Temperature dependence of the extinction coefficient of fused silica for CO_2 laser wavelengths

Anthony D. McLachlan and Fred P. Meyer

The mechanism of the interaction of CO_2 laser radiation with fused silica is determined by the absorption depth of the radiation in the material. The extinction coefficient of a number of high-purity fused silica samples has been measured by the transmission method by fabricating samples ≈30 μm in thickness. Results obtained agree with the latest reported values. In addition, samples were heated by an auxiliary CO_2 laser and the extinction coefficient determined as a function of temperature for six CO_2 laser lines. No significant difference in the extinction coefficient was observed for samples from different makers of the high-purity silica. Measurements were also conducted on a silica-rich glass Vycor and a significant difference was observed.

I. Introduction

In studying the interaction of CO_2 laser radiation with pure fused silica or silica-rich glasses it is important to know the absorption coefficient α of the radiation in the material. The value of α, which is related to the extinction coefficient k by $\alpha = 4\pi k/\lambda$ determines the depth to which the radiation is absorbed in the silica. If the absorption occurs essentially at the surface, the heat flow and thermally induced stress analysis may be treated in a relatively simple way by the assumption of surface heating. However, if absorption occurs over a significant depth, the heat flow and resultant stress analysis require considerable modification to cope with the absorption in the bulk of the material.

Literature sources[1–4] report values for the extinction coefficient k which are generally derived from reflection measurements on fused silica. These values show marked discrepancies and it was the aim of the reported work in this paper to measure the extinction coefficient accurately and, in addition, to measure its temperature dependence for temperatures up to 1800°C. No adequate information on the temperature dependence of the extinction coefficient was available.

II. Experimental

Contrary to most methods used previously,[1–4] the experimental procedure described in this paper consisted of fabricating samples of pure fused silica and silica-rich glass which were thin enough to allow significant transmission of radiation in the 10–12-μm wavelength band. From a determination of the transmittance of the sample it is possible to calculate the extinction coefficient. Samples were required to be 25-mm diameter and 25–50 μm thick with surfaces which were parallel to within a fringe of visible light. A range of samples was prepared from high-purity fused silica and silica-rich glasses obtained from a number of well-known suppliers as detailed in Table I. The techniques used in the preparation of the samples consisted of careful application of standard optical polishing procedures as fully reported elsewhere.[5] The spectral transmittance at 25°C of a typical sample (26.1-μm Suprasil), measured on a Perkin-Elmer model 580B spectrophotometer, is shown in Fig. 1. Since the surfaces were formed parallel to within one fringe of visible light, interference fringes occur between the two surfaces of the sample in the wavelength bands where there is little absorption (Fig. 1). The thickness of the sample was measured to an accuracy >1 μm by a reflecting autocollimation technique on an optical flat. It was also determined by measuring the interference fringe spacing (Fig. 1) and application of the formula

$$t = \frac{1}{2n\delta\nu},$$

where t is the thickness of the sample, $\delta\nu$ is the fringe spacing in wavenumbers, and n is the refractive index corresponding to the wavelength at which the fringe

A. D. McLachlan is with Materials Research Laboratories, Defence Science & Technology Organisation, P.O. Box 50, Ascot Vale, Victoria 3032, Australia. Fred P. Meyer is with U.S. Army Materials Technology Laboratory, Watertown, Massachusetts 02172-0001.

Received 24 December 1986.

Vol. 2
1987

Table I. Details of Makers and Thickness of Samples Prepared in the Investigation

Maker	Grade	Thickness (μm)
Heraeus-Amersil, Inc. Sayreville, NJ 08872 USA	T20 Suprasil	26.1
Dynasil Corp. of America Berlin, NJ 08009 USA	Dynasil 6000	64.4
Corning Glass Works Corning, NY 14830 USA	Fused silica 7940	38.7
Corning Glass Works Corning, NY 14830 USA	Vycor 7913	46.1
Nippon Silica Glass Co., Ltd. Somerville, NJ 08876 USA	NSG-OY	26.3

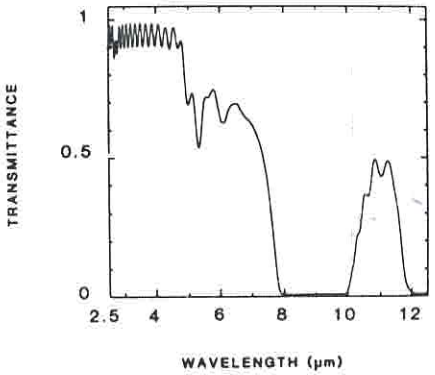

Fig. 1. Spectrophotometer transmission spectra of a sample of pure fused silica (Suprasil), 26 μm thick.

spacing was determined. Since the refractive index is known to a good accuracy up to 4 μm,[6] the thickness can be readily determined. The values so obtained by the two methods differed by <1 μm.

Samples were carefully located in a strain-free mount which permitted convenient routine handling of the samples. The spectrophotometer was fitted with a heated stage so that measurements could be conducted for sample temperatures from 25 to 175°C. To facilitate measurements at higher temperatures, and as an independent measurement of the transmittance of the samples, an experimental facility was also set up (Fig. 2). In this arrangement, the chopped output of a line-tunable CO_2 probe laser (Spectra-Physics model 941) was directed through the sample and onto a pyroelectric detector (with tuned amplifier) mounted in an integrating sphere. The CO_2 laser was operated in the TEM_{00} mode and produced a Gaussian beam whose $1/e$ radius at the sample was measured to be 4.5 mm with a power output of ~1–3 W. The power output stability of the laser was monitored by a separate power meter via a ZnSe beam splitter and its

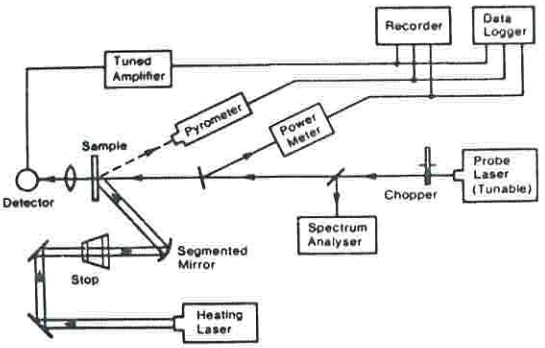

Fig. 2. Schematic diagram of the experimental arrangement for transmission measurement of fused silica at elevated temperatures.

wavelength, which was line selectable from 9.6 to 10.6 μm, was measured by a CO_2 spectrum analyzer (Optical Engineering, Inc. model 16-A).

The sample mount was devised so that the sample could be quickly translated into the beam. Monitoring of the power and wavelength output of the laser ensured that stable experimental conditions applied during the measurement. The transmittance of the fused silica sample was measured by determining the ratio of the detector signal with and without the presence of the sample in the probe laser beam.

The CO_2 laser radiation (1–3-W power) on the thin silica samples caused a temperature rise of typically 150°C and was observed with a pyrometer (Ircon model 7000) operating in the wavelength band of 4.8–5.2 μm. The emissivity of the sample was calculated from the transmission spectra on each sample (Fig. 1) following the procedure described in Ref. 7. This value, which was typically near 0.5, was checked by observing a stable blackbody at 500°C through the thin silica sample. From the observed apparent change in temperature and suitable application of Planck's Law, the effective emissivity of each sample at room temperature could be determined. Values so obtained by the two methods agreed to within 0.05.

A 40-W cw CO_2 laser beam was also directed at the sample to provide uniform irradiation over a 1-cm square area (Fig. 2). This laser beam was arranged so that none of the radiation was incident on the detector-collecting optics of the probe laser beam. The radiation was also unchopped so that the detector would reject any stray scattered cw signal by means of its tuned amplifier.

Because of the thinness of the sample and the subsequent minimum heat loss, it was possible to heat the samples to 1800°C. The pyrometer was arranged to view an area of 5-mm diameter at the center of the uniformly heated zone, coincident with the probe laser beam.

III. Results and Discussion

The extinction coefficients of the samples were determined from the measured transmittance (either spectrophotometer or experimental measurement) by

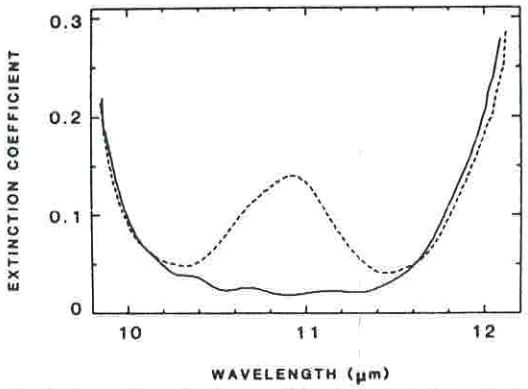

Fig. 3. Values of the extinction coefficient calculated from the data of Fig. 1: ——, pure fused silica; - - -, Vycor. The values were calculated from spectrophotometer transmittance measurements at 25°C.

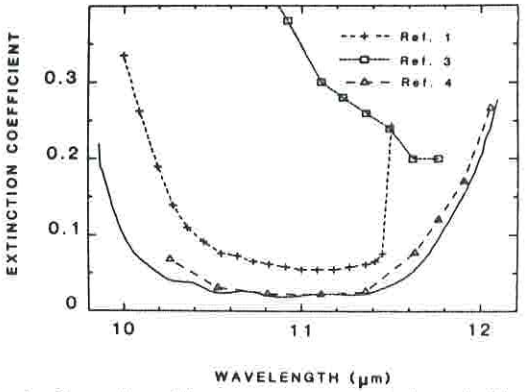

Fig. 4. Comparison of the present measurements (——) with previously reported values of the extinction coefficient of pure fused silica.

application of standard thin-film calculation procedures.[8,9] These procedures take account of multiple reflections within the sample and require the refractive index to be known to a reasonable accuracy. Within the wavelength region of interest (10–12 μm) sufficient literature values exist[1,3,4] to reliably characterize the refractive index. Variations in these literature values give rise to a maximum error of approximately ±0.003 in the extinction coefficient. The results of calculations applied to the spectrophotometer measurements of Fig. 1 are shown in Fig. 3 as well as the results obtained with the Vycor sample. The structure in the 10–12-μm transmittance spectrum of Fig. 1 is due to interference fringes. Corresponding residual minor fluctuations are still evident in the extinction coefficient in Fig. 3 and these are due to minor inaccuracies in the determination of the thickness of the sample and/or the values of refractive index assumed for the fused silica.

Figure 3 shows that the extinction coefficient of Vycor exhibits a marked difference from that of fused silica by virtue of the presence of an absorption band centered at 10.9 μm. This band can be attributed to a Si–O–B bond as a result of the boron content in the Vycor.[10] Values of the extinction coefficient of the other fused silica samples (Table I) were found not to differ significantly from the results shown in Fig. 3. These results were obtained from samples of different thicknesses (as shown in Table I) and provided a useful cross-check on the experimental procedure and calculations.

A comparison between previously reported values of the extinction coefficient of fused silica and the present results at 25°C is shown in Fig. 4. As can be seen, good agreement with the recent results reported by Philipp[4] has been obtained. The results in Fig. 4 are those for the Suprasil sample which, apart from the minor residual fluctuations, are not significantly different from the other high-purity fused silica samples.

The experimental arrangement described in Fig. 2 was used to determine the temperature dependence of the extinction coefficient by measuring the transmittance of the samples at various sample temperatures and over the range of wavelengths provided by the probe laser. A typical result is shown in Fig. 5 where each point represents an experimental measurement on the Suprasil (26.1-μm thickness) sample. Figure 6 shows the result when the extinction coefficient is calculated from the data of Fig. 5 according to the procedure described previously.[8,9] The extinction coefficient of fused silica at a wavelength of 10.26 μm is also shown in the diagram as well as the result obtained with the Vycor sample at a wavelength of 10.59 μm. A line of best fit has been fitted to the experimental points and shows that the extinction coefficient varies with temperature in a substantially linear manner. The temperature dependence of the extinction coefficient of Vycor is significantly different from the fused silica, since it is determined by the Si–O–B absorption band due to the 4% residual boron in Vycor.

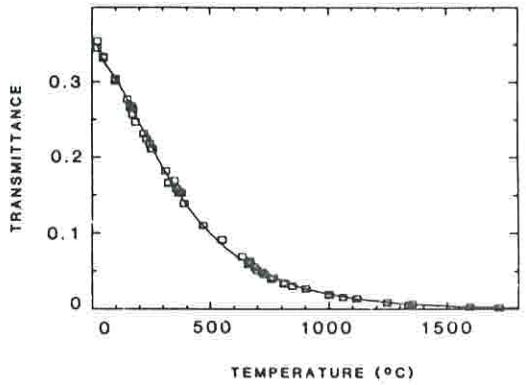

Fig. 5. Example of the temperature dependence of the transmittance of fused silica (26.1-μm Suprasil) at a wavelength of 10.59 μm.

The variation of the extinction coefficient k with temperature T for all wavelengths measured is summarized in Table II which also gives the coefficients of the line of best fit $k = a + bT$, where T is expressed in °C. It should be noted that these coefficients are

Table II. Temperature Dependence of the Extinction Coefficient Expressed for the Least-Squares Line of Best Fit $k = a + bT$ (T in °C) for the Temperature Range and Wavelengths Indicated

Material	Wavelength (μm)	a ($\times 10^{-2}$)	b ($\times 10^{-5}$)	Temperature range (°C)
Fused silica	10.61	2.25	7.71	25–400
Fused silica	10.59	1.82	10.10	25–1800
Fused silica	10.57	2.15	8.53	25–400
Fused silica	10.55	1.82	11.04	25–1600
Fused silica	10.26	3.41	16.67	25–1200
Fused silica	10.21	4.70	14.36	25–300
Fused silica	9.6	3.71	8.27	25–300
Vycor	10.61	9.74	2.79	25–400
Vycor	10.59	8.87	4.59	25–600
Vycor	10.57	8.50	3.71	25–400
Vycor	10.55	7.89	4.61	25–400
Vycor	10.26	4.04	16.63	25–600

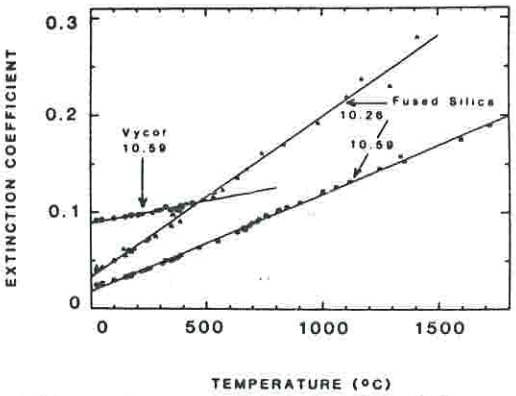

Fig. 6. Measured temperature dependence of fused silica at wavelengths of 10.59 and 10.26 μm. The temperature dependence of Vycor at 10.59 μm is also shown.

applicable for the temperature ranges indicated since measurements were not taken outside these stated ranges.

The significance of the results shown in Fig. 6 and in Table II is that the absorption depth β of the radiation, given by $\beta = \lambda/4\pi k$, varies significantly with temperature, from 34 μm at 25°C to 4 μm at 1800°C. The interaction of high power CO_2 laser radiation with fused silica thus appears to be dominated by surface heating without any substantial absorption of the radiation in the bulk of the material.

IV. Conclusion

The extinction coefficient of high purity fused silica from several suppliers has been determined in the 10–12-μm wavelength region by transmission measurements on thin (25–30-μm) samples. The values obtained were found to agree with previously reported results. In addition, the temperature dependence of the extinction coefficient was determined up to 1800°C and found to vary substantially in a linear manner. The absorption depth of CO_2 laser radiation in fused silica was found to vary from 34 μm at 25°C to 4 μm at 1800°C indicating that laser interaction processes are dominated by surface heating without any significant absorption of the radiation in the bulk of the material.

The authors would like to acknowledge the skillful work of G. N. Sprott in fabricating the samples and the technical assistance of P. Dixon in painstakingly conducting the measurements.

References

1. G. W. Cleek, "The Optical Constants of Some Oxide Glasses in the Strong Absorption Region," Appl. Opt. **5**, 771 (1966).
2. R. Hanna, "Infrared Absorption Spectrum of Silicon Dioxide," J. Am. Ceram. Soc. **48**, 595 (1965).
3. I. Simon and H. O. McMahon, "Study of the Structure of Quartz Cristobalite, and Vitreous Silica by Reflection in Infrared," J Chem. Phys. **21**, 23 (1953).
4. H. R. Philipp, "Silicon Dioxide SiO_2 (Glass)," in *Handbook of Optical Constants of Solids*, E. D. Palik, Ed. (Academic, London, 1985), pp. 749–763.
5. G. N. Sprott, "A Method of Fabricating Optically Thin, Parallel-Sided Glass Samples," MRL Technical Note, to be published.
6. I. H. Malitson, "Interspecimen Comparison of the Refractive Index of Fused Silica," J. Opt. Soc. Am. **55**, 1205 (1965).
7. H. O. McMahon, "Thermal Radiation from Partially Transparent Reflecting Bodies," J. Opt. Soc. Am. **40**, 376 (1950).
8. O. S. Heavens, *Optical Properties of Thin Solid Films* (Butterworths, London, 1955), p. 55.
9. H. A. Macleod, *Thin Film Optical Filters* (Adam Hilger, Ltd., London, 1969), p. 37.
10. A. S. Tenney and J. Wong, "Vibrational Spectra of Vapor-Deposited Binary Borosilicate Glasses," J. Chem. Phys. **56**, 5516 (1972).