

Fracture of soft cellular solids —Case of non-crosslinked polyethylene foam

Y. SHIINA, Y. HAMAMOTO and K. OKUMURA(*)

*Department of Physics, Graduate School of Humanities and Sciences
Ochanomizu University - 2-1-1, Otsuka, Bunkyo-ku, Tokyo 112-8610, Japan*

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Abstract. – Fracture mechanical properties of hard cellular solids (porous materials) with the Young modulus E larger than 3000 MPa have been well understood; scaling relations between fracture mechanical quantities and porosity are established, which can be explained by a theory based on the geometrical parameters of a cellular solid. In this study, we obtain experimentally the fracture energy of a very soft polyethylene foam with E around 1 MPa. We find scaling laws different from those for hard foams, which can be understood by considerations independent of the structural parameters of foams.

Introduction. – Cellular solid structures are widely found in nature (cork, balsa, coral, etc.) and in everyday life (bread, potato chips, industrial materials such as styrene foam, and so on). Accordingly, the mechanical properties of cellular solids or porous materials have been widely studied. It is now well accepted that the mechanical properties of cellular solids can be understood as a function of the volume fraction of the solid (*i.e.* one minus porosity) [1,2].

For hard cellular solids with the Young modulus E larger than 3000 MPa (*e.g.*, PMA, rigid PU, cellular glass), fracture mechanical properties (such as the critical energy release rate G_c and the critical stress intensity factor K_c) have been well studied; scaling relations between fracture mechanical quantities and porosity are established [1,3]. These relations can be understood by a theory based on the geometrical parameters of cellular solids, such as cell size or cell wall thickness, etc. Here, we examine a very soft polyethylene foam with E around 1 MPa to show differences in fracture mechanical properties from hard porous materials.

Structure of the foam. – The materials we examined are sheets of non-crosslinked polyethylene foam, Lightron S (Sekisuiplastics), commercially available as shock-absorbing sheets for packing. Macroscopically, they are milk-white soft sheets with millimetric thickness, suitable for wrapping fragile things such as porcelain. Microscopically, they possess a typical closed-cell foam structure: submillimetric cells are enclosed by very thin films (see fig. 1).

(*) E-mail: okumura@phys.ocha.ac.jp (corresponding author)

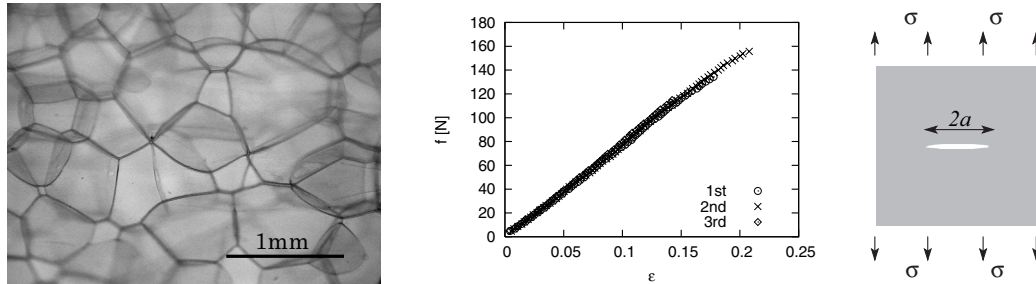


Fig. 1 – Microscopic view on Lightron S (#54), taken by a (video) camera (HC-1, Sony) with a macro lense (MSN-505, raynox) on a stand (VM-3000, raynox).

Fig. 2 – Force-strain relation of polyethylene foam, #54A. All the samples show linear elasticity up to breakage as shown here.

Fig. 3 – Griffith's problem: the scaling law in eq. (1) holds when the sample size is large compared with the half crack length a . The stress σ indicates a remote tensile stress.

In many cases, it has been assumed that the *edges of a cell* (*i.e.* the places where the film walls meet) collect a lot of liquid before solidification; as a result, the volume of the edge is often assumed to be larger than that of the walls (see, for example, fig. 5.12 of [1]). However, in our foam, the edge volume is less than or comparable (at most) to the wall volume.

Fracture measurement system. – Our fracture measurement system can hold a sheet by clamping it between two aluminum plates of 1 m long. A sample is homogeneously stretched in the direction perpendicular to the long plates through a wire connected to a driving unit. The stretching speed is set to a fixed low rate (~ 1 cm/min) via a stepping motor throughout all measurements to reduce dynamical effects. The stretching force is monitored by a digital force gauge (FCC-50B, NIDEC-SHIMPO CORP). This system has a merit over commercial universal testing machines: it allows us to determine the critical energy release rate or the fracture surface energy, directly (not via the conventional K_c test) as explained below.

Linear elasticity of the foam. – Lightron S offers a selection of thickness: we tested #41 (0.5 mm thickness), #52 (1 mm) and #54 (2 mm). Samples labeled A in table I are fabricated in winter 2005, while B are in summer 2006. Foam sheets are stretched when processed by a manufacturing machine; as a result, when not stretched, sheets are slightly wavy in the direction perpendicular to the machine direction. Experimental tensile force is always applied in the direction perpendicular to the machine direction.

All the samples show a linear-elastic stress-strain relation up to breakage. This was confirmed by setting a $50\text{ cm} \times 50\text{ cm}$ sheet (without any artificial cracks) to the measurement system and stretching it until it fails (from the clamped region), with monitoring elongation of the sheet and applied force. We repeated this three times for a given thickness of a sheet (a broken sheet is replaced by a new one for the next measurement). A representative result (#54A) is shown in fig. 2; a $50\text{ cm} \times 50\text{ cm}$ sheet typically breaks at $f \simeq 100\text{ N}$ (0.1 MPa in stress) when no artificial cracks are introduced and it behaves as a linear-elastic material up to failure. We obtained similar results for the other samples. The Young moduli obtained from these measurements are summarized in table I.

TABLE I – The Young modulus E , fracture toughness G_c , critical stress intensity factor K_c , and relative density ϕ of the sample sheets with different thickness t .

ID	t (mm)	ϕ	E (MPa)	G_c (N/m)	K_c (MN/m ^{3/2})
#41A	0.5	0.0414	1.357	452	0.025
#52A	1	0.0255	0.7495	268	0.014
#54A	2	0.0241	0.7861	326	0.016
#41B	0.5	0.0465	1.238	450	0.024
#52B	1	0.0284	0.7261	258	0.014
#54B	2	0.0248	0.7710	240	0.014

Griffith's scaling relation for foam sheets. – When a remote stress σ is applied, an elastic material without cracks accumulates an elastic energy of the order of σ^2/E per unit volume, where E is the Young modulus. When there is a crack (length $\sim a$) as in the Griffith problem illustrated in fig. 3, the uniformly distributed elastic energy is relaxed due to the crack; the amount of the released energy can be estimated by the energy which would have been stored around the crack in the volume of the order of a^2 (the plate thickness we consider here is the unit length), *i.e.*, $\sigma^2 a^2/E$. Griffith assumed that this energy is transformed into the work for the creation of a new surface, of the order of γa ; γ is the work of fracture per unit area. Balancing these two energies immediately leads to Griffith's scaling relation [4]:

$$\sigma_f = \left(\frac{2\gamma E}{\pi a} \right)^{1/2} \propto a^{-1/2}, \quad (1)$$

where a is the half crack length (the above numerical coefficient is exact under the plane-stress condition).

Originally, Griffith considered brittle materials such as glass, where the fracture surface energy corresponds predominantly to cutting of chemical bonds. This concept was extended by Irwin [5] and Orowan [6] to metals and then further to other fractures where plastic work is required in addition to bond rupturing energy to create a new surface (as shown below, *the fracture of our samples is dominated by the plastic work*). This extension can be justified (for materials with global linear elasticity) in the limit of *small-scale zone*: the zone where plastic dissipation occurs is limited around the tip region of the crack, compared with the crack size.

In our experiment, even the smallest crack size is large (5 mm) and thus the Irwin-Orowan extension is expected to work well because of *the global linear elasticity* suggested in fig. 2. We checked this expectation by the following experiments.

We introduced a short line crack in the machine direction by a sharp knife at the center of each 50 cm \times 50 cm sheet and performed a uniaxial tensile test with our measurement system until the sheet starts failing (fig. 3). We determine a failure stress from a force read from the gauge when the edge of a crack starts expanding. We repeat this three times for a given crack size (a broken sheet is replaced by a new one for the next measurement). This set of measurements are done for all the samples.

As a result, it is shown that Griffith's scaling law holds for crack sizes changed well over one order of magnitude (see fig. 4). This suggests that our experiments are in the regime of the small-scale zone. This is because 1) the fracture is governed by plasticity (as mentioned above) and 2) our material is certainly tested within the globally linear-elastic regime implied in fig. 2: when we introduce an artificial crack, the sheet always starts failing (at the edge of the crack) at a stress lower than the failure stress observed in the Young modulus experiment (done without any artificial cracks).

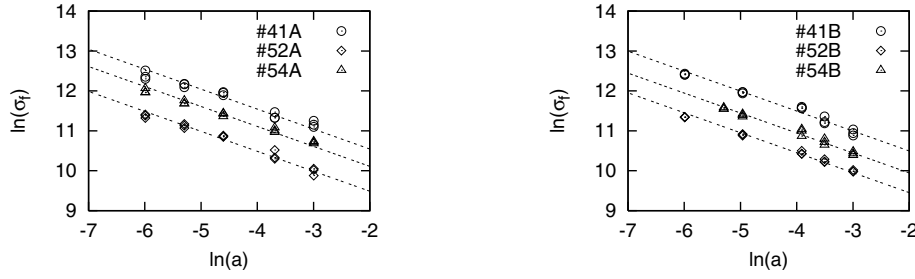


Fig. 4 – Griffith's scaling law for foam sheets. The horizontal and vertical axes are the logarithms of the half crack size (m) and the failure stress (Pa), respectively. The dotted lines, with the theoretical slope, $-1/2$, predicted by Griffith, are those fitting each data set. The data of #52 are shifted by the amount 0.5 downwards to avoid overlaps with those of #54.

We tested five sizes of cracks ($2a$) as indicated in fig. 4. The minimum size (5 mm) is limited to keep a good enough precision of the size of a crack. The maximum (10 cm) is restricted due to finite-size effect; a crack size has to be small compared with the sheet dimension.

Fracture toughness G_c of the foam. – From the intercept onto the vertical line, $\ln(a) = 0$, of a straight line (with slope $-1/2$) fitting each series of data in fig. 4 to minimize the sum of squared errors, we can *directly* estimate the fracture surface energy γ or the fracture toughness $G_c = 2\gamma$ of our foam, thanks to the exact Griffith's relation in eq. (1) for the plane stress, *i.e.*, for the present thin sheets. The results are given in table I.

Instead of what we did here, the fracture toughness G_c could be estimated from a standard K_c test on small samples incorporating the finite-size effect, with the aid of an equation [7, 8],

$$K_c^2 = G_c E, \quad (2)$$

where the numerical coefficient is exact (even under finite-size effect) for plane stress. However, softness of the samples might cause some technical problems. In addition, eq. (2), which contains an elastic constant E , requires a special care for fracture with plastic work (as in our case); such a linear-elastic relation could be used only in the limit of the small-scale zone: the constancy of K_c for changes in a should be checked carefully as in [9]. In other words, the applicability of the Irwin-Orowan extension of Griffith theory should be carefully checked each time the fracture is dominated by plastic work. In the present work, since Griffith's scaling law is confirmed well, we estimated K_c not from a standard K_c test, but from a directly obtained G_c via eq. (2), as given in table I.

In general, direct determination of the energy of a non-liquid surface is very difficult. This difficulty, for example, had long prevented a direct confirmation of the Young-Laplace relation for wetting where a JKR test was the key (Chapt. 2 in [10]). Our present study provides another unusual example where a direct determination of non-liquid surface energy is possible.

Relative density of the foam. – A dimensionless quantity $\phi \equiv \rho/\rho_s$, *i.e.*, density of the foam ρ relative to the corresponding solid ρ_s , or the volume fraction of the solid in the foam, is an important parameter to characterize properties of cellular solids [1, 2]. Although the density of a low-density polyethylene (LDPE) solid is distributed in the range 0.91–0.94 g/cm³, below ρ_s is assumed to be 0.92 g/cm³; this choice would introduce only a few per cent error in ϕ or ρ_s at most (judging from the measured Young modulus given below, the corresponding solid is not the high-density polyethylene but LDPE). The relative density ϕ was obtained from a

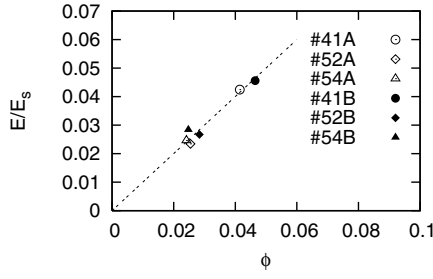


Fig. 5 – The Young modulus is plotted as a function of the relative density. The linear relation indicated by the dashed line is in accord with a brief consideration given in the text.

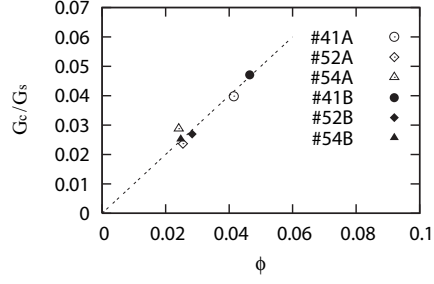


Fig. 6 – The fracture toughness G_c is plotted as a function of ϕ . The clear linear relation in the right is in accord with a brief consideration given in the text.

density measurement of the foam where the weight of a $10\text{ cm} \times 10\text{ cm}$ sample is measured. The results are also given in table I.

Young modulus and relative density. – The elastic potential energy per volume of a solid is dimensionally given as $f_s \simeq E_s \varepsilon^2 / 2 - \sigma \varepsilon$, where ε is the strain and E_s is the Young modulus of the solid. The minimization of this energy with respect to ε leads to Hooke's law: $\sigma \simeq E_s \varepsilon$. If local deformation of the foam is affine to global continuum deformation ε , the potential energy of the foam is simply given by $f \simeq \phi E_s \varepsilon^2 / 2 - \sigma \varepsilon$ (note that $E_s \varepsilon^2 / 2$ is the elastic energy per unit volume in the solid and ϕ is the solid density in the foam). This results in Hooke's law, $\sigma \simeq \phi E_s \varepsilon$, predicting that the Young modulus of foam E is expressed in terms of E_s as

$$E \simeq \phi E_s. \quad (3)$$

This relation might be understood also from the classical interpretation that an elastic constant corresponds to a cohesion energy per volume (see, for example, Chapt. 2 of ref. [11]).

Equation (3) holds well for our experimental data as shown in fig. 5. In this plot, the Young modulus E is normalized by E_s , which is obtained separately for samples A and B (see table II) by extrapolating experimental linear $E - \phi$ relations up to $\phi = 1$; these rough estimates of E_s correspond to assuming that the coefficient in eq. (3) is one and are comparable with the Young modulus of LDPE solid ($\simeq 100\text{ MPa}$) as mentioned above [1].

The difference in E_s of samples A and B thus obtained is not meaningless because the coefficient in eq. (3) is expected to be the same for samples A and B even if it is not one. This difference is possibly due to manufacturing or storage conditions (the room temperature is well above the glass transition temperature of PE); note that such difference in ρ_s is negligible because a possible error in ρ_s is less than a few per cent.

Equation (3) is consistent with a formula for closed-cell foam given in [1] because the volume of cell edges is not too large compared with the volume of cell faces (or walls) in our case (fig. 1); the formula includes a bending effect of the cell edge (proportional to ϕ^2), in addition to an extension effect of the cell face (proportional to ϕ), and the ϕ^2 -term can be neglected when the volume of the edges is not too large compared with that of walls because of the relation $\phi^2 \ll \phi$. Note that the internal pressure (p_0) term of the formula could be neglected in our case ($E_s \gg p_0$).

Fracture parameters and relative density. – Now think a foam cube of volume L^3 with voids and consider to derive the volume fraction of the foam cube by slicing this volume into

TABLE II – The Young modulus E_s , fracture toughness (critical energy release rate) G_s , critical stress intensity factor K_s of solid polyethylene corresponding to the sample foam. E_s (and, thus, K_s) are estimates with precision lower than estimates of G_s (see the text for details).

	E_s (MPa)	G_s (kJ/m)	K_s (MN/m ^{3/2})
A	$\cong 32$	11.3	$\cong 0.60$
B	$\cong 27$	9.54	$\cong 0.51$

many thin sheets of size L^2 with thickness dz . Suppose the area fraction of each sheet is f , then the net volume of each sliced sheet is just fL^2dz . Summing up all the volume of these sheets, we realize that the net volume of the L^3 cube is just fL^3 , meaning the volume fraction is f : *the area fraction of the foam is simply given by the volume fraction ϕ* (except for cases where the fluctuation of the volume fraction is too large to find an appropriate dz for which fluctuation of the area fraction of sheets is negligible). This general consideration leads a relation,

$$G_c = \phi G_s, \quad (4)$$

where G_s is the fracture toughness of the corresponding solid. We stress that the numerical coefficient of the linear relation in eq. (4) is exactly one from our argument; a similar relation was indirectly suggested in [1, 3] but without prediction on the numerical coefficient and it was justified in specific cases; their argument requires assumptions on details of the cell structure. On the contrary, our interpretation suggests that our relation with a coefficient, eq. (4), is rather general and independent of cell structures. (Note, however, that our tacit assumption of the plane crack surface *on a crack-size scale* might allow a slight correction for the coefficient in eq. (4), due to the crack surface roughness *on a cell-size scale*.)

Equation (4) seems to work well in our experiment as in fig. 6. The normalization factors G_s in this figure, given in table II, are also obtained by extrapolating experimental linear $G-\phi$ relations up to $\phi = 1$ separately for samples A and B, here with knowing the proportional coefficient of the linear relation (4).

Since eqs. (3) and (4) hold well in our experiments, eq. (2) suggests that K_c scales as

$$K_c \simeq \phi K_s, \quad (5)$$

where K_s is the critical stress intensity factor for the corresponding solids. From G_s and E_s given in table II, we obtain K_s from the relation $K_s^2 = G_s E_s$ (*i.e.* eq. (2) for the solid) from our experiment. These values given in table II are consistent with values of the polyethylene solid suggested in the literature ($K_c \sim 1 \text{ MN/m}^{3/2}$) [1, 9]. Note that these estimates of K_c may include errors associated with the above-mentioned errors in E_s .

Discussion. – The values of G_c for foams and G_s for the solids given in tables I and II are quite large compared with any kind of bond-breaking energy and dominated by plastic or dissipative work to cause rupture ($G_c \sim$ a few mN/m for van der Waals (VW) “bonds,” while $G_c \sim$ a few N/m for chemical bonds). They originate probably from the fracture surface energy of semicrystalline polymers: typically $K_c \sim 1 \text{ MN/m}^{3/2}$ and $E \sim 100 \text{ MPa}$ for LDPE [1, 9], which implies that the fracture toughness of LDPE is about 10 kN/m. This value almost coincides with the obtained G_s in table II.

The physical understanding of the fracture energy of LDPE itself is a separate important problem. While the fracture of glassy polymers with craze (as well as the fracture of weakly cross-linked polymers) is discussed, for example, in [12, 13], we could not observe any craze

along the fracture line through microscope observation for our semicrystalline polymer (our experiment was done at room temperature well above the glass transition temperature).

The above good agreement of our G_s with the typical fracture toughness of LDPE might be better appreciated with caution: 1) there is a possibility that the semi-crystalline structure (and, thus, fracture energy) of the thin polyethylene film consisting of cell faces will be rather different from that of solid LDPE. 2) The coefficient of eq. (2) could allow a slight modification due to the non-plane character of crack surface on a cell-size scale.

Conclusion. – We experimentally confirmed Griffith's scaling law without the effect of finite size in a linear-elastic polyethylene foam, using large samples with changing fracture size well over one order of magnitude. This allows us to estimate *directly* the fracture surface energy of the considerably soft foam. The obtained fracture energy of the foam is too large to be understood as bond breaking energy: it is governed by a certain dissipative process without crazing, while the plastic zone around the tip is well localized compared with the crack size. Scaling relations in the foam for Young modulus, fracture energy and critical stress intensity factor are proposed in eqs. (3), (4) and (5), based upon our experiments and theoretical considerations. These results for our soft foam are not the same as those for the well-studied cases of rigid foam ($E_s > 3000$ MPa); softness and cell structure of our samples would be in favour of our naive assumptions (such as the affine deformation) based on the continuum views. Our reasoning for these relations is independent of the details of cell structure, with a prediction of a numerical coefficient for eq. (4).

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