

Zeitschrift: Eclogae Geologicae Helvetiae
Herausgeber: Schweizerische Geologische Gesellschaft
Band: 65 (1972)
Heft: 1

Artikel: Chemically precipitated sedimentary cristobalite and the origin of chert
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DOI: <https://doi.org/10.5169/seals-164082>

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Chemically Precipitated Sedimentary Cristobalite and the Origin of Chert

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ABSTRACT

Scanning electron microscope and X-ray diffraction studies of deep sea sediments have identified cristobalite as a chemically precipitated authigenic mineral, the presence of which denotes an early diagenetic phase in the silicification of carbonate sediment and in the formation of chert. It crystallizes in the form of thin blades which grow radially, forming spherules 3-10 microns in diameter. Cristobalite has been found in significant amounts in Tertiary sediments of all major ocean basins explored to date by deep sea drilling. On the Kerguelen Plateau (Southern Ocean), it forms an essentially mono-mineralic rock unit of relatively young (Pliocene?) age. Silica for this cristobalite chert was derived from the dissolution of siliceous tests of planktonic organisms. Studies of flint clay from land sections suggest that chemically precipitated sedimentary cristobalite in the form of tiny spherules may also form in the subaerial environment.

Introduction

During their scanning electron microscope/X-ray diffraction study of a deep sea Tertiary chalk from the South Atlantic ocean basin, WISE and HSU (1971) described a late stage pore filling silicate which is sufficiently abundant in the rock to form an accessory cement. The silicate was presumed to be silica, possibly in an amorphous state (WISE and HSU 1971, p.278). Subsequent work by K. R. KELTS (WISE and KELTS 1972, in preparation) indicates that the silica is predominantly cristobalite, and WISE (1972) has suggested that its presence may represent an early stage in the formation of deep sea chert. WISE and KELTS (1971) reported this material in weakly silicified cherts associated with cherts from the Caribbean ocean basin, and WISE and WEAVER (1972) have identified similar material from Pacific and Antarctic deep sea cores. Recently, as a result of exploratory deep sea drilling in the Central Pacific, significant amounts of silicified and chertified deep sea material have become available for study, and initial examinations by light microscopy and X-ray diffraction (HEATH and MOBERLY 1971; BORCH et al. 1971) indicate further that the formation of cristobalite represents a key stage in the silicification of limestone and in the formation of deep sea chert. Although study of these phenomena in relatively young geologic strata is still in a state of infancy, we wish to present at this time a brief summary of our own recent findings, and some of the pictorial evidence on which our conclusions are based.

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Results

A fracture section through the Oligocene chalk sample (DSDP 3/22/4/1) studied by WISE and HSÜ (1971) is depicted in the scanning micrograph shown in Plate I, Figure 1. As explained previously by WISE and HSÜ, this South Atlantic chalk consists primarily of braarudosphaerid pentolith segments cemented by inorganically precipitated low magnesium calcite which forms secondary overgrowths on nanofossils and fragmented skeletal particles. The overgrowths may develop crystal faces during advanced stages of cementation. The spherical aggregates in Plate I, Figure 1, are the late stage siliceous pore fillings tentatively identified as cristobalite by K. R. KELTS. Although radiolarian tests and some volcanic glass are present in calcareous ooze samples taken immediately above this chalk, these have not been observed in the particular chalk sample we studied. It appears most likely, therefore, that the silica was derived from the tests of siliceous organisms (and possibly volcanic glass) which was dissolved during the time of chalk formation and later reprecipitated within the void areas of the rock. It has been suggested (WISE and HSÜ 1971; WISE and KELTS 1971) that the chalk was indurated shortly after deposition of the original nanofossil ooze while still in the vicinity of the sediment-water interface. Cristobalite deposition probably followed shortly after calcite cementation, and as such, represents the initial step in the silicification of the chalk. This latter process was never carried to completion, however, due to the lack of sufficiently large amounts of silica in solution. Chert stringers, however, are reported within the Oligocene ooze slightly below the chalk layer (MAXWELL et al. 1970, p.435); therefore, sufficient silica was mobilized locally during this depositional sequence to produce small amounts of chert.

Although the small (about 3 to 5 micron) siliceous spherules constitute only 5 to 10% of the chalk, they are insoluble in hydrochloric and acetic acids, and form most of the insoluble residue of the rock (Pl. I, Fig. 2). It is this residue on which the X-ray determinations have been made.

As shown in a high magnification micrograph presented by WISE and HSÜ (1971, Fig. 3), the spherical aggregates consist of thin, plate or blade-like crystals which radiate outward from the center of the spheres. In the light microscope, the spheres are extinct under crossed nichols (due to the pseudoisotropic nature of the mineral) although the many fine blades diffract nonpolarized light sufficiently to produce an apparent (but not real) high refringence.

The second most common mineral in the insoluble residue is the zeolite, clinoptilolite, an authigenic mineral which forms in deep sea sediments as an alteration product of volcanic glass. Small specimens of this lath-shaped mineral are not uncommon in the residue studied (Pl. I, Fig. 3) and are identical in form and habit to the example depicted in a transmission electron micrograph by GIBSON and TOWE (1971). A more comprehensive account of these and other accessory minerals in the residue is being prepared by WISE and KELTS (1972, in preparation).

Siliceous spherules identical to those discovered in the South Atlantic were observed by WISE and KELTS (1971) in DSDP sample 4/29B/5/cc which was recovered in the central part of the Venezuelan Basin by Leg 4 of the Deep Sea Drilling Project. In this core catcher sample, the spherules are abundant in chalky inclusions within

the cherty Middle Eocene radiolarian ooze which, at this locality, is associated with a typical Horizon A chert sequence. These spherules also form an accessory cement, and one shown in the upper portion of Plate I, Figure 4, is tightly bound to the crossbars of a calcareous nannofossil (*Chiasmolithus* sp.). The bladed structure of these spherules is not evident in the micrograph; however, the blades may be obscured by a fine coating of clay particles received when the sample was placed in suspension during specimen preparation.

The cristobalite spherules studied up to this point in the investigation occur in rock sequences which contain volcanic glass, clinoptilolite, clays, and other products of volcanism as well as tests of siliceous micro-organisms. Although we presume that siliceous tests constitute the immediate source of silica for these spherules, some silica may have been derived directly from the dissolution of volcanic ash. Within the Caribbean and North Atlantic, volcanism is thought to have been closely related to the formation of siliceous oozes and cherts during certain geologic times (GIBSON and TOWE 1971; MATTSON and PESSAGNO 1971). Volcanics are also associated with many cherts recovered by deep sea drilling in the equatorial Pacific (FISCHER et al. 1971; TRACEY et al. 1971; WINTERER et al. 1971). A purely biogenous silica source for chert, therefore, cannot be demonstrated for the examples cited above. The often stated presumption that biogenous silica is the immediate source of silica for chert formation can only be demonstrated in a depositional sequence free of volcanic products. For this reason, we feel that an opaline chert²⁾ recovered from the Southern Ocean by one of us (FMW) is especially significant. About 2 kg of the rock was sampled at the base of a white Pliocene diatom ooze cored on the Kerguelen Plateau (ELTANIN Core 47-15; latitude 57° 17.27 S, longitude 78° 48.47 E). The chert unit was sufficiently thick to prevent complete penetration by the coring device. Despite severe damage to the cutter head, however, about 12 cm of the rock was cored. The opaline chert is pure white in color and breaks with a somewhat rough fracture. Fractures through the rock show a porous texture (Pl. II, Fig. 2) with cavities lined by spherules about 10 microns in diameter. These spherules are composed of thin radiating blades with an estimated thickness of 300 to 500 Angstroms (based on measurements made from scanning electron micrographs). With growth, the fine blades in the centers of the spherules coalesce to form dense structures which appear dark on electron micrographs (Pl. II, Fig. 1 and 2) wherever fracture surfaces pass directly through individual spherules or clusters of spherules. Nevertheless, these fracture surfaces are not completely smooth, but at high magnification exhibit a slightly roughened surface when viewed at an angle oblique to the electron beam (arrows, Pl. III, Fig. 1). Aside from the spherules, only rare fragments of siliceous microfossils have been observed in this material.

X-ray diffractographs of this chert (Textfig. 1) show that it is composed entirely of silica predominantly in the form of cristobalite. The cristobalite peaks shown here are identical to those obtained by K. R. KELTS for the siliceous residue from the South Atlantic chalk. Similar peaks are noted for deep sea cristobalites examined by other workers (example, see CALVERT 1971). They are, therefore, quite characteristic for this particular material. The peaks are rather broad, however, and for this reason,

²⁾ Not a quartz-rich chert (see discussion p. 160).

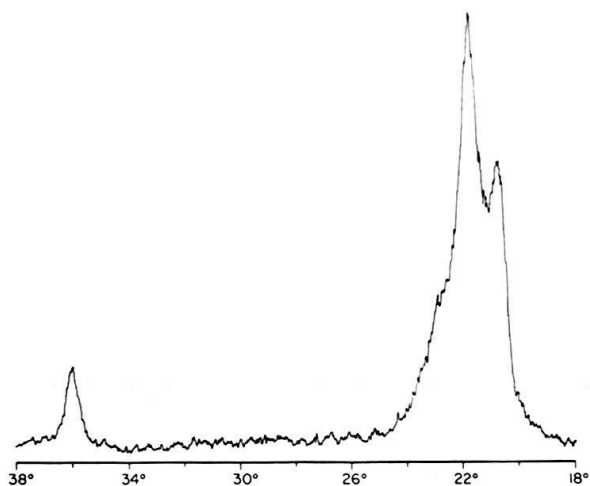


Fig. 1. X-ray diffractogram of a cristobalite chert recovered by ELTANIN Core 47-15 taken on the Kerguelen Plateau (Southern Ocean).

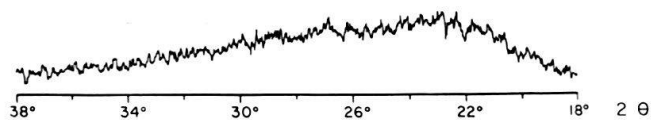


Fig. 2. X-ray diffractogram of a typical white diatom ooze from the Southern Ocean (ELTANIN Core 43-3-1, 20 cm).

this X-ray signature has been referred to as opal/cristobalite by several investigators (HERON 1969; GIBSON and TOWE 1971). A peak between 4.25 and 4.30 Angstroms is also present, and this is referred to the silica polymorph, tridymite, by some workers (CALVERT 1971). Although the peaks indicate some degree of disorder in the cristobalite structure, they are quite distinct when compared to the X-ray signature given by amorphous biogenous opal of the type which forms the diatom ooze immediately overlying the chert (Textfig. 2). Except for minor carbonate in the ooze, no peaks other than those shown occur in our diffractographs. It is clear, therefore, that both the Antarctic chert and the overlying ooze are free of montmorillonite, clinoptilolite, and other volcanic products noted in diffractographs of material from the South Atlantic and Caribbean. This, plus the stratigraphic occurrence of the chert directly beneath the ooze, strongly suggests that the silica forming the chert was derived directly from the dissolution of the diatom ooze. Following dissolution, the silica was *reprecipitated* as cristobalite.

The many recent findings of cristobalite-rich cherts in deep sea sediments is unusual in view of the fact that nearly all cherts from land sections are quartz-rich. It has been suggested (ERNST and CALVERT 1969; REX 1969; HEATH and MOBERLY 1971) that cristobalite is a relatively unstable intermediate phase in the reaction: biogenous opal \rightarrow cristobalite \rightarrow quartz-rich chert. Our preliminary examinations of quartz-rich deep sea cherts show fundamental morphological differences with the cristobalites studied. A Cretaceous quartz chert from the Sierra Leone Rise (DSDP 3/13A/6/cc) shown in Plate III, Figure 2, is composed predominantly of very small crystals 0.1 to 0.5 microns in diameter. The larger crystals are euhedral with prismatic and rhombohedral faces. These free growing crystals were chemically precipitated and did not form as pseudomorphs of cristobalite through a solid state reaction. Although this chert shows concoidal fracture in hand specimen, at high magnification the fracture surface appears quite rough.

WISE and HSÜ (1971) noted an apparent absence of siliceous spherules in shallow water sediments and considered them to be a product of the deep sea environment of

deposition. This observation may best be explained by the near absence of biogenous siliceous oozes in shallow water marine sediments. Chemically precipitated sedimentary cristobalite, however, has been reported in a number of different geologic settings (example, in some bentonites [GRUNER 1950]), and it is not unlikely that this mineral may exhibit the same form and habit in a number of different geologic environments. This possibility is further suggested by the discovery by one of us (BFB) of tiny cavities lined with spherules (Pl. III, Fig. 3) which occur in trace amounts in flint clays (Cretaceous-Tertiary) from the Coastal Plain of Georgia (USA). Although the concentration of these spherules is too low to permit X-ray diffraction analysis using standard preparation methods, the spherules (Pl. IV, Fig. 1) are strikingly similar in size and shape to those observed in the deep sea oozes and siliceous residues described previously. The morphologic details of the crystals composing the spherules (including the ragged edges and interpenetrating arrangement) (Pl. IV, Fig. 2) are indistinguishable from those exhibited by spherules in the Antarctic chert (WISE and WEAVER, unpublished). Although research by BFB and Mr. STEPHEN P. VAOS on the flint clays has only recently been initiated, it is suspected that the origin of the spherules is closely related to the complicated weathering history of the clay deposits.

Discussion

The significance of chemically precipitated sedimentary cristobalite to long-standing problems of silicification and chertification of carbonate rocks and deep sea oozes has long been neglected due to the relative rarity of this mineral in sediments exposed in land sections. An important exception is cristobalite in diatomaceous sediments now exposed in California (see BRAMLETTE 1946; ERNST and CALVERT 1969). Cristobalite is more often thought of as a high temperature form of quartz with a stability range beginning well above 1,400°C. For these reasons, the recent discoveries of significant quantities of alpha-cristobalite in deep sea sediments (and possibly subaerial weathering environments) are particularly interesting. As preliminary reports indicate (WISE and HSÜ 1971; HEATH and MOBERLY 1971), geologically young deep sea deposits contain important keys for interpreting the early lithification and diagenetic histories of older more highly altered and lithified carbonate and siliceous rocks exposed on land. This task should become easier as more is learned about the morphology and physical properties of the newly discovered deep sea materials.

The morphology of cristobalite spherules is distinctive and this facilitates their recognition in even trace amounts in sediment reasonably free of clay minerals. Thus, through scanning electron microscopy, it is possible to detect incipient stages of silicification of carbonate rocks. X-ray diffractometry of residues of carbonate rocks provides a second sensitive technique for detecting the presence of cristobalite. These two methods, together with standard petrographic analysis, will provide new insight in understanding the complex phenomena of silicification and chertification.

It is evident that the cristobalite we have observed is an authigenic mineral. It forms by the precipitation of silica from interstitial fluids. Therefore, the possibility that it forms from biogenous opal through a solid-solid reaction can be ruled out conclusively for the material we studied. Because this cristobalite does form through chemical precipitation, the migration of cristobalite-forming silica through distances

of centimeters to meters from its point of origin to its site of deposition as suggested by HEATH and MOBERLY (1971, p. 1003) is entirely feasible.

The well-formed blades of the cristobalite spherules indicate a reasonably high order of crystal structure. This crystallinity is also evident in our X-ray diffractographs which may be compared with those which JONES et al. (1964) prepared to show the range in variability in natural opals (these grade from amorphous silica to almost perfect alpha-cristobalite). As suggested by ERNST and CALVERT (1969, p. 121), there does seem to be an upper limit to the size of the crystalites. This seems to be a function of the size of the spherules which do not exceed 12μ in the material we have examined.

A high porosity is evident in electron micrographs of the cristobalite chert of ELTANIN Core 47-15. This is consistent with the unusually light weight of the rock. It should be noted that the pores are not molds of siliceous microfossils. Such molds are common in some porcelanites (BRAMLETTE 1946, p. 46).

Because ELTANIN Core 47-15 did not penetrate and sample ooze below the cristobalite chert unit, the age of this chert cannot be established with certainty. The age of the overlying diatomite is Pliocene (Gauss); therefore, the chert may also be as young as Pliocene. This suggests that the formation of cristobalite chert in the deep sea sediment may not necessarily require long periods of geologic time, and may begin at any time if favorable geologic conditions exist.

The term "chert" has been used with a wide degree of literary license by drillers, field geologists and petrographers alike, and only recently have renewed efforts been made to draw distinctions between opaline and quartz-rich varieties of this rock. REX and MURRAY (1970) distinguish "cristobalite chert" (REX 1970) from "quartz chert". CALVERT (1971) follows the lead of BRAMLETTE (1946) and includes cristobalitic cherts under the heading "porcelanite" while reserving the term "chert" for quartz-rich varieties. The cristobalite recovered in ELTANIN Core 47-15 may be referred to as "cristobalite chert" or as "porcellanite" depending on which of the above usages is followed.

Acknowledgments

We are indebted to Professor K. Jinhwa Hsü and Mr. Kerry R. Kelts (Department of Geology, Swiss Federal Institute of Technology [ETH], Zürich) for their continued collaboration on this study, and to Mr. Kelts for unpublished X-ray data on DSDP sample 3/22/4/1. Micrographs for Plate I were taken by SWW on a Cambridge Model IIA instrument kindly made available by Dr. Hans-Ude Nissen of the ETH; P. Cattori, H. E. Franz and R. Wessicken provided valuable technical assistance. SWW was supported at the ETH by an NSF Postdoctoral Fellowship; Prof. Dr. Hans M. Bolli (host professor) kindly provided laboratory facilities. Micrographs for Plates II-IV were taken by Mr. Ronald D. Parker, Department of Biology, Florida State University (FSU) on a Cambridge Model IIA instrument made available by Prof. Pasquale P. Graziadei of the Biology Department. Samples of flint clay were prepared by Mr. Stephen Vaos, Department of Geology, FSU. Partial support for FMW was provided by the National Science Foundation, Office of Polar Programs. We thank Drs. Lawrence A. Frakes and George W. DeVore for helpful discussions and encouragement. Mr. Dennis S. Cassidy (Department of Geology, FSU) assisted in preparing the plates, Mrs. Rosemarie Raymond drafted the diffractograms, and Mrs. Cynthia Curtiss Wise typed the manuscript. All Deep Sea Drilling Project samples were supplied by the National Science Foundation (USA).

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Plate I

All figures are scanning electron micrographs.

- Fig. 1 Oligocene chalk from the South Atlantic ocean basin (Deep Sea Drilling Project sample 3/22/4/1). This weakly silicified chalk, shown here in fracture section, is composed predominantly of disaggregated segments of the calcareous nannofossil, *Braarudasphaera rosa*, which have been cemented by low magnesium calcite overgrowths. The rounded aggregates (3 to 5 microns in diameter) nestled among the carbonate particles are spherules of a siliceous material deposited subsequent to carbonate cementation. 2,350 × .
- Fig. 2 Insoluble residue of the chalk shown in Figure 1 above. Most of the residue consists of the silicious spherules described above. X-ray analysis of this material by K. M. KELTS shows the spherules to be cristobalite. 1,000 × .
- Fig. 3 View at higher magnification of the insoluble residue shown in Figure 2 above. The rectangular lath-shaped crystal (upper portion of figure) is identified by WISE and Hsü (1971) as the zeolite mineral, clinoptilolite. 4,300 × .
- Fig. 4 Middle Eocene chalky ooze from the Venezuelan Basin (DSDP sample 4/25B/5/cc. Two siliceous spherules are shown, one of which partially obscures the center crossbars of a calcareous nannofossil (*Chiasmolithus* sp.) (upper portion of figure). 6,600 × .

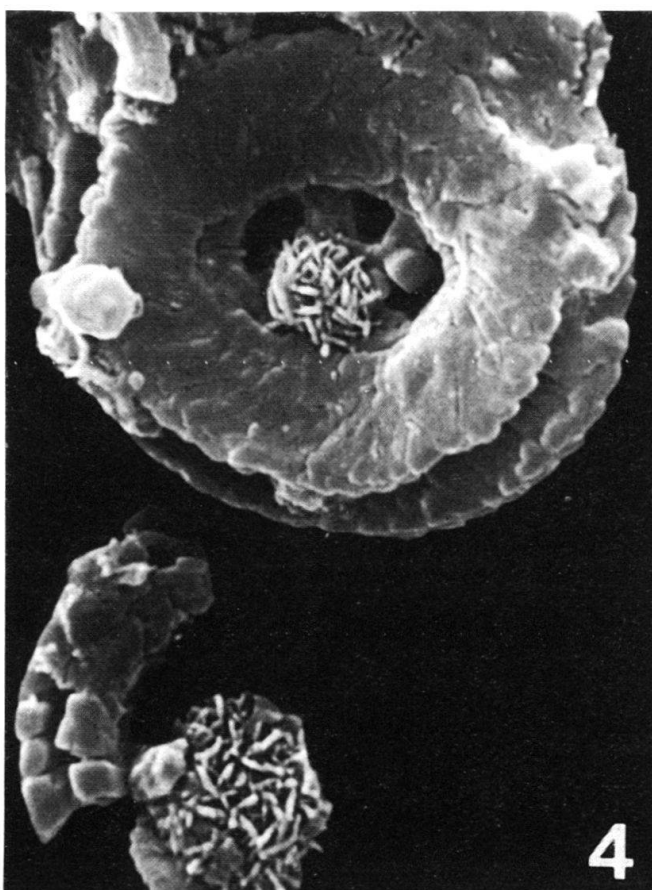
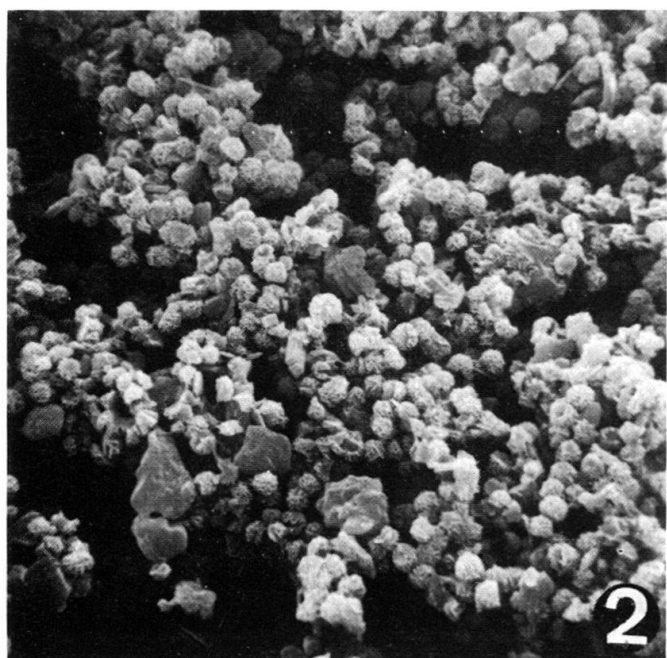
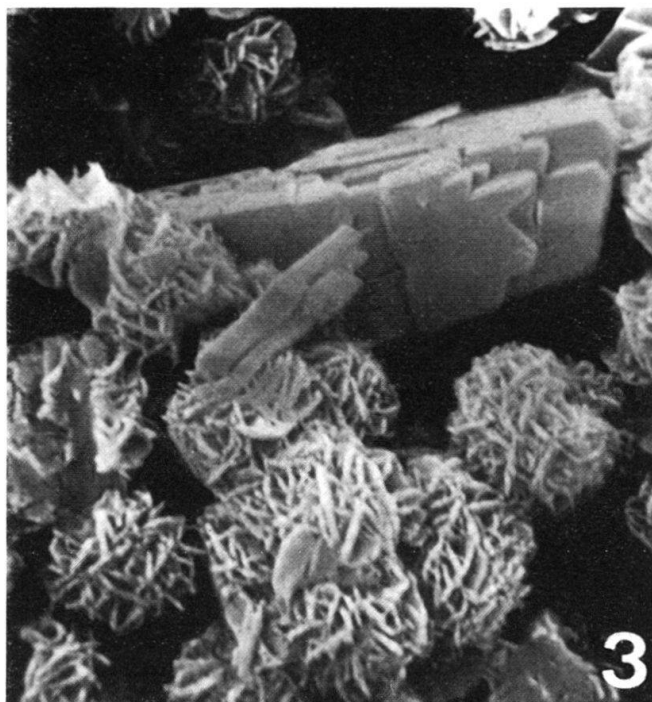
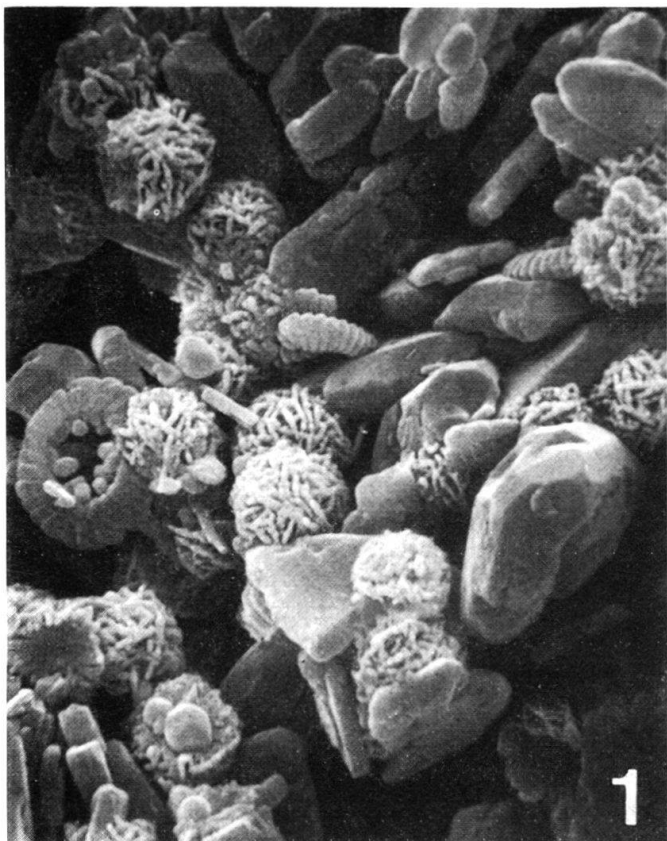


Plate II

All figures are scanning electron micrographs.

- Fig. 1 Pliocene (?) cristobalite chert from the Kerguelen Plateau (Southern Ocean: ELTANIN Core 47-15). Fracture section reveals many cristobalite spherules (up to 10 microns in diameter) lining pore spaces within the rock. Dark surfaces represent fracture planes which pass through individual spherules or clusters of spherules. 320 \times .
- Fig. 2 View at higher magnification of the cristobalite chert figured above. The spherules are composed of small (300 to 500 Angstroms thick) blade-shaped radiating crystals. The cristobalite silica was derived from the dissolution of siliceous microfossils. 1,250 \times .

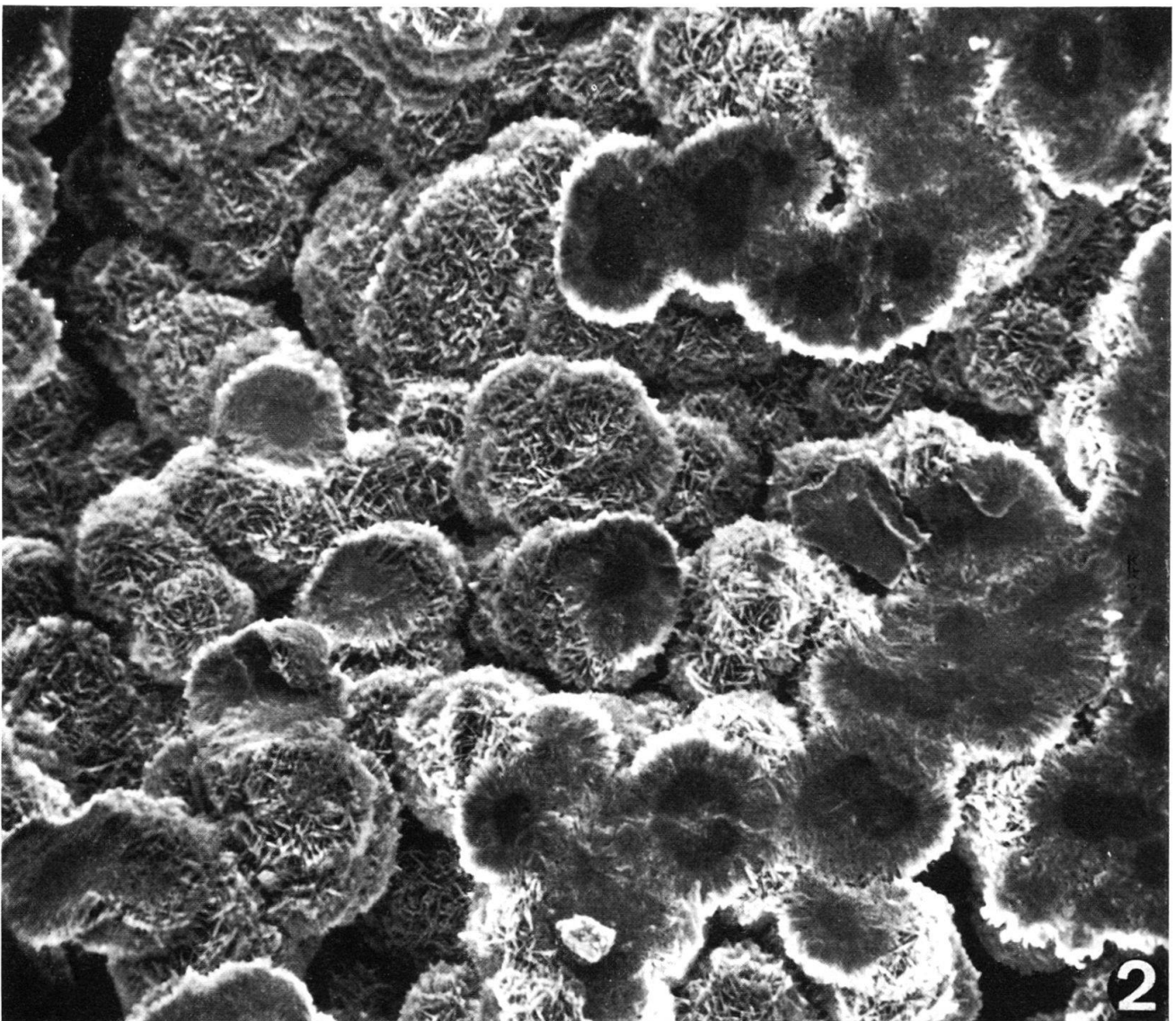
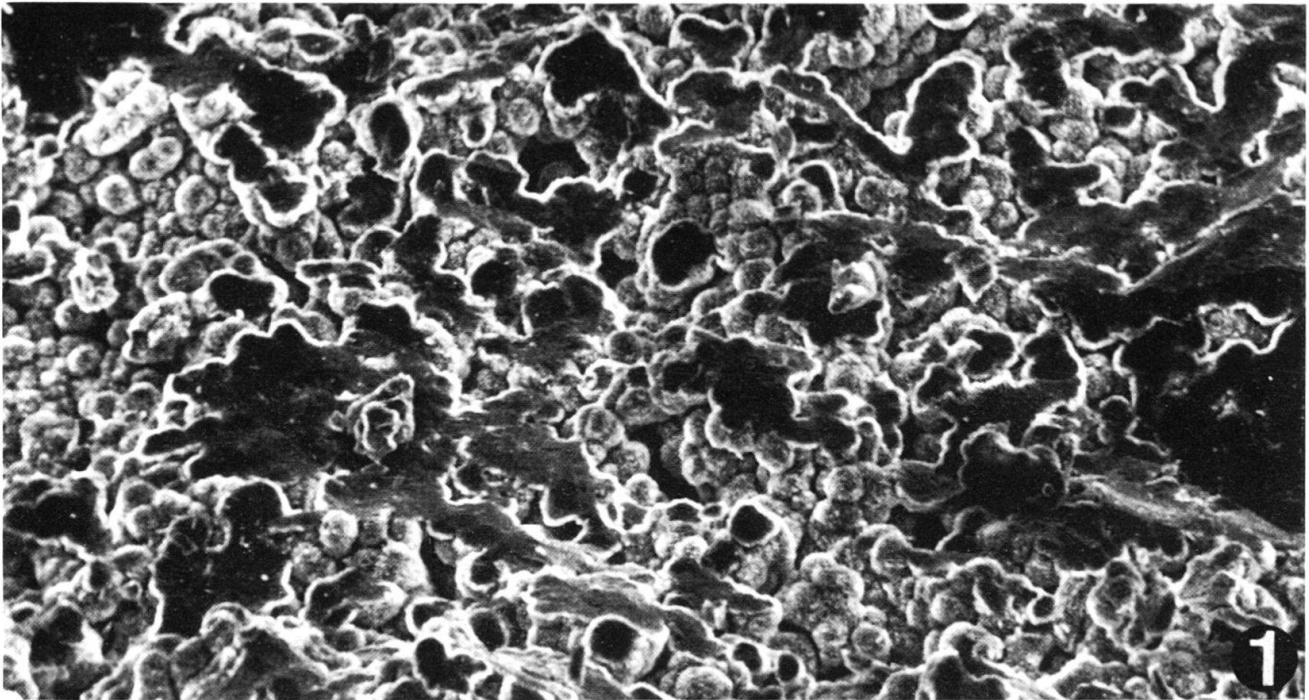


Plate III

All figures are scanning electron micrographs.

- Fig. 1 Cristobalite chert from ELTANIN Core 47-15 (figured in previous plate), oblique view. Fracture through spherules exhibits slightly roughened, fibrous surfaces. 1,050 ×.
- Fig. 2 Cretaceous quartz chert from the Sierra Leone Rise (Atlantic Ocean: DSDP sample 3/13A/6/cc). Quartz crystals revealed by this fracture surface are small (0.1 to 0.5 micron), elongate, and exhibit well-formed prismatic and bipyramidal faces. 900 ×.
- Fig. 3 Cretaceous-Tertiary flint clay from the Coastal Plain of Georgia (USA). Microcavities shown here within the clay are lined with siliceous spherules about 6 microns in diameter. 770 ×.

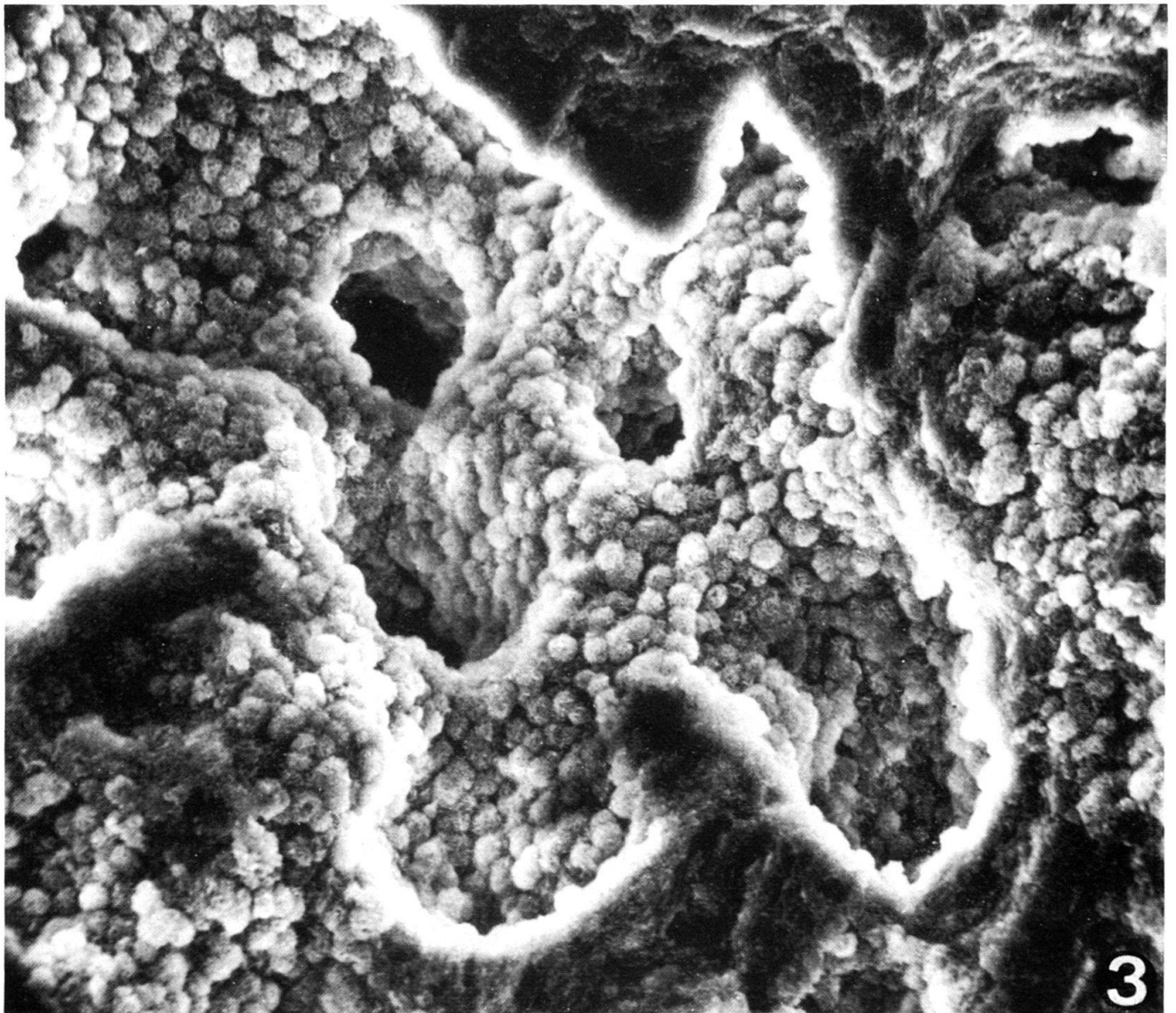
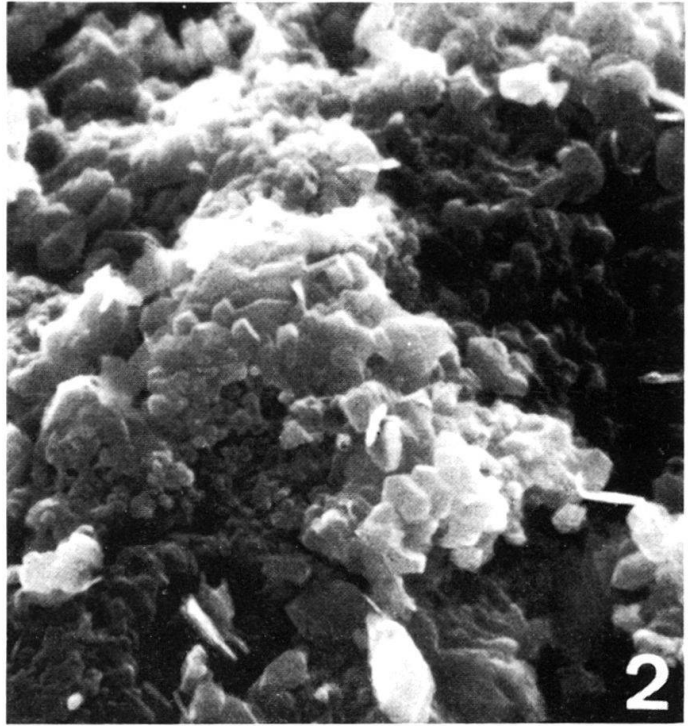
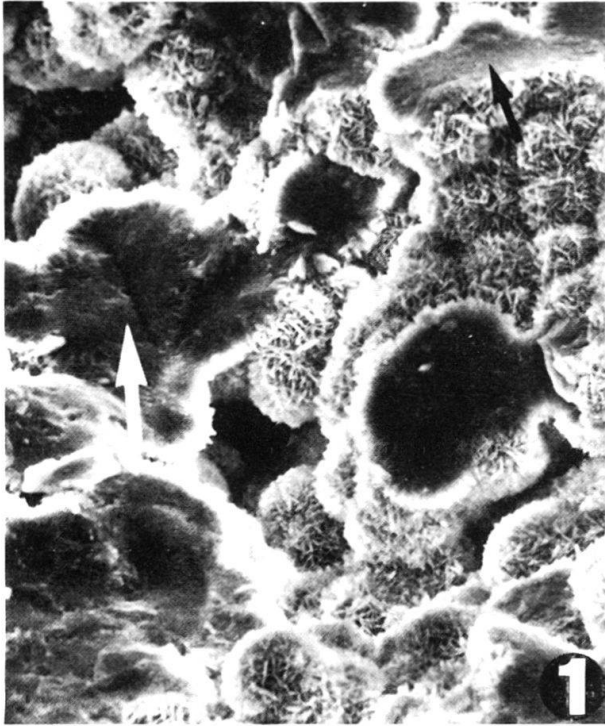


Plate IV

All figures are scanning electron micrographs.

- Fig. 1** Cretaceous-Tertiary flint clay from the Coastal Plain of Georgia (USA), enlargement of central portion of Plate III, Figure 3. Spherules are similar in size and shape to the cristobalite spherules depicted in Plates I and II. 2,700 × .
- Fig. 2** View at higher magnification of siliceous spherules figured in previous two micrographs. Material available is insufficient to allow a determination of the mineralogy of this material; however, its morphologic similarity to cristobalite suggests the possibility that cristobalite spherules may form in subaerial as well as deep sea environments. 13,000 × .

