mechanical interface interlocking increases strength and toughness.

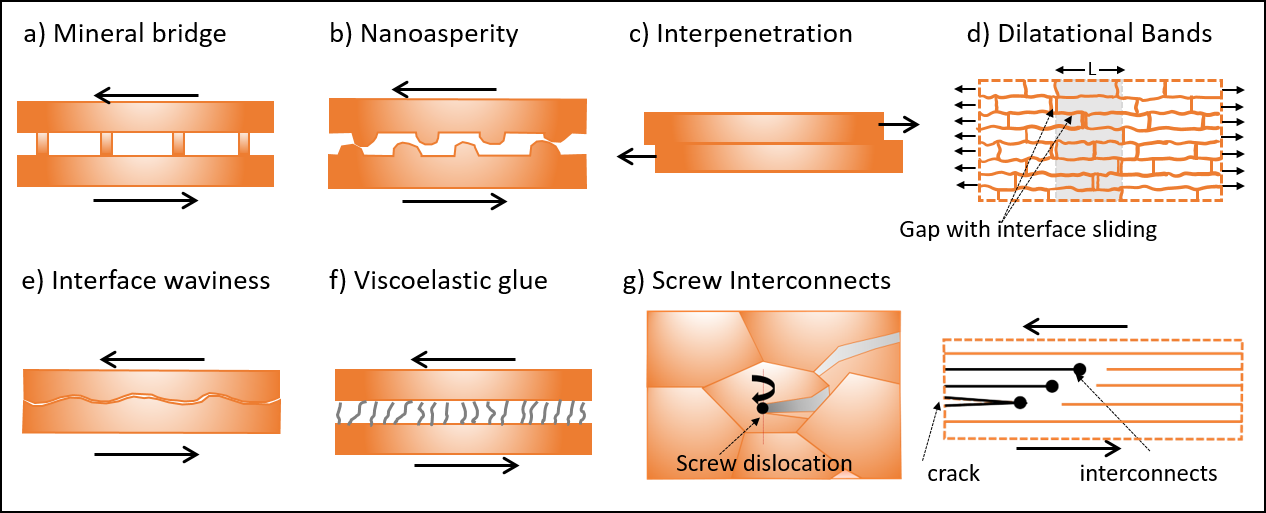
The mechanical interlocking of tablets may occur by several different mechanisms, as depicted in Fig. 4. These mechanisms include mineral bridges, nanoasperity, tablet interpenetration, dilatational bands, tablet waviness, tablet curvature, viscoelastic glue, and screw-like microscopic interconnects. These interlocking mechanisms improve structural integrity and modify the composite mechanical response and are reviewed in multiple sources [61,64,106–108].

Mineral bridges (Fig. 4a) are believed to be sites for mineralization during the growth stage of tablets [90,109] and were among the early mechanisms identified for stiffening in nacre-like materials [58,62,110]. Later experiments showed mineral bridge concentration was present only in the middle regions of tablets, limiting their role to adjacent vertical stacks of tablets rather than the whole nacre level response [65]. Other research instead identified alternative mechanisms such as nanoasperity [24,58] and tablet interpenetration for interlocking [111,112] (Fig. 4b and c). Nanoasperity increases shear resistance by increasing contact sites between adjacent tablets and improves fracture toughness by providing additional surfaces for energy dissipation [24,58] but only under small sliding to about 15 to 20 . The steady-state sliding post hardening can also be explained by the tablet sliding over the nanoasperities [24,58]. The sizeable nonlinearity and strain hardening observed in nacre under tension and increased shear stiffness under sliding can also be attributed to the formation and interconnection of multiple dilatational bands [24] (Fig. 4d). Mineral bridging, nanoasperities, and tablet interpenetration sites can contribute to the increase in the pull-out force of the structure under sliding [61,62,110–112] but cannot explain strain hardening, which was attributed to the presence of tablet waviness. Tablets surfaces have waviness (Fig. 4e) up to 200 in amplitude [66]. This waviness further improves physical interlocking, localizes damage, and increases surface areas for energy dissipation, resulting in strain hardening and increased toughness [66,67,106]. Structural waviness for interlocking has been observed in various other shell structures as well [107,113]. The polymer present in the interface and interphase regions acts as a “viscoelastic glue” by adhering to interconnecting tablets [69,73,92,114] (Fig. 4f). The polymer phase is also responsible for the enhancement of a range of composite properties, which are discussed comprehensively later.

Screw dislocation interconnects that provide biomineralization pathways during growth are another mechanism identified for tablet interlocking in several laminated structures including nacre and WO [75,75,77–79] (Fig. 4g). These interconnect are believed to interlock tablets and offer additional surfaces for energy dissipation, thus localizing damage and improving strength and toughness [75,76,79]. A theoretical analysis of the effect on fracture energy dissipation in the presence of a pair of screw dislocations was performed for the exoskeletal of WO [44] and is given by equation 3. In simple laminates, only mode of energy dissipation will exist, which is the energy dissipation along adjacent mineral interfaces. For WO laminated structure with screw dislocations, additional energy dissipation occurs due to the presence of the inelastic deformation zone ahead of the crack tip (, the formation of vertical surfaces along fracture lines (), and the opening of surfaces along fracture lines () as represented in the equation. The above theoretical analysis predicted the energy dissipation in the exoskeleton of WO having larger tablet surfaces as two orders of magnitude higher compared to simple mineral laminated structures. In contrast, energy dissipation from screw dislocations will be nominal in smaller tablet sizes, such as those of nacre [75].

equation [3]

where = energy dissipation density of mineral layer (), = energy dissipation density of mineral layer (), = surface energy along the cleavage plane of calcite ( ), t is the thickness of mineral layers (m), = spacing between two adjacent fracture lines, = inelastic deformation zone (m), and .

**Fig. 4**: Common mechanical interlocking mechanisms for tablet interfaces which include (a) mineral bridging, (b) nanoasperity, (c) tablet interpenetration, (d) dilatational bands, (e) tablet waviness, (f) viscoelastic glue, and (g) screw interconnects.

## Interpenetrated organic phases provide structural integrity, plasticity, and toughness.

The organic phase is mechanically weak compared to the mineral phase but shows viscoelasticity, deformation strengthening, and large strain to failure [89–91] (Fig. 3). Given its low strength and small volume contribution (< 5 ), many of the earlier studies focused primarily on the mineral phase, limiting the role of polymer interphases only as “viscoelastic glue” for structural integrity and shear strength modifications [69,73,92,114]. Studies have now revealed a broad role of organic phases in natural composites from crystal formation to hygromechanical property modification [23,90,98,100,108,114–118]. For example, the brittle response of nacre in the absence of hydration indicates that most of its shear load is carried by the hydrated organic phase [92]. The presence of stiff and coiled component chitin and folded structure of protein lustrin A, along with the presence of nanopores 5 to 50 in diameter within the organic phase, can explain the adhesion, stiffness, and large deformation of nacre [68,90,100,119].

Fig. 5 shows some of these mechanisms, highlighting the important and wider role of the organic layer. The intra-tablet distribution of the organic layer increases energy dissipation pathways by crack deflection [61,120,121] (Fig. 5a), thus affecting nacre toughness. Nanogranularity and interspersed organic matrix within the tablet together allow for nanograin rotation under load, increasing plasticity deformation and energy dissipation paths for toughening of individual tablets [59,122] (Fig. 5b). Nanogranularity also increases energy dissipation and flow tolerance within tablets by creating a zigzag path for crack motion [123] (Fig. 5c). Other mechanisms contributing to toughening and large strains of the tablets include varying density distribution of the interspersed polymer for elastic modulus gradient [72,103,104] and anisotropic lattice distortion [124,125]. The "saw-tooth" pattern in pull tests reveals the presence of sacrificial bonds within the organic layer (Fig. 5d), which results in significant deformation and stiffening [98,100,118]. These bonds are reversible and hence also provide self-healing properties to some natural composites [118]. Finally, various physiochemical interactions in the presence of water are also said to be responsible for adhesion and hygromechanical sensitivity [126] (Fig. 5e). Increasing hydration results in polymer softening and provides increased plasticity and viscoelasticity effects [127–130]. Similarly, a significant loss in toughness and tribological wear is observed under lower levels of hydration [126,131]. Beyond the apparent impact of polymer plasticization, the presence of physicochemical interactions influences the hygromechanical sensitivity in natural composites.

**A screenshot of a video game

Description automatically generated with medium confidenceFig. 5:** Common mechanisms highlighting the role of organic materials beyond acting as viscoelastic glue, which includes (a) pathway for viscoelastic energy dissipation, (b) nanograin rotation; (c) zigzag path for crack, (d) sacrificial joints, and (e) physiochemical interactions of polysaccharide and proteins structure in the presence of water.

## Deformation Twinning can be prominent for damage localization.

Though less common than dislocation and grain boundaries, twinning is a defect in crystalline structures like metals, allowing local plastic deformation [132,133]. Recently, nanoscale deformation twinning has been identified as a damage localization mechanism in some natural composites [113,134–137]. For example, nanoscale twinning 2 to 20 thick was found within aragonite lamella of conch shells (*Strombus gigas*) and was recognized as an essential mechanism for toughening and localization of damage [113]. Deformation twin ≈ 50 thick was formed around the damage zone of indentation in the exoskeleton of WO at the earliest stage of deformation causing damage localization at the early stages, followed by other mechanisms such as interface opening and micro/nano cracking for toughening [137]. Nanoscale twinning is also present in nacre though it has a much lower influence on mechanical response [137].

## The tablet aspect ratio influences the transition from tougher to stiffer response.

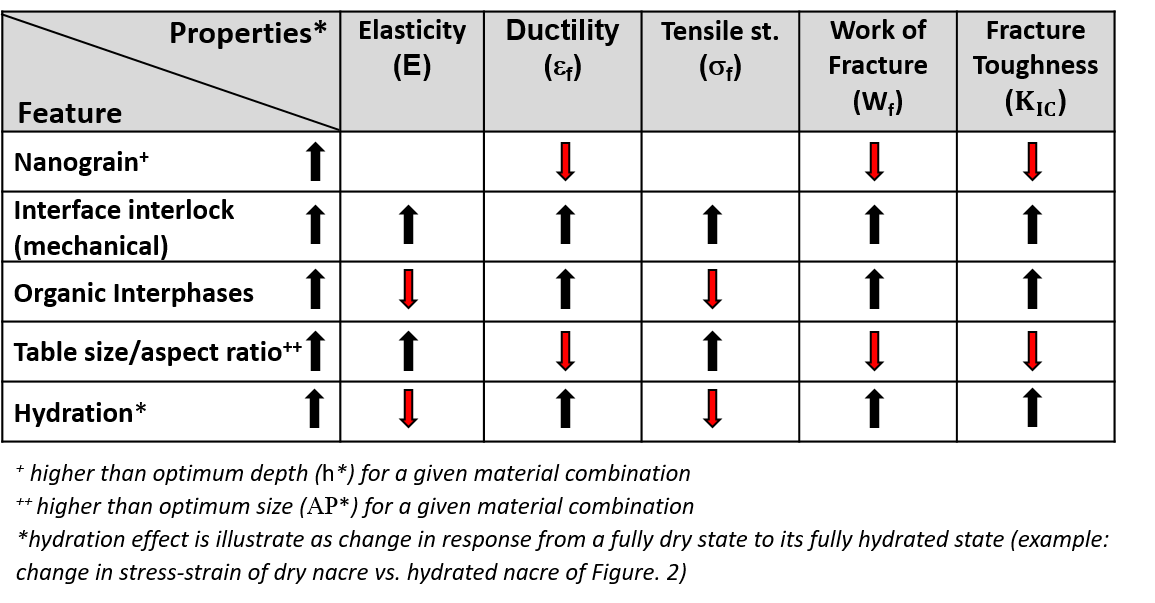
The tablet sizes and geometry vary among the different composites. Nacre tablets are polygonal 5 to 20 in diameter with an aspect ratio () from 20 to 100. Those of the exoskeleton of WO are diamond-shaped with a maximum length of 140 and of approximately 500, much higher than nacre. Finally, in the extreme case of DCMS, the “tablets” reduces to nanoscale size. The aspect ratio is an essential feature of natural composites determining the relative contribution of different deformation mechanisms [67,105,138,139]. For example, larger aspect ratios and staggered alignment can increase strength at the cost of ductility [139,140]. Screw-like interconnects can significantly increase strength for larger tablets like that of WO but have a negligible effect for smaller tablets like that of nacre [75]. In contrast, larger tablets are more susceptible to pull-out damage and fracture developing within their surface. These, in turn, indicate that an optimal tablet size or geometrical features should be guided by the functional demand of the composites and can be predicted by theoretical analysis of idealized composites [105,128,141,142].

Based on the laminar model architecture consisting of staggered mineral platelet in a protein matrix discussed earlier (equations 1 and 2) and assuming simple force distribution between mineral and protein, a critical value of aspect ratio () was derived for optimum strength and toughness [105]. The value is given by equation 4 under the simultaneous failure of the mineral and polymer phase, which shows that the optimum aspect ratio is inversely proportional to . It implied that thinner tablets need higher aspect ratios for a similar stiffening effect. An optimum of approximately 25 was obtained for typical values for mineral and organic constituents (= 1 and = 40 ) [75]. Values higher than the optimum values will have higher stiffness at the cost of its toughness and vice-versa. A similar prediction was made for general 2D composites where ductile matrix deformation dominates at lower than a critical value of while platelet fracture mode results at larger values. The exact value of will depend on the tensile strength of reinforcement [128].

equation [4]

where = failure stress of the mineral phase, = shear failure stress in the organic phase, and and are as defined in equation 1.

Based on the above theoretical analysis, the exoskeleton of WO with higher should address higher stiffness applications. At the same time, a smaller of nacre or bone should be designed for greater toughness. This prediction matches the understanding of their comparative performance and application. As a further confirmation, the toughness of nacre is three orders larger than its mineral constituent, and that of the exoskeleton of WO is only two orders larger. Another tablet size-related design feature is the thickness of the organic interphase between tablets. However, the thickness of the organic phase cannot be treated independently, but it will depend on the volume contribution of the organic phase and the size of the tablets. A smaller thickness (≈ 2 ) is present in the exoskeleton of WO which has a relatively small volume contribution of the organic phase (≈ 1%) and larger tablets. In comparison, a larger thickness (≈ 20 ) is present in nacre which has a more considerable volume contribution of the organic phase (≈ 5%) and smaller tablets. The discussion on the impact of various design features of mineralized 2D composites on the mechanical response is summarized in Table 1 below.



**Table 1**: Summary of design rules of natural composites and their impact on common mechanical properties. The arrows can be read as follows for a representative feature: as dimensions of nanograin increase, it leads to a lowering in ductility, work of fracture, and fracture toughness.

# Bioinspired design features and processing of MXene-polymer composites

The review above and Table 1 summarize the critical design features and structural organization of mineralized 2D materials to address mechanical demands for a tough armor-like response. Many of the same design features are also optimized for other functional demands of these structures, such as surface smoothness, iridescent, and optical transparency for parasitic defense for most shells [76,143–145], and high strain impact resistance for hammer-like function for DCMS [26,80]. In general, the bioinspired design space is exhaustive [22,146–150], with many nature-mimetic designs engineered for a range of functional demands from damage tolerance [8,150–156], tunable transparency [128,143,157], improved aerodynamics [148,158–161], thermal management [128,158,162–164], and as a gas barrier [130].The section below discusses the bioinspired design features in the context of MPC, illustrates examples of recent MPCs processed, and finally summarizes and identifies some key challenges ahead.

## Bioinspired design features

Fig. 6 shows an example of layered MPC film and associated design parameters such as flake thickness and length, choice of a polymer matrix, matrix thickness, and interactions and surface design. These design parameters can be guided by the bioinspired design rules summarized in Table 1. For example, controlling flake size and aspect ratio is needed for balancing strength with toughness to mimic tablet influence. Tablets in natural composites are highly organized and relatively uniform. Tablets of larger > 100 provide stiffness, but such tablets negatively impact the toughness. MXene flake sizes are highly dependent on process parameters and can vary significantly for the same technique. For example, MXene/TAEA, sheets processed by layer-by-layer self-assembly resulted in 80 to 500 [165]. In another study, acid etching resulted in flakes with a much higher of 6000 [42]. Even though almost 30 different kinds of MXenes have been synthesized so far and the field is rapidly growing [16,166–168], controlling the processing parameters for targeted flake geometry remains immature. The understanding of the impact of tablet size and aspect ratios is thus helpful in directing the research towards targeted and uniform-sized flake processing.

Another bioinspired feature of significant influence is interface interlocks. The various tablet interlock summarized in Fig. 4 increases structural properties from stiffness to toughness. Interface design such as via interlocking using surface waviness can occur within MXenes flakes during deposition or can be induced via external mechanisms such as the addition of microcapsules between layers [169]. Many of the earlier mechanical strategies for creating tablet connections in other materials have shown severe limitations. Methods like tablet stitching and creating nanotube forests improve out-of-plane stiffness and pull-out forces but lead to defects and damage within the original laminates due to the formation of stress concentration sites [170–173]. Other interface mechanisms, such as stitching via the formation of screw-like dislocation centers inspired by the exoskeleton of WO, roughened interfaces, surface waviness, and other alternatives for interface design can impact response but have remained unexplored for MXene structures.

Another bioinspired design feature is tablet interlock via polymer interphases similar to the role of organic polymers in natural composites. The organic polymeric material is crucial for structural integrity, plasticity, and shear rigidity of the mineralized 2D natural composites through multiple mechanisms, as highlighted in Fig. 5. Multiple synthetic and natural polymers are available to mirror the mechanical properties of the organic phase. Some synthetic polymers used in the nacre-mimetic design include acrylic foam tapes [149], PVA [174,175], poly-diallyl dimethylammonium chloride (PDDA) [176], poly-methyl methacrylate (PMMA) [175,177], polyacrylic acid (PAA) [178], and polyelectrolyte multilayer (PEM) [179]. Natural polymers used include chitosan [180], silk fibroin [155,181–184], alginate, and CNF [174,185]. While many of these polymers can capture the mechanical stiffness and viscoelasticity of the organic phase, a comprehensive public database of the electromechanical and thermal properties of these readily available polymers could help identify the most suitable choice for a particular set of MXene applications.

A screenshot of a video game

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**Fig. 6:** List and schematic of available parameters for the design of layered MXene-polymer composite.

## MXene composite film processing

While controlling the parameters identified above is essential for the bioinspired design of MPCs and remains an open challenge, multiple MPCs are rapidly emerging, targeted toward specific applications. Examples for MXene and MPCs films for EMI shield were discussed earlier in section 2. Below presents additional examples of MPCs for applications beyond EMI to illustrate the current state of the art.

The first flexible high-capacity conductive films were manufactured with /PVA and /PDDA via mixing of the polymer in a colloidal solution of MXene, having potential applications as supercapacitor electrode for energy-storage applications [31]. Another example /graphene oxide was mixed in the presence of cationic PDDA solution for electrostatic self-assembly to achieve layered ordered structure, with applications as a flexible supercapacitor [186]. Layer-by-layer self-organization in the presence of a small molecule, tris(2-aminoethyl) amine (TAEA), resulted in highly conductive multilayered flakes with an alternate arrangement of MXene/TAEA with flake thickness of 2.5 and the in-plane dimension of 200 to 1200 with improved electrochemical performance [165].

In other work, flexible supercapacitive properties with high strength and toughness were obtained in MXene ( functionalized graphene oxide sheets crosslinked with biomolecules 1-aminopyrene-disuccinimidyl suberate or AD [187]. Nacre-inspired brick-and-mortar MXene structure was fabricated using MXene-SilverNanowire-PDS () gel-ink through screen printing, with applicability for strain sensing [188]. A bamboo-inspired mechanically robust structure was assembled using /CNF/polydopamine (PDA), showing hygroscopic actuation capacity for soft robotics applications [189]. Simulation-guided manufacturing of -epoxy composites was undertaken for improving epoxy stiffness and toughening at a low concentration (1 ) of MXene [190]. An in-situ synthesis using ultrasonic was used to simultaneously create hybrid nanosheets made from and metal-organic (NiCo-PTA) sheets with an improved electrochemical response for energy storage and supercapacitor applications [191]. A MXene-based flexible wearable strain-sensor with long-term sensing stability was demonstrated, where the preparation method involved dip-coating polyurethane (TPU) fabric with MXene/cellulose nanocrystal (CNC) [192]. A multilayered MXene based film was produced using MXene sheets () and Ag microparticles by layer-by-layer spraying followed by vacuum pressing for energy harvesting applications [193]. Nitrogen derived from chitosan was used for preparing N-doped MXene via high-temperature calcination for its potential use as a supercapacitor [194]. A MXene//PANI (polyaniline) composite structure was prepared, first by growing on the MXene surface and then by covering the layered structure with PANI in a redox reaction [195]. The prepared structure showed good electrochemical performance over multiple charge-discharge cycles of testing.

## Challenges and opportunities for bioinspired MXene films

The above highlights several examples and processing for MXene and ultrathin MPCs. While several MXene and MPCs have emerged where layered geometry and constituent phase are replicated, a more significant hindrance has been in replicating interface interactions beyond viscoelastic glue similar to the various polymer-based mechanisms illustrated in Fig. 5. In the context of modifying interface interactions via surface functionalization, chemical and physical bonding, physical interactions, and photochemical or thermal-induced interactions are some of the available strategies. For example, increased stretchability in graphene-based composite was obtained by ionic bonding through the addition of ions [183,184]. Hydrogen bonding between the slightly hydrophobic amine-terminated silane () with oxygen atoms of chitosan increased the polymer's cohesion to alumina tablet for properties comparable to nacre [180]. Hydrogen bond interactions have been used in many systems, including interface engineering of MXene-based composite [196]. Electrically charged polymers such as PEM have also been used to duplicate bond formation and breakage [179]. The higher strength and flexibility in the bamboo-inspired MXene-composite were achieved through the use of biomacromolecule PDA as the adhesive for the integration of 2D MXene with one-dimensional cellulose nanofibers [189]. The use of PDA provides strong hydrogen bonding among constituents, leading to higher strength, extreme flexibility, and hydrophilicity for actuation. A similar moisture-based actuation with highly electrical conductivity was also achieved in a homogenous MXene sheet utilizing the hydrophilic nature of its surface termination layers [197].

Overall, tunable surface terminations, the presence of reactive terminates such as the hydroxyl functional group (-OH), and the high aspect ratio of MXene flakes all provide an exciting opportunity for targeted functionalization of MXene surfaces. While theoretical calculations have provided good evidence of the effect of surface functionalization on MXene optical and thermoelectrical properties [198–200], limited experimental work has shown control of surface functionalization. Using an aqueous colloidal solution, the MXene surfaces were functionalized by catechol [46]. Hydrogen bonding, metal-oxygen bonding, or a mixture of these were identified as the possible mechanism for catechol binding to MXene surfaces. Such an approach has the potential for larger-scale assemblies by using these functionalized surfaces as sites for polymer grafting [46]. In another study, an autonomous self-healing MPC-based sensor was processed through esterification and hydrogen bonding of serin modified MXenes with serin-grafted epoxidized natural rubber [201]. These examples illustrate mechanisms by which controlled MPCs can be generated using reactive surface bonding groups and appropriate reactive polymer groups, though only thin flakes in the nanometer range have been produced so far. Significant growth and research are needed to explore such aspects via integrated theoretical and experimental studies to develop MXene-based films of desired thickness and functionality.

Finally, another option that could be explored for MXene-polymer design is interface toughening through nanogranularity or through a functional stiffness gradient within the MXene sheets. A functional gradient of mechanical properties is observed in many other natural systems such as in the elastic modulus gradient in the dentin-enamel junction of teeth [202–204]. A functional elastic and plastic property gradient has also been replicated in engineered systems for crack suppression and improved fatigue [205–209] and is another potential direction to be explored for MXene composites.

To summarize the above discussion, the brick-mortar organization of nacre and WO discussed earlier provides one of the most straightforward systems for replication compared to other more complex hierarchical organizations, such as in bone and plants [210]. However, translating the bioinspired design features to engineered systems offers many unique challenges in general and some specific to MXenes. Some of these challenges and the current state-of-the-art in MXene and MPCs were discussed in this section in the context of structural design and processing. The above discussion leads to the following critical challenges for the future of bioinspired MXene-polymer composite.

1. Though more than 30 types of MXenes have been produced so far, the synthesis control over lateral size, thickness, and surface termination of MXene flakes and overcoming oxidation are yet to be resolved. A comprehensive public database of the electromechanical and thermal properties of readily available synthetic and natural polymers could help identify the most suitable polymer choice for a particular MXene application to take the field forward.
2. Many bioinspired mechanical interface mechanisms identified either cause failure by stress concentration or remain unexplored for MXene systems such as screw connection and wavy interfaces. Developing an understanding of the relative impact of the mechanical interface mechanisms on key performance metrics will be critical to selecting appropriate configurations of MXene-based composites.
3. A more significant challenge comes from replicating various MXene-polymer interactions such as tailored surface chemistry (covalent bonding, controlled grafting density, and specifically designed secondary interactions to tailor the property) and nanogranularity, which are essential for both inter and intra-tablet toughening. Identification of application-specific suitable interactions together with the evolution of processes for controlling the repeatable synthesis of MXene-based materials will permit the fabrication of optimal interactions and configurations.
4. Related to the challenges above, while targeted surface functionalization and identification of appropriate reactive polymers is the way to move forward for developing MPCs of desired thickness and functionality, such work requires close integration between theoretical modeling and processing expertise to make significant progress ahead to study and identify compatible combinations of polymers and processing reagents.

# Summary

The recent entry of MXenes in the 2D engineering space provides many opportunities and challenges to move the field of multifunctional composites forward. The paper focused on the highly mineralized structure of 2D natural composites as a pathway forward for the bioinspired design of MXene-based composites.

After briefly introducing the topic, sections 2 and 3 present the structure of MXenes and MXene-based composites for EMI application and highly mineralized layered natural composites (nacre, exoskeleton of WO, and outer coating of DCMS), respectively, to draw physical similarities in the structural organization between the two systems. Section 3 also presents the unique properties of the natural system different from its dominant brittle mineral phase, emphasizing the role of their structural organization and interspersed soft organic phase. Section 4 then reviews the literature of 2D natural systems to provide a mechanistic basis for the superior properties of the layered natural system. In the process, it identifies several underlying design rules that determine the balance between toughness and stiffness (Table 1). Some key design traits identified include (a) nanogranular organization, (b) mechanical interlocking of tablet interfaces, (c) organic interphase zone, (d) tablet aspect ratio, and (e) hydration. For example, a higher aspect ratio of the layered tablet increases the stiffness and negatively impacts the ductility and fracture toughness.

In section 5, the focus shifts back to MXene-based composites to identify the relevance of the various bioinspired lessons to guide the design of the engineered system. In the process, it clearly identifies critical challenges and opportunities in the current state-of-the-art in MXene and MPCs design and processing. These include (a) control over lateral size, thickness, and surface termination of MXene flakes, (b) improved understanding of the relative impact of mechanical interlocking mechanisms such as screw connection vs. wavy interfaces, (c) need for a public database of the electromechanical and thermal properties of readily available synthetic and natural polymers, and challenges in processing controlled MXene-polymer mechanical interfaces either (d) mechanically or (e) through surface functionalization.

Finally, Fig. 7 below summarizes an Ashby-style property space of strength and toughness available in the literature for a large range of materials discussed in this paper. These include 2D natural system (nacre) and its constituents (organic polymer and mineral), 2D engineered systems (MXenes, graphene, and nanoclay), and polymer-based composites including MPCs. The figure further highlights the large gap in mechanical properties of the engineered 2D systems from their bioinspired counterparts, which further stresses the need for research and growth in that area.

A picture containing bubble chart

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**Fig. 7**: Ashby style property space of 2D natural and engineered systems. Polymer-based composites are primarily polymers with < 5% of stiffening phase (graphene, MXene, nanoclay). MXene-based composites have > 50% MXene. The ellipses are plotted using mean and standard deviations of maximum and minimum values of the properties reported [22,24,42,42,50,63,69,75,76,80,84–92,94–97,106,211–223].

# Conclusion and future perspective

While the paper highlights the significant progress made, it also identifies several challenges and opportunities ahead for research and development. Identifying and overcoming these challenges can enable achieving the theoretical limits of mechanical response in the engineered 2D systems and provide multifunctional capabilities far above those of more traditional materials. Below are some perspectives on the future ahead for MXenes and MPCs including opportunities for stitching growth in physical synthesis with theoretical analysis, multiscale modeling, and physics-informed machine learning (ML) towards a new and comprehensive bioinspired multifunctional MXene-based composite.

With the rapid advancement in MXene research, a large family of MXenes has been synthesized for many different applications [16,166–168]. However, given the wide choice of elements for base and surface termination layers and multiple synthesis pathways, there is a tremendous opportunity for systematic integration of atomic-level simulation with experiments to guide materials and process choices and provide an integrated development, testing, and validation platform. Such simulations are currently very limited but show their predictive capability. For example, recently, molecular dynamics (MD) simulations on titanium-carbide (TiC) MXene predicted its tensile elastic modulus between 500 to 600 , the nonlinear transition at < 1% strains, high strain to failure (6 to 9 %), and temperature and strain-rate sensitivity [42,213,224,225]. The binding energy was obtained between 0.8 to 2 [226]. Indentation experiments on revealed a lower elastic modulus of 300 [42,213] than those predicted from these simulations. Such atomistic simulation studies need to be extended to create a comprehensive database relating MXene compositions and surface terminations with its electromechanical and electrochemical properties and explain the discrepancies between atomistic predictions from in-situ values. This will enable improved fidelity of modeling and create a validated platform to guide developments and reduce the effect of experimental unknowns (surface roughness, interface/interphase chemistry, lateral sizes, and thickness distributions, etc.) [227,228].

Atomic simulations can provide structural and interface properties of pure MXene sheets, which are otherwise not possible through experiments, and can also act as a bridge between density functional theory (DFT) predictions and experimental data. Growth in MD simulations will need progress in realistic representation of geometry to capture atomic variability in surface terminations and development of relevant and accurate potentials for bulk and interface atomic interactions. Furthermore, integrating atomic models with higher-order analysis is essential for modeling larger realistic geometries and processing to support the rapid growth of MXene-based applications [229,230]. Specifically, MD analysis can be combined with multiscale continuum mechanics-based finite element analysis (FEA) to address the size limitations of atomistic simulations. Such higher-order continuum analysis not only requires the stress-strain response of MXene sheets to create constitutive models of its progressive damage, but they also need a measure of interface strength (surface interaction energies, frictional/shear response) to model slippage and adhesion of MXene flakes and MXene-polymer surfaces. Integrated MD/FEA simulation thus serves the dual purpose of being able to use realistic interactions and material properties. In the absence of such material-specific information, FEA application for MXene-based composites design will be severely limited to elastic response and rigid contact interactions such as those used in recent works [231–233].

A simpler theoretical analysis using idealized geometry has driven an understanding of design features for natural composites [67,105,138,139]. For example, the presence of screw dislocation increases surfaces for energy dissipation [75], and increasing tablet aspect ratios can predict the transition of the composite from low tolerance to stiffer response [67,105,138,139]. However, these models do not incorporate the role of interfaces. Modifications to the existing model of natural composites or new theory and model development to include polymer/mineral interface or interactions can provide a more accurate prediction of their role in the mechanical response and correspondingly guide future developments toward the bioinspired design of engineered systems including MXene and MPCs

Beyond simulation and theoretical modeling, the development of new surface chemistries for precise control of molecular energy dissipation and tunable and sacrificial bond interactions are also needed to control interface interactions. For example, incorporating small molecules during the MXene exfoliation process, MXene layering, or introducing different surface chemistries can impact its interfacial behavior, such as adhesion and separation energies. Another aspect of surface chemistry control comes from environmental stability, especially in applications where surface oxidation may adversely affect its multifunctional performance. Humidity can lead to oxidation of MXene surfaces as well as degrade the mechanical properties of natural polymers [127–130]. Finding pathways for stability against humidity damage becomes critical when using natural polymers such as chitin and CNF. Stabilization methods may involve new interactions development via chemical crosslinking, switching mechanism, or thermal curing [128,234–238].

Furthermore, while MXenes-polymers are new material systems with limited data available for their design and simulation, progress/knowledge gained in this space can also come from other relevant material systems such as fiber/matrix composites, other natural composites such as bone, and previously studied 2D materials such as graphene. Traditional fiber/matrix composites used for aerospace applications are laminated structures formed from anisotropic fibers for strength and stiffness surrounded by a nearly isotropic matrix material to bind the fibers and provide a path for load transfer. Geometric flaws, such as fiber wrinkling or matrix porosity result in a large variation in their mechanical response [239–241]. While these structural features and flaws are orders of magnitude larger than in MXenes, the extensive body of research into laminated fiber/matrix composite laminates offers a suitable starting point for the simulation of MXenes and MPCs. On a related note, the future of air transportation requires a search for new materials for lighter and more durable designs [242]. Hence, MPC also has future potential in the design of aerospace structures. The traditional composites can also provide some insight into the chemical compatibility of MXene-polymer interfaces. The development of compatible materials and fiber functionalization and the role of environmental factors have been critical for effective load transfer in laminated composites [243,244] and fiber-reinforced structures [245,246]. This knowledge will also be essential and transferrable for MXene systems. In the same regard, lessons learned from clay-based polymer nanocomposite and their organic modifications [27,29,156,174,218] find important relevance to MPC design. Bone is another natural composite that has been extensively investigated [247,248]. Studies focusing on molecular mechanism and mineral-collagen interface via MD and FEA simulations are particularly relevant here [248–250]. Similarly, numerical approaches such as MD, atomistic FEA, and FEA analysis applied to various graphene structures [251–257] can also guide MXene system modeling.

Finally, ML is rapidly becoming an impressive tool in new materials discovery and materials design, including composites [258–263]. Integration of ML with MD or FEA simulation is also emerging [264–267]. Learning from recent composites literature, ML can also facilitate the accelerated development of MXene-polymer composites for targeted structural applications. For example, an opportunity in the MXene-polymer composite space is shown in Fig. 8, where several regression-based ML methods, including artificial neural networks (ANNs) as well as active-learning methods such as Gaussian process regression (GPR) (especially if the training dataset is small), could be employed to better understand the correlation between input composite features and elastic/failure structural properties (strength, toughness, moduli). Here input features could be described via a set of descriptors using intrinsic properties of MXenes, polymer and their interfaces (such as mechanical response of pristine systems, MXene/polymer interface strength) as well as morphological properties (MXene concentration, some measure of dispersion quality, orientational anisotropy, etc.). The trained models may assist in exploring and exploiting multi-dimensional parameter space in a much more efficient manner and help identify optimal input features for a given structural requirement much faster. Overall, the integration of multiscale modeling (MD, FEA) and experimental data of synthesized MXenes provides an exciting opportunity for growth into physics-informed ML models.

To conclude, the recent introduction of the MXene family with exceptional property combinations has led to its rapid expansion into various applications from electronics to biomedical sensors, exhibiting its immense potential as a versatile material of future society. Despite tremendous interest in these materials systems, a succinct summary of the underlying mechanical performance of these composites was missing. Here we have taken a pre-design approach guided by natural design principles of similarly structured highly mineralized 2D natural materials to address the challenges of MXene-based hierarchical composite for its varied need of strength, toughness, and surface interactions across applications with a focus on layered organization for EMI shielding. It is realized that for the full potential of MXene to be realized at an industrial scale for its many applications, it requires careful consideration of the opportunities and challenges ahead, as identified in the paper. The future also points to an integrated role of synthesis with multiscale modeling and physics-derived machine learning approaches as a step forward to enable its rapid growth for real-world applications.

Diagram

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**Fig. 8**: Integrated platform for accelerated development of multifunctional MXene-based composite.

# Abbreviations

2D : Two-dimensional

ANN : Artificial neural networks

CNF : cellulose nanofibers

DCMS : Dactyl club of mantis shrimp

DFT : Density functional theory

EMI : Electromagnetic interference

FEA : Finite element analysis

GPR : Gaussian process regression

HAP : Hydroxyapatite

MD : Molecular dynamics

MMT : Montmorillonite

MPC : MXene polymer matrix composite

PAA : Polyacrylic acid

PDDA : Poly-diallyl dimethylammonium chloride

PEDOT:PSS : poly(3,4-ethylenedioxythiophene)–poly(styrenesulfonate)

PEM : Polyelectrolyte multilayer

PMMA : Poly-methyl methacrylate

PVA : Polyvinyl alcohol

SE : Shielding efficiency

TAEA : tris(2-aminoethyl) amine

TMD : Transition metal dichalcogenides

WO : Windowpane oyster

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