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THE EFFECT OF TENSILE STRAIN ON HELIUM PEEMEATION OF GLASS AND SEVERAL REFRACTORY CERAMICS

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Berkeley, California

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ABSTRACT

A mass spectrometer was used to measure helium permeation through the walls of tubes made of Pyrex glass, fused silica, refractory mullite, and high-alumina ceramics. Application of tensile strains near the breaking point gave an instantaneous and reversible increase in steady-state helium permeation, on the order of 1%. The strain dependence of the permeation in Pyrex, fused silica, and mullite could be expressed as

$$K_e = K \exp[M\epsilon/RT]$$
,

where K_{ϵ} and K are the strained and unstrained permeability coefficient, respectively. M is the strain coefficient of the activation energy of permeation, ϵ is the total strain, and R and T are respectively the universal gas constant and absolute temperature. The observed strain dependence of permeation in these materials was similar and presumably due to uniaxial strain in the glassy phase with a resultant decrease in the activation energy of helium permeation. The high-purity alumina showed no detectable helium permeation in the stressed or unstressed state within the temperature range investigated.

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i. INTRODUCTION

Recent developments in nuclear technology have included the use of refractory ceramic materials as fuel and structural members in reactors. ¹ Ceramic materials are also used as vacuum tight envelopes in many electronic applications. It is therefore important to know the permeability of the ceramic to gases. Usually permeability is determined experimentally as a function of temperature. However, mechanical and thermal stresses are often imposed on ceramic components during use. In metals elastic strain can cause considerable change in the self-diffusion coefficient. ² Therefore, it is possible that the distortion of a ceramic by elastic strain may produce significant changes in the gaseous permeation rate. A study of the stress dependence as well as the temperature dependence of gaseous permeation through ceramic materials would provide a useful and a more thorough understanding of these materials.

The study presented here made use of a mass spectrometer to measure the temperature and stress dependence of the steady-state permeation rate of helium through several ceramic materials.

II. PERMEABILITY COEFFICIENT

For the case of the steady-state isothermal permeation of helium through a membrane, the amount (q) per unit time (t) permeating a membrane is given by 3

$$\frac{\mathbf{q}}{\mathbf{t}} = \frac{\mathbf{K} \mathbf{A} \Delta \mathbf{P}}{\ell},\tag{1}$$

where

K = permeability coefficient $\frac{\text{atom}}{\text{dec-cm}}$ A = area of the membrane $\frac{\text{cm}^2}{\text{cm}^2}$.

AP = gas pressure difference across the membrane (atm).

 $\ell =$ thickness of the membrane (cm).

For a given membrane through which a gas is permeating it is evident that a change in the permeability coefficient, K, will result in a change in the permeation rate, q/t. Therefore, the dependence of K upon temperature and strain is an important consideration.

A. Temperature Dependence

In glasses and polycrystalline ceramic materials normally considered vacuum tight. K. usually has an exponential dependence on temperature represented by

$$K = K_0 \exp - (\Delta H_K/RT), \qquad (2)$$

where

Ko = constant (atom/sec-cm).

ΔH_E = activation energy of permeation (cal/mole),

R = gas constant (cal/deg-mole),

T = absolute temperature (°K).

If ΔH_K is independent of temperature, a plot of logarithm of K against reciprocal temperature yields a straight line with a slope proportional to ΔH_K .

The permeability coefficient K is defined by

$$K = DS, (3)$$

where

D = diffusion coefficient (cm²/sec),

S = solubility coefficient (atoms/cc).

Normally, within experimental errors,

$$D = D_0 \exp(-\Delta H_D/RT)$$
 and $S = S_0 \exp(-\Delta H_S/RT)$, (4)

where

 ΔH_D = activation energy of diffusion (cal/mole),

 ΔH_S = heat of solution (cal/mole),

 $D_0 = constant \quad (cm^2/sec),$

 $S_0 = constant$ (atoms/cc).

It follows, then,

$$K = K_0 \exp[-(\Delta H_D + \Delta H_S)/RT], \qquad (5)$$

where

$$K_0 = (D_0)(S_0).$$

In the diffusion of helium through glass there is essentially hard-sphere interaction of atoms. Diffusing helium atoms can be pictured as squeezing between barriers in the glass network in passing from one solution site to another. The activation energy of diffusion is the thermal energy the helium atom must have to pass through the constrictions between solution sites. The activation energy of solution is the energy taken up or lost by the helium atom as it passes from the state of a free gas atom to a state of solution on the surface of a solid.

B. Strain Dependence

Carman has generalized that for diffusion of rare gases in glass, values of the permeability coefficient afford a fairly good relative measure of the diffusion coefficient. ⁵ For instance, there is a difference of several orders of magnitude between the permeability coefficients for helium and neon diffusion in vycor glass, whereas the solubility for those two gases differs by only a factor of two. ⁶ From Eq. (3) it is apparent that the change in permeability coefficient would be due to a change in the diffusion coefficient. The larger size of the neon atom evidently decreases the value of the diffusion coefficient. A similar phenomenon is observed for diffusion of helium in various glasses as the

constriction between solution sites are plugged with alkali and alkaline earth oxides.3.7

Therefore, it can be assumed that for rare gas diffusion in glass only the diffusion and permeability coefficients will be changed significantly if the constrictions between solution sites are dilated by tensile strain. Using the preceding assumption and a theory dealing with diffusion in strained systems developed by Girifalco and Grimes, one can obtain the expression⁸

$$K = K \exp(M\epsilon/RT).$$
 (6)

where

 K_{ε} = permeability coefficient measured for diffusion in a strained material.

K = permeability coefficient measured for diffusion in an unstrained material.

M = a constant which is a function of $(\partial \phi/\partial c)$, where ϕ is the potential energy of the system, and

e = total strain imposed on a system.

The total strain for the case of uniaxial tensile stress in a homogeneous isotropic system can be expressed as

$$\epsilon = (1-2\mu) \sigma_{\mathrm{T}}/\mathrm{E},$$
 (7)

where

μ = Poisson's ratio,

 $\sigma_{\rm cp}$ = uniaxial tensile stress, and

E = Young's modulus at the temperature of interest.

Comparison of Eq's. (2) and (6) shows that M is essentially a strain coefficient of the activation energy of permeation. Furthermore, from Eq. (6) it follows that

$$\ln (K_{\epsilon}/K) = M\epsilon/RT.$$
 (8)

If Eq. (6) is valid for a given system and K_{ϵ} and K are measured at several temperatures and strains, a plot of $\ln K_{\epsilon}/K$ against ϵ/T would yield a straight line having a slope equal to M/R and passing through the origin.

III. EXPERIMENTAL PROCEDURE

The temperature and tensile-strain dependence of helium permeation of several ceramic materials was studied by use of the specimens, apparatus, and procedure as described below.

A. Specimens

The specimens' composition and physical properties are given in Tables 1 and II. An x-ray-diffraction study of the mullite materials showed noullite and alumina present in the material designated mullite 1 but only mullite in mullite 2. Figure 1 shows photomicrographs of the mullite and alumina materials.

The specimens were obtained commercially as 9-in. -long tubes of about 0.5 in. o.d. By use of a lathe and a diamond saw, a 0.5-in.-long reduced section was ground into the middle of each of four specimens of mullite 1 and two of alumina 2. In two additional specimens of mullite 1 there was a slightly different modification consisting of two 1/8-in. -long reduced sections at the middle of the tube 1/4 in. apart (the tube between being of the original diameter). The different modifications of mullite 1 specimens were used to determine if the stress concentration at the fillets influenced the data. The wall thicknesses of the specimens are given in Table 1.

B. Apparatus

The apparatus consisted of a resistance-heated vacuum furnace, a loading device, and a mass spectrometer. A schematic diagram of the entire apparatus is shown in Fig. 2 and a photograph of the assembled furnace and loading device in Fig. 3.

The furnace could be evacuated to 10⁻⁴ mm Hg or filled with 1 atmospher of helium. The heating coil had a constant-temperature zone of about 0.5 in. for most operating temperatures. Power was supplied through a voltage stabilizer and a manually controlled auto transformer. The temperature was measured by a Pt/Pt-10%Rh thermocouple at the center of the constant-temperature zone and touching the inside wall of the specimen.

Essentially uniaxial tensile stress was induced in the specimens because of the ball and socket relationship of the grips and the pressure plates. The load on the specimen was varied by control of the air pressure in the diaphragm cylinder.

The mass spectrometer was a Consolidated Electrodynamics Corp. helium leak detector, model 24-120. Continuous monitoring of the helium partial pressure inside the specimen allowed measurement of the transient and steady-state permeation at various temperatures and stress levels. The mass spectrometer was calibrated, after each specimen had been tested, by use of a known helium flow rate.

C. Procedure

Techniques for employment of the mass spectrometer for permeation studies have been discussed elsewhere. 3,7 The temperature dependence of the helium permeation rate through unstressed mullite 1 was determined in four specimens with a thin-wall reduced section at the center. The specimens were placed consecutively in the apparatus with the reduced section in the constant-temperature zone of the furnace. With the furnace evacuated, a steady state of specimen temperature and mass spectrometer background was obtained; then the furnace was filled with 1 atmosphere of pure helium. The temperature was maintained within ± 5°C by adjusting the auto transformer. After a short time interval the mass spectrometer output would start to rise. Usually in less than an hour the output would cease to rise; steady-state permeation through the reduced section had been achieved. From the difference between steady-state readings and from a calibration curve for the mass spectrometer, the permeation rate of helium could be determined. This technique was used at various temperatures between 500 and 1000°C.

Shortly after steady-state permeation through the reduced section had been achieved, a very slow increase in permeation rate would begin, which continued until a second steady state was reached in 12 to 18 hours. The second increase in permeation rate (usually about 5% of the total) was due to permeation through the thick-walled portion of the specimen.

The tensile-stress dependence of permeation was studied, at various temperatures, in one specimen each of mullite 1 and Pyrex and two specimens each of the other four materials with unmodified configuration. In addition, two specimens of mullite 1 with a 0.5-in, reduced section and two with two 1/8-in, reduced sections were also tested.

With all specimens the technique described previously was used to obtain final steady-state permeation. In the mullite and glasses the second steady-state flow of helium through the tube wall was achieved in about 18 hours.

After steady-state permeation had been attained, a tensile load was rapidly applied to the specimen. The mass spectrometer output immediately increased to a new steady-state level, indicating a greater helium flow in the stressed condition. Removal of the load resulted in an equal decrease in flow rate. One-minute loading cycles were repeated from 12 to 15 times at each load setting and the average values of the flow rate in the stressed and unstressed conditions were calculated. Several different loads were applied at various temperatures between 214 and 347°C for the Pyrex and fused silica and between 553 and 939°C for the mullite. Tests were conducted on the alumina 2, similar to those described for the mullite and glass specimens, at temperatures between 25 and 963°C. Tests of alumina 1 were conducted as for alumina 2, but were limited to temperatures below 300°C because of its greater wall thickness and thermal conductivity, which resulted in high temperatures at the silicon rubber O-ring seals between the tube and the furnace.

Loading times varying from 0.1 second to 15 minutes were used to study the loading-rate dependence of the permeation rate. Specimens usually failed in tension at the center of the furnace at stress levels of about 3000 psi for glass, 6000 psi for mullite, and 12,000 psi for alumina.

After a specimen had failed the wall thickness was measured with a ball micrometer and the cross-sectional area calculated. Stress levels were calculated by dividing load by the cross-sectional area.

IV. RESULTS

The data obtained for helium diffusion through the four unstressed specimens of mullite 1, each containing a reduced section, is represented in Fig. 4 as a plot of ln K against 10,000/T. From the slope of a line drawn through the data points an activation energy of 8550 cal/mole was calculated.

The data obtained from tests of specimens of mullite 1 and 2 without reduced sections is represented in Fig. 5 as a plot of $\ln K_c/K$ against ϵ/T . The same parameters were used to represent data obtained from tests of specimens of mullite 1 which had a reduced section, Pyrex and fused silica as shown in Fig. 6. The linear relationships shown in Figs. 5 and 6 indicate the validity of Eq. (6) for helium permeation of the materials tested. The maximum strains applied caused increases in the permeation rate on the order of 1%.

The calculation of the permeability coefficients of Figs. 4, 5, and 6 were carried out with the assumption that most of the helium passed through the portion of the specimen at maximum temperature as indicated by the thermocouple. Strain (c) in the specimens was calculated by using the temperature dependence of Young's modulus for the materials as shown in Fig. 7.

No loading-rate dependence was found in any specimen. The effect of the stress concentration at the fillets of the reduced section of mullite 1 specimens was negligible.

No helium permeation of the alumina materials was detected under any of the experimental conditions.

V. DISCUSSION

The linearity of the data of Fig. 4 indicates that an activated permeation process was dominant. Finding a nonlinear T^{1/2} dependence of permeation rate supported the conclusion that there was no significant contribution from Knudsen flow through cracks and other channels. Diffusion in polycrystalline ceramic materials occurs predominantly in the boundary that usually exists between grains. 12 It is reasonable,

therefore, to assume that permeation of helium through the polycrystalline mullite occurred primarily by way of the continuous glassy phase and the closed porosity.

Alternose has developed an empirical relationship between the activation energy of permeation of helium and the amount of glass-forming compounds in a glass. 7 The expression is written as

$$\Delta H_{K} = -260m + 30 \times 10^{3} \text{ cal/mole,}$$
 (9)

where m is the amount of glass former in mole %.

The activation energy of permeation of helium through mullite 1 was determined to be 8550 cal/mole, and from Eq. (9), m was found to equal 82.5 mole % glass former. Within the uncertainty of the measurements involved it is reasonable that the glassy phase of mullite 1 may have had a glass-former content of that magnitude in the form of SiO₂. Therefore, the rate-determining step in the permeation process was probably diffusion through the glassy matrix.

The activation energy of helium permeation in mullite 2 was not determined, but qualitative observations at various temperatures indicated that an activated permeation process was occurring. Furthermore, the permeability coefficient, K, of mullite 2 was smaller than that of mullite 1. This was indicated by a much smaller helium flow rate per unit thickness through mullite 2 than mullite 1 at a given temperature. A higher activation energy in mullite 2 may contribute to the findings; however, the most reasonable explanation is that because of the well-developed crystalline phase, as shown in Fig. 1, the effective path length for diffusion through the glassy phase is much longer in mullite 2 than in mullite 1.

The activation energies of helium permeation in fused silica and Pyrex glass are reported to be much lower than that measured for mullite 1. The lower activation energy and the absence of a relatively impermeable crystalline phase caused the helium permeation rates through the glasses at lower temperatures to be comparable to those in mullite at much higher temperatures.

There is a linear relationable between data points at any one temperature (Fig. 5), but none between points for different temperatures. This situation was caused by errors introduced in measurements and assumptions made. However, the errors were systematic and the linearity of data at one temperature supports the conclusion that the permeation rate does have an exponential dependence on strains.

Data points obtained at different temperatures had two major sources of scatter. First, the temperature gradient was different at each indicated temperature. Since only the indicated temperature at the center of the specimen was used in calculations, scatter of data from tests at different temperatures resulted. Secondly, the Young's modulus temperature dependence obtained from the literature was a rather crude approximation because the mullite material used for the modulus determination probably differed in microstructure and composition from materials used in our work. The Young's modulus changes rapidly in the temperature range of the experiments on the mullite materials, so that assumption of an incorrect temperature dependence would result in discrepancies between data taken at different temperatures.

The first source of scatter was reduced by using specimens of mullite 1 with a reduced section that could be held within ± 10°C of the indicated temperature. Significant stress levels existed only in the reduced section. The second source of scatter was reduced by the assumption of a hypothetical temperature dependence of Young's modulus slightly different from but reasonably close to that reported by Wachtman as shown in Fig. 7. These reductions in scatter produced a more linear correlation of data for mullite 1 at different temperatures as shown in Fig. 6.

Straight lines have been drawn through the origin of Fig. 6 passing through the data points for mullite 1, Pyrex, and fused silica. The validity of Eq. (6) was indicated by the linearity of the data. The M/R values obtained from the slopes were 6.54× 10⁴ °K for mullite 1, 1.66×10⁴ for Pyrex, and 1.29×10⁴ for fused silica.

Figure 5 shows the similarity of strain dependence of helium permeation of both mullite 1 and 2. Therefore, to a first approximation, the M/R value in those materials

was assumed to be essentially the same.

The relatively small temperature interval used in tests of Pyrex and fused silica resulted in much less scatter of data than obtained from mullite without reduced sections. The M/R values of the four widely different materials tested differed by only a factor of about 4. This fact suggests that the effect measured stems from a common mechanism. Since the glassy structure is the only common component in the materials, the observed strain dependence of permeation rate may reasonably be attributed to strain in the glass structure.

The values of M/R obtained in this work and values reported for diffusion in several other materials under compressive strain are shown in Table III. The reported M/R values for diffusion by greatly different mechanisms agree within two orders of magnitude and bracket the values obtained in this investigation. Since change in interatomic forces by induced strain is a reasonable explanation for the observed strain dependence of diffusion in the other systems, it seems reasonable from the discussion above that the same may be true for helium diffusion in glass.

The mechanism envisioned for helium permeation in the glassy phases was successive jumps of the helium atom from one site of low potential energy to another through constrictions formed by strongly bonded ions where atomic interaction was maximum. 3, 7 A tensile strain induced in the glass would result in an overall increase in the distance between bonded ions and would decrease interaction with the helium atom. Therefore, the activation energy of permeation would be reduced.

No deterioration of the materials after repeated cycles of high stress levels was detected; the data obtained were independent of the strain history of the specimen. Furthermore, no loading-rate dependence of permeation was detected in any of the materials.

McAfee studied the effect of tensile and compressive strain on helium permeation of Pyrex glass at room temperature. Tensile strains calculated as greater Company of the Company

than 2×10⁻⁴ caused a very rapid increase in permeation rate. At static strains near the breaking point the permeation rate increased rapidly after an incubation time of several hours. McAfee concluded that submicroscopic flaws in the glass propagated when strained past a critical point and became gas-conducting paths. However, he did not rule out the possibility of permeation by an activation process through the glass structure for at least part of the way through the membrane.

The maximum longitudinal strains obtained in materials of this study were normally about 5×10⁻⁴; however, no increase in permeation rate after the initial change was observed even after 20 minutes of static strain. One possible explanation for this difference from McAfee's results is that although submicroscopic cracks do exist in the glassy material their operation as gas conductors is dependent on the thickness of the membrane. The wall thickness of specimens in this work was about eight times that that used by McAfee. Gracks may have been opened but the tortuosity and chance for discontinuity in the channels would have been much greater than in McAfee's specimens.

Permeation of helium could not be detected in the alumina materials even under a static stress level of 12,000 psi at a temperature of 825°C. Studt has observed bursts of helium gas permeating thin membranes of alumina at room temperature upon rapid application and removal of tensile stress. ¹³ His observations were partly explained by postulating the existence of cracks connecting closed porosity with the surfaces of the membrane. Under transient conditions of stress the cracks were opened and helium flowed through. After many cycles of stress, adsorption of helium atoms on the channel walls blocked further flow and caused the effect to disappear.

The negative results of this experiment, in contrast with Studt's observations, may have been due to the use of thicker membranes and alumina materials of different microstructure.

VI. SUMMARY AND CONCLUSIONS

The temperature and tensile strain dependence of permeation of helium through two types of glass and several polycrystal multiphase alumina and mullite materials was measured. Helium permeation of the glass and mullite materials

apparently occurred by an activated diffusion process through the glassy phase in the strained and unstrained condition.

The permeation rates of helium through the mullite and glass materials showed similar exponential temperature and strain dependence. The change in permeation rate upon stress application was evidently due to expansion of the glass network that was present in all the materials. This expansion reduced the activation energy of permeation of helium. The decrease in activation energy caused the helium permeation to increase rate on the order of 1% when tensile strains near the breaking point were induced in the materials.

All the effects measured were reversible, and neither load-rate dependence nor deterioration of the specimen under repeated application of tensile loads was detected.

No permeation of helium through two high-purity alumina materials was detected in either the stressed or unstressed condition.

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FOOTNOTES AND REFERENCES

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At the time this work was done, the writers were, respectively, Graduate Research Assistant and Associate Professor of Ceramic Engineering, Department of Mineral Technology, University of California. O. M. Stansfield is now research engineer, Solar Aircraft Company, San Diego, California.

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Table I. Physical properties of specimens.

| Material | Specimen wall thickness (in.) | | וכן | nysical pr | | | |
|-------------------------------|-------------------------------|------------------------------|-------------------------------|---------------------------|------------------|--|--------------------|
| | as received | at the reduced section | appdrent density (g/cc) | cloded porosity (%) | open porosity | Young's modulus at 25°C (psi×10 ⁻⁶) | Poissoris ratio |
| alumina 1 | 0.080 | 44 48 | 3.77 | 6 | 0.0 | ₩ 43 ₩ | *** |
| alumina 2 | 0.060 | 0.030 | 3.69 | 6 | 0.0 | *** | 40 00 |
| mullite i | 0.120 | 0.040 | 2.44 | 18 | 0.0 | (b) 20.7 | 0.25 |
| mullite 2 | 0.117 | *** | 2.74 | 13 | 0.0 | (b) 20.7 | 0.25 |
| fused silica | 0.041 | | 2.21 | 0 | 0.0 | (c) 10.6 | (c) 0.17 |
| Pyrex glass (Corning 7740) | 0.036 | *** *** | 2.19 | 0 | 0.0 | (c) 9.2 | (c) 0.23 |

⁽a) Determined by a Beckman air comparison pycnometer, analytical balance, and mercury displacement volumeter.

⁽b) Reference 9.

⁽c) Reference 10.

Table II. Composition of specimens.

Chemical composition (Wt %) (analysis supplied by manufacturers)

| Material | A1203 | SiO2 | Fe ₂ O ₃ | TiO2 | Na ₂ O | CuO | MgO | B ₂ O ₃ |
|---------------|---------------|-------|--------------------------------|-----------------------------------|-------------------|-----------------|----------|-------------------------------|
| alumina 1 | 99.7 | 0.2 | ஷ். கூரு | and the sepa | 0.1 | 100 min 475 | an an en | |
| alumina 2 | 95.2 | 4.0 | 0,2 | *** | 0.1 | 0.2 | 0.3 | at to the |
| müllite l | 54 | 43 | Commence and the second | Taraka Marahagan, J. S., 1989 - 1 | <u> </u> | · | | 20 mi mi mi |
| mullite 2 | 58.9 | 36.5 | 0.9 | 1.2 | 1.2 | 1.1 | 0.4 | dip one air |
| fused silica | date date det | ≈ 100 | and sup issue | *** | 40 th 40 | 85 96 46 | * * * | |
| Pyrex glass | 2.0 | 80.6 | 40 m. di | nd 40 da | 4.4 | | | 11.9 |
| (Corning 774) | 0) | | | | | | | |

Table III. Comparison of values of M/P for various solids.

| Material | Diffusing species | Type of strain | M/P (°K) | Reference |
|----------------------------|-------------------|-----------------------------------|-------------|-----------|
| Mercury(liquid) | mercury | isostatic | 1,970 | 8 |
| Gallium (liquid) | gallium | compression isostatic compression | 1,970 | 8 |
| Sodium | sodium | isostatic compression | 10,130 | 8 |
| Fused silica | helium | uniaxial tension | 12,900 | this work |
| Pyrex glass (Corning 7740) | helium | uniaxial tension | 16,600 | this work |
| Refractory mullite | helium | uniaxial tensile | 65,400 | this work |
| Silver bromide | silver | isostatic compression | 73,340 | 8 |
| Lead | lead | isostatic compression | 73,590 | 8 |

FIGURE LEGENDS

- Fig. 1. Photomicrographs showing grain and pore structure of (a) alumina 1.

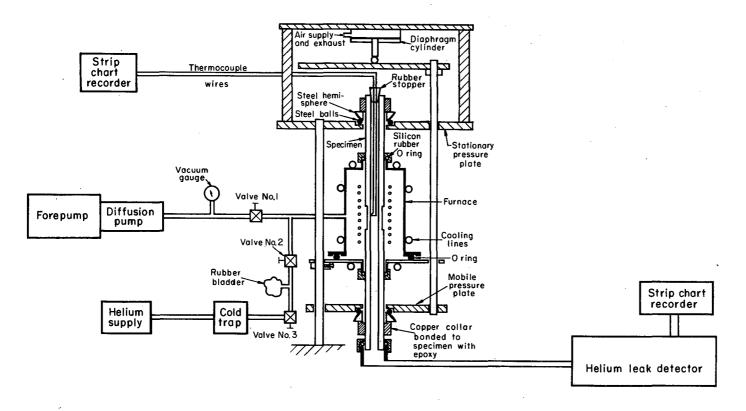
 (b) alumina 2, (c) mullite 1, and (d) mullite 2.
- Fig. 2. Schematic representation of the apparatus for the permeation study.
- Fig. 3. Photograph of the loading device and furnace with a specimen in place.
- Fig. 4. Temperature dependence of the helium permeability coefficient for mullite 1.
- Fig. 5. Logarithm of the fractional change in the helium permeability coefficient as a function of strain and reciprocal temperature for mullite 1 and mullite 2.

 These data were obtained from tubes as received from the manufacturer, therefore no permeation area at a uniform temperature could be determined.
- Fig. 6. Logarithm of the fractional change in the helium permeability coefficient as a function of strain and reciprocal temperature.

O: mullite 1; A: Pyrex glass; A: fused silica.

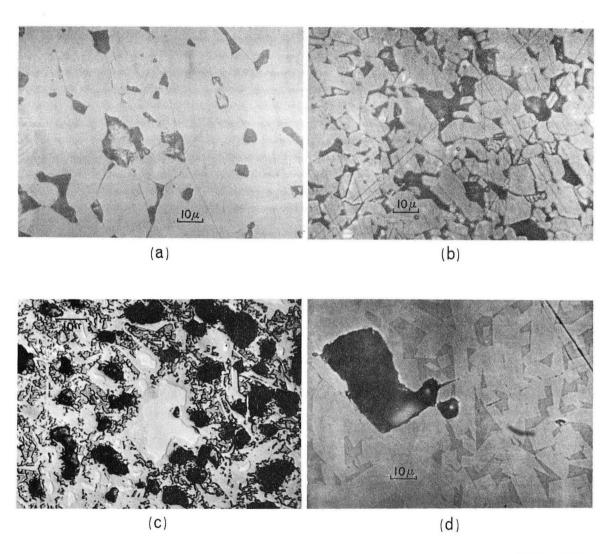
Fig. 7. Fractional change in Young's modulus as a function of temperature for several ceramic materials.

| - | fused silica (Ref. 11) |
|-------------------------------|---|
| Authors 44 46 valence -100-de | Pyrex glass (Ref. 11) |
| | Mullite (Ref. 9) |
| *** | assumed for mullite 1 specimens with reduced sections |



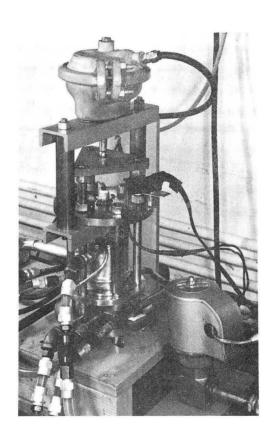
MUB-1835

Fig. 1.



ZN-3788

Fig. 2.



ZN-3726

Fig. 3.

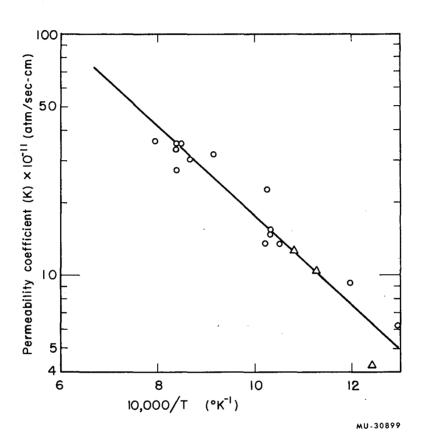


Fig. 4.

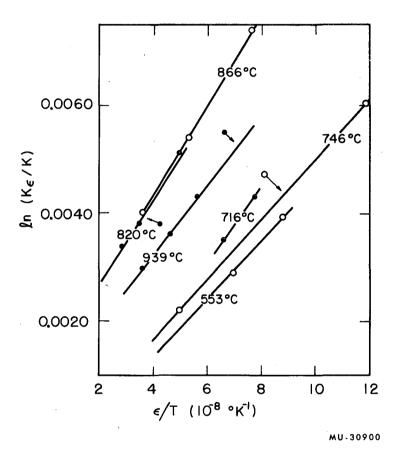


Fig. 5.

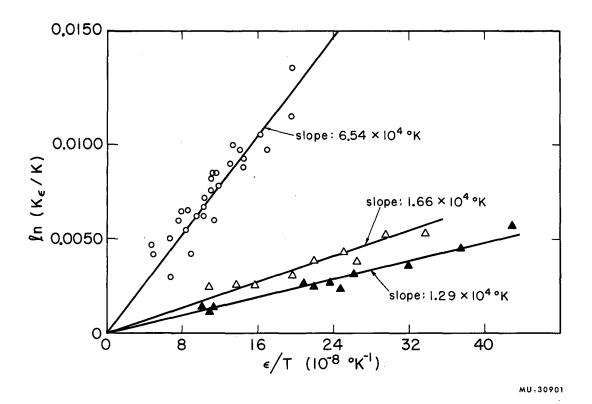


Fig. 6.

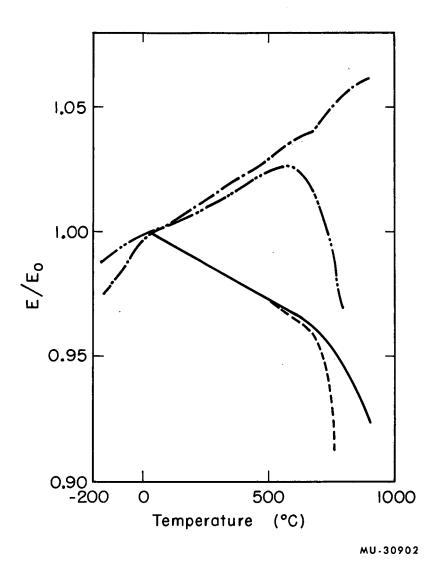


Fig. 7.

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