

Fig. 2. Diagram indicating a way of terminating the continuous lattice.

the two formulations lies in the amount of electron correlation included.

The structure proposed, which is potentially unlimited in its spatial extent, could be terminated in the manner shown in Fig. 2. As for all other oxygen atoms (and hydrogen atoms) in the lattice, the formal charge on the terminating oxygen atoms is zero since they are forming one two-electron bond and two one-electron bonds to protons. It should be noted that the terminating oxygen atom is joined to two hydrogens of the main structure in such a way that the tetrahedral angle of HOH is maintained. That is, at the terminating oxygen atom the

H × O × H

angle remains tetrahedral as required by the overall structure, one normal electron-pair bond is formed, and the orbitals for the other six electrons remain noncoincident without any distortion from the tetrahedral conformation for either of the spin-sets.

This model would therefore suggest that polywater consists of micelles having the internal structure shown in Fig. 1. These would presumably be contained in a small amount of ordinary water to which they would be hydrogen bonded in a conventional way. So the material would be colloidal in character, which is a description used by Willis et al. (7). This model accounts for the spectroscopic properties, the density, and the viscosity. Perhaps the surface initiation of the formation of polywater results from the presence of some oxygen atoms in the surface of the silica or silicate glass which have structures in which the two spin-sets of electrons are staggered with respect to one another. Such atoms could provide centers for initiating the structure indicated in Fig. 1. Moreover, once formed the micelles would be expected to be stable because the individual bonds are strong and this model requires that there would have to be a considerable cooperative change to disintegrate a micelle. On the other hand, the micelles would also not be easy to form particularly if ions or molecules were present which held the water molecules by conventional hydrogen bonds. This feature results from the fact that the structure proposed which is depicted by formula 1 is essentially different from those depicted by formula 3.

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"Polywater": A Hydrosol?

Abstract. Measurements of the dielectric constant and the effective parallel conductance of a specimen of anomalous water suggest that anomalous water is a hydrosol consisting of finely divided particulate matter suspended in ordinary water. Scanning electron micrography confirms the presence of particulate matter. These new experimental data provide an alternative explanation for the properties of anomalous water.

Condensate of nominally pure H₂O, allowed to form below atmospheric pressure in freshly drawn quartz or Pyrex capillaries, has been shown to possess physical properties very different from those of ordinary water (1). Lippincott et al. (2) have recently proposed that this condensate (known as anomalous water) contains a polymer consisting of H₂O monomeric units-polymeric water or "polywater." The possible existence of a previously unknown, stable, polymeric form of water has stimulated much interest in the properties of anomalous water.

Anomalous water has thus far been produced only in extremely small quantities. The lack of a sample large enough for detailed chemical analysis has hindered investigators from performing those experiments which would allow them to unequivocally determine the nature of anomalous water. Thus the "polywater" proposal must be considered as one possibility among many. To aid in assessing these possibilities, we report here new measurements of some of the physical properties of anomalous water. Our study reveals the presence of highly dispersed particulate matter in anomalous water. Hydrosols (3) (aqueous systems containing highly dispersed particulate matter) exhibit many of the physical properties of anomalous water. Hence, our experiments suggest that a suitable alternative explanation for the unusual properties of anomalous water may be found in the phenomenon of sol formation.

Anomalous water was prepared by the condensation of water vapor in freshly drawn Pyrex capillaries $\lesssim 50$ μ m in diameter (1, 2). Pyrex tubing was rinsed in flowing distilled water for 3 hours before being drawn into capillaries. The resulting capillaries were then allowed to stand for several days suspended over distilled water in a partially evacuated system. Condensate that formed within these capillaries was removed by centrifugation. Two microliters of fluid were thus obtained. This fluid possessed the mechanical properties previously reported for anomalous water (1). Furthermore, analysis by transmission infrared spectroscopy in the region from 1000 to 4000 cm^{-1} indicated absorption bands characteristic of both normal H₂O and "polywater." In particular, the characteristic "polywater" absorption near 1400 cm⁻¹ [as reported by Lippincott et al. (2)] was observed. Therefore, we proceed on the basis that the sample under discussion is indeed anomalous water and that it contains, in dilute form, that material which has been proposed to be polymeric water. All experiments described below were performed on the same specimen.

Dielectric measurements are a powerful tool for the determination of the symmetry of molecular species; the dielectric constant of a substance is, in general, a measure of the dipole moment of its constituent molecules. Hence, application of techniques for

the measurement of dielectric properties to anomalous water would be expected to yield useful information on its internal molecular structure. Measurements were performed by means of a specially designed, variable-separation parallel-plate capacitor in conjunction with reactance bridges operating at frequencies from 10 to 107 hz. Air, pure water, and dilute HCl were used as working standards to check the performance of this apparatus. The dielectric constant ε and effective parallel conductance G of our anomalous water specimen are shown as a function of frequency in Fig. 1. The magnitude and frequency dependence of these data are quite unusual and these dependencies differ in character from those observed for true solutions, or for other singlephase liquids including polymers (4). Single-phase liquids have dielectric properties qualitatively similar to those shown in Fig. 1 for H₂O. There is,



Fig. 1. Dielectric properties of several liquids as a function of frequency. The data for anomalous water (circles) strongly resemble data obtained on suspensions of Pyrex spheres 2μ m (squares) and 20μ m (triangles) in diameter in 0.005M potassium chloride (4). To illustrate this resemblance straight-line segments are drawn through the data as appropriate. For comparison, data are shown for pure water (dashed line).

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however, a class of two-phase systems, the colloidal suspensions or sols, that possess dielectric properties (5) remarkably similar to those we have observed for anomalous water. To illustrate this similarity, data (5) on twosol systems are also shown in Fig. 1. Thus it appears that our anomalous water specimen is a hydrosol consisting of finely divided particulate matter suspended in ordinary water.

This particulate matter was further investigated by scanning electron microscopy. The anomalous water specimen was vacuum-dried prior to micrography to remove all free water. Two representative micrographs of the resulting residue (Fig. 2) reveal substantial quantities of particulate matter. The particles appear "fluffy" and range in size from 800 Å to several micrometers. The electrical conductivity of these particles is low, as was indicated by the appreciable charging observed during scanning electron micrography. This charging somewhat degrades the resolution capabilities of the scanning electron microscope.

No extraordinary precautions were taken to assure the absence of casual particulate contamination (as from dust and the like) of our specimen. The possibility of contamination was investigated by taking scanning electron micrographs of a vacuum-dried tap water specimen handled in the same manner as the anomalous water specimen. These micrographs gave no evidence of particulate matter. In addition, the dielectric constant of tap water is virtually indistinguishable from that of distilled water. Hence, casual contamination does not yield the properties observed for our sample of anomalous water.

Electron microprobe analysis of the elemental composition of the particulate matter observed in our anomalous water specimen (6) indicates the presence of substantial quantities of both potassium and chlorine. The presence of small quantities of other elements including sodium and sulfur was also indicated. No appreciable amount of silicon was found. The microprobe used was not sensitive to elements lighter than fluorine, and hence the presence of such elements as oxygen and nitrogen could not be determined by this technique. These elements are likely to be present because the characteristic "polywater" absorption band at approximately 1400 cm⁻¹ resembles strongly the absorption spectrum of KNO₃.

To realistically interpret the role of particulate matter in the "polywater" phenomenon, it is necessary to carefully consider the experimental techniques involved in the production and analysis of anomalous water. Anomalous water has thus far been produced primarily in freshly drawn quartz or Pyrex capillaries. Freshly drawn capillaries possess, on a submicroscopic scale, a large, highly reactive free surface which may serve as a source for potential sol formers. We propose that sol formers, in highly hydrated form, are responsible for the properties of our anomalous water specimen. The presence of even trace quantities of sol formers will entirely alter the properties of water. For example, the commercial sol-forming material Macaloid (7),



Fig. 2. Scanning electron micrographs of anomalous water. (a) Low-resolution micrograph showing the edge of the residue left after vacuum drying. A large number of particles comprise this residue. (b) Higher resolution micrograph showing the region set off by the small dashed white circle near the upper center of (a). Many smaller particles are evident. Micrographs of vacuum-dried tap water show no such particulate matter.

when added in concentrations of approximately 1 percent by weight, will transform a beaker of water into a thixotropic paste which exhibits many external physical properties similar to those of polywater.

We conclude that our anomalous water specimen is a two-phase system, that is, a hydrosol, consisting of finely divided particulate matter suspended in ordinary water. This conclusion is consistent with the reported properties of anomalous water. As previously noted, thoroughgoing analyses of all kinds will be required before sufficient data is amassed to unequivocally establish the nature of anomalous water. There is presently little evidence to suggest that the anomalous water studied by others (1, 2) is not a hydrosol. We therefore suggest that proposed "polywater" specimens be examined for the presence of particulate matter. The presence of such matter offers an alternative explanation for the unusual properties of anomalous water. This alternative does not require the postulation of a polymeric form of H₂O.

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Alpha Radioactivity of the Lunar Surface at the Landing Sites of Surveyors 5, 6, and 7

Abstract. Evidence has been obtained for a radioactive deposit on the lunar surface at Mare Tranquillitatis with a total intensity of 0.09 \pm 0.03 alpha disintegration per second per square centimeter. The presence of polonium-210 in amounts that are close to equilibrium indicates a continuous turnover rate of lunar material at this site of less than 0.1 micrometer per year. The lack of such a deposit at two other lunar sites suggests lower local concentrations of uranium there.

The possibility of an alpha-emitting radioactive deposit on the lunar surface, caused by the decay in space of radon isotopes diffusing out of lunar surface material, was suggested by Kraner et al. (1). Recently Yeh and Van Allen (2) have set upper limits on the amount of such alpha radioactivity by the use of data from the Explorer 35 satellite orbiting the moon. The alpha-scattering experiment performed at three locations on the moon in 1967-68 by Surveyor spacecraft has provided evidence for such an alpha-active deposit in Mare Tranquillitatis. No such evidence was found at Sinus Medii or outside the crater Tycho.

The active deposit from radon (222 Rn, $t_{1/2} = 3.825$ days) should contain, at equilibrium, daughter products that emit alpha particles of energies 6.00, 7.69, and 5.31 Mev. The deposit from thoron $(^{220}$ Rn, $t_{1/2} = 54.5$ seconds) should be less intense if the source rock has a Th/U concentration ratio in the usual range and should emit alpha particles of energies 6.78 (1), 6.05 (0.33), and 8.78 (0.67) Mev, where the numbers in parentheses refer to the relative intensities within the series. The deposit from both of these radon isotopes, as well as from the even less abundant actinon ²¹⁹Rn, should be on the very top of the undisturbed lunar surface. The ²¹⁸Po and ²¹⁴Po daughters of ²²²Rn and the ²¹⁶Po, ²¹²Bi, and ²¹²Po daughters of ²²⁰Rn, because of their relatively short half-lives and those of their precursors, should come to equilibrium with their noble gas ancestors within a day or less. The formation of ²¹⁰Po is held up, however, by the 22-year half-life of its grandparent ²¹⁰Pb.

Although not designed for this purpose, the Surveyor alpha-scattering experiment (3, 4) provided information on this question of an active deposit on the lunar surface. Its sensitivity was limited by the short operating time in certain stages of the experiment, by the presence of a small amount of 254 Es (T = 6.44

Mev) placed close to the alpha detectors to provide a check of the energy scale of the instrument, and by the presence of a small "background" produced by the scattering of uncollimated alpha particles from the gold-lined interior of the instrument. The cosmic-ray-produced background in the alpha detectors was very low.

In the second stage of operation of the experiment (3), data were recorded while the instrument was suspended about 56 cm above the lunar surface. In this position the alpha detectors should have measured any long-lived (for instance, ²¹⁰Po, $T_{\alpha} = 5.31$ Mev) alpha activity on approximately 7000 cm² of lunar surface underneath the instrument. They should also have measured the rate of the deposition of active products of ²²²Rn through the amounts of the short-lived progeny-that is, ²¹⁸Po (6.00 Mev) and ²¹⁴Po (7.69 Mev). Because of shadowing by the spacecraft and by the overhanging instrument (30 cm diameter), the observed rate of deposition is estimated to be only about 0.74 of the rate expected on an open lunar surface. The proton detectors of the instrument should have been sensitive only to the 7.69-Mev (214Po) alpha particles because of the gold absorbers over the detectors. The degraded alpha spectrum in this mode is expected, however, to be too smeared to be identifiable.

In this stage of operation, the Surveyor 5 experiment at Mare Tranquillitatis gave moderately convincing evidence for alpha particles of energy 5.31 and 6.00 Mev (see Fig. 1). The presence of 5.31-Mev alpha particles indicates that at least part of the surface near the spacecraft had not been disturbed by the landing. In addition to the evidence in the alpha spectrum, the overflow channel of the pulse-height analyzer, which recorded events of energy greater than 7.3 Mev and therefore should have recorded the 7.69-Mev alpha particles also, showed an excess number of events when the in-