samples from St. Paul's Rocks, such as those on rare earth elements (*32*), and strontium isotopes (*33*), evidently were done on the spinel peridotite mylonites. Taken alone, neither the spinel peridotite mylonites nor the brown hornblende mylonites have compositions thought to be appropriate for the average composition of the mantle. The former, as a whole, are much too low and the latter much too high in alkalies and probably in U and Th to be appropriate for parental basalt materials, or to be consistent with oceanic heat flow. The pargasite-enriched mylonite (18-900, Table 1) and the estimated average of the mylonites are both more in accord with suggested average mantle compositions.

Clearly, additional analytical studies are needed to evaluate the idea that, as a whole, St. Paul's Rocks material is acceptable as compositionally representative of the mantle. We are continuing our study of this interesting intrusion, both on the suites of samples from the islets and on the extensive suites of samples obtained by dredging about the intrusion.

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References and Notes

1. Located at 0°56′N, 29°22′W: consists of five larger islets and four sizeable rocks separated by narrow channels; combined subaerial surface little more than 0.01 km^2. Published reports of St. Paul's Rocks were summarized by W. L. Tressler, S. Bershad, and W. H. Berninghausen in *U.S. Navy Hydrographic Office Technical Report HO 31, 1-63* (1956). We have followed their nomenclature for the individual islets.
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5. We landed on and sampled each major islet [Northwest, Northeast, Southeast, Southwest (Main), and South Islets, and the larger of the Southwest Rocks].
6. We have recently summarized some other aspects of the St. Paul's survey [*Oceanus* **12**(4), 2 (1966) and, in an abstract, *Geol. Soc. America, Program 1966 Meetings*, p. 138]. A complete cruise report and several articles are in preparation.
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14. The allanite is not metamict but rather well crystallized, shows strong yellow to reddish-brown pleochroism, and is strongly zoned. Some allanites contain considerable amounts of P or Mg, but this contains only 0.1 percent P and about 0.7 percent Mg. From this mineral in SE-13 (Table 2), the microprobe obtained high count rates for $L\alpha_1$ radiation of Ce and La; about 10 percent rare earth oxides, mainly Ce_2O_3 and La_2O_3, as well as between 0.5 and 2 percent ThO_2, are commonly found in allanites.
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18. $K\alpha_1$ radiation from Cl excitation was about 40 times as intense from the blue as from the brown hornblende; adequate standards are being sought for quantitative Cl determinations. The chloro-hornblende, pale blue to yellow pleochroic prisms, rims typical brown hornblende augen, especially commonly at contacts of brown hornblende with post-mylonitization scapolite-chloro-apatite veinlets.
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35. The samples described here are being entered into the collections of the U.S. National Museum under the following numbers: 110685 (7-327), 110686 (7-479), 110687 (18-900), and 110393-13 (SE-13).
36. The Brazilian government gave permission for the landings. For their help, we thank the officers, men and scientists of cruises 35 (1963) of R.V. *Chain* and 20 (1966) of R.V. *Atlantis II*. Harold Banks gave technical assistance and R. F. Fudali critically reviewed the manuscript. Eugene Jarosewich was the analyst for Table 1. At the Smithsonian Institution this study was assisted by Smithsonian research grants and by NASA; at the Woods Hole Oceanographic Institution, support came from the AEC, the ONR, and the NSF. All this support is gratefully acknowledged. This is contribution No. 1896 from the Woods Hole Oceanographic Institution.

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Diffusion of Water in Zeolites

Abstract. *The self-diffusion coefficient* D *of water occluded in samples of near-faujasite has been determined by pulsed field-gradient spin-echo nuclear magnetic resonance. The value of* D *in square centimeters per second* × *10^5 at 30°C is 1.34, 1.65, and 1.88 in the following zeolites, respectively: Na* X, *Ca* X, *and Ca* Y (X *and* Y *being an indication of ratio of silicon to aluminum in the zeolites). By comparison, the value of* D *in pure water at 30°C is 2.5* × *10^{-5} cm^2/sec. Arrhenius activation energies for* D *are 6.9, 6.8, and 5.6 kilocalories per mole, respectively, for the three faujasites and 5.0 kcal/mole for pure water. Thus, there appears to be little difference in diffusion behavior between free water and water occluded in faujasite.*

How the properties of a liquid are modified in the vicinity of a solid is a subject that has often been discussed. An interesting class of solids lending itself to an investigation of this question is that of the porous crystalline aluminosilicates known as zeolites or molecular sieves. An interesting property of a liquid, related to its structure, is diffusion. Since water normally fills the cages of a zeolite in equilibrium with the atmosphere, it seems natural to find that several studies have been devoted to self-diffusion of water in zeolites (*1, 2*).

Generally, it was found that the self-diffusion coefficient D of water occluded in these crystals was substantially smaller than that of pure water. Thus, Barrer and Fender (*2*) report a value of D equal to 1.26×10^{-7} cm^2/sec for water in chabazite at 45°C. This is more than 100 times less than the corresponding value for pure water.

These low figures are easily explained by the hindrance to motion of water molecules as they diffuse through the very small channels of the zeolites under study. However, one structure exists that presents wider channels and larger cavities than are found in the crystals studied thus far (*3*). This is faujasite (or near-faujasite in the synthetic form), a structure resulting from the stacking of SiO_4 and AlO_4 tetrahedra in a cubo-octahedron arrangement. Thus, cavities about 12 Å in diameter are formed that contain about 30 water molecules when the zeolite is

Table 1. Self-diffusion coefficient of water, $D \times 10^5$ cm^2/sec. T, temperature.

	Faujasites			Bulk water	
T (°C)	Na X	Ca X	Ca Y	This work	Simpson and Carr (*10*)
0				1.04	
10		0.74	0.81	1.44	1.36
20	1.00				1.85
25				2.23	
30	1.34	1.65	1.88		2.46
40	2.11	2.41	2.78	3.40	3.14
60	4.10	4.61	5.47	5.47	4.82
69	5.30				
70					5.78

equilibrated with the atmosphere. Four windows about 8 Å in diameter lead from each cavity to four similar adjacent cavities. Charge-compensating cations (for example, sodium) occupy various positions in the structure.

Because of the presence of these large accessible cavities, faujasite has recently received a great deal of attention as an adsorbent and catalyst. It was decided to use this material in order to reexamine the possible effect of the solid on liquid structure and to measure self-diffusion of water in faujasite by means of pulsed gradient spin-echo nuclear magnetic resonance (NMR), as described by Stejskal and Tanner (*4*). This method, which evolved from the original one of Carr and Purcell (*5*), has many advantages of speed and simplicity that make it much more desirable than the conventional techniques previously used in such work.

The apparatus we used has been described in detail (*6*). The basic spin-echo spectrometer was the model ELH-15 constructed by Magnion Inc., Burlington, Massachusetts. Magnetic field gradients G were applied by winding a pair of magnetically opposed coaxial coils on tapered forms with their axis parallel to the laboratory field. The coils were designed so as to produce as large a region of homogeneous gradient as possible. They were calibrated with pure water by assuming a value for D equal to 2.51×10^{-5} cm^2 sec^{-1} (*7*) and by determining G with the use of the expressions given by Abragam (*8*) which also relate D to the spin-echo amplitude data. The gradient switch was activated by the pulse output of two Tektronix type 163 pulse generators, which were referenced to the transmitter pulse sequence. Two near-faujasite samples were obtained from the Linde Company. They were in the sodium form and designated as Na X and Na Y, the main difference between the two being in the Si:Al ratio (approximating 1 for X and 2 for Y). The Ca X and Ca Y samples were prepared from those by ion exchange.

The self-diffusion coefficients of pure water and of water in the various zeolites are given in Table 1. Eight different gradients were used at each temperature, and the indicated value for D is an average of the eight values obtained. The data of Table 1 yield straight lines in plots of ln D versus $1/T$. Activation energies E_D corresponding to $D = D^* \exp(-E_D/RT)$ are 6.9, 6.8, 5.6, and 5.0 kcal/mole for water in Na X, Ca X, Ca Y, and bulk water, respectively. These minor differences in temperature coefficient are further compensated by corresponding minor variations in preexponential factors D^*, so that values of D for water in faujasite are only slightly lower (by less than a factor of 2 at low temperature) than the value of D in pure water.

This then is the main result of our investigation: as revealed by very similar values of D, the motion of water molecules in faujasite resembles very much that of pure water. It appears that the cavities and windows in the structure are large enough so that the influence of the crystal lattice becomes minor. It is important to note that the spin-echo NMR technique concerns itself with motion of protons over large distances that cover many cages and windows. Indeed the duration of a gradient pulse is of the order of 10^{-3} second, and during this time a water molecule will have diffused over a distance of the order of 10^{-4} cm.

Barrer *et al.* (*9*) have remarked that molecular sieves provide a macromolecular framework not unrelated to that found in many biological systems. If this is so, the results of this work may be of interest not only to those concerned with molecular sieves as adsorbents and catalysts. Finally, for studies of diffusion in these and other porous materials, the NMR method used here seems to have very wide applicability.

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Crystallization of Human Lysozyme

Abstract. *Lysozyme, isolated from the urine of patients with monocytic leukemia, has been crystallized as the chloride at* pH *4.5 and at* pH *10.5. The crystal forms of the human enzyme show certain similarities as well as distinct dissimilarities compared with the crystal forms of lysozyme chloride from hen's egg white.*

There are large quantities of lysozyme in the serum and urine (up to 4 g per day) of patients with monocytic and monomyelocytic leukemia (*1*). The enzyme has been isolated from the urine of several patients; physicochemical and immunochemical analyses indicate that it is a low-molecular-weight (≅ 14,000) basic protein with an isoelectric point at about *p*H 10.5. The lysozyme present in these serums and urines is apparently identical to that of normal human tears, saliva, leukocytes, and serum; but it is structurally different from the lysozyme of hen's egg white (*1*). Whereas the human and egg-white lysozymes are of similar molecular size and basicity, they differ significantly in amino acid composition, tryptic peptide ("fingerprint") patterns, antigenic structure, and enzymatic activities. When assayed with heat-killed *Micrococcus lysodeikticus* organisms, the activity of the human enzyme has been found to be 3 to 12 times greater than that of twice-crystallized egg-white enzyme.