An exact comparison of results from samples subjected to hydrostatic pressure with results from samples subjected to pressure in the uniaxial pressure transmitting device is not possible since densification of SiO_2 glass cannot be recorded below 20 kilobars in the uniaxial pressure-transmitting device. However, one point obtained from a sample of silica glass subjected to 20 kilobars pressure in the uniaxial pressure-transmitting device at $\sim 700°$ C is shown in Figure 6. The curve for the data obtained from runs subjected to hydrostatic pressure has been extrapolated to 20 kilobars and seems to be in the same region as the 20 kilobar point from the sample subjected to pressure in the uniaxial pressure-transmitting device.

Additional results from runs of silica glass samples which were subjected to hydrostatic pressure are tabulated in Table 2. These results establish beyond doubt that silica glass can be substantially densified when subjected to hydrostatic pressure at temperatures greater than approximately 500° C.

One further experiment was made to see if a large sample of glass could be densified. A glass sample ($0{\cdot}12 \times 0{\cdot}25 \times 0{\cdot}60$ in) of approximate window glass composition was wrapped in gold foil and subjected to nine kilobars hydrostatic pressure at 600° C for 18 hours. The index of refraction and density were measured in the laboratories of Bausch & Lomb, Incorporated, Rochester, New York. The initial index of refraction at the sodium D line and density were respectively 1·51721 and 2·5280 g/cm^3, whereas these values for the densified sample were respectively 1·52981 and 2·5925 g/cm^3.

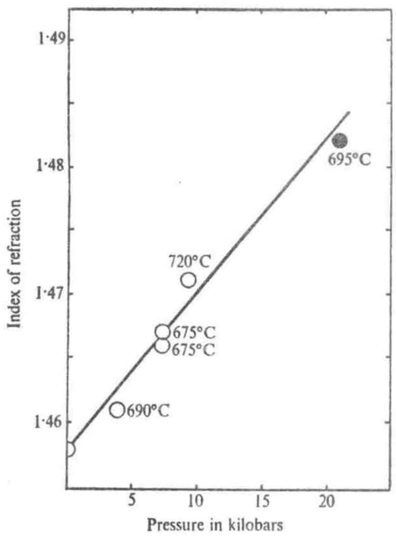

Figure 6. *The densification of SiO_2 glass which has been subjected to hydrostatic pressure*

○ Hydrostatic pressure-vessel
● Uniaxial pressure-transmitting device

Table 2. *Results from SiO_2 glass samples densified under hydrostatic pressure*

Pressure (Kilobars)	*Temperature (° C)*	*Index of refraction* of sample after removal from the pressure vessel*
4·0	300	1·458
4·0	400	1·458
3·9	690	1·461
4·9	876	1·469
6·8	612	1·462
6·7	847	1·470
7·2	600	1·464
7·3	10	1·459
7·3	675	1·467
7·4	675	1·466
7·9	600	1·469
9·3	720	1·472

*The index of refraction was measured at the sodium D line. The values are accurate within $\pm 0{\cdot}002$ of a unit.

Kinetics of densification

At a given temperature and pressure the densification proceeds very rapidly and the glasses attain a density which is apparently characteristic of the pressure–temperature conditions to which they are exposed. For silica glass this 'characteristic' density is attained in less than one minute. This was shown by a series of runs from 15 seconds to several days. Typical results are tabulated in Table 3. The initial refractive index of silica glass is 1·458.

The sample was first heated to the temperature indicated in Table 3, the pressure applied, the sample quenched, and the pressure released. The 'Duration of the run' refers to the time which elapsed between the application of the pressure and the quench.

The alkali silicate glasses which were studied also densified rapidly, although not quite as rapidly as silica glass. The rate of densification of $Na_2O.0{\cdot}1MgO.2{\cdot}9SiO_2$ glass is shown in Figure 7. The index of refraction for glasses of this composition which were subjected to 26 kilobars pressure at 300 and 500° C is plotted as a function of the time of the run. It is seen that the rate of densification is more rapid at 500 than 300° C.

Reversibility of densification

In order to prove that there is a metastable equilibrium structural state for glass which is a function of the

Table 3. *Rate of densification of silica glass*

Pressure (Kilobars)	*Temperature (° C)*	*Duration of run*	*Refractive index after pressure is released*
40	25	15 seconds	$1{\cdot}470 \pm 0{\cdot}005$
40	25	22 hours	$1{\cdot}470 \pm 0{\cdot}006$
40	25	5 days	$1{\cdot}470 \pm 0{\cdot}005$
30	600	15 seconds	$1{\cdot}475 \pm 0{\cdot}005$
30	600	2 minutes	$1{\cdot}480 \pm 0{\cdot}006$
30	600	6 minutes	$1{\cdot}480 \pm 0{\cdot}005$
30	600	50 minutes	$1{\cdot}482 \pm 0{\cdot}005$

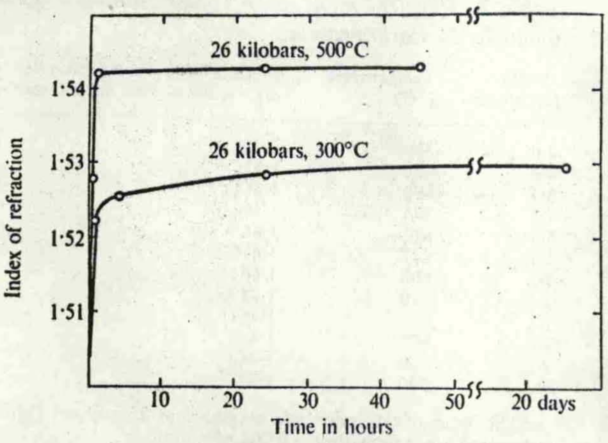

Figure 7. *The rate of densification of $Na_2O.0{\cdot}1MgO.2{\cdot}9SiO_2$ glass*

pressure–temperature environment to which the glass is exposed, it would be necessary to show that the curves in Figures 1–6 could be approached by samples whose initial densities, before application of pressure, were higher and lower than the assumed equilibrium density. However, the densified SiO_2 glass samples were extremely 'stable', and it was not possible to decrease the density of densified SiO_2 glass by subjecting it to pressures below which the glass was originally densified. This is, of course, analogous to the difficulty of causing a dense crystalline phase to revert to a less dense one at low temperature. Two samples of silica glass which had an index of refraction of 1·52 were subjected respectively to 20 and 30 kilobars pressure at 600° C for 12 hours; however, there was no perceptible change in the indices of refraction for these samples.

Densified samples of $Na_2O.Al_2O_3.4SiO_2$ glass could, however, be caused to revert to a lower density under pressure. Samples whose initial refractive indices were $1{\cdot}500 \pm 0{\cdot}002$ were subjected to 80 kilobars pressure at 600° C to increase their indices of refraction to $1{\cdot}565 \pm 0{\cdot}002$. These samples were then subjected respectively to 20 and 40 kilobars pressure at 600° C, their indices of refraction decreased respectively to $1{\cdot}527 \pm 0{\cdot}002$ and $1{\cdot}542 \pm 0{\cdot}002$. From Figure 3 it is seen that samples whose initial indices of refraction are $1{\cdot}500 \pm 0{\cdot}002$ will increase their indices of refraction to $1{\cdot}522 \pm 0{\cdot}003$ and $1{\cdot}542 \pm 0{\cdot}003$ when subjected respectively to 20 and 40 kilobars pressure at 585° C.

Physical properties of densified glasses: the relationship between the refractive index and density of progressively densified glasses

The relationship between the index of refraction and density of some progressively densified glasses in the system $Na_2O.Al_2O_3.SiO_2$ is shown in Figure 8. The relationship for SiO_2 glass and glasses in the series $R_2O.0{\cdot}1MgO.2{\cdot}9SiO_2$ has already been reported, and is linear within the experimental error.[17] The relationship was determined by measuring the density of a cube of approximately one millimetre of the respective specimens and then measuring the refractive index of the same piece. The value of the refractive indices represents an 'average' value approximately centred around a spread of $\pm 0{\cdot}005$ of a unit for SiO_2 glass and $\pm 0{\cdot}003$ of a unit for the alkali silicate and sodium aluminosilicate glasses respectively. It is noted that the densified alkali silicate and sodium aluminosilicate glass samples were more homogeneous than the densified SiO_2 glass samples. The spread of the index of refraction in the respective glasses would reflect a spread in the density of approximately $\pm 0{\cdot}02$ g/cm^3. Thus, the measured density of each specimen is actually an 'average' density.

Infra-red absorption spectra

Infra-red absorption spectra in the region from 2–25 μ were obtained for progressively densified SiO_2 glass and glasses in the series $(2\text{–}X)Na_2O.XAl_2O_3(6\text{–}2X)SiO_2$. The spectra from 2–15 μ are shown in Figures 9 and 10, and the spectra from 11–25 μ are shown in Figures 11 and 12.

Two significant changes occur in the SiO_2 absorption band in the 9 μ region and are shown in Figure 9. Firstly, the position of the maximum absorption shifts to higher wavelengths as the density increases; secondly, there is a very marked broadening of the

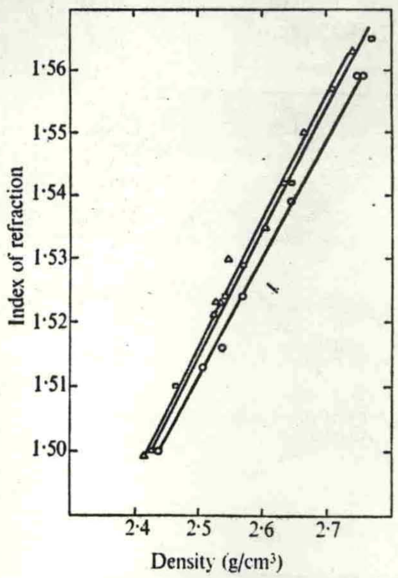

Figure 8. *The relationship between the density and index of refraction of glasses in the series $(2\text{–}X)Na_2O.XAl_2O_3.(6\text{–}2X)SiO_2$*

○ X = 0·50
△ X = 1·0
□ X = 1·1