Two scaling domains in multiple cracking phenomena

U. A. Handge,[1] Y. Leterrier,[2] G. Rochat,[2] I. M. Sokolov,[1] and A. Blumen[1]
[1]*Theoretical Polymer Physics, University of Freiburg, Hermann-Herder-Strasse 3, 79104 Freiburg im Breisgau, Germany*
[2]*Laboratoire de Technologie des Composites et Polymères (LTC), École Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland*
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The fragmentation of two-phase composite systems such as coatings on substrates and fibers in matrices under uniaxial tension shows two regimes: the evolution of the mean fragment length with applied strain displays different power laws for small and for large strains. From theoretical arguments, we find that in both cases the scaling exponent depends on the shape parameter of the strength distribution. The exponent for large applied strains depends also, as we show, on the way (linear or nonlinear) in which the stress transfer between the two constituents takes place. We present experimental data on the sequential cracking of SiO_x coatings on thermoplastic substrates that indeed show the two regimes; the detailed analysis of the data supports our theory.

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The statistical aspects of failure phenomena are closely associated with the random distribution of defects in materials [1]. The interplay between these statistical aspects and the elastic properties of solids influences strongly the formation of cracks. In a bulk material under tension, the stress enhancement that originates at a single defect may result in a crack propagating through the whole sample and may cause the failure of the material. In such a situation we observe only one crack event. By contrast, in composite materials such as coatings on substrates or fibers in matrices, we often deal with one phase that carries the load, while the other phase cracks sequentially [2,3]. The analysis of the evolution of these repeated fracture events provides much insight into the behavior of the system and particularly into the properties of the interface between the two constituents.

Experimental [4,5] and theoretical studies [6–8] have revealed different stages in the fragmentation of thin brittle coatings adhering to ductile substrates under uniaxial tension. For many systems, the fragmentation kinetics, i.e., the evolution of the mean fragment length $\langle L \rangle$ with the applied strain ε, shows different power laws both at early and at late stages. This scaling behavior, when plotted in double logarithmic scales (see, e.g., Fig. 1), displays two straight lines. The two regimes are related to the correlation length ξ [6,7,9–11] (also denoted as stress transfer length [8] or as screening length [12]) and to its magnitude with respect to the mean fragment size $\langle L \rangle$. In the initial breaking stage, $\xi \ll \langle L \rangle$ holds and the strain $\epsilon_{xx}(x)$ in each segment equals the applied strain ε in the substrate. This ''isostrain'' situation [13] leads to randomly located cracks, practically independent of their position in the sample. After several breaking events, at larger strains $\xi \gg \langle L \rangle$ holds, and the strain $\epsilon_{xx}(x)$ attains a scaling, universal shape with a clear peak at the center of each fragment [14]. At this stage, the new cracks form near to the fragments' centers (''midpoint cracking regime'' [4]). Eventually, at very large strains, one notices that the segments start to debond from the substrate (adhesive failure) and the crack density saturates.

In what follows, we focus on the first two fragmentation stages (random cracking and midpoint cracking) and pay attention to the scaling exponents in the power-law domains. We show that these exponents are related to the strength distribution and thus are not independent. Hence their measurement is expected to provide important experimental information about the strength distribution. Note that such a two-stage scaling behavior with different scaling exponents at initial and at late stages was also found in the time dependence of photocurrents in glassy semiconductors and modeled within the continuous-time random walk approach by Scher and Montroll [15].

We start first from a linear elastic stress transfer mechanism between the two components. Then we compare the theoretical results with measurements of the fragmentation process of SiO_x coatings on thermoplastic substrates under uniaxial tension. The analysis of the experiments reveals that both scaling regimes lead (within experimental error) to the same shape parameters. In a second step, we also address nonlinear elastic stress transfer. It turns out that now the scaling exponents depend both on the shape parameter and on the exponent of nonlinearity. Consequently, both parameters can be determined from the scaling exponents of the initial and of the advanced stages of cracking.

Our approach follows the analysis of (scalar) network models [6,7,14], which is similar to the shear-lag analysis [8,16]. We assume that the deformation of the substrate (or of the matrix) is uniform and that the strain in this component equals ε. This assumption is an approximation, since nonuniform substrate deformation may be important. In addition, ductile materials have a yield point and may exhibit nonlinear deformation, facts that we do not take into account. The x axis is parallel to the loading direction. Applying a ''weakest link'' approach [17], we divide the brittle component (coating or fiber) of length L_0 into N breakable elements of length ΔL, so that $N = L_0/\Delta L$ holds. In the initial stage, the strain $\epsilon_{xx}(x)$ in the coating (or in the fiber) equals the applied strain ε, and the local strain in each discrete element is simply ε. The strength of a breakable element of length ΔL is a random function, which obeys the probability density $f(\varepsilon)$. The corresponding cumulative distribution func-

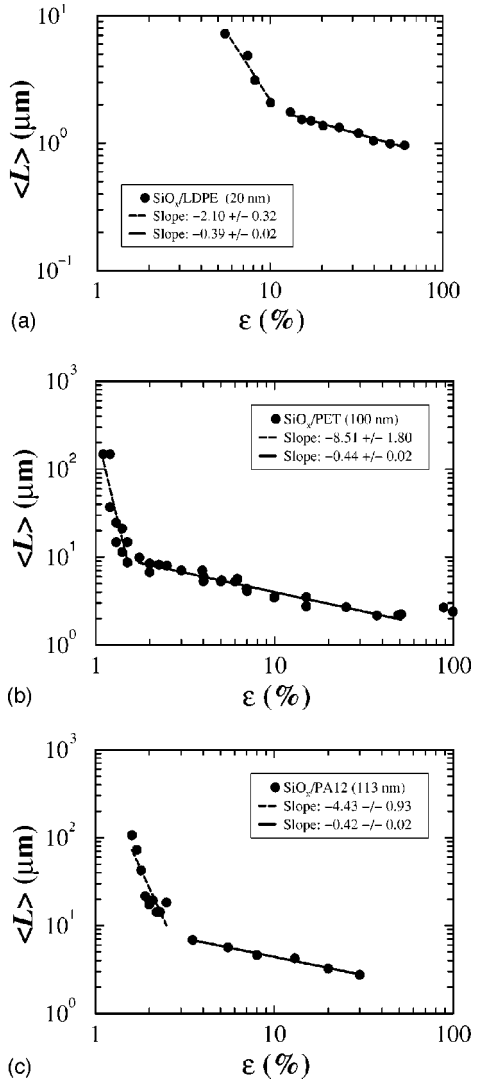

FIG. 1. Mean fragment length $\langle L\rangle$ as a function of the applied strain ε for a SiO_x coating (a) on a LDPE substrate, (b) on a PET substrate, and (c) on a PA12 substrate in double logarithmic scales. The thickness of the coating is indicated in brackets. The data for the LDPE and the PET substrate are taken from Refs. [30,31]. The circles denote the experimental values. The dashed and the solid lines indicate least-squares fits in the initial and in the advanced stage, respectively.

tion is denoted by $F(\varepsilon)=\int_0^\varepsilon f(\tilde{\varepsilon})d\tilde{\varepsilon}$. We remark that $F(\varepsilon)/\Delta L$ is the so-called ''hazard rate'' [18]. Now the probability $P(\varepsilon)$ that the coating (or the fiber) fails under the applied strain ε is $P(\varepsilon)=1-[1-F(\varepsilon)]^N=1-\exp\{N\ln[1-F(\varepsilon)]\}$, so that for large N,

$$P(\varepsilon)=1-\exp[-NF(\varepsilon)] \tag{1}$$

holds. If $F(\varepsilon)$ is a power law, say $F(\varepsilon)=(\varepsilon/W)^\alpha$ for $0\leqslant\varepsilon\leqslant W$ [19], we obtain the Weibull distribution [20]:

$$P(\varepsilon)=1-\exp\left[-\frac{L_0}{\Delta L}\left(\frac{\varepsilon}{W}\right)^\alpha\right]. \tag{2}$$

In the initial stage, the correlation length ξ is much smaller than the dimensions of the sample L_0, and the strain $\epsilon_{xx}(x)$ is, apart from small regions around cracks, uniform and equal to the applied strain, i.e., $\epsilon_{xx}(x)\equiv\varepsilon$. On the average, taking the cracks as being independent, we have $\langle n\rangle=L_0F(\varepsilon)/\Delta L=NF(\varepsilon)$ cracks. Then (neglecting crack opening) the mean spacing between cracks is $\langle L\rangle=L_0/(\langle n\rangle+1)\approx L_0/[NF(\varepsilon)]$. With $F(\varepsilon)=(\varepsilon/W)^\alpha$, we obtain as in Refs. [8,18,21–26] a scaling law for $\langle L\rangle$:

$$\langle L\rangle\propto\varepsilon^{-\kappa_1}, \tag{3}$$

where $\kappa_1=\alpha$ holds.

We turn now to the later stages of fragmentation, for which $\xi\gg\langle L\rangle$. We consider first that the stress transfer between the two constituents is linear elastic. Then the strain $\epsilon_{xx}(x)$ in a fragment of length L can be readily evaluated based on scalar network models [6,7,27] or a shear-lag analysis [16]; thus for a fragment centered at the origin, with the loading direction along the x axis, it follows that

$$\epsilon_{xx}(x)=\varepsilon\left(1-\frac{\cosh(x/\xi)}{\cosh[L/(2\xi)]}\right), \tag{4}$$

where ξ depends on the elastic properties of the two components. For instance, for fiber/matrix systems we have $\xi=R_f\sqrt{(1+\nu_m)E_f\ln(R_m/R_f)/E_m}$, where ν_m is Poisson's ratio of the matrix, E_f and E_m denote, respectively, Young's moduli of the fiber and of the matrix, R_f is the fiber radius, and R_m is the equivalent matrix radius [16,28]. For $\xi\gg L$, the function $\epsilon_{xx}(x)$ is parabolic:

$$\epsilon_{xx}(x)=\varepsilon L^2(1-4x^2/L^2)/(8\xi^2). \tag{5}$$

Note that in advanced stages of fragmentation, having a fragment of length L implies that we know that its elements have already withstood the whole previous stress history without breaking; this changes in a complex manner the actual cumulative distribution function [8]. Disregarding for a moment this ''censoring'' effect, the probability $P(\varepsilon)$ that a fragment of length L breaks under the applied strain ε is $P(\varepsilon)=1-\Pi_{i=1}^{L/\Delta L}[1-F(\epsilon_{xx}(x_i))]\approx 1-\exp[-\int_{-L/2}^{L/2}F(\epsilon_{xx}(x))dx/\Delta L]$, where x_i denotes the position of the ith breakable element of length ΔL. Then the mean value $\langle\varepsilon\rangle$ of the elongation at failure is given by

$$\langle\varepsilon\rangle=\int_0^\infty\varepsilon P'(\varepsilon)d\varepsilon, \tag{6}$$

where the prime denotes differentiation with respect to ε. Using Eq. (5) yields

$$\langle\varepsilon\rangle=8\xi^2W(\Delta L/A_{1,\alpha})^{1/\alpha}\Gamma(1+1/\alpha)L^{-(2\alpha+1)/\alpha} \tag{7}$$

with $A_{1,\alpha}=\sqrt{\pi}\Gamma(\alpha+1)/[2\Gamma(\alpha+3/2)]$ and $\Gamma(t)$ being the gamma function. As before, inverting the dependence between $\langle\varepsilon\rangle$ and L leads again to a scaling law, $\langle L\rangle\propto\varepsilon^{-\kappa_2}$, now, however, with [6,7]

$$\kappa_2=\frac{\alpha}{2\alpha+1}. \tag{8}$$

Consequently, not only in the initial stage, but also in advanced fragmentation stages $\langle L\rangle$ scales with ε, albeit with another exponent. In previous works [7,14,29], direct simu-

lations of the full fragmentation process were performed, which led precisely to Eq. (8). Hence when scaling is concerned, ''censoring'' seems to play a secondary role. This may be due to the fact that cracks form next to highly ''proof-tested'' regions: in the cracks' close vicinity, the tensile stress and strain are very close to zero, so that there the probability for an additional failure is small anyhow.

In what concerns three-parameter distributions such as $F(\varepsilon)=[(\varepsilon-\varepsilon_{\min})/W]^{\alpha}$ with $W/\varepsilon_{\min}$ small, previous studies have shown [6,14] that they lead to $\kappa_2=1/2$, i.e., the same behavior as given by Eq. (8) in the limit $\alpha\to\infty$.

Our analysis has shown that both for small and for large substrate strains, the average fragment length $\langle L\rangle$ scales with ε. Furthermore, since the two scaling exponents κ_1 and κ_2 depend only on α, one can either write κ_2 as a function of κ_1, or use both κ_1 and κ_2 for an internal check on α.

Such a check is provided by the progressive fragmentation of thin SiO_x coatings on thermoplastic substrates under uniaxial stress. The experiments and discussion of substrate-yielding are reported in detail in Refs. [31,24,30]. The substrate materials are (a) low-density polyethylene (LDPE), (b) poly(ethylene terephthalate) (PET), and (c) polyamide12 (PA12). The resulting experimental $\langle L\rangle$ versus ε curves are depicted in Fig. 1, where two scaling regimes are evident. A least-squares fit in the initial regime leads to $\alpha=2.10\pm0.32$ for the LDPE substrate. Hence Eq. (8) implies $\kappa_2=0.40\pm0.01$. The experimental κ_2 value obtained from the advanced stage is given by $\kappa_2=0.39\pm0.02$, which is very close to 0.40. In the case of the PET substrate, a least-squares fit to the data in the initial stage leads to $\alpha=8.51\pm1.80$. Equation (8) yields then $\kappa_2=0.47\pm0.01$, which does not differ much from the experimental value $\kappa_2=0.44\pm0.02$. In the case of the PA12 substrate, we find $\alpha=4.43\pm0.93$ for the initial stage and thus $\kappa_2=0.45\pm0.01$ for the advanced stage, which is again close to the experimental value $\kappa_2=0.42\pm0.02$. In conclusion, the fragmentation of SiO_x coatings confirms our theoretical results.

We now show how to extend our results to the case of nonlinear stress transfer between the two phases. The elastic shear stress τ at the interface depends on w, the difference between the displacement $u(x)$ of the coating (or the fiber) and the displacement $v(x)$ of the substrate (or the matrix) at the interface; one has $w(x)=u(x)-v(x)$. Thus $w(x)$ is proportional to the shear strain at the interface. We now introduce the auxiliary function $G(z)$ and write

$$\tau(w)=\mathrm{sgn}(w)\left.\frac{dG(z)}{dz}\right|_{z=|w|}, \tag{9}$$

where the sgn function takes into account the direction of the shear force. In Ref. [29], $G(z)$ was taken as $G(z)=cz^{m+1}$, with c and m being parameters. Such a form includes among others both Hooke's law ($m=1$) and also a constant interfacial shear stress ($m=0$).

In the advanced fragmentation stages, the length L of a fragment is small with respect to ξ, and the displacement of the coating is negligible. Then one finds $w(x)\approx-\varepsilon x$ [29], where the center of the x axis is in the middle of the fragment. The balance of forces implies [8,29,32]

$$\frac{d\sigma}{dx}=\frac{\tau}{h_c}, \tag{10}$$

where h_c denotes the thickness of the coating. Inserting $w(x)=-\varepsilon x$ into Eq. (10) yields for $x\geqslant0$

$$\frac{d\sigma}{dx}=-\frac{1}{h_c}\left.\frac{dG(z)}{dz}\right|_{z=\varepsilon x}=-\frac{1}{h_c\varepsilon}\frac{dG(\varepsilon x)}{dx}. \tag{11}$$

The stress vanishes at the fragment's edges. Therefore, we have $\sigma(-L/2)=\sigma(L/2)=0$, and the solution of Eq. (11) is

$$\sigma(x)=[G(\varepsilon L/2)-G(|\varepsilon x|)]/(h_c\varepsilon). \tag{12}$$

Thus the tensile stress in the coating is a simple function of $G(z)$. Equation (12) leads for $G(z)=cz^{m+1}$ to known former results: For $m=0$, Eq. (12) yields $\sigma(x)=cL(1-|2x/L|)/(2h_c)$, i.e., a triangular form for $\sigma(x)$ when the interfacial shear stress is constant [8,32]. Linear stress transfer $G(z)=cz^2$ recovers, when inserted into Eq. (12), the parabolic form of Eq. (5). Generally, $G(z)=cz^{m+1}$ leads to

$$\sigma(x)=\frac{c\varepsilon^m L^{m+1}}{h_c 2^{m+1}}(1-|2x/L|^{m+1}). \tag{13}$$

Equation (13) reproduces the corresponding result of Refs. [14,29]. This expression shows scaling. From Eq. (13), the strain $\epsilon_{xx}(x)$ in a fragment follows for a linear elastic coating from $\epsilon_{xx}(x)=\sigma(x)/E_c$, where E_c denotes Young's modulus of the coating.

Analogous to our previous calculations, the mean elongation $\langle\varepsilon\rangle$ at failure follows readily. The probability $P(\varepsilon)$ that a fragment of length L fails under the strain ε is again $P(\varepsilon)\approx1-\exp[-\int_{-L/2}^{L/2}F(\epsilon_{xx}(x))dx/\Delta L]$. This leads to

$$\langle\varepsilon\rangle=(W/B)^{1/m}(\Delta L/A_{m,\alpha})^{1/(m\alpha)}\times\Gamma[1+1/(m\alpha)]L^{-[(m+1)\alpha+1]/(m\alpha)}, \tag{14}$$

where $A_{m,\alpha}=\int_0^1(1-t^{m+1})^{\alpha}dt$ and $B=c/(E_c h_c 2^{m+1})$ hold. Hence we again find $\langle L\rangle\propto\varepsilon^{-\kappa}$ with

$$\kappa=\frac{m\alpha}{(m+1)\alpha+1}, \tag{15}$$

as already derived in Refs. [14,29]. In the case of nonlinear stress transfer, the scaling exponent κ is a function both of α and of m. For $m=1$, Eq. (15) simplifies to Eq. (8). Since from κ_1 we have $\alpha=\kappa_1$, we can use κ_2 to determine m.

In summary, we have analyzed the fragmentation of two-phase composites under uniaxial tension where the cracking component is brittle and the load-carrying part is ductile. Our theoretical analysis reveals that $\langle L\rangle$ scales with ε both in the initial and in the advanced fragmentation stages, the exponents being, however, different. For linear elastic stress transfer, both exponents depend only on α, the shape parameter of the strength distribution. Consequently, α can be obtained from two independent experimental results. As displayed, the fragmentation of SiO_x coatings on thermoplastic substrates indeed shows two scaling domains; moreover, the determination of the exponents in several experimental runs confirms the theoretically established relations. Finally, we

have also considered nonlinear elastic stress transfer; in this case, the scaling exponents depend both on α and on the nonlinearity parameter m. Hence the measurement of the scaling exponents κ_1 and κ_2 allows us to determine both α and m.

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