Toughness of Double Network Hydrogels: The Role of Reduced Stress Propagation

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Double network hydrogels show remarkable mechanical performance, combining high strength and fracture toughness with sufficient stiffness to bear load, despite containing only a low density of cross-linked polymer molecules in water. We introduce a simple mesoscale model of a double network material, detailed enough to resolve the salient microphysics of local plastic bond breakage, yet simple enough to address macroscopic cracking. Load sharing between the networks results in a delocalization of stress such that the double network inherits both the stiffness of its stiff-and-brittle sacrificial network and the ductility of its soft-and-ductile matrix network. The underlying mechanism is a reduction in the Eshelby stress propagator between sacrificial bonds, inhibiting the tendency for the plastic failure of one sacrificial bond to propagate stress to neighboring sacrificial bonds and cause a follow-on cascade of breakages. The mechanism of brittle macroscopic cracking is thereby suppressed, giving instead ductile deformation via diffusely distributed microcracking.

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Soft materials are central to numerous important technologies, including biomedical tissue engineering [1], stretchable electronics [2], and soft robotics [3]. Such applications typically demand large material deformations, making stretchable polymer networks such as hydrogels and elastomers promising candidates. However, these materials often also exhibit brittle failure via crack propagation under only modest loads, limiting their use in practice. A key technological challenge is to design soft materials that combine high strength and fracture toughness with sufficient mechanical stiffness to bear load.

This combination can be achieved in composite materials based on a *double* network structure, including double network hydrogels [4], elastomers [5,6], and macroscopic composites [7]. In pioneering work, Gong *et al.* [8] combined a stiff and brittle "sacrificial" network in low molar ratio with a soft and ductile "matrix" network. The resulting double network hydrogel was both stiff and ductile. Indeed, such hydrogels can achieve remarkable mechanical performance, combining hardness (elastic modulus 0.1–1.0 MPa), strength (failure stress 1–10 MPa, failure strain 10–20), and toughness (tearing energy 100–1000 Jm^{-2}), independent of deformation rate, and despite comprising up to 90% water [4,9,10]. This makes them promising candidates for artificial tendons, ligaments, and cartilage [11]. Indeed, many biological networks are themselves multicomponent, including the cell cytoskeleton [12] and extracellular matrix [13]. Biomimetic composites have also been shown to display increased stiffness and strength *in vitro* [14,15].

The remarkable toughness of double network materials remains poorly understood theoretically. Lacking in particular is a physical understanding of how a double network structure inhibits the propagation of a macroscopic crack of the kind that would cause catastrophic brittle failure in a single network. Molecular simulations provide important insights at the microscopic level [16–20], but struggle to access the system sizes needed to address macroscopic crack propagation (or inhibition). Continuum models naturally address macroscopic scales [21–28], but do not capture the detailed underlying microphysics.

Recent experiments have provided valuable insights. Probing the length scale of internal nonaffine deformations by small angle neutron scattering suggests that stress is delocalized in a double network [29]. Birefringent imaging shows that nonoptimized double networks have localized stresses whereas optimized ones have more uniformly distributed stress [8]. Dynamic light scattering shows the fluctuations of the two networks to be strongly coupled, indicating a mutual entanglement and suggesting that they can share load [30]. A higher molar ratio of matrix network predisposes the creation of multiple microcracks (ductile behavior) instead of the propagation of a single macroscopic crack (brittle failure) [31]. Multinetwork elastomers have wide-spread molecular damage and delocalized strain concentration near a crack, compared with single network materials [32].

Here we introduce a simple mesoscale model of a double network material, detailed enough to resolve the salient microphysics, yet simple enough to address macroscopic

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cracking. To understand its predictions, which capture all the experimental observations described above, we build on recent progress elucidating yielding in amorphous materials [33–36], in which a plastic relaxation event localized in one part of a sample causes an elastic Eshelby stress propagation to nearby regions [37], triggering a follow-on cascade of plastic events and macroscopic cracking. We show how that physics is modified in a double network, with the effective Eshelby propagator between the weak but stiff sacrificial bonds reduced by load-sharing with the matrix network, for a sufficiently high fraction of matrix. The plastic failure of a sacrificial bond then no longer causes nearby sacrificial bonds to break. This inhibits stress localization and macroscopic cracking, favoring instead ductile deformation via the formation of multiple diffusely distributed microcracks. The double network then inherits both the stiffness of its stiff but weak sacrificial component and the increased failure strain of its soft but tough matrix.

Model—A random double network is prepared by adapting a method commonly used to prepare single networks [38]. First, we randomly initialize an ensemble of N_S disks of bidisperse radii R_s and $1.4R_s$ in equal numbers at area fraction $\phi = 1.0$ in a square box, with periodic boundary conditions. With a soft Hookean repulsion between any pair of overlapping disks, the packing is evolved to equilibrium. Each overlapping pair then has their centers connected by a spring of stiffness μ_s and breakage strain λ_s, with length l initialized to its equilibrium length l_0. The disks are then removed to leave a network of springs. This forms our sacrificial network, Fig. 1(a). The matrix network is created likewise, but now starting from N_M disks with radii R_m and $1.4R_m$, and insisting that one disk of this new ensemble is pinioned to each node of the sacrificial network during equilibration, so as to become a node common to both networks once the disks are then replaced with matrix network springs of stiffness μ_m and breakage strain λ_m. We take $M \equiv R_s/R_m > 1$, so that the sacrificial network (red) is sparse compared with the matrix (blue). This leaves finally a double network in which each sacrificial node overlaps with a matrix node, Fig. 1(b). Their respective network coordinations are $z_s = 5.42$ (sacrificial) and $z_m = 5.33$ (matrix). The nodes common to both networks may represent bonds or entanglement points between them. Either results in load sharing, as seen experimentally [39].

The double network is then stretched along the y direction at constant area L_xL_y with biperiodic boundary conditions, starting from a square of side $L_{x0} = L_{y0}$. See Figs. 1(c) and 1(d). The extensional strain $\epsilon \equiv L_y/L_{y0} - 1$. Any bond breaks immediately once its own strain $l/l_0 - 1$ exceeds a breakage threshold, λ_s or λ_m for sacrificial or matrix bonds, respectively. The stretching proceeds in strain steps of size $\Delta\epsilon = 10^{-3}$ by default, unless more than one spring would immediately break after such a step, in which case we iterate $\Delta\epsilon$ to find its value, $\pm 10^{-8}$, that ensures just one bond is (initially) unstable to breakage. We

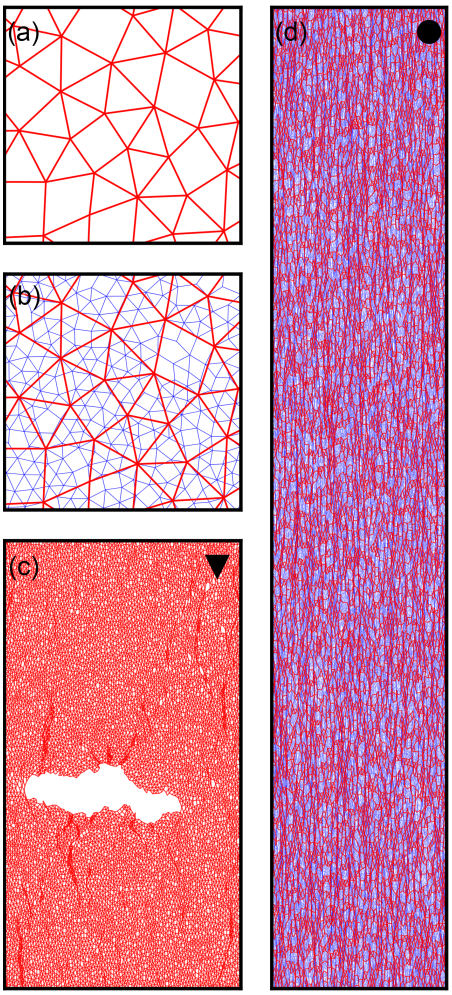


FIG. 1. Portion of an (a) single and (b) double network, with sacrificial bonds in red and matrix bonds blue. (c) Stretched single network at a location on the stress-strain curve shown by the triangle in Fig. 2(a). A macroscopic crack has propagated across the network, causing catastrophic material failure. (d) Stretched double network at a location on the stress-strain curve shown by the circle in Fig. 2(b). Damage has arisen diffusely via many microcracks in the sacrificial network, which do not propagate macroscopically. This allows the double network to stretch much further than the single network without failing.

then allow any ensuing cascade of bond breakages to run to completion, and for the system to equilibrate after it, with rms node force $< 10^{-6}$, before moving to the next step. We thereby implement quasistatic stretching. The component $\Sigma_{yy} \equiv \Sigma$ of the network's stress response is calculated using the Irving-Kirkwood formula [40,41].

We choose our stress unit $\mu_s = 1.0$, and our length unit as the typical sacrificial bond length, by setting $L_{x0} = L_{y0} = \sqrt{N_S}$. We set our strain scale $\lambda_s = 0.5$ and define the typical number of matrix bond lengths per sacrificial bond length $M = R_s/R_m$. Larger M corresponds to a sparser sacrificial network, with more matrix bonds in between. A single sacrificial network is defined to have $M = 0$. Consistent with the experimental literature, we

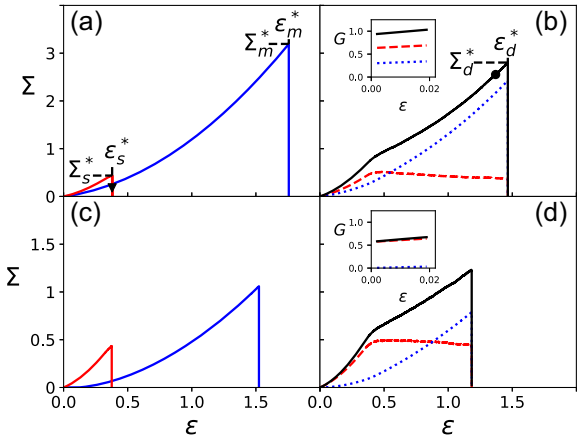


FIG. 2. Stress-strain curves. (a) Single sacrificial network (red solid line) and single matrix network (blue solid line). (b) Composite double network (black solid line), with the contributions of the component sacrificial network (red dashed line) and matrix network (blue dotted line) shown separately. Inset: modulus $G = d\Sigma/d\epsilon$ at small strain. Triangle and circle show the location of the snapshots in Fig. 1. Parameters: $M = 3.0$, $\mu_m = 0.2$, $\lambda_m = 3.0$. (c) and (d) show the effect of reducing the matrix network connectivity to $z_m = 3.5$, as discussed in the penultimate paragraph of the main text.

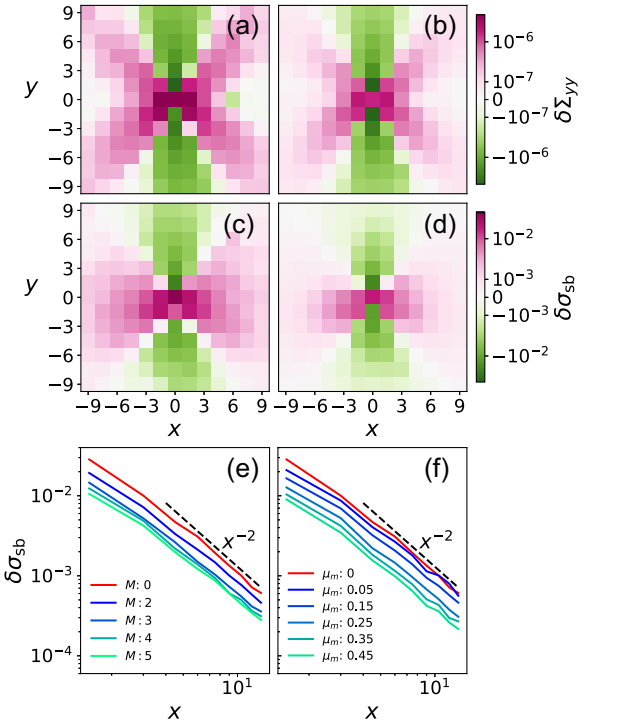


FIG. 3. Stress propagation to neighboring sacrificial bonds from the breakage of a sacrificial bond at the origin: (a),(c) in a single sacrificial network and (b),(d) in a double network with $M = 5.0$, $\mu_m = 0.2$. Color scales show the change due to stress propagation of (a),(b) the yy component of the Kirkwood stress $\delta\Sigma_{yy}$; and (c),(d) the sacrificial bond stress $\delta\sigma_{sb}$, with $\sigma_{sb} \equiv \mu_s(l - l_0)/l_0$. Each quantity is averaged over all bonds at each grid square. (e),(f) Show the change in bond stress as a function of distance along x at $y = 0$ for (e) several M at fixed $\mu_m = 0.2$, and (f) several μ_m at fixed $M = 3.0$. Lengths are expressed in units such that $\Delta x = 1$ corresponds to the typical sacrificial bond length. The propagator of a single sacrificial network is recovered for $M = 0$ or $\mu_m = 0$.

explore parameters for which, in comparison with the matrix, the sacrificial network is sparse ($M > 1.0$), stiff ($\mu_m < 1.0$), and brittle ($\lambda_m > 0.5$). We take our system size $N_M = 320^2$, which also specifies N_S via M.

Fracturing has been studied previously in network models of single component materials [42]. A double network model was studied in Ref. [43], but initializing both a sacrificial bond and a matrix bond between each pair of neighboring nodes on a triangular lattice. Accordingly, it did not capture the reduction in Eshelby stress propagation as a function of distance between sparse sacrificial bonds with increasing M, which is the key physics reported in what follows. Fracture in a double network model with a sparse concentration of unbreakable springs, opposite to the case in real double network hydrogels, has also been studied [44].

Results—Our key finding is summarized as follows. A single network fails at only modest strain via spatially localized macroscopic cracking, Fig. 1(c). In contrast, a double network can be stretched to much larger strain without globally failing. Instead, sacrificial bonds break in numerous microcracks that arise diffusely throughout the network, Fig. 1(d). This distinction is reflected in the corresponding stress-strain curves: Fig. 2(a) for the single network shows a sharp stress drop at only modest overall strain (red curve), whereas Fig. 2(d) for the double network shows greatly increased toughness (black curve).

This can be understood as follows. In a single network stretched along y, the stress lost locally when one bond breaks is propagated to neighboring bonds along x, putting them under increased stress, Figs. 3(a) and 3(c). These neighboring bonds then also break, propagating stress to their next neighbors in turn. The resulting knock-on cascade causes a crack to spread macroscopically along x [Fig. 1(c)]. In contrast, when a sacrificial bond breaks in a double network, its stress is partly absorbed by load sharing with nearby matrix bonds, which are tough enough to resist failure. Stress propagation to neighboring sacrificial bonds is thereby reduced, Figs. 3(b) and 3(d). In a well designed network, with model parameter values optimized as described below, this reduction is sufficient to avoid those neighbors then also breaking, eliminating the knock-on cascade of sacrificial bond breakage seen during macroscopic cracking in a single network.

We now explore this more quantitatively. As discussed above, a well designed double network has two key properties. (i) It inherits the stiffness of its stiff and brittle sacrificial component, with modulus $G_D \approx G_S \gg G_M$. [G_S, G_M, G_D are defined via the initial stress-strain slopes in Fig. 2(b)]. (ii) It inherits the ductility of its soft and ductile matrix component, with a failure strain $\epsilon^*_D \approx \epsilon^*_M \gg \epsilon^*_S$. [Failure

strains and stresses are also defined in Figs. 2(a) and 2(b).] Our basic hypothesis is that (i) and (ii) stem from the presence of sufficient matrix to reduce the stress propagated between neighbouring sacrificial bonds; but sufficient sacrifical network to confer high stiffness. We now explore parameter values M, μ_m, and λ_m that achieve both (i) and (ii).

Figure 3 shows that stress propagation between the stiff but breakable sacrificial network bonds indeed decreases in a double network as the number of soft but strong matrix bonds between them, set by M, increases at fixed matrix bond stiffness μ_m; or as μ_m increases at fixed M. This is to be expected: part of the stress is instead absorbed by the matrix, to an extent that increases with increasing amount of matrix M or increasing matrix stiffness μ_m. (Conversely, for $M = 0$ or $\mu_m = 0$ the level of propagation between neighboring sacrificial bonds reverts to that in a single network.) These soft matrix bonds can stretch significantly without breaking. In this way, the breakage of a single sacrificial bond at an origin site has a reduced tendency to cause knock-on breakages in neighboring sacrificial bonds.

As a result of this reduced stress propagation, we expect a double network to be tougher than a single network, with a failure strain ϵ_D^* and stress Σ_D^* that increases with M or μ_m. This is confirmed in Figs. 4(a)–4(d). Indeed, as $M \to \infty$ or $\mu_m \to \infty$, a double network fully inherits the toughness of the matrix, with failure strain $\epsilon_D^* \to \epsilon_M^*$, achieving property (ii) above. This limit is however trivial: the double network effectively just *becomes* a single matrix network in that limit, inheriting also its softness, so failing to achieve (i). Figures 4(e) and 4(f) confirm this: the sacrificial network contributes progressively less to the modulus G_D of the double network as M or μ_m increases.

Designing a double network that inherits both the stiffness of its stiff but brittle sacrificial component and the toughness of its soft but ductile matrix component therefore requires a trade-off. In practice, we have found the region of parameter space exemplified by the values $M = 3.0$, $\mu_m = 0.2$, $\lambda_m = 3.0$ in Figs. 1 and 2 to be a sensible compromise. The single sacrificial network is stiffer than the single matrix network, $G_S > G_M$, but also more brittle, $\epsilon_S^* < \epsilon_M^*$ [Fig. 2(a)]. Combining them then gives a double network with a stiffness comparable to that of the sacrificial component and significantly greater than that of the matrix, $G_D = 1.47G_S = 3.11G_M$ and a toughness comparable to that of the matrix component, $\epsilon_D^* \approx \epsilon_M^* \gg \epsilon_S^*$, Fig. 2(b). Its failure stress Σ_D^* greatly exceeds that of the sacrificial network Σ_S^* and is of the order of that of the matrix, Σ_M^*, consistent with a fraction $f = 0.41$ of sacrificial bonds having broken diffusely across the material by the time the double network fails, with the sacrificial network no longer percolating.

The model studied here is clearly oversimplified with respect to real double network hydrogels. Indeed, we have taken a $d = 2$ dimensional approach and considered bonds with only central forces, without any bending penalty or

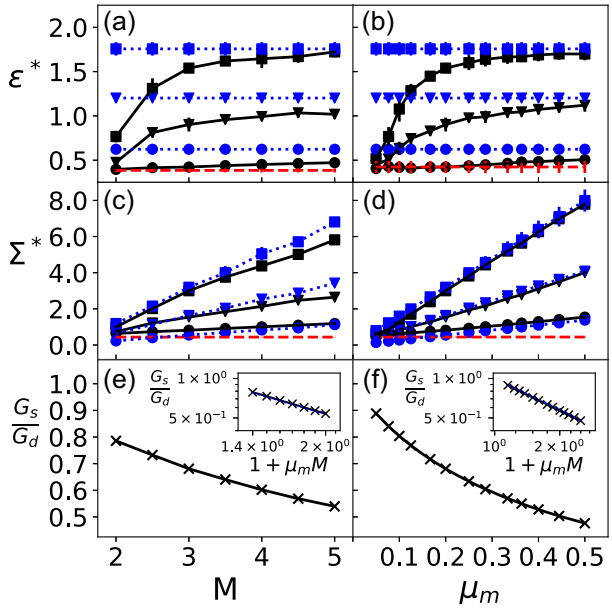


FIG. 4. Top: failure strain of double network (black solid lines) as a function of (a) the amount of matrix M and (b) matrix bond stiffness μ_m, shown in each case for matrix bond breakage thresholds $\lambda_m = 1.0$, 2.0, 3.0 in curves bottom to top, marked by circles, triangles, and squares, respectively. Failure strains of the corresponding single sacrificial network and single matrix network are shown by red dashed and blue dotted lines respectively. Middle: as in (a) and (b) but now showing failure stresses. Bottom: modulus of the sacrificial network relative to that of the double network. Matrix bond stiffness $\mu_m = 0.2$ in (a),(c),(e). Amount of matrix $M = 3.0$ in (b),(d),(f).

prestretch, in networks with average coordination $z > 2d = 4$ to ensure rigidity according to Maxwell's criterion. Real gels inhabit $d = 3$ dimensions and typically have subcritical coordination numbers $z < 2d = 6$, with a higher density of cross-linkers in the stiff, strongly cross-linked sacrificial network compared with the soft, weakly cross-linked matrix network. Furthermore the sacrificial network, which is synthesized first, is naturally prestretched by the synthesis of the second matrix within it. We have instead used a single stiffness parameter μ for each network, with $\mu_s > \mu_m$, as a proxy for the greater stiffness of the sacrificial network due to its prestretch and denser cross-linking. This simplified approach has enabled us to identify the key physics at play with the most minimal ingredients possible.

A first step to exploring these more detailed features is taken in Figs. 2(c) and 2(d). Here the matrix network's initial connectivity is reduced to $z_m = 3.5$, with other parameters as before. The double network is still much tougher, with a greatly increased failure strain compared with the single sacrificial network. However, its stiffness (modulus G) is now overwhelmingly dominated by that of the sacrificial matrix, as seen experimentally, due to the now subcritical connectivity of the matrix $z_m < 4.0$.

In summary, we have proposed a simple model of double network hydrogels, and shown by numerical simulation

that it captures the key features observed experimentally in these materials. Load sharing between the two networks leads to a delocalization of stress, such that the double network inherits both the stiffness of its stiff and brittle sacrificial component and the ductility of its soft and ductile matrix component. The underlying mechanism is a reduction in the Eshelby stress propagator between neighboring sacrificial bonds. This inhibits the tendency for the localized plastic failure of a sacrificial bond to propagate stress to neighboring sacrificial bonds and cause a follow-on cascade of breakages. The mechanism of brittle macroscopic crack formation is thereby suppressed, favoring instead the formation of multiple diffusely distributed microcracks. The trade-off between stiffness and ductility as a function of model parameter values suggests pointers to ongoing material design, both in the protocol explored here and in others such as cyclic shear [45–47].

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Data availability—The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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End Matter

In the main text, we performed simulations using an algorithm that implements the limit of quasistatic shear. Finally, we show in Fig. 5 that the introduction of a finite shear rate does not qualitatively change our key finding of a greatly improved toughness in a material with a double network structure.

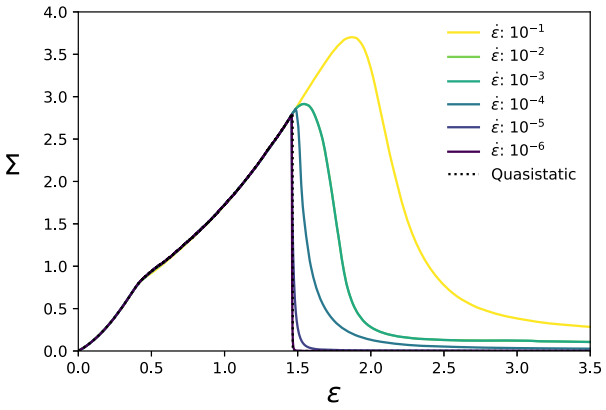


FIG. 5. Stress-strain curves for a double network stretched at a finite rate $\dot{\epsilon} > 0$, showing also the approach to the quasistatic limit $\dot{\epsilon} \to 0$ explored in the main text. Parameters otherwise as in Fig. 2(a).