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Long-term mechanical behaviour of optical fibres coated with a u.v.-curable epoxy acrylate

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Long-term strength tests have been performed on glass fibres drawn from both Suprasil rods and graded index pre-forms in a laser furnace and in a graphite resistance furnace and coated in both cases with a u.v.-curable epoxy acrylate. Results of stress-free ageing tests in humid environments over a period of 18 months show that the fibres weaken at a rate that depends not only upon humidity level but also upon which heating source is used for fibre drawing. The weakening phenomenon has been ascribed to stress-induced reactions between silica and adsorbed water at the glass–polymer interface which occur spontaneously during the tensile test (dynamic fatigue). Static fatigue tests in 90% r.h. show that the fatigure behaviour deviates from that predicted by the theories of Charles and of Charles and Hillig for uncoated glasses; specifically plots of log (stress) versus log (time-to-failure) and stress versus log (time-to-failure) show downward curvatures which become more pronounced as the applied stress is lowered. In the absence of fatigue data for uncoated fibres, however, it is not possible to determine whether such non-linear behaviour is brought about by the presence of the organic coating layer.

1. Introduction

One important characteristic of glass fibres or other glass structures is their tendency to weaken under the combined influence of water vapour and applied load. With the advent of optical communications technology in which glass fibres are used as optical waveguides, long-term retention of fibre strength is of paramount concern. Although recent efforts on fibre drawing and coating techniques have led to the production of strong fibres [1,2], successful application of these fibres to high-strength areas requires reliable data on long-term performance in hostile environments. In this paper we present and compare results of long-term tests performed on fibres drawn with a graphite resistance furnace and with a CO_2 laser and subsequently coated with a u.v.-curable epoxy acrylate. We place special emphasis on the influence of the organic coating layer on the long-term behaviour and examine whether the static fatigue behaviour could be accounted for by the stress-corrosion mechanisms of Charles [3] and of Charles and Hillig [4] originally derived for uncoated glasses.

2. Experimental

The glass fibres were drawn from 8 mm graded-index pre-forms and 7 mm Suprasil-2 (Amersil, Inc.) rods using either a 250 W CO_2 laser or a graphite resistance furnace. Prior to drawing into fibres, the Suprasil-2 (SS-2) rods were fire-polished with an oxy-hydrogen torch to reduce the possibility of surface damage that may result from handling. Results of fire-polishing on fibre strength have been reported previously [1, 2]. The pre-forms (with germania-doped silica cores) were prepared by a modified chemical vapour deposition technique using an Amersil TO-8 fused quartz (14 mm o.d., 12 mm i.d.) as the starting material [5].

Fibres were drawn vertically to a diameter of 110 μm and coated in-line with a u.v.-curable epoxy-acrylate [6] containing 0.5 wt % of a styryl-functional silane (Dow Corning Z6032). Ap-

plication of the coating was accomplished by means of a flexible applicator [7] which allowed the deposition of a nearly concentric coating on the moving fibre without damaging the glass surface. Curing was performed between the applicator and the take-up drum using a pair of 200 W $in.^{-1}$ medium pressure u.v. lamps.

Fibres were drawn at a speed of 0.4 m sec^{-1} and the viscosity of the coating resin was maintained at about 50 P by regulating the resin temperature at about 40° C [8]. However, in one of the coating runs involving a furnace-drawn, graded-index fibre, the resin viscosity was intentionally increased to about 500 P (resin temperature 17° C) to assess its effect on the long-term strength of the fibre. Presumably, fibres coated with viscous resin would be more susceptible to attack by moisture because of poorer wetting at the glass–resin interface [9]. The organic coating was approximately 60 μm thick and had an extremely smooth surface.

The ageing studies included evaluation of residual tensile strengths upon exposure to three different environments: (1) water, 22° C; (2) 90% r.h., 32.6° C and (3) 50% r.h., 22° C, while the static fatigue tests were conducted in an atmosphere of 90% r.h., 32.6° C. Although the main objective here was to study the long-term effects of H_2O on the fibre strength, it was found necessary, in the case of 90% r.h., to raise the temperature somewhat to allow a close control of humidity level over the extended period of ageing. Variations in humidity and temperature were maintained at less than ±3% r.h. and 1.5° C, respectively, of the specified values throughout the ageing period. Tensile tests were performed with an Instron testing machine at a strain rate of 21% min^{-1} and a gauge length of 0.61 m. Specimens (aged and unaged) were conditioned in the test environment of 50% r.h. and 22° C for about 15 min prior to the tensile tests.

Static fatigue tests were performed on 0.61 m gauge length specimens in an atmosphere of 90% r.h. and 32.6° C using (50) standard dead-weight loading units individually equipped with electric timers. Prior to application of loads, specimens were stored in the test chamber for about 15 min to allow equilibration of the glass fibre with the test atmosphere. To avoid vibration in the test chamber, the humidity and temperature control system was installed in a separate structure and connected to the test chamber via two rubber tubings.

Specimens for each test were taken from kilometre-long fibres at random intervals. In the case of water-immersion tests, specimens were sealed at both ends with a water-resistant epoxy to prevent ingress of water from fibre ends during the tests.

3. Results and discussion

3.1. Initial strength

The results of short-term tensile tests (gauge length 0.61 m) are plotted on a Weibull probability scale in Fig. 1 where the numbers in parentheses indicate the number of data points used in the plots. (For the sake of clarity some points have been left out at the upper ends of the curves.) It is worth noting that the distributions of the two laser-drawn fibres, regardless of whether they were drawn from a graded-index pre-form or an SS-2 rod, are extremely narrow and unimodal while, by contrast, the distribution of the furnace-drawn fibre is rather broad and multimodal. The low strength dispersion in the furnace-drawn fibre has been ascribed to damage inflicted on the fibre surface by particulate contaminants generated from furnace components [10, 11].

The slope of the data plot, which is equivalent to the shape parameter, m, in the Weibull distribution function [12] Equation 1, is about 40 for the laser-drawn SS-2 fibre, 35 for the laser-drawn TO-8 clad fibre, and 36 and 5.8 for the two highest

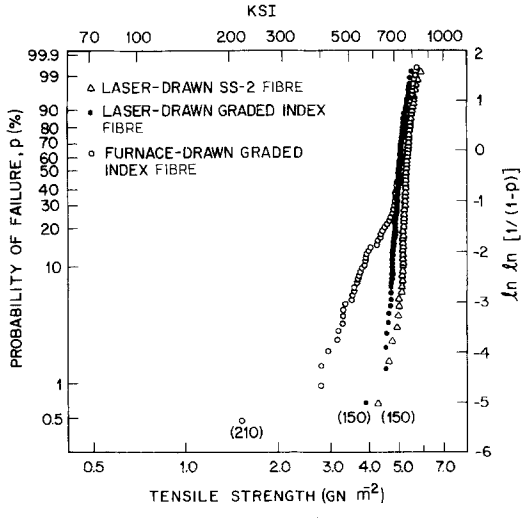

Figure 1 Weibull plots of initial strengths for the three fibres coated with epoxy acrylate.

strength modes of the furnace-drawn TO-8 clad fibre.

$$p = 1 - \exp\left[-l\left(\frac{S}{S_0}\right)^m\right] \quad (1)$$

Here p is the probability of failure, l is the specimen gauge length, S is the fibre strength, and S_0 and m are constants. The slopes for the two laser-drawn fibres are much larger than the value reported previously (about 25 [1]) for a laser-drawn SS-2 fibre at 20 m gauge length indicating that either the shape parameter, m, may be length dependent or the distribution is narrowed as a result of further improvement in the drawing and coating techniques (e.g. better control of fibre diameter, etc.).

3.2. Stress-free ageing tests

The results of stress-free ageing tests in water and 90% r.h. are shown in Figs. 2 and 3, where the median strengths of the aged fibres have been normalized in terms of the corresponding initial median strengths to facilitate comparison. (Results of ageing tests in 50% r.h., 22° C are not plotted here because all three fibres showed no perceptible changes of median strength over the same test period.) Each median (of the aged fibres) was determined from twelve measurements in the case of the two laser-drawn fibres, and 30 or more in the case of the furnace-drawn fibre on account of the latter's wide scatter in the initial strength.

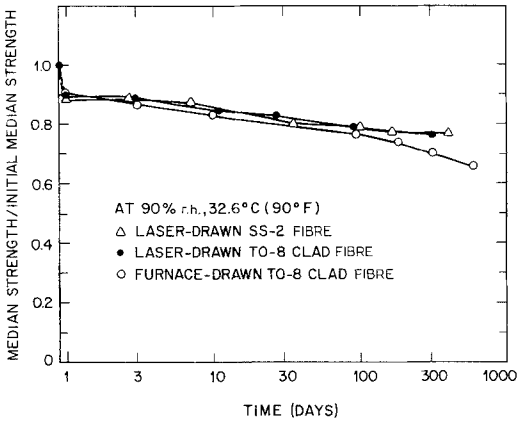

Figure 2 Ratio of median strength to initial median strength versus time (days) of exposure to an atmosphere of 90% r.h., 32.6° C. Each median of the aged specimens was determined from 60 measurements in the case of the furnace-drawn fibre and 12 measurements in the case of the two laser-drawn fibres. The standard deviation was about 9% of the mean in the former and 2.5% in the latter.

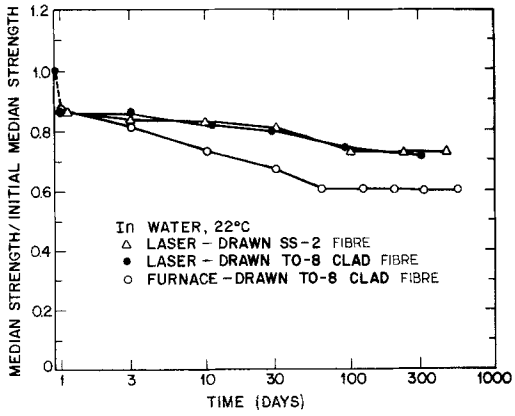

Figure 3 Ratio of median strength to initial median strength versus time (days) of exposure in distilled water, 22° C. Each median of the aged specimens was determined from 30 to 60 measurements in the case of the furnace-drawn fibre and 12 measurements in the case of the two laser-drawn fibres. The standard deviation was about 8% of the mean in the former and 2% in the latter.

Although all three fibres weaken on exposure to both environments, the furnace-drawn fibre degrades most rapidly especially in the water immersion tests. In addition, the responses of the two laser-drawn fibres are almost identical, suggesting that the ageing behaviour is less sensitive to the compositional difference between the SS-2 fibre and the TO-8 clad fibre than to the nature of the heat source used in fibre drawing.

Proctor *et al.* [13] have recently studied the ageing behaviour of pristine fused silica fibres and found the strength to be solely a function of moisture content in the test atmosphere and independent of the humidity to which the fibre was exposed prior to the tests. They suggested that the changes of strength in the test atmosphere were caused by stress-induced reactions between silica and the atmospheric moisture during the tensile test (dynamic fatigue [14]).

The fact that the pristine fibre and the coated fibre behaved differently is perhaps not surprising since in the latter the fibre is bounded by a layer of organic material which may retain some of the water absorbed during the ageing treatment. Thus it is quite possible that the strength losses observed in the coated fibres were brought about by reactions of this entrapped water with silica during tensile testing. This is supported by the observation [15, 16] that upon drying in a vacuum oven (33° C, 1/3 atm.) over a dessicant ($CaSO_4$), the

aged fibre displayed a smaller loss of strength, depending on the length of drying treatment and the time of exposure to the tensile test environment (50% r.h., 22° C) prior to the test. (In fact, the median strengths of all three coated fibres exceeded their initial values by about 14% when tested within 15 min after 10 days of the drying treatment). Clearly, if the strength loss resulted from some sort of reactions during ageing, the loss should be permanent and subsequent drying should not cause recovery of strength. The water-absorption-induced dynamic fatigue mechanism is also in agreement with the fact that the coated fibres did not display any change in strength when aged in the tensile test environment (50% r.h., 22° C) over an extended period (~18 months).

Since the tensile test normally lasts only a few seconds, the stress-activated reaction should be more sensitive to water available in the immediate region surrounding the glass surface (glass–polymer interface) than to water contained in the bulk coating layer. In principle, the level of water concentration at the glass–polymer interface is determined by the kinetics of sorption which involves not only the regular diffusion mechanism but also other processes such as swelling of the polymer by water, debonding of the coating layer from the glass surface, and clustering of water molecules into liquid water at the interface [17]. However, since molecular water can diffuse readily through the polymer (diffusion constant $\sim 10^{-7}$ cm^2 sec^{-1}) [18] it is likely that the initial rapid drop of strength in both environments is largely due to the diffusion process, while the slow weakening trend in the later stage of ageing is brought about by further build-up of water concentration due to the other factors. Accordingly, the gradual levelling-off in the strength curves is explained by the saturation of water level at the interface which depends both on the state of interfacial bonding and the humidity level in the test environment.

The above discussion suggests that formation of a strong interfacial bond is essential for long-term retention of strength in the humid environment. To form such a bond the resin must be initially of low viscosity to allow proper wetting of the glass surface [9]. Therefore we would expect the ageing behaviour to be sensitive to the viscosity or temperature at which the coating is applied to the fibre. To assess this effect a furnace-drawn graded-index fibre was coated with resin

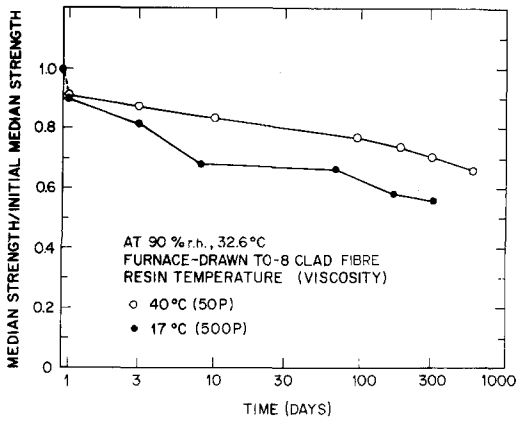

Figure 4 Strength plots of furnace-drawn fibres coated with resin maintained at two different viscosities. Each median of the aged fibre was determined from 60 measurements. The standard deviation was about 8% of the mean in the fibre coated at 17° C and 9% in the fibre coated at 40° C.

maintained at 17° C (500 P) for comparison with fibres coated in a regular run (resin viscosity 50 P). Results of ageing tests in 90% r.h. and 32.6° C are shown in Fig. 4, where it can be seen that the fibre coated at a higher viscosity sustained a greater strength loss throughout the entire test period. It is also worth noting that the difference between the two curves does not become significant until after one day when the influence of interfacial bonding becomes dominant.

3.3. Static fatigue

The individual static fatigue data obtained for the three fibres in 90% r.h. and 32.6° C are listed in Tables I, II and III along with their medians, averages and standard deviations. Significantly, the time-to-failure varies widely at each stress level even in the two laser-drawn fibres which had extremely narrow distributions in the initial strengths. An example of distribution plots for the fatigue data at 2.068 MPa (300 KSI) is shown in Fig. 5.

The broadening of the time-to-failure distribution can be readily rationalized if it is assumed that the static fatigue failure arises from slow growth of pre-existing cracks at the glass surface in accordance with the power law [19]

$$V = AK_I^n \qquad (2)$$

where V is the rate of crack growth, K_I is the stress intensity factor at the crack tip, and A and n

TABLE I Static fatigue data (time in min) of laser-drawn SS-2 fibre under different stresses

Fatigue data	Stress (KSI, MPa in brackets)							
	275 (1896)	300 (2068)	325 (2241)	350 (2413)	375 (2586)	400 (2758)	425 (2930)	475 (3275)
Time (min)	32 377	6048	3257	2175	497	140	47.2	8.2
	48 075	11 097	4076	3197	562	174	58.6	9.8
	65 738	15 899	6215	3489	662	207	67.3	10.2
	76 907	16 380	7867	3966	764	275	68.2	11.2
	79 772	23 669	8683	4423	788	350	85.3	11.6
	80 032	28 151	10 798	4741	888	352	106	11.9
	84 617	30 384	13 494	5644	1115	589	123	12.8
	97 435	31 522	14 845	5692	1284	640	141	12.9
	97 489	36 698	15 101	5915	1614	669	142	17.6
	141 567	40 845	18 007	5923	1684	832	160	18.7
Median	79 902	25 910	9741	4582	838	351	96	11.8
Average	80 401	24 069	10 234	4516	986	423	100	12.5
Standard deviation	29 665	11 412	5023	1300	423	241	40	3.3

TABLE II Static fatigue data (time in min) for laser-drawn TO-8 clad fibre under different stresses

Fatigue data	Stress (KSI, MPa in brackets)							
	200 (1379)	225 (1551)	250 (1724)	275 (1896)	300 (2068)	350 (2413)	400 (2758)	425 (2930)
Time (min)	109 477	57 477	35 708	22 002	4421	158	10.2	7.6
	141 401	107 070	35 799	23 411	5976	170	12.2	7.9
	149 249	121 173	53 628	24 567	7218	200	19.6	7.9
	176 965	123 177	61 576	27 671	9497	231	27.9	8.9
	240 480	145 051	61 793	28 450	14 233	236	30.0	9.2
	244 800	149 204	62 547	32 309	18 324	272	31.6	9.6
	254 950	202 512	63 098	35 080	25 755	317	37.8	10.8
	259 200	208 061	72 650	40 273	26 190	346	45.8	12.4
	295 771	278 747	74 409	40 400	26 463	374	58.0	14.3
	385 920	290 786	102 394	50 991	34 422	485	79.3	17.7
Median	242 640	147 128	62 170	30 380	16 279	254	30.8	9.4
Average	225 821	168 315	62 360	32 515	17 250	279	35.2	10.6
Standard deviation	82 983	75 402	19 285	9231	10 513	102	21.3	3.3

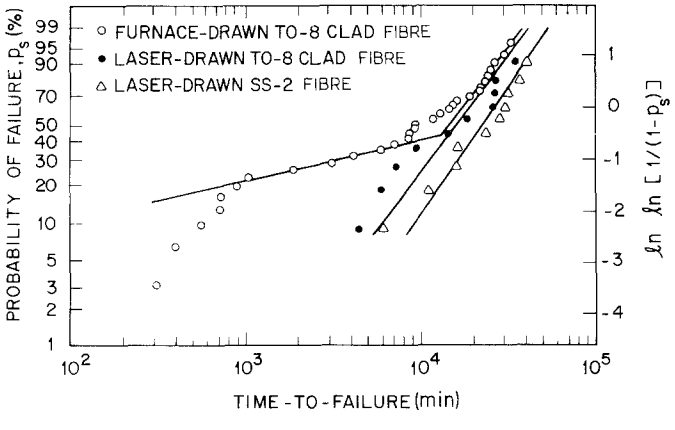

Figure 5 Weibull plots of times-to-failure for the three fibres tested under an applied stress of 2.068 MPa (300 KSI) and in an atmosphere of 90% r.h., 32.6° C.

TABLE III Static fatigue data (time in min) for furnace-drawn TO-8 clad fibre under different stresses

Fatigue data	Stress (KSI, MPa in brackets)					
	250 (1724)	300 (2068)	325 (2241)	340 (2344)	375 (2586)	400 (2758)
Time (min)	2889	329	72.1	40.4	8.1	2.0
	4231	396	90.7	49.5	12.4	2.5
	5723	552	103	61.1	15.3	3.1
	7496	703	114	69.0	18.6	3.2
	11 223	718	141	98.8	21.5	3.4
	18 547	898	241	136	27.1	4.3
	30 397	1049	328	185	32.3	4.4
	30 412	1872	514	270	41.1	5.5
	32 706	3122	514	276	54.7	7.0
	38 504	4155	545	301	66.6	8.1
	41 417	5905	821	340	76.9	9.3
	45 172	7052	959	407	86.4	10.5
	45 843	8442	1059	481	97.8	12.1
	51 444	8676	1172	524	117	15.1
	55 624	9220	1279	581	129	16.7
	59 370	9338	1320	723	132	18.6
	63 880	11 764	1510	760	143	19.0
	69 124	12 899	1649	838	158	20.4
	73582	14 605	1719	945	169	21.2
	84 411	15 404	1804	1017	171	23.1
	85 148	16 092	2130	1145	184	24.5
	97 203	18 993	2415	1289	201	25.0
	119 509	21 813	2711	1419	213	27.6
	135 420	22 002	2873	1618	226	27.7
	141 293	23 459	2900	1737	240	30.1
	152 084	24 126	3286	1813	241	30.2
	155 272	25 112	3979	1978	286	35.9
	179 219	26 750	4121	2202	302	41.2
	181 633	30 235	4527	2411	372	50.6
	187 403	32 894	5296	2502	405	77.1
Median	57 497	9 279	1415	652	131	17.7
Average	73 539	11 953	1673	874	142	19.3
Standard deviation	57 541	10 105	1465	766	109	16.7

are constants. Thus if one assumes the Weibull form, Equation 1, for the initial strength distribution, the probability of static fatigue failure, p_s, can be expressed in the following form [19–21],

$$p_s = 1 - \exp\left[-l\left(\frac{S}{S_0}\right)^{mn/(n-2)}\left(\frac{t}{t_0}\right)^{m/(n-2)}\right] \quad (3)$$

where t is the time-to-failure under an applied stress, S, and t_0 is defined as [21],

$$t_0 = \frac{2}{n-2}\frac{1}{Y^2}\left(\frac{K_{Ic}}{S_0}\right)^2 AK_{Ic}^{-n} \quad (4)$$

Y being a constant.

In Equation 3 the exponent, $m/(n-2)$ of t is a measure of dispersion of the time-to-failure distribution and is inversely proportional to the range of scatter. Since the value of n usually ranges between 15 and 50 for most glasses including fused silica [22], $m/(n-2) \ll m$. Consequently, for a fibre with a given scatter in the initial strength (which is proportional to $1/m$), the distribution of time-to-failure at any stress level is broadened by a factor of $(n-2)$.

In order to compare the results of the three coated fibres, we note that Equation 3 is essentially a statistical expression of Charles' equation for delayed fracture in glasses [3]. Thus for a fixed p_s and l, Equation 3 reduces to the familiar form of Charles,

$$S^n t = \text{constant}; \quad (5)$$

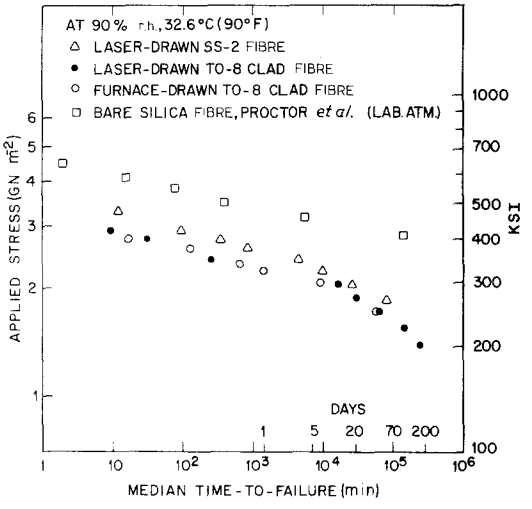

Figure 6 A log–log plot of the static fatigue data for the three fibres tested at 90% r.h., 32.6° C.

accordingly, a log–log plot of applied stress against time-to-failure should yield a straight line with a slope equal to $-1/n$. Such a plot is shown in Fig. 6 for the three fibres using the median time-to-failure, t_{50}, as the abscissa.

There are several points of interest on inspection of Fig. 6. Firstly, the results for the two TO-8 clad fibres are almost identical indicating that the median fatigue behaviour is relatively insensitive to the chemical environment in which the fibres had been drawn. Secondly, although the SS-2 fibre displays a significantly greater resistance to fatigue than the two TO-8 clad fibres at high stresses, the effect tends to diminish as the stress is reduced. In fact, the plots for all three fibres tend to converge as the stress is reduced below $2\,\mathrm{GN\,m^{-2}}$. The last point of interest is the non-linear trends in the plots which become more pronounced as the stress is lowered. Proctor *et al.* [13] have recently reported the static fatigue data of pristine fused silica fibres drawn from Vitreosil rods (Thermal Syndicate Limited) in an oxy/coal-gas flame and tested in the laboratory atmosphere. A replot of their data on log–log axes (indicated by square symbols in Fig. 6) shows that the plots follow closely a linear relationship ($n \sim 24$) as predicted by Charles [3]. Thus if it is assumed that the same behaviour prevails at higher humidity levels, the departure from linearity (Fig. 6) would seem to be associated with the presence of the organic coating layer. However, this conclusion should be accepted with caution in view of the fact that the pristine fibres of Proctor *et al.* and the glass fibres used in the present study were prepared under somewhat different conditions.

Alternatively, the results of the fatigue tests may be examined in a stress versus log (time-to-failure) plot in accordance with the refined stress corrosion theory of Charles and Hillig [4]. Charles and Hillig have derived an expression, Equation 6, for the static fatigue in uncoated glasses by assuming the rate of crack growth to be an exponential function of the applied stress.

$$\log t = -\beta S + f(S) \qquad (6)$$

where β is a constant and f is a function of stress whose contribution is significant only at stresses near the fatigue limit. Accordingly, in the S versus $\log t_{50}$ plot, their theory predicts a curve which is linear (with a negative slope) in the stress range far removed from the fatigue limit and concave upward as it approaches the fatigue limit. A replot of our fatigue data on semi-logarithmic axes, Fig. 7, shows that while the plots are more rectified in comparison with those shown in Fig. 6, the downward curving trends (in the form of a sharp bend near 10^4 min) still persist in all three fibres. By contrast, the plots for the pristine fibres of Proctor *et al.* (also shown in Fig. 7) display an upward curvature and tend to level off to a fatigue limit ($\sim 2.75\,\mathrm{GN\,m^{-2}}$ [13]) as predicted by Charles and Hillig. Consequently, while the precise role of the coating material cannot be understood at the moment, it is clear that the fatigue mechan-

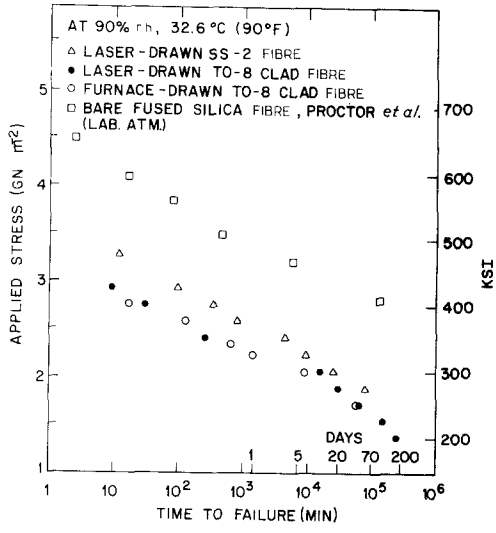

Figure 7 A semi-log plot of the fatigue data for the three fibres tested at 90% r.h., 32.6° C.

ism of the coated fibres is more complex than the models suggested by Charles [3], and Charles and Hillig [4] for uncoated glasses.

4. Conclusion

The two laser-drawn fibres were found to weaken at a slower rate than the furnace-drawn fibre when aged in water and in an atmosphere of 90% r.h. However, all three fibres remained intact on exposure to the tensile test environment (50% r.h.) over the same test period. The weakening phenomenon has been explained in terms of adsorption of water molecules to the glass–polymer interface during ageing which subsequently interact with the silica fibre during tensile testing.

Static fatigue tests in 90% r.h. showed that all three fibres behaved in a similar manner but plots of the data on both log–log axes and semi-log axes showed a downward curvature which became more pronounced as the applied stress was lowered. The results deviated from the stress-corrosion theories of Charles and Charles and Hillig, but in the absence of comparable data for the uncoated fibre it is not clear if this deviation represents an hitherto unsuspected inherent property of glass and whether or not it is exaggerated or diminished by the presence of the coating.

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