

Supplementary material for “How super-tough gels break”

Itamar Kolvin^{1,2}, John M. Kolinski^{1,3}, Jian Ping Gong⁴ and Jay Fineberg¹

¹*The Racah Institute of Physics, The Hebrew University of Jerusalem, Jerusalem, 91904, Israel*

²*UC Santa Barbara, Santa Barbara, California, 93106, USA*

³*École Polytechnique Fédérale de Lausanne, Lausanne, 1015, Switzerland*

⁴*Faculty of Advanced Life Science and Soft Matter GI-CoRE, Hokkaido University, Sapporo, 001-0021, Japan*

NUMERICAL VALUES OF THE DATA PRESENTED IN FIG. 5

L [mm]	ℓ [mm]	λ_y	$U_{el}L$ [kJ/m ²]	v [m/s]
63.13	22.12	1.98	7.09	8.87
69.01	35.14	2.17	8.9	9.64
63.5	16.98	1.75	5.81	7.45
61.44	11.35	1.69	4.28	5.31
23.54	5.88	1.66	1.89	1.63
23.36	5.54	1.69	2.08	1.8
25.01	11.39	1.99	3.2	6.03
30.53	12.12	1.94	3.12	5.87
28.05	14.92	2.10	3.78	7.03

Table I. The data used in Fig. 5

DN GEL SMALL STRAIN RESPONSE

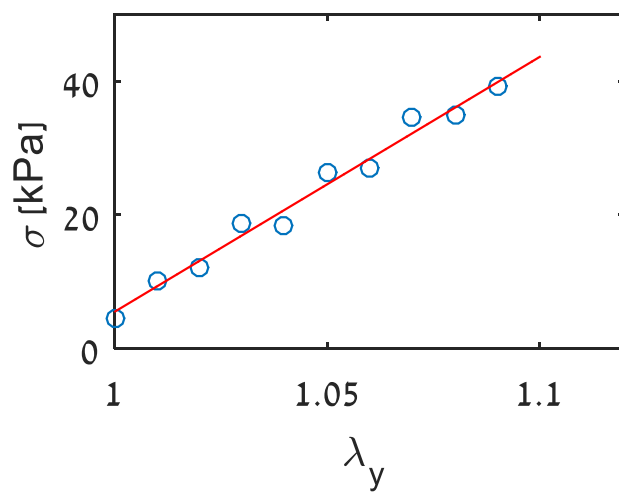


Figure S1. Stress-strain curve at small strains taken by averaging the curves of Fig. 1(b). The Young modulus $E = 380 \pm 40$ kPa is obtained via linear regression (red line). We estimate the shear modulus to be $\mu = E/3 = 130 \pm 10$ kPa.

RESIDUAL STRAINS BEHIND A PROPAGATING CRACK

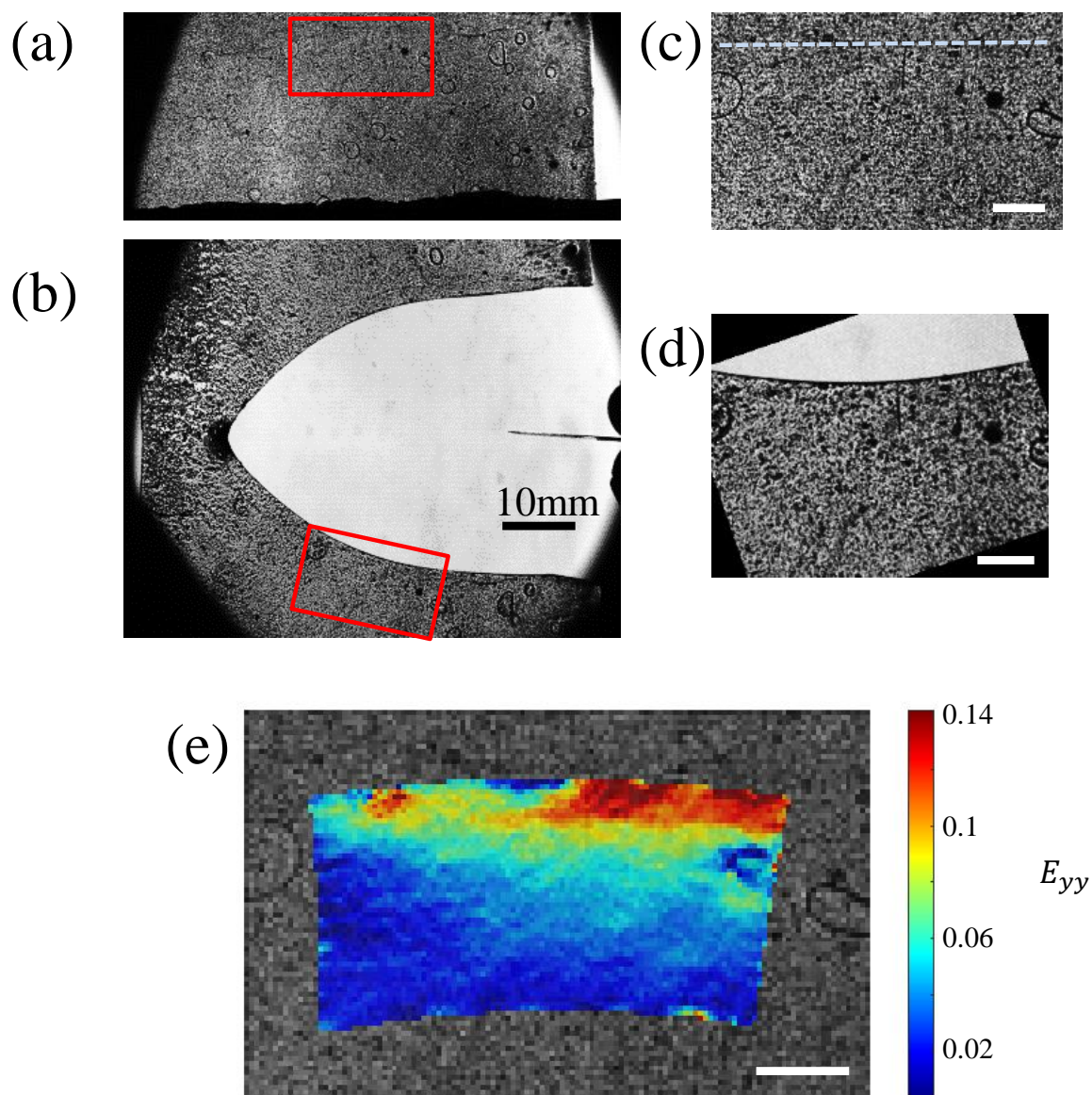


Figure S2. Residual strains within the tail of a running crack. (a) The gel sheet at its rest state. (b) A snapshot of the running crack under an imposed stretch of $\lambda_y = 1.64$. (c,d) Zoomed sections highlighted in the red boxes in (a,b). The dashed line in (c) marks the approximate crack path in the non-stretched frame. (e) The residual E_{yy} strain within the tail of the crack extracted through DIC of (c) and (d). White scale bars are 3 mm long.

CTOD SCALING AT LARGE STRAINS

In the main text we have observed that dynamic cracks in double-network gels have a CTOD of the shape $x \sim |y|^b$, where $b = 1.64$. Here we give a heuristic argument for how non-parabolic CTOD's may arise due to large strain elasticity.

The elastic response of an isotropic material to deformation is derived from a strain energy density e , which depends on rotation invariants of the strain tensor. In 3D, there are three invariants [1]. Denoting the displacement of a point in the reference frame by u_i , and the strain tensor by $E_{ij} = \frac{1}{2}(\partial_j u_i + \partial_i u_j + \partial_i u_k \partial_j u_k)$, the first invariant is $I = E_{ii}$. Here i and j run over the three spatial coordinates (X, Y, Z) in the material rest frame, and we use the Einstein summation convention. We will assume that the contribution of the second invariant to e is negligible, since it is second order in E_{ij} . The third invariant, which is related to volume changes, is a constant for incompressible materials (a good approximation for gels). We therefore remain with $e = e(I)$. To obtain the CTOD scaling we note that one may construct a path independent integral around the crack tip, namely the J-integral [2, 3]

$$J = \int_{\Gamma} (e(I)dy - S_{ij}n_j \partial_X u_i ds) \quad (S1)$$

where Γ is a contour surrounding a crack that lies along the negative X axis, S_{ij} is the nominal stress tensor, n_j is the outward normal to the contour and ds is the arc length element. The path-independence of J amounts to the existence of a $1/r$ singularity of the integrand as the radius $r = \sqrt{X^2 + Y^2}$ approaches 0. Since both terms in the integrand must scale in the same way ($S_{ij} \sim \partial e / \partial(\partial_j u_i)$), the energy density scales as $e(I) \sim 1/r$.

For simplicity, let's assume the strain energy has a power-law dependence $e(I) \sim I^n$. For incompressible materials $\det(\delta_{ij} + \partial_i u_j) = 1$, which translates to $\partial_i u_i + \frac{1}{2}(\delta_{ij}\delta_{kl}\partial_i u_j \partial_k u_l - \delta_{ij}\delta_{kl}\partial_i u_k \partial_j u_l) + \det \partial_i u_j = 0$. The strain invariant is therefore $I = \partial_i u_i + (\partial_i u_k)^2 = (\partial_i u_k)^2 - \frac{1}{2}(\delta_{ij}\delta_{kl}\partial_i u_j \partial_k u_l - \delta_{ij}\delta_{kl}\partial_i u_k \partial_j u_l) - \det \partial_i u_j$. The first term in I is quadratic in the displacement gradient, and all others involve double and triple cross-products of displacement gradient components. Close to the crack faces, $\partial_X u_Y$ becomes the most dominant component, and thus $I \sim (\partial_X u_Y)^2$. Then,

$$(\partial_X u_Y)^{2n} \sim \frac{1}{r} \quad (S2)$$

And hence $u_Y \sim (-X)^{1-1/2n}$ or $x \sim y^{2n/(2n-1)}$ where x and y are a lab frame coordinate system with origin at the crack tip, and $y = u_Y(X, Y = 0, Z)$. That is,

$$b = \frac{2n}{2n-1}. \quad (S3)$$

This expression for the CTOD power-law recovers the parabolic shape $b = 2$ for the Neo-Hookean case $n = 1$ [4] and approaches $b = 1$ for large values of n . Our observed value of $b = 1.64$ would arise in this model for $n = 1.28$. Long, Krishnan and Hui [5] solved analytically the fracture problem in the context of the Generalized Neo-Hookean material model. This model is defined as $e(I) \sim \epsilon^{-1}\{(1 + \epsilon n^{-1}(I - 3))^n - 1\}$, where ϵ and n are parameters.

This model approaches the Neo-Hookean behavior for small strains or small ϵ . Their solution leads to a CTOD with $b = 2n/(2n - 1)$, the same as derived by the scaling argument above.

Both the power-law model $e(I) \sim I^n$ and the Generalized Neo-Hookean model are, to an extent, toy models for nonlinear elasticity. To test whether the CTOD scaling changes in more realistic models, we solved for the displacement fields surrounding a crack in an Arruda-Boyce material model using finite-elements software (Abaqus). Briefly, the Arruda-Boyce model is defined by $e(I) = \mu\lambda_m^2 f(\sqrt{I/3}/\lambda_m)$ where μ is an elastic modulus and the function $f \rightarrow \infty$ when $I \rightarrow 3\lambda_m^2$ (see [6] for the definition of $f(\cdot)$). The latter property makes λ_m the maximal stretch that the material can sustain.

We solved for the displacement fields surrounding a crack of length 35mm in a rectangular sample of $L = 65$ mm and $W = 70$ mm. We assumed that $\mu = 100$ kPa, and applied a constant displacement of 21.1mm to the y edge of the sample. Taking advantage of the symmetry of the problem, we solved it only in the upper half of the sample, assuming zero displacement along the x axis, ahead of the crack tip. The solution used an adaptive triangular mesh of ~ 800 nodes.

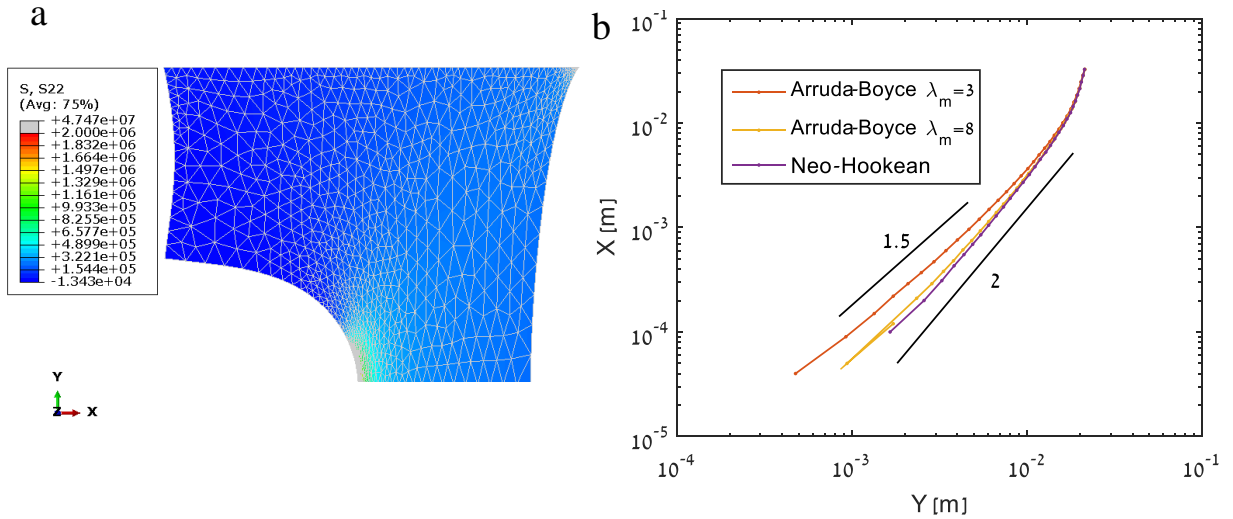


Figure S3. (a) The S_{yy} stress component surrounding a crack in an Arruda-Boyce material. Only half of the sample is shown. (b) CTOD profiles for Neo-Hookean and Arruda-Boyce materials.

Fig. S3 shows an example of a solution and the CTOD profiles extracted from solutions obtained using three different materials. The Neo-Hookean model results in a parabolic CTOD, as predicted by the scaling argument. An Arruda-Boyce model with $\lambda_m = 8$ shows a very small deviation from a parabola. Decreasing λ_m to 3 results in a decrease of the CTOD power-law exponent to ~ 1.5 .

In all of the models considered here, the exponent b depends directly on the steepness of the stress-strain curve (n in the power-law and Generalized Neo-Hookean models and $1/\lambda_m$ in the Arruda-Boyce model). We anticipate that a

future model that takes into account the anisotropy of our gels as well as nonlinear elasticity will be able to explain our observations.

-
- [1] M. Marder, *J. Mech. Phys. Solids* **54**, 491 (2006).
 - [2] J. Rice and G. F. Rosengren, *J. Mech. Phys. Solids* **16**, 1 (1968).
 - [3] J. W. Hutchinson, *J. Mech. Phys. Solids* **16**, 13 (1968).
 - [4] T. Goldman-Boué, R. Harpaz, J. Fineberg, and E. Bouchbinder, *Soft Matter* **11**, 3812 (2015).
 - [5] R. Long, V. R. Krishnan, and C. Y. Hui, *J. Mech. Phys. Solids* **59**, 672 (2011).
 - [6] M. C. Boyce and E. M. Arruda, *Rubber chemistry and technology* **73**, 504 (2000).