

THE BEHAVIOR OF NOBLE GASES IN SILICATE LIQUIDS: PRELIMINARY RESULTS. Gayle Lux, Dept. of Physics, University of California, Berkeley, CA 94720.

Studies of noble gas systematics in terrestrial and extraterrestrial samples are an important source of information in formulating and constraining geo- and cosmochemical models. Therefore, an understanding of the basic processes involved in noble gas incorporation and transport in natural samples is fundamental to deriving useful conclusions from these studies. Despite the importance of noble gases to scientific inquiry of the earth and solar system, our understanding of their behavior in silicate samples is incomplete. This report describes preliminary results of an attempt to determine the solubility of the noble gases in a range of silicate melts as a function of the intensive thermodynamic properties. Diffusion coefficients for the five noble gases in a tholeiite basalt at 1350°C are also reported.

Experimental Procedures. A suite of wet-chemically analyzed samples consisting of an andesite, leucite-basanite, tholeiite basalt, alkali-olivine basalt and ugandite were equilibrated in Pt crucibles for 72 hours at 1 atm in a vertical Pt resistance furnace with a noble gas mixture (49.97% each Ar and He; 0.02% each Ne, Kr, and Xe) and varying amounts of oxygen and nitrogen over the temperature range 1275-1500°C. The oxygen partial pressure was varied with temperature in order to maintain constant Fe^{3+}/Fe^{2+} in each melt (1). Also, the oxygen pressure was high (0.05-0.4 atm) to prevent loss of Fe from the melt into the Pt crucible. After equilibration the melts were quenched to a glass in the gas atmosphere of the experiment. The compositions of the samples were determined by electron microprobe to ascertain that they did not vary with experimental conditions. In addition, several experiments were conducted in the same manner described above except that the samples were suspended on Pt wire loops instead of in crucibles. Upon quenching, these experiments yielded samples that approximated spherical beads. For the diffusion experiment, a sample that had previously been purged of its noble gases was suspended in a cylindrical Pt crucible and run at 1350°C for 8 hours in the gas mixture. After quenching, the sample was cut in serial sections normal to its length. For all samples, noble gases released at 1600°C were analyzed mass spectrometrically using the isotope dilution technique.

Results and Discussion. Figure 1 shows solubility to be a strong function of melt composition, at least for the heavier gases Ar, Kr, and Xe. Helium is not plotted because it is suspected that it is lost from the glasses, either during quench or the time elapsed prior to analysis. The Henry's constants for noble gas solubilities fit the equation $K_i = a \exp(-br_i^2)$ where a and b are constants and r_i is the gas kinetic radius (2,3). The Henry's constants reported here are somewhat higher than those reported by other workers (3-5).

Figure 2 shows the same correlation holds for the diffusion coefficients as for the Henry's constants ($D_i = a \exp(br_i^2)$). These results indicate that at least at high temperatures, relative values of D among the noble gases do not differ by the several orders of magnitude previously believed (6); therefore, models invoking large differences in diffusion between noble gases to account for fractionation may not be valid.

The effect of temperature on solubility is generally weak for all the gases analyzed but does vary depending on the particular gas and melt composition. In general, however, solubility increases with increasing temperature, yielding positive heats of solution.

Figure 3 shows Henry's constants for samples of tholeiite basalt equilibrated at 1350°C on Pt wire loops compared to those run in crucibles. The crucible samples ($S/V=0$) had their surface removed prior to analysis. These results indicate that there is an apparent "surface effect": except for He, the greater the surface/volume, the greater the solubility of the gas. The effect decreases from Ne to Xe, which is opposite the trend induced by adsorption or disequilibrium. Helium shows the opposite trend as the other gases, a preferential loss of gas for greater S/V . This surface effect is especially strong at neon and may be a significant process of gas incorporation under appropriate conditions.

A good deal of attention has been given to elemental abundance patterns in marine basalts with the intention of characterizing the nature of mantle noble gas reservoirs. A number of workers have noted a characteristic elemental abundance pattern that consists of an enrichment of Ne and Xe and a slight depletion of Kr relative to ^{36}Ar when compared to atmospheric abundances (6-11). Some prefer to interpret this pattern as evidence for solar type gases in the mantle (7,11), while others see evidence for a planetary component (6,8). The incorporation of noble gases in igneous rocks erupting to the surface is certainly affected by multiple processes (crystal/liquid distribution coefficients, degree of partial melting, partial gas loss, contamination with atmospheric gases, etc.); however, simple models are sometimes useful. An obvious question is whether solubility fractionation alone can account for the enrichments. Figure 4 shows the effect of the solubility fractionation factor on three melts (andesite, tholeiite basalt, ugandite) in equilibrium with gas reservoirs (assumed effectively infinite) of solar, planetary, and air compositions. The pattern given by the basalts (6) is also shown. All three melts in equilibrium with a planetary reservoir yield dissolved gas compositions that generally agree with the measured basalt trend. Melts equilibrated with solar or atmospheric gas reservoirs show substantial disagreements at the heavier gases. Thus, for this simple model where the observed basalt patterns are related to a single parent gas by equilibrium solubility, the best agreement is obtained for a planetary reservoir. However, the significant scatter exhibited by the data requires either mixing of additional gas reservoirs or the operation of other fractionation processes than that considered here.

References. (1) Sack R.O. *et al.* (1980) *Contrib. Mineral. Petrol.* 75, 369. (2) Blander M. *et al.* (1980) *J. Phys. Chem.* 63, 1164. (3) Kirsten T. (1968) *J. Geophys. Res.* 73, 2807. (4) Hayatsu A. and Waboso C.E. (1982) *Abst. V Intl. Conf. on Geochron., Cosmochron., and Isotope Geol.*, Nikko, Japan, 139. (5) Jambon A. *et al.* (1982) *EOS* 63, 451. (6) Ozima M. and Alexander E.L. (1976) *Rev. Geophys. Space Phys.* 14, 385. (7) Dymond J. and Hogan L. (1973) *Earth Planet. Sci. Lett.* 20, 131. (8) Fisher D.E. (1974) *Geophys. Res. Lett.* 1, 161. (9) Kirsten T. *et al.* (1981) *Meteoritics* 16, 341. (10) Ozima M. and Zashu S. (1983) *Earth Planet. Sci. Lett.* 62, 24. (11) Hart R. *et al.* (1983) *Nature* 305, 403.

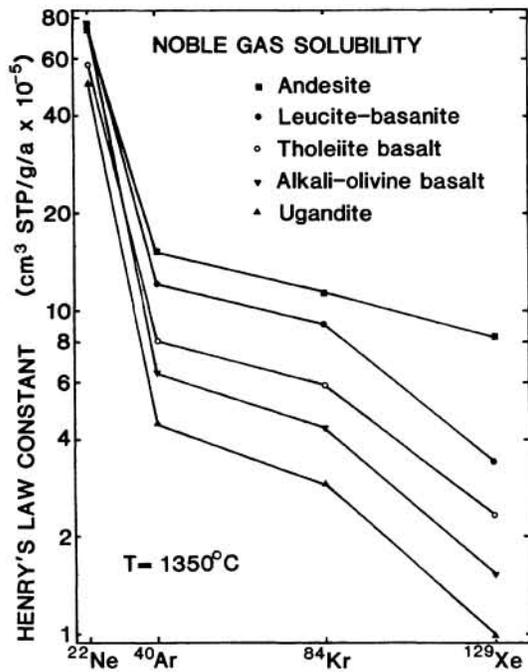


Fig. 1. Noble gas solubility in five melt compositions at 1350°C.

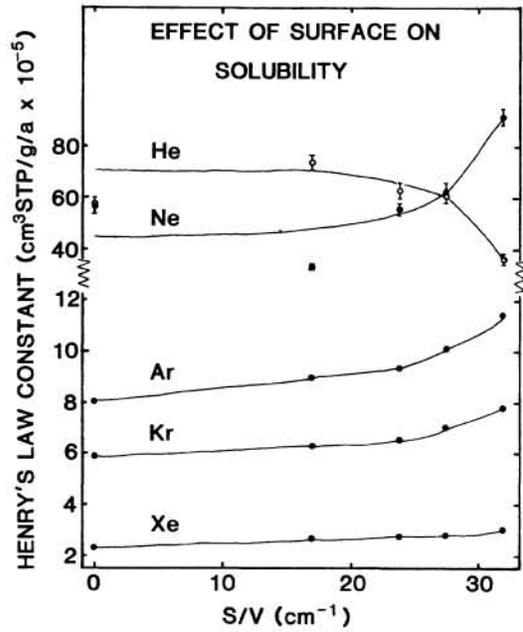


Fig. 3. Effect of surface/volume on measured gas solubility in a tholeiite basalt at 1350°C. Sample with S/V = 0 was equilibrated in a crucible and the surface was cut off after quenching. Samples with S/V > 0 were equilibrated on Pt wire loops; after quenching they approximated spheres.

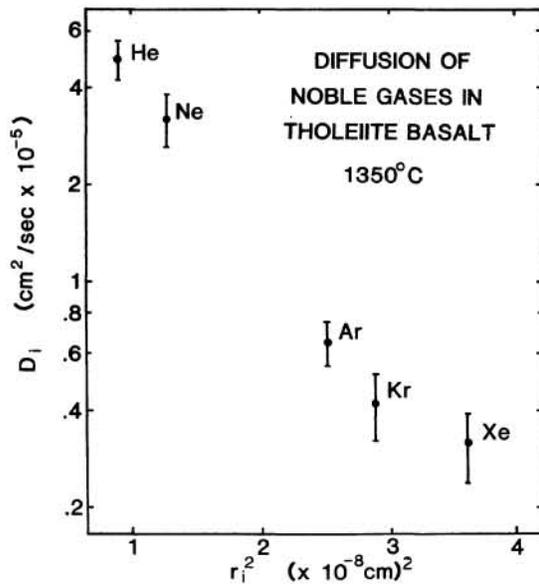


Fig. 2. Diffusion coefficients for the five noble gases in a tholeiite basalt at 1350°C.

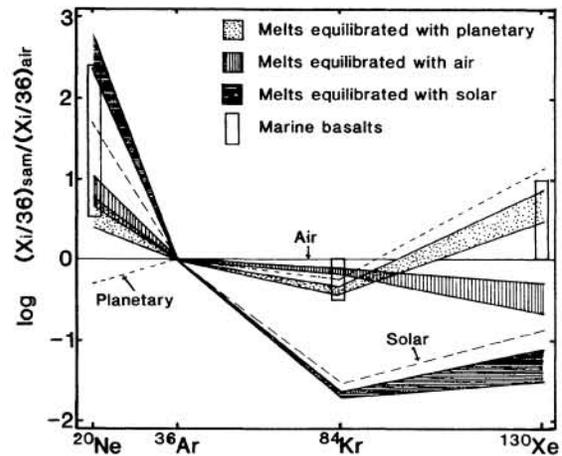


Fig. 4. Noble gas fractionation patterns (shaded) obtained by equilibrating the sample melts (K_V/K_{Ar}) with gas reservoirs of planetary, air and solar composition. Planetary, air and solar gas abundance patterns (lines) are also shown.