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Note: Diffusion constant and solubility of helium in ULE glass at 23 °C

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Among other applications, ultra-low expansion (ULE) glass is commonly used as material for laser cavities. ULE glass is a trademark of Corning Incorporated and consists for the most part of SiO₂ with additionally about 9% TiO₂. ULE cavities were recently proposed to measure helium refractivity for a new fundamental realization of the pascal.^{1–3} In Ref. 1, it was reported that the diffusion of helium into ULE glass caused a length change of the ULE spacer. Another challenge in this application may be a pumping effect of ULE for helium or outgassing of previously absorbed helium. To estimate the impact of helium absorption, we have measured the diffusion constant and solubility of helium in ULE near 23 °C.

We received a sample of ULE glass code 7972 (charge no. 12 204 ANT1) from the Corning company and cut this sample into rectangular plates. For this investigation, we used three plates of different dimensions, as described in Fig. 1. The plates were loaded with helium in a previously evacuated load lock chamber at a pressure of $p_{\text{load}} = 21.1$ (1) kPa. The loading and all experiments were performed at 23 (1) °C. Preliminary experiments indicated that a loading time of 170 h should be sufficient for a thickness of 2 mm and 680 h for a thickness of 4 mm to obtain a homogeneous helium concentration. The plates had been loaded for a minimum of 262 h and 771 h, respectively.

After loading with He, the plates were moved into the sample chamber to measure the outgassing rate vs. time by a quadrupole mass spectrometer (QMS), namely, Transpector from the Inficon company in our outgassing rate measurement system.⁴ The mathematics used to describe the diffusion process and calculate diffusion and permeation coefficients as well as solubility of the material has been discussed in many articles and books.⁵ Here we refer to Ref. 6 where the outgassing rate vs. time is calculated with a one-dimensional approximation for an infinite plane, i.e., $d \ll a, b$. The time dependence at a temperature T is characterized by the constant

$$t_{\text{out}} = \frac{d^2}{\pi^2 \times D}, \quad (1)$$

where D is the diffusion constant at T . For times $t > t_{\text{out}}$, the specific outgassing rate q_{out} deviates less than 1% from the

simple exponential function⁷

$$q_{\text{out}} = \frac{4 \times D}{d} \times c_0 \times \exp\left(-\frac{t}{t_{\text{out}}}\right), \quad (2)$$

with c_0 being the initial concentration of helium in the plate. Hence, the diffusion constant D can be determined from Eq. (1) and c_0 can be obtained by extrapolation of the exponential curve Eq. (2) to $t = 0$. The measured total outgassing rate Q_{out} happens on both sides of the plate, which is

$$Q_{\text{out}} = 2 \times A \times q_{\text{out}}. \quad (3)$$

Equations (2) and (3) require a traceable and absolute measurement of Q_{out} which is possible with our apparatus.⁴ We should note that the QMS was calibrated *in situ* and the interference with other gases present in the system (mainly hydrogen, water vapor, CO, and CO₂) was found to be negligible at our measurement conditions. In addition, we considered the “edge effect,” i.e., gave up the assumption that the plate’s area is infinite.⁸ The changes of t_{out} and c_0 due to this effect are in the low percent region and presented in Table I. Finally, the solubility S can be determined by using Henry’s law,

$$c_0 = S \times p_{\text{load}}. \quad (4)$$

Figure 1 shows the result for the helium outgassing rate vs. time. The measurements were interrupted at some time intervals because of a limit of the recording time of the QMS software. It can be seen that deviations from the mono-exponential decay decrease quickly. After about half a time constant t_{out} , they are no longer recognizable in Fig. 1. To estimate t_{out} , the values between one and at least four times the expected value of t_{out} were considered.

D was determined according to Eq. (1), where the uncertainty is dominated by the one of t_{out} . The values determined for $Q_{\text{out}}(t=0)$, c_0 , and S for each plate were determined according to Eqs. (2)–(4), respectively, and can be found together with t_{out} in Table I.

The values from the three plates agree within their uncertainties so that we calculated the following results by the weighted means: $D = (4.22 \pm 0.06) \times 10^{-8}$ cm²/s and $S = (1.82 \pm 0.14)\%$. The permeation coefficient K was calculated from

$$K = S \times D = (7.68 \pm 0.59) \times 10^{-10} \text{ cm}^2/\text{s}. \quad (5)$$

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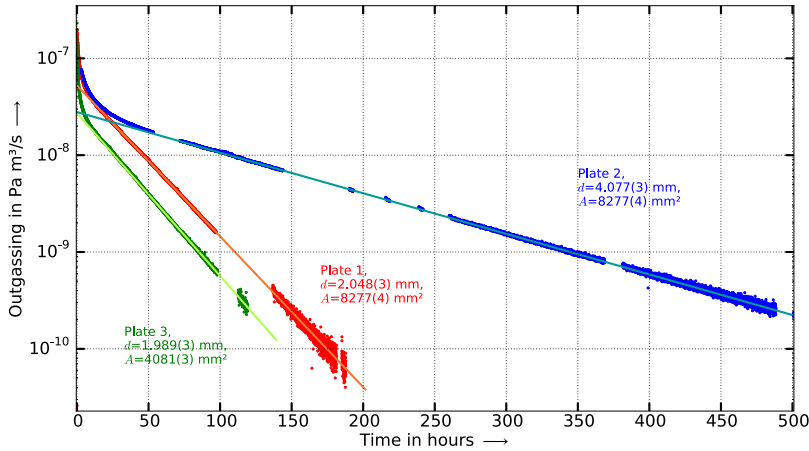


FIG. 1. Measured helium outgassing rates Q_{out} vs. time t for three different plates made from the same ULE glass. Straight lines indicate exponential decays.

TABLE I. Results for t_{out} , $Q_{\text{out}}(t=0)$, c_0 , and S for ULE glass at 23 (1) °C. Corrections are due to the edge effect (giving up the assumption of infinite 2-dimensional plate).⁸

Name	Correction for t_{out} (%)	t_{out} in h	Correction for $Q_{\text{out}}(t=0)$ (%)	$Q_{\text{out}}(t=0)$ in Pa m ³ /s	c_0 in Pa m ³ /m ³	S in %
Plate 1	+1.2	28.3(7)	+0.1	5.15(72)	384(54)	1.82(26)
Plate 2	+2.6	108(4)	+0.2	2.63(48)	377(69)	1.79(33)
Plate 3	+2.2	26.4(6)	+0.5	2.70(34)	379(55)	1.84(26)

The values for D , S , and K are of the same order of magnitude like the results for other types of glasses obtained by permeation measurements carried out in the 1950s and 1960s.^{9–13}

For example, for Lithosil (SiO₂) from the Schott company, values are $S_{\text{Lithosil}} = 2.5\%$ and $D_{\text{Lithosil}}(25^\circ\text{C}) = 2.4 \times 10^{-8} \text{ cm}^2/\text{s}$, leading to $K_{\text{Lithosil}}(25^\circ\text{C}) = 6 \times 10^{-10} \text{ cm}^2/\text{s}$.¹⁴

From the values obtained in this paper, one can calculate that the initial diffusion of helium into a helium-free ULE glass causes a sink rate R (pumping speed or volume speed per area) of

$$R = 10^{-6} \times S \times \sqrt{\frac{D}{\pi \times t}} = \frac{2.13 \times 10^{-12}}{\sqrt{t/(s)}} \text{ m}^3 \text{ s}^{-1} \text{ cm}^{-2}, \quad (6)$$

i.e., $1.2 \times 10^{-13} \text{ m}^3 \text{ s}^{-1} \text{ cm}^{-2}$ at 5 min, $3.6 \times 10^{-14} \text{ m}^3 \text{ s}^{-1} \text{ cm}^{-2}$ at 1 h, and $1.1 \times 10^{-14} \text{ m}^3 \text{ s}^{-1} \text{ cm}^{-2}$ at 10 h.

New optical pressure standards are targeting uncertainties of several ppm.^{1–3} If He is used for a refractivity measurement in combination with ULE glass, the changes of the size of the ULE cavity spacer caused by He-absorption, but also the pumping speed of the ULE surfaces, need to be considered.

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