Fracture at the two-dimensional limit

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More than a century ago, A.A. Griffith published the seminal paper establishing the foundational framework for fracture mechanics. The elegant theory creatively introduced the concepts of elastic energy and surface energy to the science of fracture, and solved the problem of brittle fracture of glass materials. Many subsequent milestone studies in fracture mechanics were motivated by the real problems encountered in different materials. The emergence of two-dimensional (2D) materials provides an exciting opportunity to examine fracture processes at the 2D limit. An important question to be addressed is whether the classic Griffith theory is still applicable to 2D materials. Therefore, recent progress in both experimental and theoretical studies of fracture of 2D materials will be briefly reviewed, with new developments and discoveries in relevant techniques and theories highlighted. Given the early stage of exploring fracture behaviors in 2D materials, more emphasis will be placed on challenges and opportunities for this budding field.

Introduction

Few theories have had the kind of impact in both technology and science as Griffith's,[1] stating the universal fact that "a crack will propagate when the reduction in potential energy that occurs due to crack growth is greater than or equal to the increase in surface energy due to the creation of new free surfaces."[2] Mathematically, it is often expressed (with G being the energy released per unit crack advance, and γ the surface energy) as:

$$G = 2\gamma. \quad (1)$$

The basic concept of balancing the energy released due to crack advancement (e.g., creating new surfaces) with the energy necessary to facilitate such a process, provides a general framework for modeling all sorts of fracture phenomena, including complex dissipation mechanisms (e.g., dislocations, crack shielding, etc.).[3,4]

Indeed, the prevention of fracture has become a key engineering design objective, and it is prevalent across domains and industries, from buildings to computer chips to biomedical devices. The scope of what engineers build has shifted over the years, and moved to more complex, smaller, and extreme designs at the level of molecular machines.[5] The materials research community has expanded on the early successes of fracture mechanics focused on the macroscale, and moved increasingly to understand fracture at the nanoscale, and across scales and modalities. Strikingly, the energy-based concept introduced by Griffith holds across these scenarios, underscoring its universal appeal.

Griffith's fracture theory has seen numerous applications over the years and invoked many studies as the materials field embraced nanomaterials starting a few decades ago.[6] The powerful concept of Griffith's approach has resulted in insights especially at the bio-nanomechanics interface, revealing important concepts such as flaw tolerance[7] and superior adhesion.[8] While phenomena at these scales had been simulated with atomistic modeling before, the door for engineering applications opened when Griffith's concepts enabled translation into the mechanics field. Questions explored include if the Griffith model holds at the nanoscale, what it can teach us about biomaterials design, and what type of scaling behavior can be deduced from an engineering science perspective of biophysical phenomena.

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Meanwhile, two-dimensional (2D) materials, such as graphene, hexagonal boron nitride (h-BN), and molybdenum disulfide (MoS_2), have exhibited exceptional electrical, thermal, and mechanical properties[9,10] in the past two decades since their discoveries. They hold great promise for a number of functional and structural applications, as next-generation nano-electromechanical systems (NEMS),[11] pressure sensors and barriers,[12] nanocomposites,[13] and more. Understanding mechanical properties, in particular fracture behaviors, of these novel 2D materials is essential for their reliable integration into future electronic, composite, and energy-storage applications.[14,15] Particularly, the property of fracture toughness describes the ability of a material containing a crack to resist fracture and therefore, is one of the most important material mechanical properties for many engineering applications.[1,16] However, 2D materials are atomically thin membranes with nanometer scale thickness, which creates significant technical challenges in quantitatively measuring their mechanical properties.

The pioneering mechanical testing of graphene was conducted by Lee et al.[14] through nanoindentation of freely suspended graphene films using an atomic force microscope (AFM). They reported Young's modulus and "intrinsic strength" of mechanically exfoliated pristine graphene as 1 TPa and 130 GPa, respectively, placing graphene about five times stiffer and more than 200 times stronger than stainless steel.[17] Although mechanical exfoliation remains one of the most reliable fabrication techniques to obtain high-quality small-area 2D materials for lab-scale experiments, large-scale production methods including liquid exfoliation[18] and chemical vapor deposition (CVD)[19] have also been developed. One important feature of large-area 2D materials is that they often contain defects such as vacancies or grain boundaries, especially for those prepared by CVD. It is well known that properties of polycrystalline materials are most likely dominated by the size of their grains and the nature of grain boundaries. These effects are expected to be more pronounced in 2D materials, because even a line defect like a dislocation could disrupt a 2D crystal due to its reduced dimensionality.[20] Therefore, the useful strength of large-area 2D materials with engineering relevance is better represented by its fracture toughness,[21] rather than the "intrinsic strength" that dictates the uniform rupture of atomic bonds.[14] These technological advances in materials science provide the opportunity for Griffith theory to meet its 2D limit a century after its birth.

In this article, we focus on the applications of Griffith theories to 2D materials, where a host of new fracture phenomena have been discovered. In the following sections, a brief overview will be provided for both advanced experimental studies and theoretical/modeling efforts on several representative 2D materials, including graphene, h-BN, MoS_2, etc. This will be followed by a brief discussion on some novel aspects of fracture behaviors in the 2D limit. We end by outlining some current challenges and future opportunities for the study of 2D materials fracture.

Experimental studies of fracture of 2D crystals

Thanks to rapid advancements in fabrication, manipulation, and testing capabilities, recent decades have witnessed a continuous surge in experimental studies of 2D materials.[22–24] In this section, we review some representative studies focusing on fracture behaviors in different 2D materials, starting with graphene and going beyond to other emerging 2D materials.

Graphene and its derivatives

As the first 2D material isolated, graphene is by far the most studied 2D material. Therefore, we begin by discussing the fracture of graphene, and by extension graphene oxide. One of the first experimental studies of mechanical properties of graphene found it to be the strongest material ever tested using an AFM-based nanoindentation method (**Figure 1**a).[14] However, the fracture process, especially atomic details of crack morphology and crack–microstructure interactions, was not revealed in this study. Subsequent transmission electron microscopy (TEM) observations found that cracks propagate along armchair or zigzag directions of graphene and that cracks could cross over grain boundaries (Figure 1b) instead of aligning with them.[25] Meanwhile, bulge tests allowed high-speed camera observations of crack propagation in monolayer CVD graphene, showing that cracks can be arrested by folds in 2D materials and that cracks can bifurcate likely due to environmental stress corrosion.[26] These valuable qualitative studies seem to suggest the brittle nature of fracture in graphene, which calls for more quantitative assessment.

When deformation and fracture of 2D materials is concerned, AFM is the most commonly adopted method for its relative simplicity and efficiency in collecting large amounts of data without causing superfluous damage via E-beam irradiation. However, AFM nanoindentation introduces a complex stress state with large gradients and only reflects local properties, which makes it non-ideal for applying Griffith theory and identifying key fracture properties. To overcome this limitation, uniaxial tension via microelectromechanical systems (MEMS) devices in the SEM have been developed for 2D materials and become the gold standard thanks to quantitative strength measurements, images of the samples during the test, and the ability to directly apply Griffith theory.

Guided by the Griffith theory,[1] mono- and bilayer polycrystalline graphene fracture toughness were carefully measured under uniaxial tension with a precrack created by a focused ion beam (FIB) using an *in situ* SEM nanoindenter-driven microfabricated device.[27] A critical stress intensity factor (SIF) of 4.0 ± 0.6 MPa $\sqrt{m}$ and the equivalent critical strain energy release rate of 15.9 J m^{-2} were found for the brittle graphene.

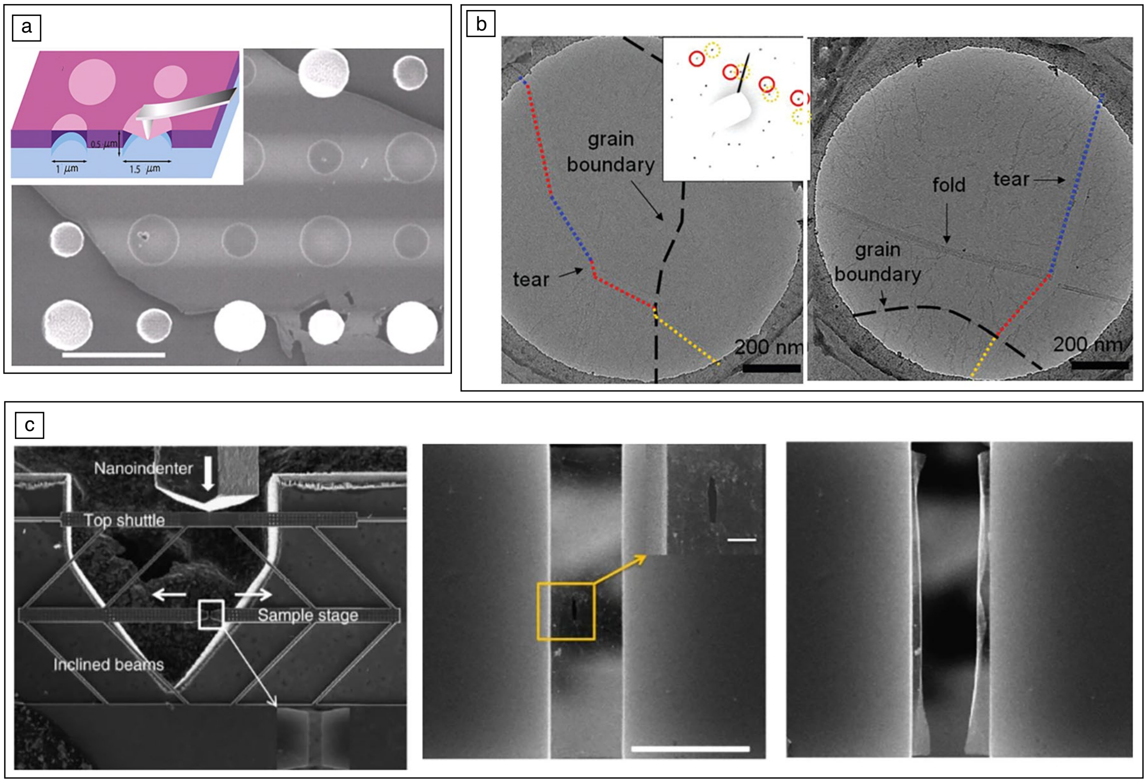

Figure 1. Experimental studies of the fracture of graphene. (a) A scanning electron microscope (SEM) image of graphene over silicon holes in preparation of atomic force microscopy (AFM) indentation with inset of a diagram of the AFM indentation of 2D samples.[14] (b) Transmission electron microscope (TEM) images of the tearing direction of graphene; inset of the diffraction pattern around the grain boundary showing two sets of hexagonal patterns from two adjacent tilt grains, where the hexagonal pattern marked with red and dashed yellow circles corresponds to the grain in the left and right sides, respectively. The blue dotted lines represent tear lines in the zigzag direction. The red and yellow dotted lines represent tear lines in the armchair direction.[25] (c) An SEM image of a push-to-pull microelectromechanical systems device that studied the fracture toughness of graphene; the graphene and a precrack in the sample and the fracture surface after failure.[27] Scale bars: (a) 3 μm, (c) 5 μm, and the inset 500 nm.

This signified the first experimental evidence that the Griffith theory of brittle fracture could apply to 2D materials, and provided proof that defects dictate the strength of the strongest known material (Figure 1c). Subsequent work explored the fracture toughness of monocrystalline pristine trilayer graphene,[28] and reported the effects of interlayer slippage, which will be discussed in a later section.

Given the brittle nature of the fracture process in graphene, it becomes important to explore effective ways to toughen it for engineering applications. One way to increase the toughness of graphene is by integrating nanotubes as reinforcement. Specifically, carbon nanotubes were integrated into graphene and the resultant so-called "rebar graphene" has demonstrated enhanced toughness compared with graphene due to active crack diverting and bridging characteristics (**Figure 2**a).[29] Another method that has shown the ability to arrest crack advances and prevent catastrophic failure is increasing the defect density.[30] Purposefully increasing the defect density led to a weaker overall strength but was able to confine the crack propagation in graphene, as shown in Figure 2b. Although not necessarily toughening graphene itself, monolayer amorphous carbon (MAC), a 2D carbon allotrope, exhibits both plastic deformation and damage tolerance as shown in Figure 2c.[31]

Based on the measurements of graphene, it is apparent that 2D materials, such as graphene, are not immune to the famous "strength-toughness" tradeoff that is commonly observed in bulk materials. In contrast to graphene, its closest derivative graphene oxide (GO) is unique for its relatively high strength and high fracture toughness, allowing it to resist failure better than graphene.[32,33] A study of the fracture strength of monolayer GO found that samples with a higher ratio of carbon to oxygen (i.e., compositionally closer to graphene) exhibit a higher strength.[33] Even with lower strength, multilayer GO was observed to have a nonlinear fracture toughness over two times greater than graphene and, unlike graphene, an ability to arrest crack growth as shown in Figure 2d.[32] This crack arresting ability is attributed to the asynchronous cracking among layers and the strain fields created by

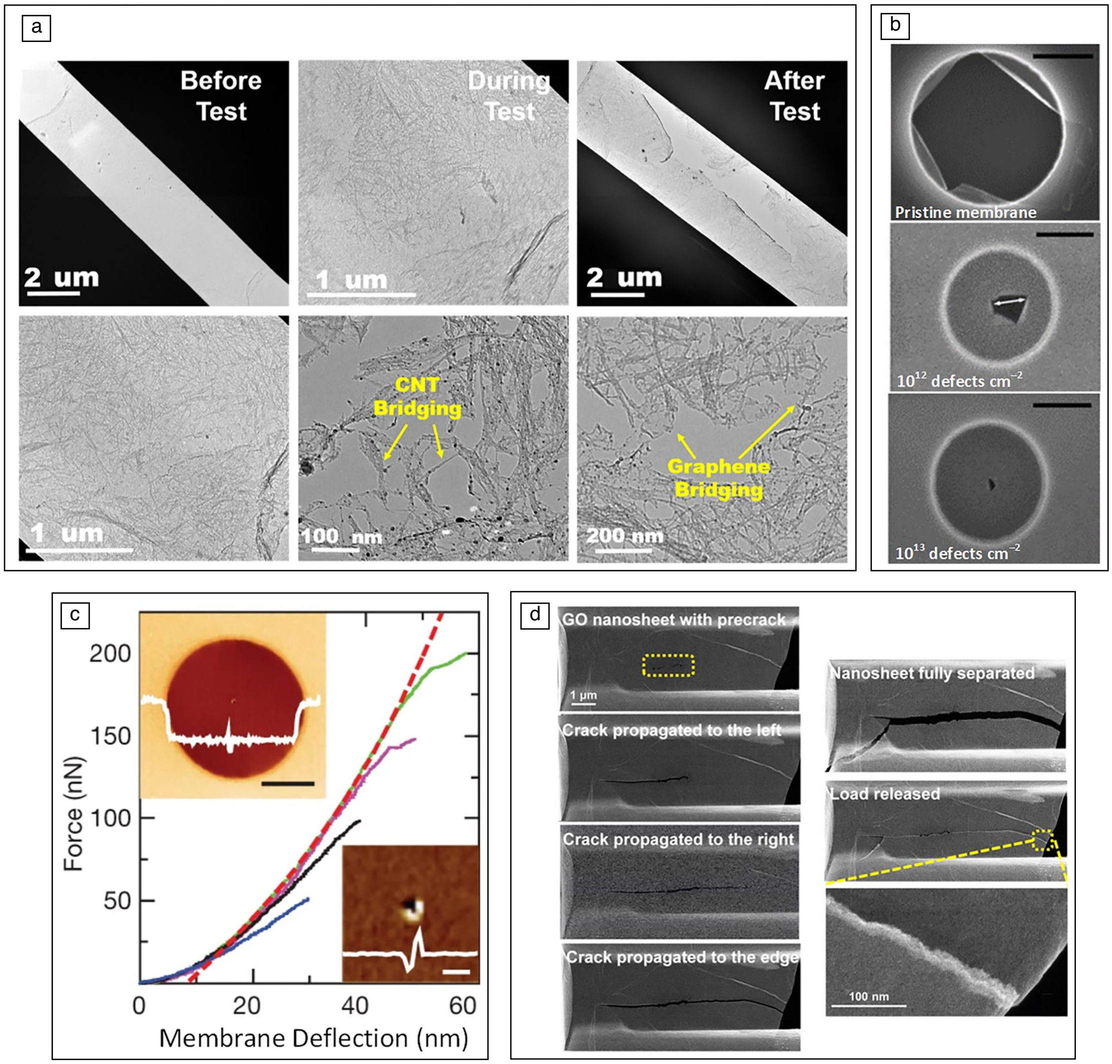

Figure 2. Toughening Graphene. (a) Transmission electron microscope images of a rebar graphene uniaxial tensile test with crack-deflecting capabilities highlighted.[29] (b) Graphene post atomic force microscopy indentation with different levels of defects in descending order: pristine membrane, defect density 10^{12} defects cm^{-2}, defect density 10^{13} defects cm^{-2}. The double arrow in panel b illustrates the tearing length.[30] (c) Monolayer amorphous carbon post indentation as well as the force versus membrane deflection of multiple samples.[31] (d) Scanning electron microscope images of crack growth in multilayer graphene oxide (GO).[32] Scale bars: (b) 500 nm, (c) top inset 1 μm, bottom inset 100 nm. CNT, carbon nanotube.

functionalized carbon atoms.[32] Moreover, it is hypothesized that the functionalization of other multilayer 2D materials should lead to an increase in their fracture toughness compared to nonfunctionalized counterparts.[32] Work that studied the fracture of thin and thick films of GO found that thin films (<30 nm) of GO failed due to intraplanar crack propagation; meanwhile thicker films (~70 nm) failed due to interlayer crack propagation.[34] The functionalization of GO, in the form of interlayer hydrogen bonds, again plays a factor, as molecular dynamics (MD) simulations found that they transfer loads between layers.[34]

Beyond carbon: Hexagonal boron nitrides, transition-metal dichalcogenides, and more

Hexagonal boron nitride (h-BN) possesses a honeycomb atomic structure very similar to graphene. The only difference is that boron and nitrogen atoms are adjacent to each other and form B-N covalent bonds instead of C–C bonds. Therefore, h-BN has an ultrahigh intrinsic strength (about ~100 GPa) and Young's modulus (about ~1 TPa). For most 2D materials, fracture normally occurs in a brittle manner as discussed earlier in the case of graphene, where a catastrophic failure happens in the early stage of crack propagation. This brittle nature of 2D materials such as graphene greatly restricts their potential for engineering applications. Surprisingly, it has recently been discovered that h-BN exhibits unique fracture behaviors and intrinsic toughening mechanisms owing to its asymmetric lattice structure. Using *in situ* SEM and TEM tensile tests of monolayer polycrystalline h-BN, large elastic strain up to 6.2% and 5.8% were achieved for defect-scarce samples and samples containing voids of about 100 nm, respectively.[35] Using *in situ* SEM tensile tests on monolayer monocrystalline h-BN with a natural precrack, as shown in **Figure 3**a, an extremely high fracture toughness was reported.[36] The effective energy release rate of h-BN was found to be 172 J m^{-2}, which is one order of magnitude higher than both its Griffith energy release rate and that reported for graphene. Due to the asymmetric edge polarization and threefold symmetry, crack deflection and branching occurred repeatedly during the crack propagation, which consumed a large amount of energy and thus contributed to the enhanced fracture toughness.[36] It is likely that many 2D materials with alternating bonds such as this have similar fracture behaviors.

Transition-metal dichalcogenides (TMDs) are layered materials with stoichiometry of MX_2, where M represents the transition-metal element and X represents the chalcogen species, such as S and Se. The quantitative study of the fracture toughness of 2D TMDs remains rare given their extreme sensitivity to flaws as demonstrated in the *in situ* SEM study of $MoSe_2$.[37] $MoSe_2$ was found to fracture brittlely, as shown in Figure 3b. From DFT calculations, the fracture toughness of $MoSe_2$ in terms of fracture energy was calculated to be ~3.1 J m^{-2}, which partly explains the challenging nature of such measurements.[37] An inverse analysis based on the Griffith theory suggested that fracture-producing preexisting defects in monolayer $MoSe_2$ could be on the order of tens of nanometers, which is hard to avoid during the material preparation or transfer. Instead of actively applying controlled tensile loading, studies of the fracture behavior of MoS_2 popped by E-beam found that cracks are either atomically sharp or edge reconstructed, as shown in Figure 2c, and that cracks predominantly propagate along the zigzag direction.[38] Moreover, MoS_2 was found to fracture brittlely until the defect density increased past a certain point;[38] recent work via AFM indentation also observed the same phenomenon

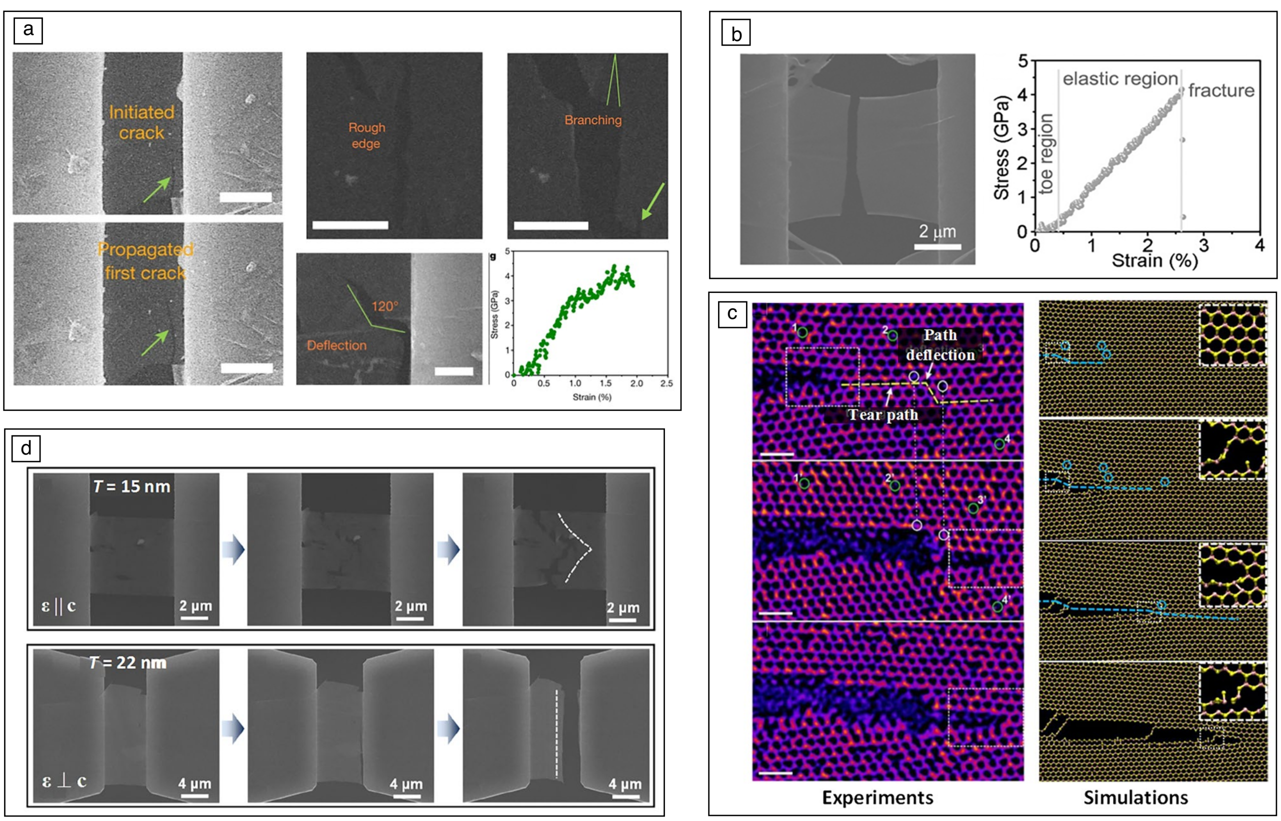

Figure 3. Experimental studies of fracture in other 2D materials beyond graphene. (a) Single-crystal monolayer h-BN that showed intrinsic toughening. Scanning electron microscope (SEM) images of the sample in the push-to-pull device, fracture edges, and the toughening mechanisms and the stress–strain response of h-BN.[36] (b) Brittle fracture of $MoSe_2$ SEM image and the stress–strain curve.[37] (c) Aberration-corrected transmission electron microscope images of crack propagation over time in monolayer MoS_2 with simulations modeling the observed behavior.[38] (d) SEM images of the anisotropic fracture behavior of 2D selenium. In the top figures the strength is higher and the failure strain is lower due to tension along the strong covalent bonds of 2D selenium, whereas in the lower figures the strength is lower and the failure strain is higher due to tension along the weak van der Waals bonds.[39] Scale bars: (a): 2 µm, (c): 1 nm.

by irradiating MoS_2 to increase defect density and then fracturing samples.[40] Anisotropic mechanical behaviors have also been noted in 2D fracture such as few-layer selenium, as shown in Figure 3d,[39] and few-layer black phosphorus.[41] The anisotropic mechanical behavior in few-layer materials is due to anisotropic structures. 2D selenium is comprised of covalently bonded chains that are held together with the weak van der Waals force making it strong along the covalent bonds and weak along the van der Waals interactions (Figure 3d). Meanwhile, black phosphorous contains "atomic puckers," similar to corrugation, which give it compliance when the puckers are perpendicular to the tensile load and thus flattened before failure and stiffness when the tension is parallel to the puckers so they do not contribute to the stiffness.

Theoretical studies of fracture of 2D crystals

Accompanied with the rapid advancement of experimental investigations, theoretical studies have played an important role in unveiling the fundamental mechanisms of various fracture processes in different 2D materials. In this section, we review some important developments in theoretical studies by outlining the multiscale nature of modeling fracture in 2D materials and highlighting the emerging applications of machine learning (ML) approaches to enhance the prediction and optimization capabilities of fracture events in 2D materials.

Multiscale modeling of fracture in 2D materials

By nature, fracture processes often involve events across different scales, ranging from bond rupture at the crack tip, possible atomic reconstructions in local process zones, to the continuum deformation across the material. Two-dimensional materials possess less confined geometry, various chemical compositions, and complicated constitutive relations. Thus, modeling and simulations at different scales and their combinations have proven to be important tools in studying and understanding the fracture characteristics of different 2D materials. Here, we only outline some main methodologies among them and refer the interested readers to specific reviews or studies for detailed discussions.

At the atomic scale, MD simulations are often adopted to study the detailed fracture process with atomic resolution (see more discussion in the next section). Importantly, special attention has been paid to validating the reliability of the adopted force field in capturing the right fracture behaviors. For example, the cutoff parameters in AIREBO/REBO potential[42,43] need to be modified to avoid nonphysical stiffening near bond rupture.[27] Various types of force fields, including conventional empirical interatomic potentials (IAPs)[44,45] and newly emerged ML-based ones,[46] have been developed for graphene and other 2D materials, which by itself forms an active research direction. When reliable force fields are not available, first-principle-based methods, like density functional theory (DFT) and tight-binding methods, have been utilized to predict the complex deformation and fracture behavior at the crack-tip region in 2D materials.[36]

Beyond atomic scales, phase-field modeling has been used for studying defect engineering in graphene.[47,48] Peridynamics[49] has also been applied for fracture simulations in graphene, given its lower computational costs compared to MD simulations.[50] At the continuum level, the finite element method (FEM) is the most used tool whose applications in graphene have been summarized comprehensively in another review paper.[51] Besides monolayer graphene sheets, multiscale modeling is also essential to investigate graphene-based materials. The massive literature works have been covered in different review papers about graphene-based layer materials,[52] and graphene-based polymer/metal composites.[53]

Machine learning models toward 2D fracture

Although physics-based multiscale simulations have provided invaluable insights about the fundamental mechanisms of fracture behaviors in 2D materials, serious obstacles still exist in applying those methods for large-scale samples in realistic time scales. For example, due to the high computational costs, MD simulations are typically performed for very short time scales with high strain rates,[54] which may not always reflect conditions of interest. There remains a longstanding demand for alternative avenues of tackling material fracture with higher efficiency. Recent breakthroughs in artificial intelligence (AI) and growing surges of applying ML-based approaches to various physics and material problems have opened new doors to study fracture phenomena in 2D materials. Here, we review some of the relevant directions, including fracture characterization, modeling and material design, with an emphasis on the unprecedented potentials in combing ML approaches with fracture studies in 2D materials.

ML-driven fracture characterization

Spurred onward by developments in the fields of feature recognition and image processing, fracture detection models have grown into applicable maturity across many contexts. In engineering, ML classification models can identify regions of ductile versus brittle fracture in images of structural steels with pixel-level fidelity.[55] In geology, deep neural nets have been utilized to recognize and identify fracture paths from 2D images of rocky outcrops, with the ability to be applied at scale in the field.[56] In medicine, deep image recognition models have been applied to augment human diagnoses of fracture in rib bones from 2D CT images.[57] Tools such as these have successfully learned on data sets of 2D images, and would be generalizable to the direct study of 2D materials provided a proper data set.

In the context of 2D materials themselves, there are varied ML efforts to predict fracture properties beyond just identification from an image. Using MD simulations as a base, supervised learning models have been developed to predict fracture strain, fracture strength, and Young's modulus of 2D materials such as $MoSe_2$[58] and WS_2,[59] as a function of material chirality, temperature, and strain rate. In doing so, a limited number of costly MD simulations can be leveraged to quickly gain greater insights. Furthermore, ML feature recognition from optical microscope images of graphene can successfully characterize fracture strength, outstripping efficiency of manual characterization by over an order of magnitude without sacrifice of accuracy.[60]

ML-driven fracture modeling

Developing models that can capture dynamic and mechanistic progressions of fracture remains a challenging area of investigation, but some progresses have been accomplished.

At the FEM scale, graph-based models have been used to represent a fracture with nodes as locations of damage within the material, edges as crack coalescence between those locations, and virtual edges as paths of potential cracking. Dynamic graph evolution predicted by a convolutional neural network, trained on high-fidelity FEM, thus acts as an effective model for fracture propagation.[61,62] As a result, the method can predict fracture evolution of a 2D material given multiple initial crack flaws of various sizes at various locations within the structure, and yield the time of material failure along with the final fracture path. Aside from a graph-based approach, an ML-aided phase-field method has been recently reported to predict both 2D and 3D fracture, wherein an extended support vector regression model with Dirichlet feature mapping is used to nondeterministically predict the probability of failure under a given load condition.[63] This approach yields both critical loads and predicted crack paths for a given material and has been demonstrated across both numerical and experimental tests.

At the MD scale, ML models have been implemented as surrogate fracture models. Viewing fracture propagation as a sequential classification problem, where each subsequent step of fracture is a function of the crack pattern that came before, allows for the implementation of a deep neural network utilizing a long short-term memory (LSTM) module to predict fracture propogation.[64] After training on MD simulations, these ConvLSTM models have succeeded in predicting fracture not only for representative 2D structures utilizing a Lennard-Jones potential, but also for predicting the qualitative fracture paths and quantitative fracture energies of specific materials like graphene[65] and MoS_2.[66] The rapidity of these predictions allows one to fully map out the fracture energy as a function of grain orientation in bicrystals and identify structural trends in more complicated polycrystalline structures.

Two-dimensional material design

With the advent of ML models that can quickly predict properties of interest, engineers can understand, discover, and synthesize structures in 2D materials toward their intended goals[67]—design for fracture behaviors being one of these topics of interest.

Through a combination of generative and evaluative models iterated by a genetic algorithm, a property such as shear crack resistance can be optimized with dramatically lower cost than brute-force methods.[68] Similar work has been done to optimize other properties including toughness,[69] and resilience to defects,[70] and obtain specific fracture paths[66] in 2D materials. ML models allow for directed exploration through an otherwise intractable design space and enable inverse design in previously unprecedented ways. The successes outlined thus far are no doubt just the beginning of an even greater understanding of, appreciation for, and control over 2D material fracture as we look toward the next 100 years.

Fracture behavior at the 2D limit

With ultrathin thickness and unprecedented mechanical properties, 2D materials have emerged as a new playground to study various fracture phenomena in solids and led to a series of novel discoveries about fracture behaviors and crack interactions that are rarely observed in bulk materials. In this section, we review some recent progresses along those directions, including crack–defect interactions, size effects, out-of-plane effects, edge effects, and interlayer interactions, with special attention to the comparison between fracture in 2D materials and conventional 3D bulk solids.

Crack–defect interactions

Crack–defect interactions are key in understanding fracture behaviors and constructing effective toughening mechanisms in various bulk materials, including metals,[71] ceramics,[72] and diamonds.[73] Inspired by these successes, in 2D materials the crack interactions with different kinds of defects, including vacancies,[74] Stone–Thrower–Wales (STW) defects,[75] dislocations, and grain boundaries (GBs),[76] have been studied via comprehensive methods. For example, combining MD simulations and continuum theory, researchers[74,77] demonstrated that nanoscale vacancies can alter the crack-tip field and crack path in graphene by changing the stiffness distribution (**Figure 4**a). Via such crack–vacancy interactions, the fracture strength of graphene can be tuned by strategically arranging nano-holes around the crack tips. Using MD simulations, scientists[75,78] have studied mechanical properties and failure morphology of graphene with STW defects and discovered the fracture toughness of graphene can be enhanced by defect-induced crack bridging (Figure 4b); Meng et al. [79] have shown that the nonlocal residual stress associated with dislocations in graphene can lead to the dislocation shielding effect on a crack tip, which agrees with the linear-elastic fracture mechanics prediction (Figure 4c). Beyond single crystals, the effects of GBs and their joints on fracture behaviors of polycrystalline 2D materials have also been investigated and several potential toughening mechanisms have been identified. For example, with MD simulations, Jung et al.[80] demonstrated that irregular GBs can reduce stress concentration and create branches near the crack tip, thus increasing the critical energy release rate for crack propagation by about 50% (Figure 4d). By studying a large number of random samples of various grain sizes, Shekhawat and Ritchie[76] have shown that the statistical variation of the toughness of polycrystalline graphene can be explained by the weakest-link statistics. Interestingly, by simulating graphene samples with well-shaped hexagonal networks of GBs, Song et al.[81] have discovered a pseudo Hall–Petch relation between the fracture strength and grain size that can be explained with a dislocation-pileup model (Figure 4e).

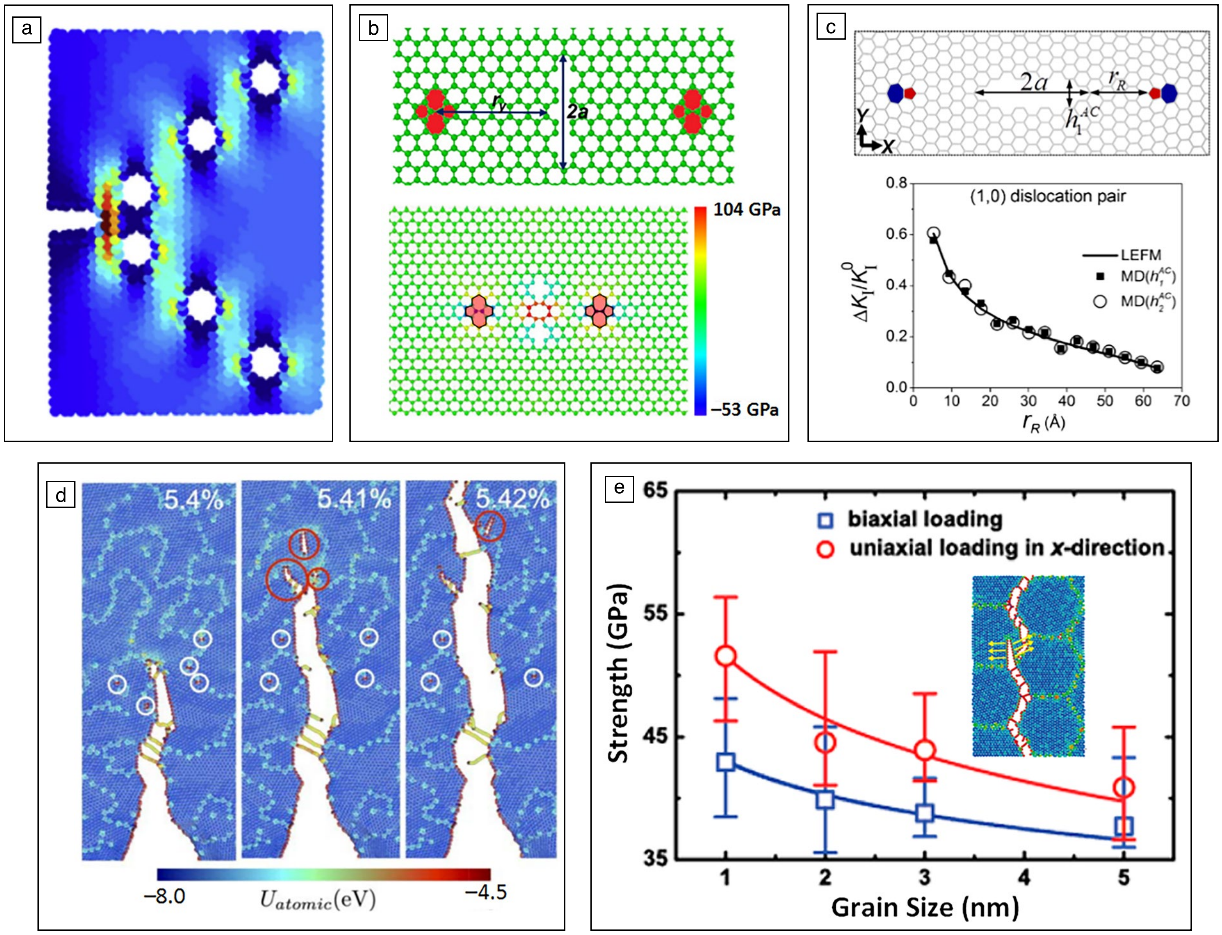

Figure 4. Crack–defect interactions in 2D materials. (a) Crack–vacancy interaction.[74] (b) Crack bridging induced by Stone–Thrower–Wales defects.[75,78] (c) Shielding effect of dislocations on a crack.[79] (d) Toughening effect of irregular grain boundaries in polycrystalline graphene.[80] (e) Pseudo Hall–Petch relation in polycrystalline graphene.[81] LEFM, linear-elastic fracture mechanics; MD molecular dynamics.

While some crack–defect interactions in 2D materials show similarities to those in the bulk materials and can be well captured via the Griffith theory and conventional fracture mechanics, a series of studies have highlighted a few unique aspects of fracture phenomena in 2D materials, which distinguish them from their bulk counterparts or predictions of conventional theories. Some are reviewed in the following.

Size effects

Griffith theory and conventional fracture mechanics were initially developed for macroscopic systems under continuum assumptions.[82] Identifying the critical dimension where continuum theory breaks down is of great theoretical significance for predicting fracture behaviors in nanomaterials including 2D materials. At the same time, the discovery of “smaller being stronger” in natural and man-made nanomaterials, such as nacres[7] and nanopillars,[83] raises great engineering interests in finding or fabricating stronger/tougher materials using 2D materials at the right scale. Motivated by these scientific questions and engineering applications, the size dependence of the failure mechanisms and the flaw tolerance phenomena in 2D materials have been studied by combining theories, simulations, and experiments. For instance, using MD simulations and theoretical analysis, Yin et al.[84] have demonstrated that the energy-based Griffith fracture criterion remains valid in graphene for cracks above 10 nm while a local strength-based failure criterion needs to be adopted for shorter cracks as the continuum assumption of a sharp crack diminishes under such small scale (**Figure 5**a). Taking advantage of the competition between the energy-based fracture and the strength-based bond rupture, Zhang et al.[85] proposed a nanocrystalline graphene strip model by introducing various defects and demonstrated that under a critical width, its failure becomes no longer sensitive to the presence of preexisting flaws, which agrees with the flaw tolerance theory[86] (Figure 5b).

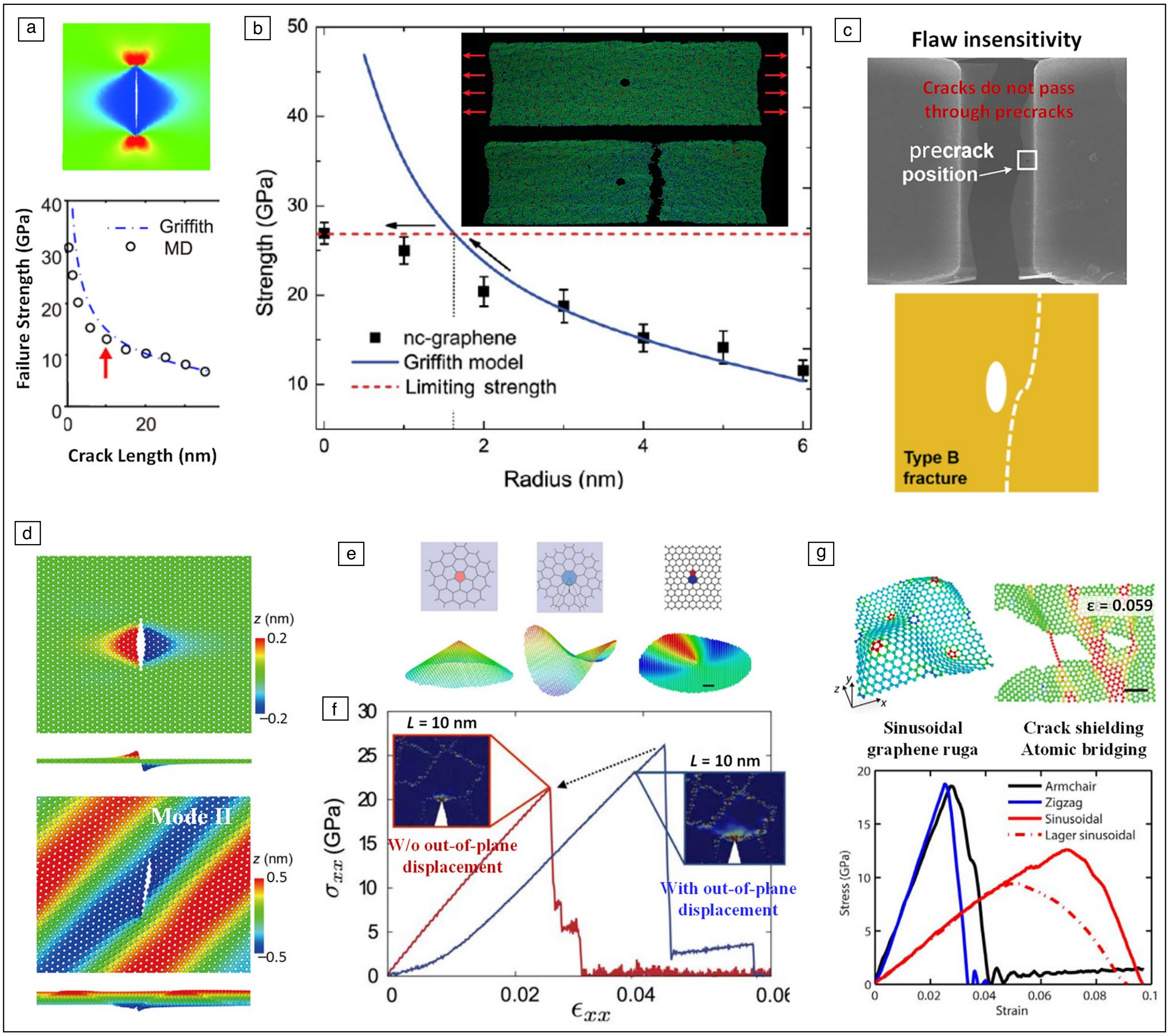

Figure 5. Size effects and flaw tolerance in 2D materials (a–c) and out-of-plane effects on the fracture behaviors of 2D materials (d–g). (a) Griffith theory overestimates fracture strength in graphene for nanocracks shorter than 10 nm.[84] (b) Flaw tolerance in nanocrystal graphene strip.[47] (c) Flaw tolerance in 2D covalent organic frameworks.[87] (d) Out-of-plane distortion of Griffith crack under modes I and II.[89] (e) Topological defects in graphene lead to out-of-plane displacements.[90] (f) Out-of-plane relaxations contribute to the toughening effect of grain boundaries in polycrystalline graphene.[80] (g) Sinusoidal graphene demonstrates enhanced fracture toughness compared with pristine graphene.[47] MD, molecular dynamics; nc, nanocrystal.

Recently, by adopting a newly emerged 2D covalent organic framework (COF), Fang et al.[87] have demonstrated experimentally that this new 2D material can remain flaw-tolerant with a strip width reaching beyond 2 μm (Figure 5c). These discoveries provide encouraging evidence on engineering robust nanomaterials out of 2D materials by taking advantage of the size effect and flaw tolerance and thus call for further studies of the size dependence of failure mechanisms in other 2D materials.

Out-of-plane effects

Because 2D materials are crystal layers of atomically thin thickness they often have very low bending resistance,[88] which can make out-of-plane deformation an energetically affordable or even favorable option to accommodate deformation. This out-of-plane deformation freedom distinguishes 2D materials from the predictions of conventional 2D in-plane theories. As such, fracture studies have continuously explored the novel out-of-plane effects in 2D materials. For example, combining MD simulations and theoretical analysis, Song et al.[89] have demonstrated that compressive in-plane stress in the Griffith crack field can lead to localized out-of-plane buckling in mode I and delocalized wrinkling in mode II, making the 2D Griffith theory overestimate the critical load for crack propagation in graphene (Figure 5d). Topological defects such as dislocations and GBs can also introduce out-of-plane distortions in 2D materials[90] (Figure 5e). It has been demonstrated that the toughening effect introduced by irregular GBs is strongly related to the out-of-plane relaxation as it decreases significantly when out-of-plane motion is forbidden in the same simulation[80] (Figure 5f). Taking advantage of this coupling between topological defects and out-of-plane deformations, Zhang et al.[47] constructed a sinusoidal graphene ruga model with distributed declination quadrupoles and demonstrated that it shows toughening mechanisms such as nanocrack shielding and atomic-scale bridging and results in a nearly twofold enhancement in fracture energy compared with pristine graphene (Figure 5g). Besides defect-induced out-of-plane effects, folds, wrinkles, and corrugations in non-flat regions of graphene have also been experimentally observed to act as barriers to crack propagation and arrest cracks (see discussions in the previous experimental section). These out-of-plane effects reveal the unique coupling between in-plane and out-of-plane deformations in 2D materials and open doors to complex fracture behaviors and novel toughening mechanisms. Interested readers may refer to specific reviews[91] for more discussions on this topic.

Edge effects

In 2D materials, the crack surfaces/edges can also affect the fracture process in a way that is rarely observed in bulk materials. For instance, the experimentally measured fracture energy release rate of single-crystal monolayer h-BN is one order of magnitude higher than its surface energy,[36] thus defying Griffith's theory (**Figure 6**a). DFT calculations revealed that the symmetry-breaking crack edges (boron/nitrogen-dominant ones) in h-BN generate asymmetrical edge stress and elastic properties, which is rarely observed in bulk materials and different from conventional surface elasticity theory that assumes symmetrical edge states.[92,93] This asymmetric edge effect results in a mode II stress intensity factor (SIF) that automatically tracks the crack tip from behind and leads to repeated crack branching and deflections as the crack edges swap during the propagation. This edge-enabled intrinsic toughening mechanism makes h-BN maintain high strength as well as high toughness. Besides toughening, the edge effect can also reduce the effective toughness in 2D materials. In 2D rhenium disulfide (ReS_2), Huang et al.[94] have experimentally observed that plastic deformation due to lattice reconstructions can initiate from the post-crack edges (instead of the crack tip as bulk materials usually do)[92] and superpose an opening strain to the crack tip, reducing the effective fracture toughness (Figure 6b). The crack edge properties in 2D materials can also be tuned by chemical functionalization. For example, via simulations, it has been predicted that chemical additives (e.g., oxygen) can affect the crack path in graphene under tearing[95] (Figure 6c), and hydrogen passivation enhances the fracture toughness of h-BN under mode I[96] (Figure 6d). Evidenced by these examples, special attention may need to be paid to the edges when studying fracture phenomena in various 2D materials.

Effects of interlayer interactions

Going beyond monolayers, fracture in multilayered 2D materials can be affected by interlayer interactions. For example, cracks can propagate asynchronously (Figure 6e) along dissimilar paths (Figure 6f) in trilayered graphene due to interlayer slippage.[28] At the same time, the interlayer interactions in multilayered 2D materials are mainly governed by dispersive van der Waals (vdW) interactions and sensitive to the detailed interlayer stacking order, in-plane and out-of-plane deformations. Currently, understanding the properties of such interfaces in 2D materials is an active research field by itself.[97,98] Under such interlayer interactions, complex fracture behaviors have been observed. For instance, combining *in situ* TEM and MD simulations, Jung et al.[99] have studied the fracture behaviors in a bilayer MoS_2 system under electron beam and observed that the initial crack can propagate, get blocked or branched in the original layer, or a new crack can initiate in the neighbor layer due to the initial crack (Figure 6g). The complex fracture behaviors are revealed to be closely related to the highly variable interlayer friction, which is sensitive to the interlayer stacking order and in-plane loading conditions. Similar effects of interlayer interactions on the fracture behavior have also been observed in GO systems (see more in the earlier experimental section). As the interlayer interactions in GO can be affected by vdW interactions, H-bonding, and interlayer covalent bonding via functionalization, more complex fracture behaviors are expected within and between layers thus calling for in-depth studies on this topic.

Challenges and opportunities

Despite the great progress made in the past decade to understand fracture of 2D materials from both theoretical and experimental fronts, much remains to be explored. The unique features of 2D materials including its diminishing thickness dimension and the combination of extraordinary physical and chemical properties, provide both great challenges to investigate their unique fracture behaviors and a fertile ground to develop exciting synthesis-structure–property-application relationships at the 2D limit potentially extending our knowledge for the science of fracture beyond the Griffith theory. In this section, we will highlight a few areas that we believe could benefit from synergistic and collaborative efforts from the community.

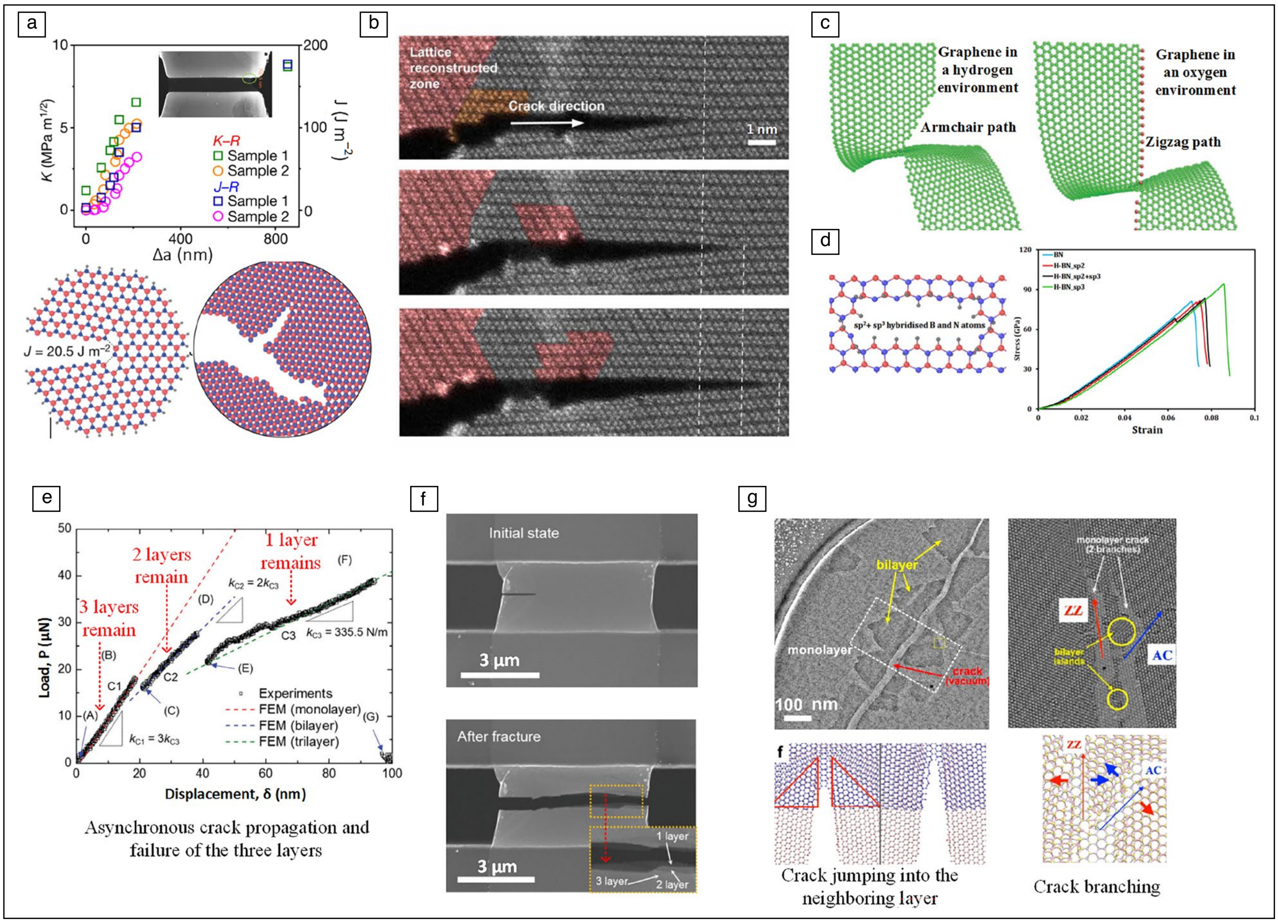

Figure 6. Edge effects (a–d) and effects of interlayer interactions (e–g) on the fracture behaviors of 2D materials. (a) Asymmetric edge stress and elastic properties in hexagonal boron nitride (h-BN) lead to crack branching, deflection, and stable crack propagation.[36] (b) Lattice reconstruction initiates from crack edges behind the crack tip in 2D rhenium disulfide.[94] (c) Chemical functionalization affects the crack path in graphene under tearing load.[95] (d) Hydrogen passivation enhances the fracture toughness of h-BN.[96] (e, f) Cracks in a trilayer graphene propagate along dissimilar paths asynchronically.[28] (g) Crack paths in bilayer MoS_2 samples are affected by the interlayer frictions.[99] FEM, finite element method.

Effects of defects on fracture

As discussed earlier, the importance of various types of defects on affecting fracture behaviors of 2D materials has been extensively studied via simulations and experiments. However, how to precisely control the creation and distribution of specific types of defects in 2D materials remains a key challenge. Breakthroughs in this direction will benefit not only fundamental studies concerning crack–defect interactions at the 2D limit, but also robust engineering applications of 2D materials against fracture. For example, advancements in sample preparation and testing can be key in enabling more systematic studies on the novel impacts of some unique types of defects (e.g., topological defects and edge defects) on the fracture behaviors in 2D materials. Additionally, going beyond monolayers, interfacial defects are becoming increasingly important and providing exciting opportunities for property tuning in the fast-growing family of van der Waals solids with heterostructures. Addressing questions like how we can engineer interfacial interactions to moderate fracture behaviors in multilayered 2D materials will surely open new possibilities for both engineering applications and scientific quests.

Complex loading conditions in 2D fracture studies

There are a few methodologies developed at this point for obtaining the fracture strength, Young's modulus, and fracture toughness of 2D materials. But these experiments only exist for room temperature, quasi-static, tension/point loading. Therefore, there is much that is still unknown and to be explored about the fracture of 2D materials. (1) How does the strength and fracture of 2D crystals relate to the temperature, or strain rate, or some combination of the two? Virtually no experimental work exists in this area beyond impact.[100]

(2) How do different loading conditions affect the fracture of 2D materials? To date no quantitative shear, torsional, or biaxial tension experimental methods exist. (3) What is the impact of mode II failure of interlayer bonds on overall failure behaviors of 2D materials? Some methods have explored this property;[101] however, a 2D lap shear style test has yet to be performed. Understanding these fracture conditions will help us improve the connection to modeling/simulations, better utilize 2D crystals in applications, and discover potential unique properties such as the difference between graphene and h-BN fracture.[36]

Multi-physics studies

At the 2D limit, one very exciting aspect is "multi-physics" studies that seek to understand the intersection of fracture mechanics with other disciplines. For example, chemical functionalization of the abundantly available surfaces, interfaces, and even edges could alter the fracture behavior in a more profound way compared to bulk materials. Electrochemical energy-storage and conversion systems, such as electrode–electrolyte interfaces in batteries, can be an area where studying the interplay between electrochemical reactions and fracture properties in 2D materials-based systems is highly needed. On the other hand, the ability to control the highly concentrated stress/strain field ahead of a crack tip could be used to modulate the electronic structure of 2D materials to an extent not yet achieved via strain engineering in the semiconducting industry. Similar types of modulation could be realized for other properties such as optical and thermal, opening an under-explored area of fracture-enabled functional property modulations.

In situ experimental analysis

Most current *in situ* fracture studies of 2D materials were performed under an electron microscope (SEM or TEM). Although such studies provide important insights into the fracture processes, there is still ample room for improvement. For example, how to achieve a quantitative fracture study at the atomic resolution while minimizing the electron-beam damage is a challenge that is just beginning to be addressed.[102] Following the discussion of multi-physics studies, can we globally or locally probe different functional properties to correlate them with quantitative fracture in 2D materials? On the other hand, extending the probing modules beyond electron microscopy, with sufficient temporal and spatial resolutions, will be very important to enrich the *in situ* fracture study toolbox. Super resolution optical microscopy/spectroscopy and different types of ion-based microscopy/spectroscopy techniques in conjunction with mechanical testing platforms discussed earlier could greatly expand our capabilities to study fracture and related phenomena at the 2D limit.

Interatomic potentials development

The reliable force fields for 2D materials are essential for studying fracture behaviors using MD simulations. Facing the rapid progress in synthesizing new and complex 2D materials and their assemblies, some novel directions for current IAP development have emerged. For example, most of the current IAPs haven't been optimized to capture the subtle interlayer interactions in multilayer 2D materials. Given the rise of magic-angle twisted bilayer graphene[103] and vdW heterostructures, the development of reliable interlayer potentials has emerged as a promising research direction. Also, continuously developing IAPs for novel 2D materials (e.g., COFs and MXenes) with various chemical compositions and structural diversities is another important research direction. Additionally, facing these growing complexities in constituents and configurations, AI and ML tools can be helpful in providing room for models to go beyond conventional paradigms (e.g., empirical functional forms or explicit physical relations). Although it can be a naïve thought, the ultimate goal to develop a unified potential framework for all types of 2D materials might only be possible by training a large-scale ML model.

Deep learning in 2D material fracture

The past decade has witnessed an explosion of applications of deep learning (DL) models in various fields. Excitingly, the specific applications of these tools, such as image processing models, to 2D material fracture have only just begun, with many unexplored pathways on the horizon. For example, for multiscale modeling of fracture, there exists a current divide between the atomic detail of MD simulations and the scalability of FEMs. Although we have started to see how ML models can accelerate 2D material fracture modeling (as discussed in the earlier subsections), we have yet to see a full implementation of finite element scale systems treated with an ML approach that has learned MD-level behaviors. The ability to see across multiple length scales simultaneously is one longstanding problem that DL methods may finally allow us to breach. In doing so, we may be able to identify and understand precisely how properties at the macroscale emerge from the collective properties and behaviors at the micro- and nanoscale.

Novel 2D materials by design

Despite the growing understanding of mechanical properties of 2D materials, there remains plenty of challenges in understanding fracture at such 2D limits and tuning the materials for optimized performances. With heterostructures, topological defects, kirigami, and COFs, broad spaces for designing novel 2D material systems of improved properties and better functionality are waiting to be studied and explored. For example, with COFs, what is the underlying relationship between elementary structures (i.e., pore geometry or pore shape and flexibility of the skeleton) and overall fracture properties? Can we design such 2D polymeric materials with stronger noncovalent interlayer bonds (e.g., interlayer hydrogen bonds or electrostatic force) to achieve higher fracture toughness? The designability of 2D COFs is just one material platform that has a highly promising potential for gaining a fundamental understanding of structure-fracture-property relationships in 2D materials. At the same time, the combination of predictive and generative DL models and advanced genetic algorithms has great potential in providing practical pathways to navigate and explore the broad design space to rapidly accelerate novel 2D materials design far beyond the current pace.

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Conflict of interest
On behalf of all authors, the corresponding authors state that there is no conflict of interest.

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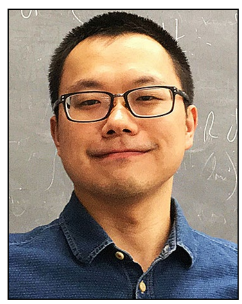

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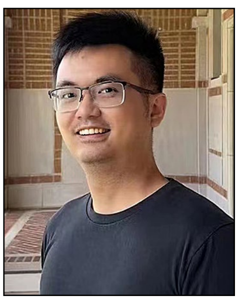

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