record (Fig. 1) are due to lightning strokes. (Although perhaps ten times as many appear on some of our records, their integrated energy never exceeded a small fraction of a degree of antenna temperature.) The large deflection on the 1415-Mhz record is due to the passage of the plane of the galaxy through the antenna beam as the earth rotates. Its daily presence on the records provided a convenient check of our sensitivity and timekeeping. (At 16 Ghz this effect is undetectably small, given the sensitivity of our equipment.)

Although the observed increases in 1415-Mhz antenna temperature are clearly not of the magnitude predicted (*1*), the fact that any effect appears on the record is not otherwise easily accounted for (*3*). Indeed, standard calculations of attenuation by rain at 1415 Mhz lead to antenna temperature changes of only a small fraction of a degree (*4*). Such a calculation gives 5×10^{-4} for the ratio of attenuation at the two frequencies or only about one tenth of the peak value observed. It is our conclusion that the observed effect, although not as high as the value predicted by Sartor and Atkinson, does fall within the range of possible values discussed in their paper, given the uncertainty of the parameters upon which the calculations are based.

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References and Notes

1. J. D. Sartor and W. R. Atkinson, *Science* **157**, 1267 (1967). References to previous work are given in this paper. See also J. D. Sartor, "Electricity and rain" in *Physics Today*, August 1969.
2. We thank D. C. Hogg who called this problem to our attention.
3. Two other possible ways of producing an increase in apparent system temperature correlated with rain rate were investigated and excluded. The first was accumulation of water on the waveguide window or walls of the antenna throat. Therefore, care was taken to make sure that critical components were kept free of water during observations. The second possible effect was rain scattering of man-made interference into our beam. Investigation of oscilloscope and chart records, on which spurious radar signals are normally readily discernible when present, failed to show evidence of such a signal. Furthermore, azimuthal scans along our horizon failed to reveal any interfering source.
4. D. E. Setzer, unpublished data.

25 March 1970

Ice Sandwich: Functional Semipermeable Membrane

Abstract. *At a temperature slightly below the freezing point of an aqueous solution, the functions of a nearly perfect semipermeable membrane can be simulated with two nonselective filters with ice sandwiched between them. Potential applications include production of potable water from brackish sources or of highly purified water from tap water.*

Reflection on the process of frost heaving in soil revealed that an extraordinarily simple device should simulate the behavior of a nearly perfect semipermeable membrane. This device could have important applications in the purification of liquids, particularly water. Crude experiments showed that the device worked more or less as expected and suggested a configuration for optimum performance.

Functionally, the behavior of a semipermeable membrane for aqueous solutions can be simulated with a pair of rigid nonselective filters with ice maintained in a narrow space between (an "ice sandwich") held at a temperature slightly below the freezing point of the solution. This assembly can be used to simulate both osmosis and reverse osmosis. The ice phase provides the solute barrier, and its movement (engendered by concurrent freezing and melting at opposing faces in contact with the respective filters) provides for solvent transport. The filters serve only as phase barriers, confining the ice phase to its allotted place even though supercooled water, or supercooled solution, fills the pores of the filters and adjoining spaces. The accompanying diagram depicts the system in schematic form (Fig. 1).

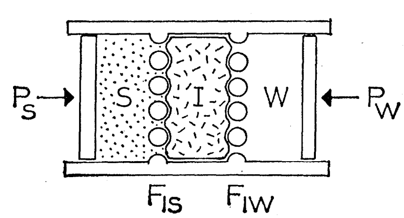

Fig. 1. The ice sandwich osmometer. One chamber contains an aqueous solution, *S*, at pressure P_s; another contains water, *W*, at pressure P_w; the intervening chamber contains ice, *I*, with the common walls being rigid filters, F_{IS} and F_{IW}. The osmometer is maintained at a temperature slightly below the freezing temperature of the solution.

The filters have two critical requirements. The first is that they be wettable by the liquid phase, with surface interactions that sustain a liquid film between the ice and the filter substance at subfreezing temperatures—a characteristic of common filter materials (and of soil particles). The second requirement concerns maximum pore size. Surface tension of the interface between ice and water tends to exclude ice from the filter pores at subfreezing temperatures, the maximum permissible pore size, r, depending upon the operating conditions. If, for example, the osmometer in Fig. 1 is at equilibrium, P_w is atmospheric pressure, P_s is adjusted to the osmotic pressure of S, and the temperature, T, is held below the freezing temperature of the solution, T_f,

$$(r_{IS})_{max} = \frac{2\,T\sigma}{d_w L(T_f - T)}$$

and

$$(r_{IW})_{max} = \frac{2\,T\sigma}{d_w L(T_0 - T)} \simeq \frac{5 \times 10^{-6}}{T_0 - T}\ \text{cm}$$

where σ is the surface tension of the interface [30 dyne/cm is a good working value (*1*)], d_w is the density of water, L is the latent heat of fusion, and T_0 is the ice point. If the process is driven at a finite rate in either direction, these values shift slightly, reflecting local shifts of temperature, pressure, and concentration. Note that $(r_{IS})_{max}$ is greater than $(r_{IW})_{max}$.

A parameter of practical interest is the apparent permeability, k_w, of the ice sandwich. This is a complex quantity, but one can estimate its upper limit under the simplest possible circumstances, that is, when S and W are both pure water and a steady state has been established in which the limiting process is the transfer of the heat of fusion, L, across the ice layer of thickness l cm with this being the only heat transport (that is, the temperature on the high pressure side is uniformly higher than that on the low pressure side). We now write standard transport equations for the flow of water and heat, respectively, neglecting the impedance of the filters to water movement:

$$Q_w = k_w \frac{(P_s - P_w)}{l}$$

and

$$Q_h = k_h \frac{(T_s - T_w)}{l}$$

where Q_w is the mass of water transported per second per square centimeter, Q_h is the transport of heat (neglecting advective transport by the moving ice), and k_h is the thermal conductivity of ice. For the most favorable estimate, we replace $(T_s - T_w)$ by $(P_s - P_w)\ T/d_I L$, and since Q_h equals LQ_w, we find that

$$k_w \leqq \frac{T}{d_I L_2} k_h$$

or

$$k_w \leqq 5.6 \times 10^{-6} \text{ g (bar cm sec)}^{-1}$$

if P is in bars. Thus if l is 0.1 mm and P_s is 10 bars, the most favorable estimate amounts to 200 liters m^{-2} hr^{-1}.

In experiments performed with distilled water on both sides of the sandwich, observed values of k_w ranged from about 4×10^{-6} at $-0.07°C$ to about 2×10^{-6} at $-0.15°C$. The decrease of k_w with decreasing temperature is tentatively attributed to sliding friction of the ice plug against the Teflon walls of the ice chamber of the test apparatus and is consistent with increasing ice pressure as temperature decreases. Extrapolation to 0°C suggests that k_w approaches the most favorable estimate as the ice approaches atmospheric pressure.

During reverse osmosis, the accumulation of solutes in the filter next to the ice will have the effect of reducing the apparent permeability, and any general expression for k_w must include terms for the back diffusion of solutes through the filter F_{IS}.

The experiments referred to were performed in 1966 with the simple cell shown disassembled in Fig. 2. Except for the brass endplates, all parts were stock commercial items. A detailed description of the experiments is not warranted here nor were the results of a quality that merit detailed presentation. Since the only filters on hand were relatively coarse (nominal pore diameter 0.22 μm), the solutions used were rather dilute, of the order of 8.5 meq of NaCl or of $CaCl_2$ per liter. The maximum pressure that could be imposed with the apparatus used was 98 cm-Hg. There was no provision for stirring other than a slight mixing effect that could be obtained by very slow bleeding of a limited quantity of solution through the input chamber. As expected, the rate of water production was very sensitive to the rate of bleeding; the highest value of k_w obtained was about half that obtained with water at the same temperature.

When the system was operated at maximum pressure, the water produced was slightly contaminated with solutes. It seems likely that the contaminants traversed the ice as occluded droplets of solution. The external filter support was a smooth stainless-steel plate with small circular holes that occupied about one quarter of the gross area. The highest concentration of rejected solutes would develop midway between these openings. If the freezing temperature of such solution reached the operating temperature, a small amount of solution could enter the ice chamber as a liquid. This effect caused complete dissolution of the ice barrier if the solution was not flushed away, provided the operating temperature was not too low and provided that the operating pressure was sufficiently high. This risk can be avoided by flushing or by respecting a maximum operating pressure defined by the operating temperature, T,

$$(P_s)_{max} = \frac{L}{d_I T}(T_o - T) = 11.1\ (T_o - T) \text{ bars}$$

This relation reflects the fact that concentration of the solution ceases when the osmotic pressure reaches the imposed pressure, and that to preserve the ice, the operating temperature must be below the freezing temperature of the solution at that concentration.

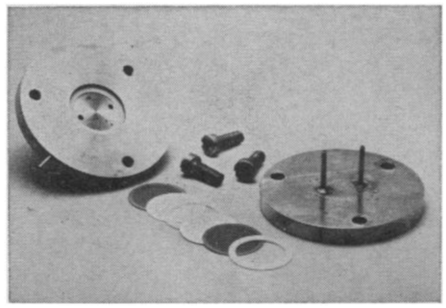

Fig. 2. Test cell used to demonstrate the ice sandwich principle. Visible are three Teflon gasket-spacers, two cellulose ester filters (diameter 25 mm with pore diameter 0.22 μm) and two stainless steel filter-support screens (catalog numbers XX 30 025 0, GSWP 0 25 00 and XX 30 025 10, respectively, Millipore Filter Corp., Bedford, Mass.). When the cell was assembled, a small hole (No. 80 drill) bored radially through the center gasket was aligned with the radial access tube visible at 8 o'clock on the base plate. This permitted nucleation of freezing in the central chamber when the cell was filled with supercooled water.

In principle, the ice sandwich has merits as a device for purifying brackish water. It is a continuous stream device, and the heat of phase transition is exchanged directly across what can be an extremely thin layer of ice, much thinner than the 0.7-mm layer in the test apparatus. As compared to ordinary semipermeable membranes, the ice sandwich may be easier to protect against fouling by suspended matter by passing the solution through an initial filter with pores no larger than those of F_{IS}. If matter that passes this filter also passes F_{IS}, it will be deposited between the ice and the filter without necessarily interfering with the process, although it might eventually fill the space intended for ice. The greatest practical disadvantage is the requirement for close control of the operating temperature; the temperature gradient in the ice is established spontaneously by the applied pressure differential.

The external support for F_{IS} used in the test cell obstructed dispersal of rejected solutes. This problem could be solved by fabricating a "quilted" combination of F_{IW} and F_{IS}. Under operating conditions P_I is greater than P_s which is greater than P_w, which means that the quilted filter would only require support on the discharge side. No rigid support would be needed if a pair of quilted filters were placed back to back on the opposite sides of a porous core through which water would be discharged. The points of attachment of F_{IS} to F_{IW} would have to be impervious to solution and their spacing would have to take into account the tensile strength of F_{IS} and the desire to keep the ice "batting" thin to promote heat exchange. The quilted filter would also eliminate the problem of sliding friction of the test cell.

A more specialized application may be found in the production of extremely pure water, with special advantages (relative to distillation) for the elimination of volatile solutes. Cascaded stages could be assembled in a single bath and would simulate the performance of a multiple zone-freezing purification process. The conductivity of ordinary distilled water was substantially reduced by a single pass through the test cell.

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References and Notes

1. R. W. R. Koopmans and R. D. Miller, *Soil Sci. Soc. Amer. Proc.* **30**, 680 (1966).
2. I thank the Norwegian Geotechnical Institute and its director, Dr. Laurits Bjerrum, for the opportunity to carry out the preliminary experiments as an adjunct to other activities, and the Cold Regions Research and Engineering Laboratory, Hanover, N.H., for support of research on frost heaving, which led to the conception of the ice sandwich as a functional semipermeable membrane.

8 June 1970