Nonlocal intrinsic fracture energy of polymer-like networks

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Connecting polymer network fracture to molecular-level chain scission remains a quandary. While the Lake-Thomas model predicts the intrinsic fracture energy of a polymer network is the energy to ~~scission~~ rupture a layer of chains, it underestimates recent experiments by ∼1-2 orders of magnitude. Here we show that the intrinsic fracture energy of polymer-like networks stems from nonlocal energy dissipation by relaxing chains far from the crack tip using experiments and simulations of 2D and 3D networks with varying defects, dispersity, topologies, and length scales. Our findings not only provide physical insights into polymer network fracture but offer design principles for tough architected materials.

Polymer networks are the molecular scaffolds that form the basis of materials in a wide range of both common and highly specialized applications, including consumer products (like tires, rubber bands, contact lenses) [1], biomedical implants [2] and soft electronic devices [3]. The lifetime of polymer networks is limited by their fracture, which is characterized by the energy required to propagate a crack per newly created surface area [4]. This energy typically has contributions from bulk dissipation and elastically active structures ~~of~~ within the material~~s~~ [5]. The contribution from the latter is known as intrinsic fracture energy [6, 7]. As established by Griffith in the 1920s [8], the intrinsic fracture energy of glass can be considered as the energy required to break a layer of atomic bonds, i.e., $\Gamma_0 = MU_{\mathrm{bond}}$, where M is the number of broken atomic bonds per unit created area and U_{bond} is the bond dissociation energy of a single atomic bond (Fig. 1a). Although this simple model ~~successfully~~ semi-quantitatively explained the fracture of some hard materials, the intrinsic fracture energy of polymer networks has been found to be several orders of magnitude larger than just that of breaking a single layer of atomic bonds [9].

Lake and Thomas explained this phenomenon in 1967 by connecting the intrinsic fracture energy of polymer networks to the rupture of covalent polymer chains [10]. This model predicts that the intrinsic fracture energy is equal to the number of broken bridging polymer chains per unit created area (M) multiplied by the work to rupture a bridging polymer chain (U_{chain}), i.e., $\Gamma_0 = \Gamma_{\mathrm{LT}} = MU_{\mathrm{chain}}$ (Fig. 1b). During crack propagation, the energy stored in the bridging polymer chains is dissipated after the chains are broken. The Lake-Thomas model has been widely applied to explain experimental data and predict intrinsic fracture energy [11–13]. Recent experiments [14, 15], however, indicate that the Lake-Thomas model has also significantly underestimated the intrinsic fracture energy of polymer networks by ∼1-2 orders of magnitude (see Table I). Modified models have been proposed to rationalize this underestimate [7, 15–17], but these models cannot fully explain the orders of magnitude discrepancy between the Lake-Thomas model and experimental results. This discrepancy implies the potential for nonlocal energy release and dissipation even within well-formed gels that are highly elastic. This is further supported by recent studies on brittle hydrogels [18, 19], which suggest the existence of a nonlocal process zone around the crack tip.

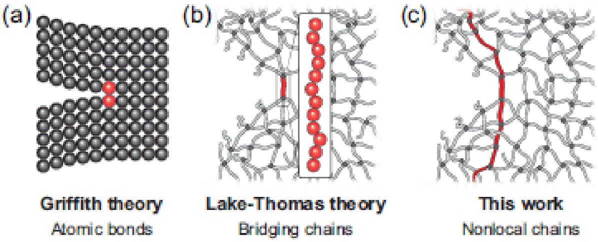

FIG. 1. **Intrinsic fracture energy.** (a) In Griffith's theory, the intrinsic fracture energy equals the energy needed to break a single layer of atomic bonds. (b) In the Lake-Thomas model, the intrinsic fracture energy equals the energy needed to break a single layer of polymer chains. (c) Our study suggests that the intrinsic fracture energy mainly results from nonlocal energy dissipation by relaxing polymer chains far away from the crack tip and is orders of magnitude higher than Lake-Thomas prediction.

	Wang et al. [20]	Lin et al. [7]	Akagi et al. [12]	Barney et al. [15]
$\Gamma_0/\Gamma_{\mathrm{LT}}$	**≈ 77**	**≈ 60**	**≈ 72**	**≈ 150**

TABLE I. The ratio of experimentally measured intrinsic fracture energy, Γ_0, to the predicted values according to the Lake-Thomas model, Γ_{LT}. These experimental results indicate that the Lake-Thomas model underestimated the intrinsic fracture energy of polymer networks by 1∼2 orders of magnitude.

~~The concept of connecting chain scission to the intrinsic fracture energy of a network should not be limited to the microscopic level [10].~~ In this letter, we ~~therefore~~ investigate the fracture of generic networks that consist of polymer-like chains across multiple length scales. We find that the intrinsic fracture energy of the network is orders of magnitude greater than the energy required to rupture a single layer of polymer chains. This discrepancy arises because energy is released and dissipated from the relaxation of polymer chains far beyond the crack tip when a bridging chain ruptures (Fig. 1c).

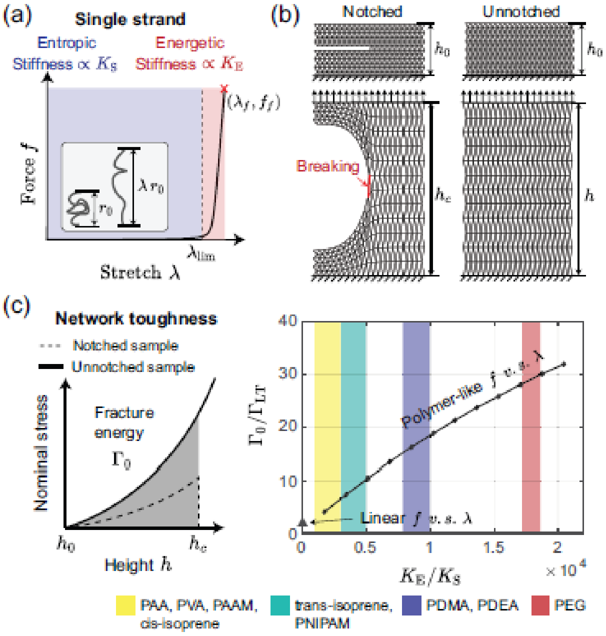

FIG. 2. **Intrinsic fracture energy of polymer-like networks.** (a) A single strand is governed by a strongly nonlinear force-stretch curve, with a long, soft entropic part characterized by a modulus K_S and a short, stiff energetic part characterized by a modulus K_E. (b) To measure the intrinsic fracture energy of polymer-like networks, a pure shear test is conducted. (c) The intrinsic fracture energy Γ_0 of polymer-like networks with different ratios of K_E/K_S (solid line) compared to a network consisting of linear chains (triangular marker). These results are obtained from numerical simulations under quasi-static loading conditions. Parameters K_S and K_E are taken from the single-molecule force spectroscopy (SMFS) experiments of different polymers: poly (acrylic acid) (PAA) [21], poly (vinyl alcohol) (PVA) [22], polyisoprene [23], poly(acryl amide) (PAAM) and poly(N-isopropyl acrylamide) (PNIPAM) [24], poly(dimethylacrylamide) (PDMA) and poly(diethylacrylamide) (PDEA) [25], poly(ethylene glycol) (PEG) [26].

We adopt the modified freely jointed chain model (m-FJC) [27] to describe the force-extension dependence of chains. The m-FJC model considers both the conformational entropic elasticity of the polymer chain and the energetic elasticity of backbone bonds (e.g., bond stretching and bending). The relationship between its stretch λ and reaction force f can be written as [27]

$$\frac{\lambda}{\lambda_{\lim}} = \left[\coth\left(\frac{f}{K_S}\right) - \frac{K_S}{f}\right]\left(1 + \frac{f}{K_E}\right), \tag{1}$$

where the parameters K_S and K_E are the soft entropic modulus (Fig. 2a, blue) and the stiff energetic modulus (Fig. 2a, red) of the polymer chains, respectively ($K_E \gg K_S$ for typical polymer chains) [28]. The parameter $\lambda_{\lim}$ is the entropic stretch limit beyond which the force increases rapidly due to the deformation of backbone bonds.

To model the fracture of polymer-like networks, we start with 2D samples with triangular lattices consisting of n nodes and e edges (Fig. 2b). Each edge is modeled by a nonlinear spring with initial end-to-end distance r_0 and force-stretch relation $f(\lambda)$ defined in Eq. (1) [29, 30]. To further capture the fracture of a polymer chain, the nonlinear spring is set to break at a force f_f with stretch λ_f. In the simulation, clamped boundary conditions are applied to the top and bottom surfaces, which quasi-statically stretch the sample from an initial height of h_0 to a height of h. The deformation of the lattices is fully described by the coordinates of every node (x_i, y_i), where $i = 1, ..., n$. At each loading step, the total energy of the system is obtained by summing the elastic energy of every spring:

$$U_{\text{total}} = \sum_{i,j} \int_1^{\lambda_{ij}} f(\lambda')d\lambda' \tag{2}$$

where $\lambda_{ij} = r_0^{-1}\sqrt{(x_i - x_j)^2 + (y_i - y_j)^2}$ is the stretch of the edge connecting node i with j. The coordinates of each node (x_i, y_i) are then numerically determined by minimizing U_{total} using Newton's method in MATLAB. Additionally, if $\lambda_{ij} > \lambda_f$, which indicates that the edge connecting nodes i and j has broken, this edge will be removed from the lattice for future steps (see numerical details in SI section S2). To measure the intrinsic fracture energy of a given network, we perform the pure shear test [4]. This includes two steps: i) load a notched sample to the point where the bridging chain breaks and record the critical height of the sample h_c; ii) load an unnotched sample and record the nominal stress as a function of sample height h (Fig. 2b). The intrinsic fracture energy of the network is then calculated as

$$\Gamma_0 = \int_{h_0}^{h_c} s\, dh, \tag{3}$$

where s is the nominal stress of the unnotched sample The value of Γ_0 is an intrinsic property of the network and is size independent, provided the network is sufficiently large. While, for polymer-like networks with $K_E \gg K_S$, a converged value of Γ_0 requires a significantly large sample, typically containing over 1000 layers of chains (see SI section S2.4 for convergence studies).

Using a sufficiently large sample with 4000 layers, we calculate the fracture energies Γ_0 of the polymer-like networks with a wide range of K_E/K_S values and compare them to the Lake-Thomas predictions, i.e., $\Gamma_{LT} = MU_{\text{chain}}$. Parameters K_E and K_S are taken from reported single-molecule force spectroscopy (SMFS) experimental results of different polymers that use the m-FJC model as the fitting function. As shown in Fig. 2c, Γ_0/Γ_{LT} is significantly larger than unity for all networks with polymer-like chains. This phenomenon is closely linked to the nonlinearity of the force-stretch behavior of the polymer-like chains, as increasing K_E/K_S ratios ~~resulting~~ gives in larger Γ_0/Γ_{LT} ratios. Notably, when we use linear springs to model the chains, the ratio Γ_0/Γ_{LT} nears the expectation of the Lake-Thomas model (~~dashed line~~ triangular marker). These results emphasize that – even without hysteresis – the force-extension relationship of chains directly affects the intrinsic fracture energy of polymer-like networks.

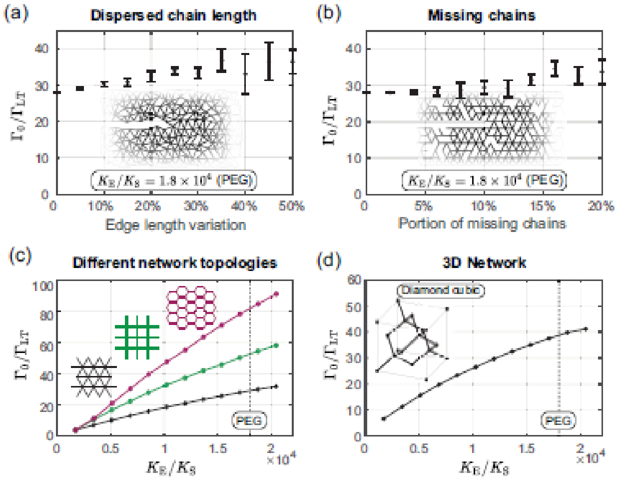

FIG. 3. **Generality of elevated intrinsic fracture energy in polymer-like networks.** (a) Intrinsic fracture energy of irregular networks with dispersed edge lengths, with variations ranging from 0% to 50%, and (b) networks with missing chains ranging from 0% to 20%. Both (a) and (b) are based on single strands with $K_E/K_S = 1.8 \times 10^4$ (PEG chains). Intrinsic fracture energy of networks with (c) different 2D topologies, including triangular, square, and hexagonal lattices, and (d) 3D diamond cubic lattices for varying K_E/K_S ratios.

To assess the generality of our findings, we conduct additional simulations that investigate the impact of inhomogeneities, defects, and lattice topologies on the intrinsic fracture energy of polymer-like networks. Specifically, we feature the PEG-like chain with $K_E/K_S = 1.8 \times 10^4$ and examine the intrinsic fracture energy of irregular networks (lattices with dispersed edge lengths, Fig. 3a) and networks with dangling chains (lattices with missing edges, Fig. 3b). Unsurprisingly, we find that these defected networks retain large Γ_0/Γ_{LT} ratios. Different topologies, including triangular, square, and hexagonal lattices, along with 3D diamond cubic lattices are also investigated with various K_E/K_S ratios. Fig. 3c shows that increasing chain nonlinearity leads to a magnifying trend of Γ_0/Γ_{LT} in different 2D lattice topologies, where the hexagonal lattice exhibits relatively higher fracture energy due to its larger loop size~~s~~ [17, 31–33]. A similar trend is also observed in the 3D diamond cubic network (Fig. 3d), which signifies that more chain nonlinearity leading to a higher Γ_0/Γ_{LT} is a universal phenomenon, regardless of lattice topology or dimensionality. These numerical results are consistent with recent experimental findings [7, 12, 15, 20] showing that the intrinsic fracture energy of polymer networks can be significantly larger than the values predicted by the Lake-Thomas model (see Table S1).

To further understand the anomalously high intrinsic fracture energy of polymer-like networks, we investigate the behavior of chains near the crack tip (Fig. 4a) by comparing networks of PEG-like chains to those with linear elastic chains. The strain energy distributions of the two networks just prior to the fracture of a chain are shown in Fig. 4b, where the color indicates the energy on each chain normalized by the work to rupture a single chain U_{chain}. Notably, the energy distribution of the PEG network is clearly nonlocal when contrasted to the network with linear chains, where the energy is concentrated in the one or two layers nearest to the crack tip. We therefore postulate that for polymer-like networks, the abnormally high ratio of Γ_0/Γ_{LT} is predominantly due to nonlocal energy dissipation by the relaxation of chains distant from the crack tip. To test this hypothesis, we analyze the released energy of each chain, which is defined by the energy difference before and after the fracture of a single chain at the crack tip. The Lake-Thomas model predicts the released energy of the broken chain contributes to the overall intrinsic fracture energy of the network. This is qualitatively true for the network consisting of chains with linear elasticity, where the major energy release is limited to a relatively confined zone around the crack tip (e.g., ∼1-2 layers Fig. 4c, top). However, for the network of PEG-like chains, even chains far away from the crack tip (e.g., $>$ 20 layers) release a significant amount of energy after the rupture of a single chain at the crack tip. The released energy is then converted into kinetic energy in the network and is eventually dissipated.

To investigate where and how the released energy is dissipated, we establish a spring-mass model to simulate the dynamic process upon the fracture of a chain at the crack tip until a new equilibrium is reached. When a bridging chain ruptures, the tension on the bridging chain suddenly vanishes, and other junctions in the network continuum are not balanced. The elastic energy stored in the continuum is then released and becomes the kinetic energy of the junctions. In real polymer net-

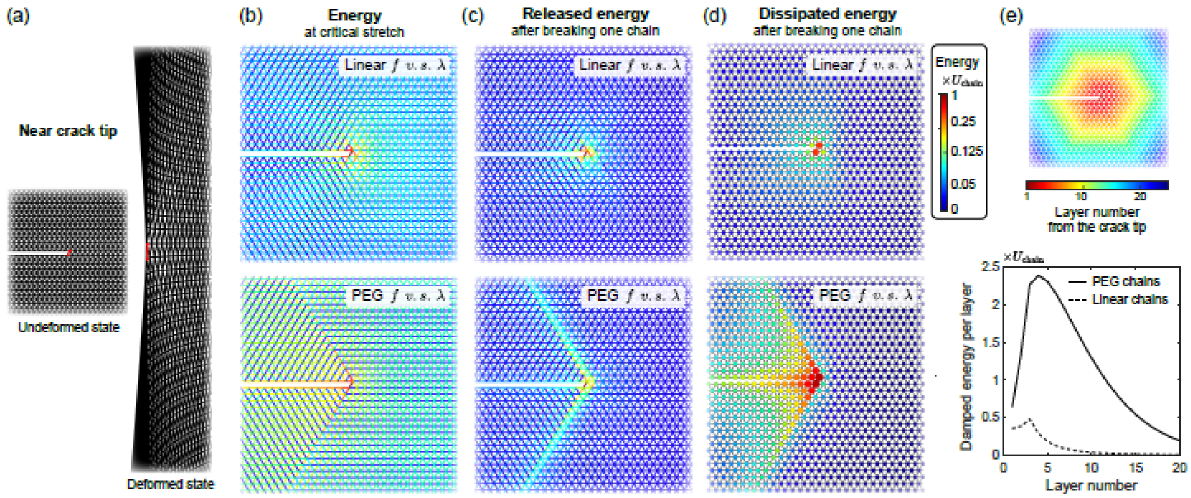

FIG. 4. **Nonlocal energy release and dissipation during fracturing of polymer-like networks:** (a) Visualization of the polymer-like network near the crack tip in the undeformed and deformed states. (b) Energy stored in each chain just before a chain breaks at the crack tip. (c) Energy released by each chain after a chain breaks. (d) Energy dissipated at each node after a chain breaks. In (b-d), the top row presents results for networks with linear chains, while the bottom row presents networks with PEG chains. The energy on each chain is normalized by the work to rupture a single chain U_{chain}. (e) Damped energy as a function of the layer number away from the crack tip for networks consisting of PEG chains (solid line) and linear chains (dashed line).

works ~~analogs~~, the energy released from the network continuum is damped by the relaxation of both broken and unbroken polymer chains, and eventually dissipated as heat. To damp the kinetic energy induced by the chain breaking in our simulated network, we implement viscous damping on every node of the lattice. Unlike dynamic fracture [34–36], we aim to investigate the critical tearing energy of polymer-like networks, under conditions by which the crack velocity approaches zero. Therefore, the network was deformed quasi-statically as previously mentioned and set to be overdamped to mitigate potential dynamic effect. The total damped energy throughout this process is recorded and shown in Fig. 4d. While the released energy maps in Fig. 4c tell us where the energy comes from, the damped energy in Fig. 4d shows where the energy goes during the fracture process. To provide a more quantitative comparison, we present the total damped energy as a function of layer number from the crack tip in Fig. 4e. In stark contrast to linear networks (dashed line) where most of the energy is damped near the crack tip, the polymer-like network (solid line) dissipates much more energy through chains far away from the crack tip; this eventually leads to the high ratio of $\Gamma_0/\Gamma_{\mathrm{LT}}$. With the same U_{chain}, a network made from polymer-like chains is much tougher than that made from chains with linear elasticity. Additionally, the ratio $\Gamma_0/\Gamma_{\mathrm{LT}}$ provides insight into the spatial extent of the dissipation zone, quantified in terms of the layers of chains (see SI section 2.3.2 for details).

Finally, in order to experimentally validate our findings, we fabricated macroscopic architected materials that consist of polymer-like strands. Specifically, each strand was produced by laser cutting (model: Epilog Zing 24 60W) an Acetal Film (semi-clear white, 0.003" thick, Mcmaster-Carr) into an initially folded ribbon. The ribbon-shaped strand is designed to mimic the force-extension behavior of polymer chains, with ~~an~~ soft initial unfolding followed by a stiff stretching upon loading [37]. The former corresponds to the "entropic stiffness", while the latter corresponds to the "energetic stiffness". This results in a ratio of energetic to entropic stiffness $K_{\mathrm{E}}/K_{\mathrm{S}} \approx 800$ (Fig. 5a). To test the intrinsic fracture energy of this architected material, ~~the~~ ribbon strands ~~is formed~~ are connected into a triangular lattice (Fig. 5b). Tensile tests are performed to obtain the force-stretch curve for the unnotched sample (Fig. 5c, black solid curve) and the critical height h_c for the notched sample (red solid line, see SI section 3 for fabrication and testing details). The measured intrinsic fracture energy of the network is significantly higher than that predicted by the Lake-Thomas model, with $\Gamma_0/\Gamma_{\mathrm{LT}} = 5.57$. Furthermore, simulations of the network using the experimentally determined force-stretch curves for each strand match the experimental results for both notched and unnotched samples, yielding a similar $\Gamma_0/\Gamma_{\mathrm{LT}} = 5.61$ with an error of only 0.7%. These experiments not only validate the accuracy of our simulations but also demonstrate a new approach to designing ultra-tough architected materials by engineering the force-stretch response of individual strands.

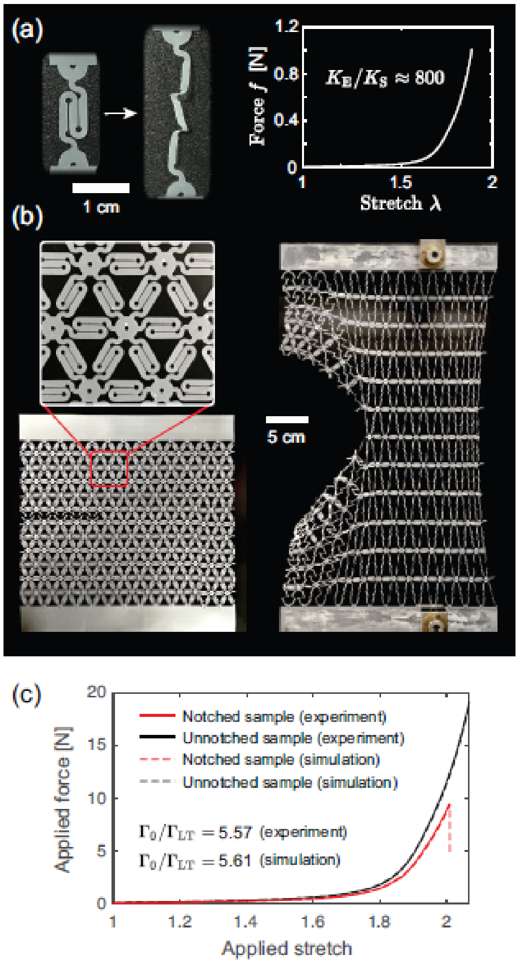

FIG. 5. **Experimental validation of the polymer-like behavior and enhanced intrinsic fracture energy in architected materials.** (a) Force-stretch response of an architected strand fabricated to mimic the behavior of polymer chains, with a ratio of energetic to entropic stiffness $K_E/K_S \simeq 800$. (b) A laser cut architected material consists of polymer-like strands. The pure shear test is performed to measure its intrinsic fracture energy. (c) Pure shear tests on the architected materials via experiments (solid lines) and simulations (dashed lines).

In this study, we have demonstrated that the Lake-Thomas model underestimates the intrinsic fracture energy of polymer-like networks by more than an order of magnitude, even if the networks are purely elastic with lattice-like topology. Our experimental and numerical analyses reveal that this discrepancy arises from significant energy release and dissipation far from the crack tip, rather than solely in the bridging chain. We show that this nonlocal intrinsic fracture energy is strongly connected to the high nonlinearity of the force-extension relation and the abrupt breaking of polymer chains. Our findings provide implications for the mechanics of real polymer networks and designing metamaterials. For real polymer networks, our results indicate that the released or dissipated energy per broken chain is inherently larger than the work to rupture a single chain. For metamaterials, results indicate that high toughness and resilience can be obtained by controlling the nonlinear mechanics of the constituent architected chains. Future directions could involve exploring the relationship between the network structure and the intrinsic fracture energy, as well as investigating the effect of defects and inhomogeneities in the network on its mechanical properties. Moreover, our findings could be extended to other materials, such as ultra-tough structures, soft matter and biomaterials, with implications for a wide range of applications, including aircraft, space vehicles, and tissue engineering.

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