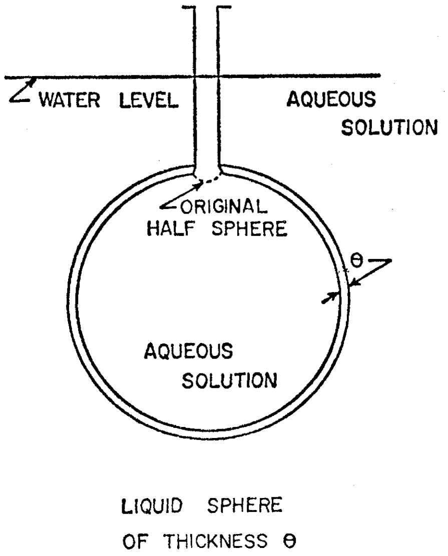

Fig. 5. Liquid sphere of thickness θ suspended in an aqueous solution with a blow tube.

air, while the interfacial tension of *o*-toluidine–salt solution at 20°C equals 30 dyne/cm, that is, about the same as our soap solutions, the efflux time for the same geometry is increased by a factor of 200.

These experiments are much more complicated to perform than those with a soap bubble and require gravity-free conditions (*8*).

The two-sphere case, shown in Fig. 4, however, is simple to demonstrate. Two spheres of *o*-toluidine, with radii *R* and *r* (between 5 and 1 cm), suspended in a solution of NaCl, were connected with a flow tube in the form of a T filled with *o*-toluidine. If the tips of the flow tube are covered with a thin layer of paraffin there is no difficulty in having the liquid spheres adhere to them; by coloring one sphere with an organic dye, I can observe the liquid flow. As in the case of soap bubbles, the larger sphere grows at the expense of the smaller one until the latter disappears and is converted into a flat film, sticking to the tip of the flow tube. The efflux time is in agreement with the theoretical one given by the integral 6, only $\sigma_i = \frac{1}{2} \cdot \sigma$.

With liquids one can also "blow" double spheres, one inside the other, as was shown by Plateau (*2*) and as seen in Fig. 5. Whereas soap bubbles have a practically infinite thinness of 10 to $\simeq 100\ \mu\mu$, the liquid double spheres can have any desired thickness, θ. By attaching a known volume, v, of, for example, *o*-toluidine at the end of a tube (by means of paraffin) and pouring water or salt solution slowly into this tube from a dropping funnel, the *o*-toluidine can be expanded first into a half-sphere (as indicated by the dotted half circle in Fig. 5) and, by adding more water, into a sphere of any desired size, surrounded by a thin sphere of *o*-toluidine of thickness, θ. By using 1.0 ml of *o*-toluidine I have been able to "blow" a water sphere 6.0 cm in diameter surrounded by a thin sphere of *o*-toluidine about 0.18 mm thick. Of course, such liquid film spheres are fragile and require great care in handling.

The efflux time of water (or any other suitable fluid) from such a thin film liquid sphere is given by Eq. 4, provided ΔP is doubled, since it has two interfacial tension surfaces. It can be more readily demonstrated and measured than any of the other liquid sphere cases, since the inside liquid, water, can be easily permitted to flow to the outside water reservoir by means of a flow tube covered on the outside with a thin paraffin layer. Care has to be taken so that its inlet is positioned in the center of the sphere; the "blow" tube also has to be progressively lowered to keep the flow tube inlet in the center of the water sphere. The observed efflux times confirm Eq. 4, although the accuracy attained was much less than that with soap bubbles.

So far I have discussed only the change with time of gaseous and liquid spheres. Such a study does not have to be limited to spheres. Plateau (*2*) and Boys (*4*) have shown that both soap-film and liquid cylinders can be produced; these are stable if their height is less than π times their diameter. In a cylinder, one of the radii of curvature is infinite. It follows from Laplace's equation that the ΔP of a soap cylinder equals $2\sigma/R$, or only one-half of that of a sphere of the same radius. As Boys has shown (*4*, pp. 52–62) the cylinder contracts to a surface of zero curvature, and thus zero pressure, or the catenoid of rotation. The efflux time of a cylinder to a catenoid has been measured; in contrast to a sphere where ΔP increases as it contracts, in this particular case the contraction becomes slower and slower, and finally stops when the zero curvature of the catenoid has been reached (*8*).

Cylinders can be connected with flow tubes to other cylinders, or spheres, or catenoids, and thus the number of combinations increase manifold. Only one combination might be mentioned since it permits the measurement of σ and η of liquids from their efflux time. If in Fig. 4 a liquid sphere of radius r is connected by a flow tube to a catenoid of rotation of suitable size, the efflux time will be given by Eq. 4 instead of the integral 6.

In summary, the measurement of time in experiments involving flows determined only by surface tension forces gives a sensitive method to study the influence of various factors, physical and particularly chemical, on these forces (*8*).

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8 February 1967; 13 April 1967

Farmington Meteorite: Cristobalite Xenoliths and Blackening

Abstract. *The Farmington chondrite contains two small xenoliths of granular cristobalite, each surrounded by a thin reaction rim of diopsidic clinopyroxene. Similarities between the blackened structure and drusy cavities, characteristic of this meteorite, and those of an experimentally heat-treated chondrite suggest that Farmington was reheated rather than shocked, but neither the exact stage in the history of this stone at which reaction rims developed around the xenoliths nor the source of the calcium necessary to form rim diopside have been established with certainty.*

Xenolithic structures are not uncommon in chondritic meteorites, but usually the xenoliths are similar in overall chemical composition and mineralogy to the material by which they are enclosed. They differ in extent of recrystallization, but their similar composition and lack of marginal reaction denote an essentially cognate relation to their hosts (*1*). My purpose is to draw attention to two small xenoliths, of quite different character, discovered on a broken surface of one specimen (*2*) of the Farmington meteorite, a black olivine-hypersthene chondrite (Fig. 1A). Each, composed entirely of cristobalite, is surrounded by a reaction rim 0.5 mm thick. A small patch of similar rim material at an edge adjacent to these two fragments indicates that a third xenolith, now broken away, belonged to the cluster.

The two xenoliths, opalescent blue-gray in color, have granular textures (average grain size, 1 mm) reminiscent of terrestrial metaquartzites. Their constituent cristobalite gives the x-ray powder diffraction pattern of the low-temperature or α modification (Table 1); but when examined with a petrographic microscope, it shows faint anisotropism and a fine plaid-like twin pattern suggestive of inversion from the β or high-temperature polymorph. Its refractive index (1.480 ± 0.001; birefringence, less than 0.001) is slightly lower than those of terrestrial and synthetic cristobalites (*3*), but no compositional abnormality is revealed by the following results of electron-probe analyses: SiO_2, 100 ± 2 percent; Al_2O_3, 0.5 ± 0.1 percent; Fe, Mn, Mg, Ca, and Na, sought but not detected (*4*).

The pale-greenish reaction rims are composed of a fine-grained aggregate of twinned clinopyroxene (grain size, 10 to 20 μ). X-ray powder photographs of material from two different points on the rim of the larger xenolith and from one point on the rim of the smaller gave only diopside patterns (Table 2), but optical study and electron-probe analyses (Table 3) indicate that a little pigeonite-clinoenstatite accompanies the diopside.

Cristobalite xenoliths do not appear on other well-known Farmington specimens (*5*); nor are they mentioned in earlier descriptions of Farmington (*6*–*8*), although a large black cognate xenolith of very different appearance (*7*) and small "amphoterite" fragments (*8*) have been reported. The only comparable material known from other stony meteorites is the grain of cristobalite, also rimmed by diopsidic pyroxene, described from the Nadiabondi meteorite, a gray, spherical, olivine-bronzite chondrite (*9*). The Farmington xenolith cluster is clearly accidental in origin, and its presence implies the availability of fragments from a highly siliceous body at some stage in the evolution of this stone, most probably when it accumulated into a solid object. The xenoliths have no known equivalents among achondritic stony meteorites.

The main portion of the Farmington chondrite is notable for its blackened character, due to finely disseminated

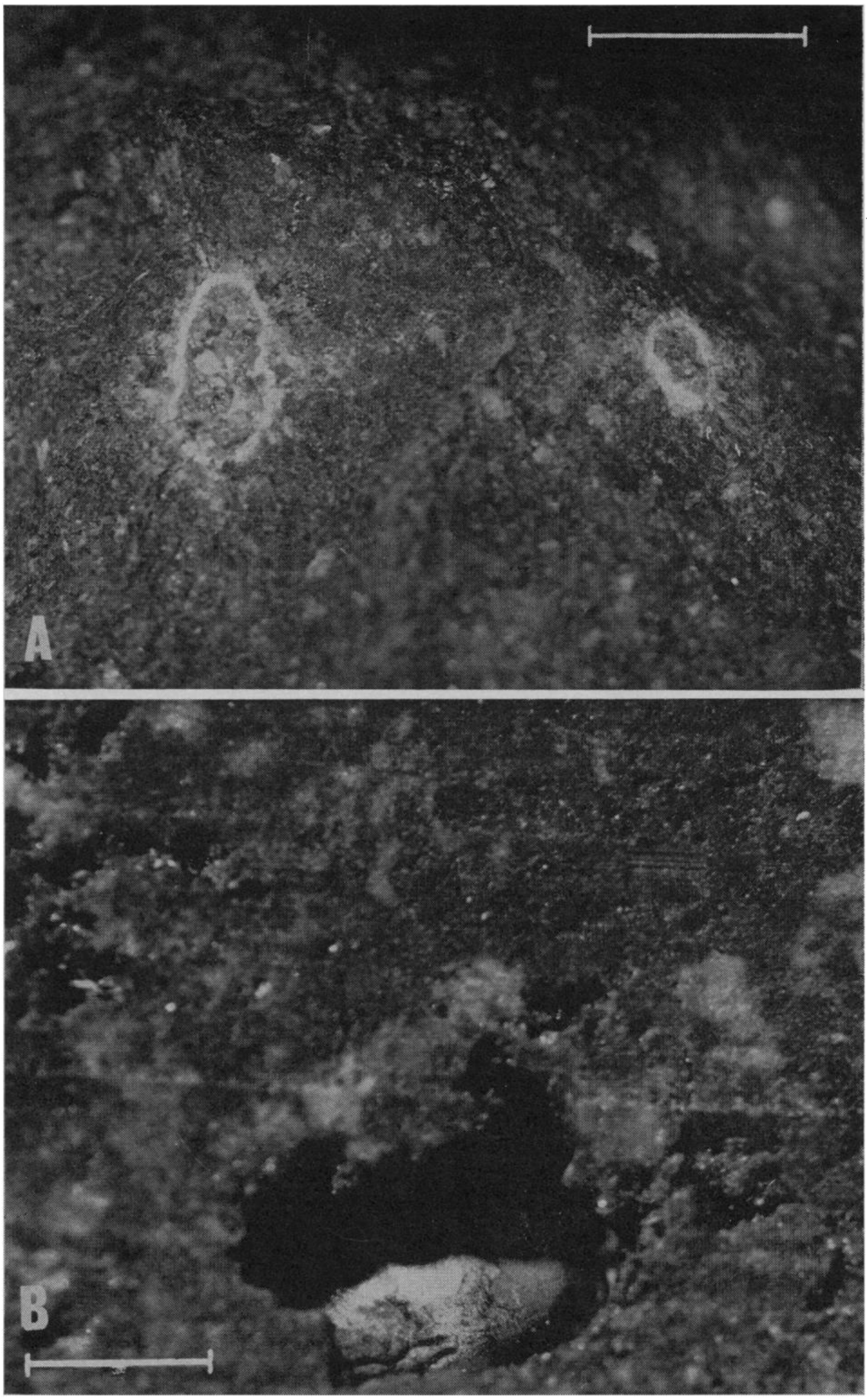

Fig. 1. (A) Two small xenoliths of granular cristobalite, each surrounded by a narrow reaction rim (white in photograph, but really pale green), on a broken surface on the Farmington meteorite. The chondritic host near the larger xenolith appears slightly leached. A third patch of rim material occurs on the edge just beyond the right of the photograph. Scale bar, 1.0 cm. (B) Irregular drusy-like cavities on a sawn face of the Farmington meteorite. A globular body of metal lies within the largest cavity, which is also partly lined by idiomorphic troilite crystals (invisible in the photograph). Scale bar, 1.0 mm.

opaque granules, dominantly of troilite. This structure is possibly related to the exceptionally low cosmic-ray-exposure age of the stone—less than 25,000 years (*10*). Two explanations, both supported experimentally, have been advanced for the peculiar characteristics of the black chondrites; the characteristics either arose from shock effects due to high-velocity cosmic imparts (*7, 11*), or resulted from reheating, possibly during close solar approach by orbiting parent bodies (*12*). In Farmington, the second alternative is favored by an abundance of drusy-like cavities; they vary considerably in shape and size (Figs. 1B and 2) and are partly lined by idiomorphic troilite crystals. Metal particles projecting into the cavities have a remarkable globular habit (Fig. 1B), appearing as though they crystallized rapidly from a molten state. There are no cracks or other indications of disturbance near the holes.

It is most unlikely that transient high-pressure shock waves [to which maskelynite, transformed metal phases, and veined or brecciation structures in other chondrites have been attributed (*11, 13*)] would have produced open cavities; it is also unlikely that shock waves could have passed through an already porous body without causing fracturing or pronounced textural irregularities in the vicinity of the cavities. However, a possible origin for the holes is suggested by the following heating experiment.

Table 1. X-ray spacing data for cristobalite from a xenolith in the Farmington meteorite (Cu radiation, 11.46-cm-diameter powder camera) and for synthetic low-temperature cristobalite (Mo radiation). Intensities (*I*) for Farmington were visually estimated; vs, very strong; s, strong; m, moderate; w, weak; vw, very weak.

Cristobalite			
Farmington		Synthetic, low-temperature (*3*)	
d(Å)	*I*	*d*(Å)	*I*
4.05	vs	4.04	100
3.14	m	3.138	12
2.85	m	2.845	14
2.49	s	2.489	18
		2.468	6
		2.342	<1
2.12	w	2.121	4
2.02	w	2.024	3
1.933	w	1.932	4
1.869	w	1.874	4
		1.756	1
		1.736	1
1.695	vw	1.692	3
		1.642	1
1.610	w	1.612	5
		1.604	2
		1.574	1
1.535	vw	1.535	2
1.496	vw	1.495	3
1.430	vw	1.432	2
		1.423	1

A sample of the Parnallee chondrite, held at 1050°C for 20 days in an evacuated silica-glass tube before quenching over a period of several seconds by immersion in water, was converted to a black porous body similar to Farmington. Most of its large metal and troilite particles were melted during the experiment, and upon cooling they left cavities lined by globular troilite bodies or fine intergrowths of troilite and taenite. Much troilite and some kamacite became dispersed as minute particles throughout the chondrules and interchondrule mesostasis. There was a slight overall loss of weight (0.05 percent), probably of sulfur vapor into the evacuated tube. Apart from fine devitrification of some brown chondrule glass, the silicate constituents of the chondrules were not affected by the heating, although electron-probe investigations to check their compositional homogeneity or inhomogeneity are still awaited. Since the experiment was carried out under evacuated conditions, some or all of the disseminated troilite was probably mobilized as a sulfide- or sulfur-vapor phase, while the fine, dispersed, metallic particles possibly resulted from redistribution of oxygen.

Although the nature of the material lining cavities was not identical, perhaps because of different vapor pressures, this experiment suggests that reheating provides an acceptable explanation of the blackening and cavitation characteristic of Farmington and certain other black chondrites (*14*). However, some unblackened chondrites (such as Richmond) contain open pores, so that it is also possible that the cavities in Farmington are original structures predating the blackening process.

Some specimens possess larger, discontinuous, joint-like holes (Fig. 2); these may represent remelted metal-troilite veins, but there is no other evidence that such veins existed in the main body of Farmington although they do occur in the large black xenolith (*7*). Replacement relations between metal, troilite, and chromite, and textural evidence suggesting that kamacite was once molten, have already been reported from the main part of Farmington (*7*), but it is by no means certain when these features were developed.

Blackened character apart, Farmington shows microscopic evidence of recrystallization of a kind common in other chondrites. This was comparatively mild: sufficient to devitrify chondrule glass, to homogenize the Mg-Fe content of its olivines and pyroxenes, and to invert most of the pyroxene to orthorhombic symmetry (clinohypersthene remains in places), but not to disturb the general textural arrangements in its chondrules. Thin-section study of Farmington and other black chondrites shows that the blackening was superimposed on this recrystallization; the two phenomena apparently are not directly related genetically.

Table 2. X-ray spacing data for pyroxene from the corona about a Farmington xenolith (Cu radiation, 11.46-cm-diameter powder camera) and for a synthetic diopside (Co radiation). Intensities (*I*) for Farmington were visually estimated; vs, very strong; s, strong; m, moderate; w, weak; vw, very weak; b, broad or diffuse line.

Farmington pyroxene		Synthetic diopside (*19*)	
d(Å)	*I*	*d*(Å)	*I*
3.34	w	3.35	10
3.23	m	3.23	20
2.99	vs	3.00	80
2.94	w	2.96	10
2.88	s	2.90	40
2.82	vw*		
2.56	m	2.57	10
2.52	vs	2.53	100
2.29	w	2.30	10
2.20	w	2.21 2.20	10 10
2.14	m, b	2.15 2.13	10 20
2.10	vw	2.10	10
2.04	m	2.04	20
2.01	vw	2.01	10
1.966	vw	1.97	10
1.825	vw	1.83	10
1.751	w	1.75	20
1.667	vw, b	1.67 1.66	10 10
1.622	s	1.62	60
1.610	vw	1.61	10
1.55	vw, b	1.56 1.55	10 10
1.528	vw	1.53	10
1.503	w	1.50	10
1.486	vw	1.49	10
1.418	m, b	1.42 1.41	40 20

* This line occurs in powder patterns of natural terrestrial diopsides.

Table 3. Results of 11 electron-probe analyses of pyroxenes in the reaction rim surrounding a cristobalite xenolith in the Farmington meteorite. Measurements were made on an imperfectly polished surface of a small chip from the reaction rim. Low totals may result partly from fine grain size and poor polish, but they may also denote presence of other elements.

Analysis	Element (wt. %)					Oxide (wt. %, adjusted to 100%)				End member (mole %)			Fe:(Fe+Mg) (mole %)
	Si	Ca	Fe	Mg	Total (incl. O)	SiO_2	CaO	FeO	MgO	Wo	En	Fs	
1	24.5	15.2	3.9	10.7	96.6	54.3	22.0	5.3	18.4	42.5	49.6	7.9	13.7
2	24.9	15.1	4.3	10.4	97.3	54.7	21.8	5.7	17.8	42.7	48.5	8.8	15.4
3	26.3	3.4	4.9	15.6	93.1	60.4	5.1	6.7	27.8	10.4	78.9	10.7	11.9
4	25.6	14.4	4.4	10.3	97.8	56.0	20.6	5.9	17.5	41.6	49.3	9.1	15.6
5	25.1	13.3	4.5	9.8	94.3	56.9	19.7	6.2	17.2	40.6	49.4	10.0	15.5
6	25.4	14.0	4.0	9.1	94.1	57.7	20.8	5.5	16.0	43.9	47.0	9.1	16.2
7	25.7	15.7	3.9	10.3	99.0	55.5	22.2	5.1	17.2	44.3	47.7	8.0	14.4
8	25.6	15.3	3.8	9.6	96.9	56.4	22.0	5.1	16.5	45.0	46.9	8.1	14.7
9	25.4	1.2	7.6	15.8	92.0	59.0	1.9	10.6	28.5	3.8	79.6	16.6	17.2
10	23.6	13.5	3.6	9.0	88.7	56.9	21.2	5.2	16.7	43.7	48.0	8.3	14.7
11	24.2	13.8	3.6	8.9	90.7	57.2	21.3	5.2	16.3	44.3	47.3	8.4	15.1

The existence of reaction rims around the cristobalite xenoliths denotes elevated temperatures some time during evolution of the Farmington meteorite. The occurrence itself of what was once β cristobalite indicates high temperatures, but, since inversion of cristobalite to other silica polymorphs is sluggish, and since it may crystallize metastably outside its true stability field (*3*), little can be deduced about the time the meteorite formed or the physical conditions then prevailing. At high temperatures, cristobalite or any other silica phase would be distinctly unstable in the enclosing olivine-bearing chondrite host, and a faintly leached zone near the larger xenolith (see Fig. 1A) indeed suggests that the adjacent chondritic material has participated in a reaction with the silica-rich fragment to form the diopside-rich coronas. The average Fe:(Fe + Mg) ratio of the diopsidic pyroxene in the reaction rim (15.0 moles percent) is appropriate to an equilibrium association at about 1000°C (*15*) with the host orthopyroxene, which has an Fe:(Fe + Mg) ratio of 20.5 moles percent (*16*), but the validity of this temperature estimate depends upon whether or not equilibrium was established between rim and host, a question to which I shall refer again. Higher temperatures are indicated by the distribution of iron and magnesium between the calcium-rich and calcium-poor pyroxenes in the rim itself, but compositional data on the latter constituent remain too limited to be useful in this respect.

Since a similar rim surrounds the cristobalite grain in the unblackened Nadiabondi chondrite, it is not likely that reaction between xenolith and host occurred during whatever process was responsible for the blackening of Farmington. The mild recrystallization suffered by Farmington before it was blackened could have induced sufficient reaction between xenoliths and enclosing material to form the diopside-rich rims, but the composition of the chondritic host is such that a calcium-poor rather than calicum-rich pyroxene would be expected from a reaction during such recrystallization.

There remains the possibility that the reaction took place at the earliest stage in the evolution of Farmington, when molten or very hot silicate and metallic material accumulated along with siliceous fragments to form the stone. Here, too, there is a problem in reconciling the calcic nature of the diopside rims with the calcium-poor chemistry of the meteorite as a whole, or of its constituent chondrules. The abundant diopside in reaction coronas surrounding quartz xenocrysts or quartzite xenoliths in terrestrial olivine basalts (*17*) can be interpreted in terms of experimental studies [as in the systems diopside-forsterite-silica or diopside-forsterite-anorthite-silica (*18*)], but, by contrast with lime-rich basaltic magmas, a melt having the composition of the Farmington chondrite would contain far too little normative diopside to produce such a corona. Instead a calcium-poor orthopyroxene or clinohypersthene should have formed. Unfortunately a microscopic section across the xenolith boundary, which may have provided some textural indication of the time or mechanism of rim forma-

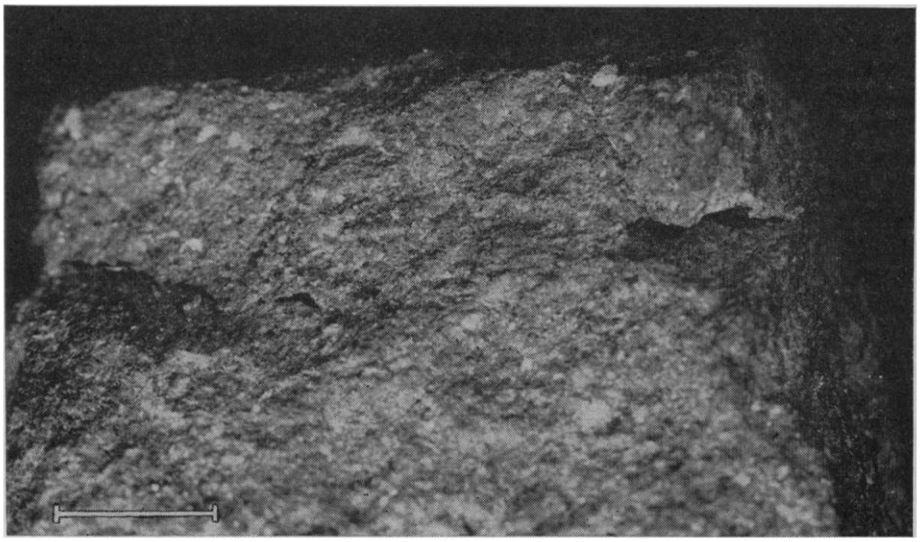

Fig. 2. A large, joint-like opening in the Farmington meteorite. The two cavities shown are roughly coplanar but are not linked by any kind of fracture or vein. Scale bar, 1.0 cm.

tion, could not be prepared without virtual destruction of the specimen. For the present, the exact origin of these diopsidic reaction rims remains obscure.

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10 March 1967

Surface Tension and Surface Structure of Water

Abstract. *The surface tension–temperature relationship for water is smooth and linear when account is taken of the expansion of the surface with temperature. This plot permits the calculation of thermodynamic properties for 1 square centimeter of surface. Molar surface quantities are derived by assuming a hexagonal water surface structure.*

A basic knowledge of the nature of water surfaces is needed in several scientific pursuits dealing with evaporation from reservoirs, growth and coalescence of cloud droplets, and solubilization of oxygen in lakes, and for water-treatment processes dependent upon reactions involving dissolved oxygen. The most well known property of the surface is its tension, yet so elementary a subject as the relationship of surface tension and temperature is poorly understood. A good understanding of this relationship is needed for the calculation of the thermodynamic properties of the surface.

A re-analysis of some earlier data on water surface tension (*1*), along with some data recently obtained in this laboratory, has been made on the basis of some concepts first developed by Eötvös (*2*) and expanded by Ramsay and Shields (*3*). This method of analysis has the unique characteristic of permitting the calculation of extensive surface properties as a function of temperature for an amount of surface that always contains the same number of molecules. The surface layer of molecules is believed and assumed to expand with temperature in the same ratio as does the interior liquid. The volume expansion of the interior liquid varies as the reciprocal of the density, d, of the liquid. The surface area of the top layer is assumed, correlatively, to expand as the reciprocal of the bulk liquid density raised to the two-third power. Thus, one should be able to calculate the extensive surface property of surface free energy for a certain fixed number of molecules by dividing surface tension, γ, by $d^{2/3}$. The number of molecules under identification and measurement is that number composing 1 cm^2 of surface at 4°C; for, in dividing by $d^{2/3}$, one implicitly assumes that the 4°C-area, when the density is 1.0000, is the fixed unit area under consideration. The quantity $\gamma/^{2/3}$ is the surface free energy ΔG_s in ergs for this number of molecules.

The three sets of surface tension data were obtained by three different methods: Moser, by the ring mehod; Teitel'baum, by the maximum bubble pressure method; and Claussen, by rise in a capillary of 0.30 mm diameter. The first two sets of data were for tensions of water in contact with air, while Claussen's is for water in contact with helium. The Moser and Claussen data were normalized linearly to agree with the Teitel'baum data at 25°C, in order to eliminate systematic errors in calibration or measurement.

The method of plotting the surface tension data follows from standard practice in chemical thermodynamics. A consideration of the equation

$$\Delta G_s = \Delta H_s - T\Delta S_s \tag{1}$$

where ΔG_s $(= \gamma/d^{2/3})$, ΔH_s, and ΔS_s are the free energy, heat, and the en-

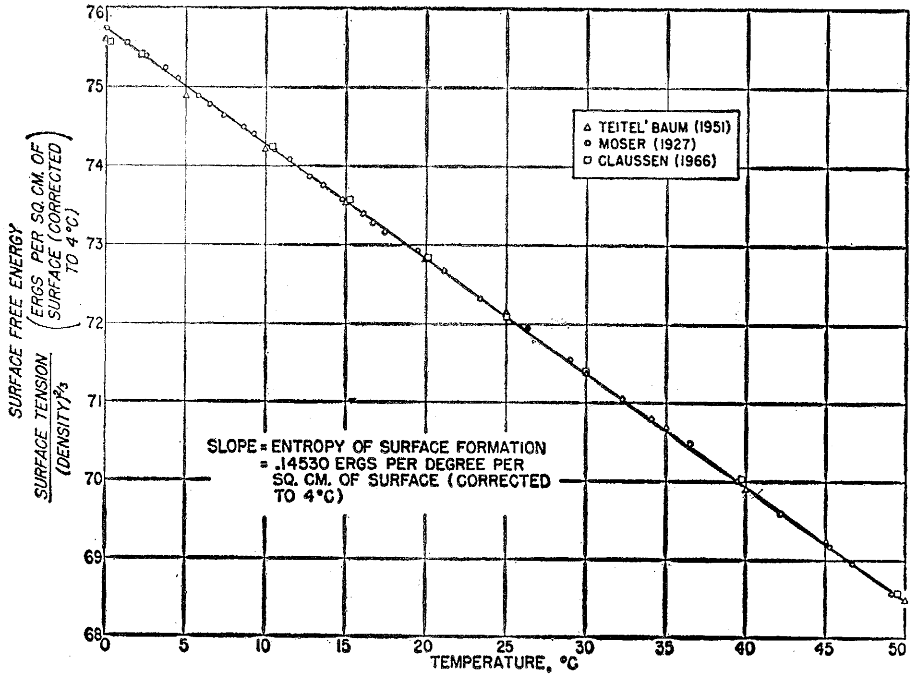

Fig. 1. Surface free energy versus temperature for liquid water.