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Intrinsic failure and non-linear elastic behavior of glasses

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Abstract

Inert failure strains of carefully prepared glass fibers, measured by using the two-point bending technique, are large. For example, the failure strain is about 18% for silica fibers. To convert such high strains to stresses, the strain dependence of the Young's modulus (i.e., non-linear elastic behavior of glass) has to be taken into account. However, experimental values of modulus for most glasses are available only for low strains (typically <2%). In this paper, we propose that a description of the non-linear elastic behavior of glass up to and including the fourth-order modulus is necessary and adequate for converting *intrinsic* failure strains to failure strengths. We examine the validity of the proposed formulation by comparing the measured fiber strengths in pure tension to the values calculated – using the proposed formalism – from measured intrinsic failure strains in two-point bending for two glass compositions (silica and E-glass) for which all necessary data are available in the published literature.

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1. Introduction

Tensile strength studies of bare E-glass fibers during 1960s and 1970s and of coated silica glass fibers during 1980s have demonstrated that carefully prepared glass fibers exhibit extremely high tensile strengths with very small standard deviations (∼1%). In addition, these strength values are fairly reproducible among several independent investigators [1,2]. For example, when the moisture-induced break down of siloxane bonds is prevented by testing at sufficiently low temperatures, such as the boiling point of liquid nitrogen (77 K), the measured average tensile strength of silica fibers is about 12.5 GPa [3,4] and of E-glass fibers is about 6.5 GPa [5,6]. Kurkjian and Paek [7] were the first to argue that measured high values for the strength of silica fibers were intrinsic property of the glass and not controlled by microscopic extrinsic flaws. Later, Kurkjian and Gupta [8] similarly argued that the measured E-glass strengths were also intrinsic. The argument that measured fiber strengths are intrinsic is based on the experimental results that (a) the measured standard deviation in strength is about two times the measured standard deviation in fiber diameter, and that (b) the measured strengths do not vary with fiber diameter. Additional indirect support is provided by estimates of flaw sizes corresponding to measured high strengths using the Irwin–Griffith equation [9]. The estimated flaw sizes are comparable to a typical size of basic structural units in glasses and are too small to be considered as stable extrinsic flaws. For example, in the case of silica, the estimated flaw size is less than 1 nm, which is about the same as the diameter of a six-membered ring made of SiO_4 tetrahedra.

Measurements of bare fiber strengths in pure tension in non-ambient environments (such as 77 K) are tedious because of difficulties in gripping fiber-ends and high probability of damaging the fiber surface. A simple

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and convenient technique – called two-point bending (TPB) – was refined around 1980 for testing silica fibers [10,11]. TPB measures the failure strain of a glass fiber bent in a U-shape as the distance between the parallel fiber arms is reduced at some constant rate. The measured TPB failure strains at 77 K for silica fibers are about 18% [12]. Recently, using TPB, failure strains as high as 23.5% are reported for sodium silicate glass fibers [13], 21% for sodium alumino-silicate fibers [14], and 13% for E-glass fibers [15]. These high failure strain values also correspond to intrinsic failure for the same reasons mentioned earlier for failure strengths in pure tension.

With the growing use of TPB technique, especially for strength measurements at 77 K, the question arises as to how to convert a high failure strain, ε_f, to failure strength, σ_f. In general, the Young's modulus, Y, is expected theoretically [16] and known experimentally [17] to be strain dependent. However, most measurements of modulus are available only for low strains. Lacking knowledge of the strain dependence of modulus, failure strains, ε_f, are frequently [18] converted to failure strengths, $\sigma_f(0)$, using the zero-strain Young's modulus (Y_0):

$$\sigma_f(0) \equiv Y_0\varepsilon_f. \qquad (1)$$

Using this equation, the value of $\sigma_f(0)$ for silica fibers ($Y_0 = 72$ GPa) agrees with the value of σ_f measured in pure tension. However, the value of $\sigma_f(0)$ for E-glass fibers ($Y_0 = 74$ GPa) is almost 50% higher than the value measured in pure tension. This disagreement, in the case of E-glass, shows the inadequacy of Eq. (1) and points to a need to include the strain dependence of the modulus. In this paper, we show that a description of non-linear elastic behavior of glass (up to the fourth-order modulus) is necessary and adequate for converting intrinsic failure strains to intrinsic strengths.

In the next section, we formulate the non-linear elastic model and derive an equation relating intrinsic failure strain to intrinsic failure stress. In Section 3, we show how to obtain the values of pertinent non-linear elastic parameters from experimental results and discuss the validity of the proposed formalism using published data for silica and E-glass fibers. Section 4 concludes this paper.

2. Non-linear description of the elastic behavior of glass

Phenomenological descriptions of non-linear elasticity are available in standard textbooks. Here, we follow the approach of Ruoff [19] to estimate the ultimate yield strength of cubic crystals. For our purpose, the behavior in pure tension can be described by the following equation where we exclude terms containing powers of true strain higher than the third:

$$\sigma(\varepsilon) = Y_0\varepsilon + (Y_1/2)\varepsilon^2 + (Y_2/6)\varepsilon^3. \qquad (2)$$

Here, Y_0 is the conventional (zero-strain) Young's modulus. Y_0 is also known as the second-order modulus. Y_1 is the third-order Young's modulus and Y_2 is the fourth-order Young's modulus. The strain dependence of the modulus, Y ($\equiv\partial\sigma/\partial\varepsilon$), can be readily obtained from Eq. (2):

$$Y(\varepsilon) = Y_0 + Y_1\varepsilon + (Y_2/2)\varepsilon^2. \qquad (3)$$

Experimental results [20] show that for most glasses, except for silica rich glasses, Y decreases with increasing strain (i.e., $Y_1 < 0$). This behavior is termed normal. Silica-rich glasses are anomalous: Y increases with strain (i.e., $Y_1 > 0$). The anomalous behavior is not unique to Y_1. Silica-rich glasses also exhibit anomalous behavior in other properties. For example, Y_0 of silica glass increases with increase in temperature, the isothermal compressibility increases with increase in pressure, and the thermal expansion coefficient becomes negative in some temperature ranges [21]. Fig. 1 shows a plot of experimentally measured values of $Y(\varepsilon)$ for pure silica (anomalous) and for E-glass (normal) fibers.

It is easy to see that inclusion of the Y_2 term is necessary in Eq. (2). Measurements of Y using high strength silica fibers [22] at high strains show that $Y(\varepsilon)$ exhibits a maximum with respect to strain; decreasing at higher strains as one would expect theoretically. The observation of the maximum in $Y(\varepsilon)$ is in agreement with independent observations of minimum in the compressibility with respect to pressure [23]. To describe such a maximum in $Y(\varepsilon)$, it is necessary to include the Y_2 term.

Using Eq. (2), a strain value can be converted to the corresponding stress provided values of the three

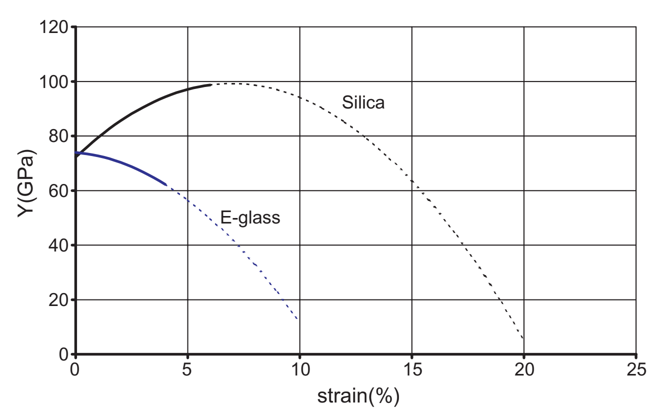

Fig. 1. Plot of Young's modulus (GPa) vs. strain (%) for silica fibers (Eq. (13)) and for E-glass fibers (Eq. (14)). The curves represent the best fit of Eq. (3) to the experimental data for silica [22] and for E-glass [29, 10 μm diameter]. The strain ranges of the solid curves indicate the range over which experimental data are available. Dashed curves are extrapolations based on Eq. (3). Notice that Y vanishes at about 20% strain for silica and at about 13% strain for E-glass. These strain values agree well with measured intrinsic failure strains of the two glasses, respectively.

Young's moduli, Y_0, Y_1, and Y_2, are known. Techniques used for the measurement of Y_1 and Y_2 are the same as those for the measurement of Y_0. The only difference is that measurements of Y need to be performed as a function of strain (or stress) up to sufficiently high strains. We discuss measurement techniques and published values of Y_0 and Y_1 in the next section. The values of Y_2 are difficult to obtain experimentally since one has to measure Y up to fairly high strains, which are difficult to achieve in brittle materials like glass.

The value of Y_2 can be expressed in terms of the *intrinsic* failure strain, ε_f. At intrinsic failure, the stress is maximum with respect to strain. In other words, $Y(\varepsilon_f) = 0$. Therefore, it follows from Eq. (3) that

$$Y_0 + Y_1\varepsilon_f + (Y_2/2)\varepsilon_f^2 = 0. \quad (4)$$

Solving for Y_2 gives

$$Y_2 = -2(Y_0 + Y_1\varepsilon_f)/(\varepsilon_f^2). \quad (5)$$

Substitution of this expression of Y_2 in Eq. (2), gives

$$\sigma(\varepsilon) \equiv Y_0\varepsilon + (Y_1/2)\varepsilon^2 - (\varepsilon^3/3)[(Y_0 + Y_1\varepsilon_f)/(\varepsilon_f^2)]. \quad (6)$$

At intrinsic failure ($\varepsilon = \varepsilon_f$), Eq. (6) simplifies to

$$\sigma_f \equiv \varepsilon_f[(2/3)Y_0 + (Y_1/6)\varepsilon_f]. \quad (7)$$

Eq. (7) is our key result. It shows that knowledge of Y_0 and Y_1 values is sufficient to convert intrinsic failure strain to intrinsic failure stress.

3. Discussion

To begin, it is important to examine whether the material remains elastic up to the intrinsic failure. This can be answered by examining the reversibility of the glass after subjecting it to high strains just short of intrinsic failure. Although, in principle, such an experiment should be possible, we do not know of any reports of such an experiment. However, reversible elastic behavior has been firmly established for silica glass under hydrostatic compression up to 8 GPa [24]. This value of hydrostatic compression corresponds to 24 GPa in uniaxial compression. This suggests that the silica glass most likely remains elastic in tension up to the intrinsic strength (<15 GPa).

3.1. Estimation of the third-order Young's modulus (Y_1)

To obtain reliable values of Y_1, it is necessary to measure Y_0 to strains of at least 2% (the higher the better). As mentioned earlier, for brittle samples like glass, it is difficult to achieve high strains in pure tension (except in the case of high strength fibers). An alternative is to measure elastic moduli in hydrostatic compression where high strains can be achieved without failure since the strength of glass is much higher in compression than in tension. As shown later in this section, using equations of non-linear elasticity for isotropic materials, the data obtained in hydrostatic compression can be used to obtain Y_1. We emphasize that, according to Eq. (2), the value of Y_1 is a property of the material at zero-strain. It is independent of the stress state and its value is the same in compression as in tension.

There are three techniques for measurement of Y_0:

(i) *Static techniques* [25]. Here, one measures the force–displacement curve of the material in some convenient configuration such as in pure tension or in bending. Since these techniques require measurements of sample dimensions, in addition to measurements of force and displacement, they typically have large uncertainties.

(ii) *Ultrasonic techniques* [26]. Here, one measures the velocity of sound waves (longitudinal and transverse) in the sample. The sound velocities can be converted to modulus provided the mass density is known. Ultrasonic techniques are extremely convenient and are used most routinely.

(iii) *Brillouin scattering technique* [24]. In this technique, which is also known as the hypersonic technique, one measures the frequency shift of light scattered by acoustic phonons. This information can be converted to sound velocity and therefore to moduli values. This is a non-contact technique and can be used to obtain modulus values as a function of stress. However, the technique has not been used extensively as the instrumentation for Brillouin Scattering, generally assembled in the laboratory, requires special expertise. In addition, one needs a high-pressure cell or a device for applying stress.

The ultrasonic and Brillouin scattering techniques provide adiabatic values of bulk modulus B and shear modulus G (or the Poisson's ratio v) while the static technique gives isothermal values. However, the difference between adiabatic and isothermal values of moduli is negligible for most solids and especially so for silicate glasses [27].

Using hydrostatic compression, a pair of elastic moduli (frequently the bulk modulus B and the shear modulus G or B and the Poisson's ratio v) can be obtained as a function of pressure P. For isotropic materials like glass, knowledge of any two moduli is sufficient to calculate the Y:

$$Y = 3B(1 - 2v). \quad (8)$$

The validity of Eq. (8) is well known for linear elastic behavior of isotropic materials. It can be shown readily that Eq. (8) remains valid in the non-linear elastic regime of isotropic materials. By taking the derivative of both sides of Eq. (8) with respect to pressure, one obtains [28]

$$\frac{\partial Y}{\partial P} = 3(1-2v)\frac{\partial B}{\partial P} - 6B\frac{\partial v}{\partial P}. \tag{9}$$

The pressure derivative can be converted to strain derivative:

$$\frac{\partial Y}{\partial P} = \left[\frac{-1}{3B}\right]\frac{\partial Y}{\partial \varepsilon}. \tag{10}$$

Combining Eqs. (9) and (10), one obtains the following expression for Y_1:

$$Y_1 = -9(1-2v)B\left(\frac{\partial B}{\partial P}\right)\bigg|_{P=0} + 18B^2\left(\frac{\partial v}{\partial P}\right)\bigg|_{P=0}. \tag{11}$$

The validity of Eq. (11) can be examined by comparing Y_1 values obtained from static tension and from either ultrasonic or hypersonic compression techniques. Silica is the only glass for which results are available from all three techniques. The values using hypersonic technique [26] agree with those from ultrasonic techniques. Table 1 summarizes these results. For static tension results, a fit of Eq. (2) to the results obtained experimentally by Krause et al. [22]:

$$\varepsilon = (\sigma/72.3) - 3.2(\sigma/72.3)^2 + 12(\sigma/72.3)^3 \tag{12}$$

gives

$$Y(\varepsilon) = 72.3 + 772.4\varepsilon - 5542\varepsilon^2, \tag{13}$$

where Y and σ are in GPa. In Eq. (12), ε is actually engineering strain [22]. It should be corrected to true strain. The correction is small and inconsequential to the main conclusion of this work.

Considering the uncertainties in the static tension technique due to measurements of fiber diameter and small values of elongation as well as the paucity of results, we conclude that the results for Y_1 shown in Table 1 – particularly the room temperature results – validate Eq. (11).

Table 2 lists values of Y_1 of some simple glasses. For E-glass, the only published results are the static tension data at room temperature [29]. Fitting Eq. (2) to these data, we obtain

$$Y(\varepsilon) = 74 - 73.15\varepsilon - 5527\varepsilon^2, \tag{14}$$

which gives $Y_1 = -73.15$ GPa for E-glass.

Table 1
Values (in GPa) of Y_0, Y_1, and Y_2 for silica glass

Temperature (K)		Static tension	Ultrasonic compression	
		[22]	[25]	[28]
298	Y_0	72.2	72.4	
	Y_1	772.4	906	1074
	Y_2	−11 084		
	B_0		37.7	
77	Y_0	72 [17]	69.1	
	Y_1	475	1549	1083
	B_0		38	

Table 2
Values of Y_0, Y_1, and Y_2 (GPa) for simple glasses at room temperature

Glass	Y_0	Y_1	Y_2	Refs.
Silica	72.3	772.4	−13 058[b], −11 084[a]	[25]
Na_2O–Al_2O_3–$2SiO_2$	74.1	−0.9		[30]
E-glass	74	−73.2[a]	−7631[b], −11 054[a]	[29]
B_2O_3	14.3	−34.3[a]		[22]
GeO_2	43.9	210.7[a]		[22]
As_2S_3	15.3	−132[a]		[22]

[a] Values from static tension results.
[b] Values calculated using Eq. (5).

3.2. Examination of the validity of the proposed formalism for intrinsic failure (Eq. (7))

Table 3 lists values of intrinsic failure stress calculated using Eq. (7) for both silica and E-glass. Also listed are the values of intrinsic failure strength measured in pure tension. The agreement is good for both glasses.

For silica, the agreement was good even without considering the non-linear elastic terms (see values of $\sigma_f(0)$ in Table 3). The fact that the agreement remains good after including non-linear terms is entirely due to a coincidental combination of Y_0, Y_1, and ε_f values. For silica, the value of (Y_1/ε_f) happens to be approximately equal to that of $(2Y_0)$.

The agreement between the calculated and measured strength values for E-glass is good only after including the non-linear terms. This provides the principal support for the validity of our formalism.

3.3. Discussion of intrinsic strength in terms of non-linear elastic parameters

For normal glasses ($Y_1 < 0$), an upper bound of σ_f can be obtained from Eq. (7):

$$\sigma_f \leqslant \varepsilon_f(2/3)Y_0. \tag{15}$$

Actually, Eq. (15) is a good approximation for the intrinsic strength of normal glasses since the term containing Y_1 in Eq. (7) is much less than the term containing

Table 3
Comparison of calculated (using Eq. (7)) and measured values of intrinsic failure strengths (σ_f) of silica and E-glass fibers

	Silica	E-glass
ε_f (2PB)	0.18	0.13
σ_f (Eq. (7))	12.8	6.2
σ_f (pure tension)	12.5–15	~6.5
$\sigma_f(0)$ (Eq. (1))	13	9.6

only Y_0. For example, for E-glass, Eq. (15) gives a value of 6.4 GPa which is comparable to the value of 6.2 GPa using Eq. (7). This comparison suggests that the 'effective secant modulus at failure strain' for normal glasses is *approximately* equal to (2/3) Y_0 and that the value based on Eq. (1) is off by at least 33%. Eq. (15) provides a lower bound for σ_f of anomalous glasses. Here, the term containing Y_1 is significant and cannot be neglected.

By solving the quadratic (Eq. (4)) for the failure strain ε_f in terms of elastic parameters Y_0, Y_1, and Y_2 and then substituting the resulting expression in Eq. (2), one can obtain an explicit expression for σ_f in terms of Y_0, Y_1, and Y_2. We have already argued in Section 2 that inclusion of Y_2 is necessary to rationalize silica data. Can one neglect the Y_2 term in the case of normal glasses like E-glass? Upon substituting the values of al three elastic parameters and the failure strain for E-glass in Eq. (2), it can be seen that the contribution of the Y_2-term is higher than that of the Y_1 term. Thus, at least in the case of E-glass, the Y_2 term cannot be neglected. This raises the question whether one should also include higher order terms (such as Y_3 etc.). The good agreement for silica and for E-glass demonstrated in this paper suggests that higher order terms are not significant.

4. Conclusions

In this paper, we have formulated a scheme to convert high intrinsic failure strains to failure stresses taking into account the non-linear elastic behavior of a glass. The non-linear elastic behavior is described by a third-order polynomial of stress in terms of strain that includes the term containing the fourth-order modulus Y_2. We show that including Y_2 is necessary. However, it is not clear a priori that terms of order higher than Y_2 can be neglected for strains as high as 20%. The results of this paper suggest that the higher order terms may not be significant.

Our formalism cannot be used for a general strain since, to our knowledge, values of Y_2 are not available for glasses (except approximately for silica and E-glass). However, we derive a relation among Y_0, Y_1, Y_2 and the *intrinsic* failure strain, ε_f. Using this relation, Y_2 can be estimated from the knowledge of Y_0, Y_1, and ε_f. We show that this estimate is reasonable by considering the silica and E-glass data.

Intrinsic failure stresses calculated using our formalism for both silica and E-glass, agree well with respective values of intrinsic failure strength measured in pure tension. This excellent agreement, especially for E-glass, supports the validity of our formalism. This implies that the intrinsic strength of a glass is determined by the values of the second, third, and fourth-order elastic moduli.

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