Reports

Artificial Microfossils: Experimental Studies of Permineralization of Blue-Green Algae in Silica

Abstract. *A technique has been developed to artificially fossilize microscopic algae in crystalline silica under conditions of moderately elevated temperature and pressure. The technique is designed to simulate geochemical processes thought to have resulted in the preservation of organic microfossils in Precambrian bedded cherts. In degree of preservation and mineralogic setting, the artificially permineralized microorganisms are comparable to naturally occurring fossil algae.*

Almost all of the Precambrian microorganisms described in recent years (*1–4*) are cellularly preserved as structurally intact organic residues in bedded, microcrystalline cherts. Most of the reported assemblages are dominated by filamentous blue-green algae, many of which are morphologically quite similar to members of extant taxa (*2–4*). Although it seems apparent that these microbiotas were silicified in situ near the sediment-water interface, the exact processes involved in their preservation are rather poorly understood and little is known of the morphological and chemical changes that occur in organisms as a result of permineralization. In an effort to investigate these problems, we are conducting a series of experiments designed to artificially fossilize algae in silica under controlled conditions.

The origin of bedded cherts has been a subject of considerable speculation and controversy (*5–7*). Only a few examples of modern chert deposition are known (*6*), and none of these seems comparable to the Precambrian occurrences. However, many of the models proposed on the basis of geologic evidence are consistent with the following sequence of events which our experimental procedure is designed to simulate. (i) Weathering of silicate rocks produces dissolved silica which is transported to a relatively restricted basin; in the absence of silica-depleting organisms (during the Precambrian) the water becomes saturated with respect to silicic acid and a silica colloid is formed. The colloid tends to accumulate on the bottom of the basin where it permeates microorganisms living near the sediment-water interface. (ii) Through evaporation of water from the basin, perhaps aided by *p*H changes associated with microbial activity or by the addition of electrolytes, the colloid becomes increasingly concentrated and is converted from the sol to the gel state; the embedded microorganisms are thus "frozen" in a sort of siliceous "Jell-O." (iii) With continued water loss through syneresis and compaction, the gel is transformed to an amorphous opaline state; subsequent crystallization produces an interlocking mosaic of microcrystalline quartz grains, forming fossiliferous, bedded chert.

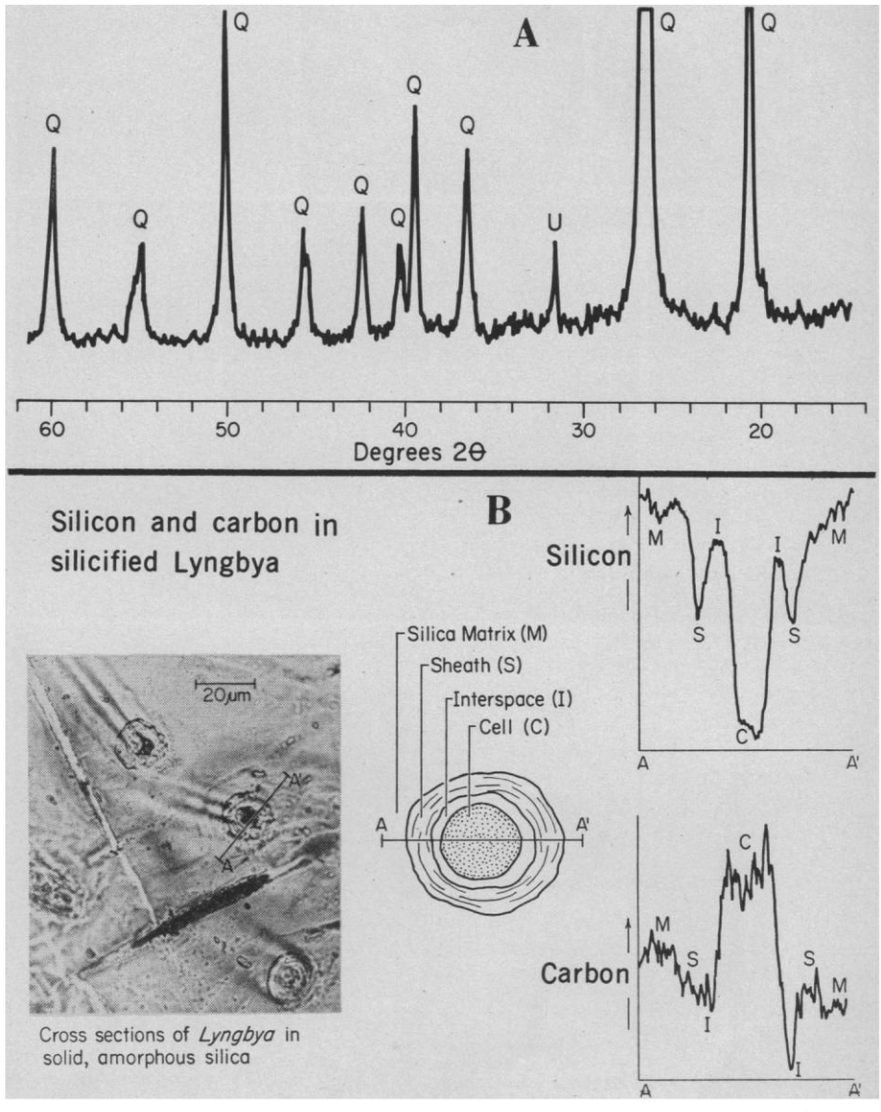
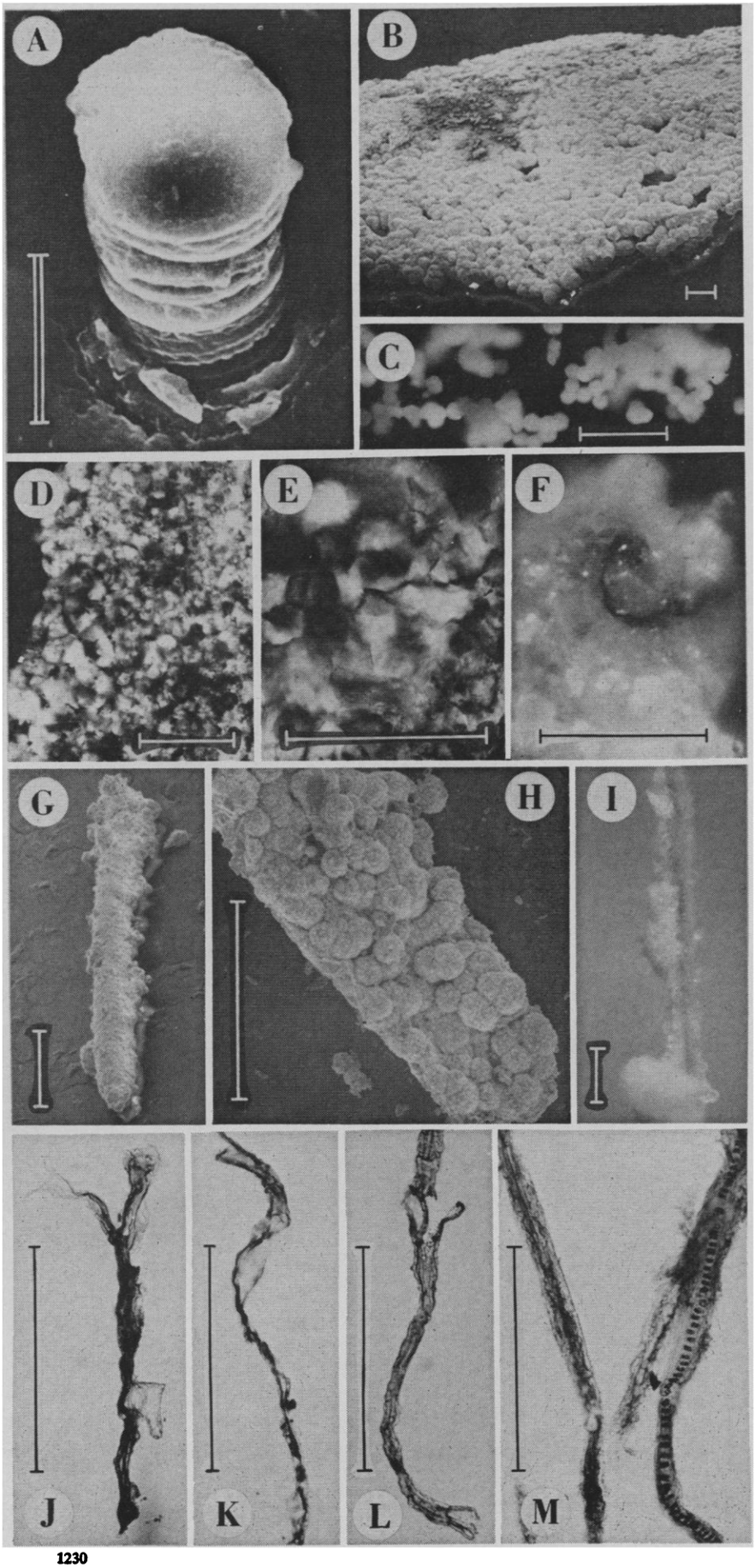


Fig. 1. (A) X-ray diffraction pattern of synthetic quartz formed from silica gel treated at 165°C and 3000 bars for 2 weeks; CuKα radiation with a LiF monochromator, 40 kv, 20 ma; Q = quartz; U = unidentified peak. (B) Distribution of silicon and carbon in artificially silicified *Lyngbya*, determined by electron probe x-ray microanalysis; 15 kv, 0.04 μa on brass.



Although this model cannot account for the genesis of some chert beds, such as those known to be of secondary or biological origin, it appears to approximate rather closely processes involved in the formation of bedded algal cherts of the type occurring in the Bitter Springs (*2*) and Gunflint (*3*) formations, perhaps the two most important fossiliferous Precambrian units now known.

Our simulation of this model involves three major stages: (i) permeation of living blue-green algae by a solution of colloidal silica; (ii) conversion of the silica colloid to a solid gel; and (iii) transformation of the amorphous silica gel to microcrystalline quartz under conditions of moderately elevated temperature and pressure.

In the first stage of the procedure, filamentous blue-green algae of the genus *Lyngbya* (*8*) are stirred in a solution of colloidal silica (*9*) for a period of 24 to 48 hours. In the second stage, the colloid is converted to a gel by neutralizing the *p*H with HCl and by adding NaCl to produce a concentration of 0.1*N* NaCl. In the third stage, standard hydrothermal techniques and apparatus (*10*) are used to crystallize the silica. Briefly, samples of the solid gel, containing embedded algae, are sealed in silver capsules which are placed in heated pressure vessels where the selected temperature and aqueous fluid pressure are maintained.

Fig. 2. (A) Scanning electron micrograph of *Lyngbya* filament in solid, amorphous silica, exposed at the surface by HF etching. (B) Scanning electron micrograph of a sheet of aggregated synthetic quartz microspheres. (C) Optical photomicrograph of single spheres of synthetic quartz (reflected light). (D) and (E) Optical photomicrographs of synthetic chert (polarized, transmitted light). (F) Optical photomicrograph of *Lyngbya* filament preserved in synthetic chert (reflected light). (G) Scanning electron micrograph of *Lyngbya* filament preserved in synthetic quartz. (H) Scanning electron micrograph of *Lyngbya* filament preserved in synthetic quartz microspheres. (I) Optical photomicrograph of *Lyngbya* filament preserved in synthetic quartz (transmitted and reflected light). (J) through (L) Optical photomicrographs of "artificial fossils" (*Lyngbya*) freed from the synthetic quartz matrix by HF maceration (transmitted light). (M) Optical photomicrograph of natural algal microfossils, with and without preserved cells, freed from Ordovician chert by HF maceration (*16*) (transmitted light). Line for scale in (A) represents 10 μm; lines for scale in (B) through (M) represent 100 μm.

The net reaction occurring under these conditions is as follows (*11*):

$$n\,Si(OH)_4 \rightarrow (SiO_2)_n + 2n\,H_2O$$

During this reaction, depending primarily on the temperature used, the algae undergo varying degrees of thermal alteration similar to the natural process of "carbonization" (*12*).

In an effort to determine optimum conditions of crystallization, hydrothermal experiments were conducted at temperatures ranging from 100° to 300°C and pressures between 1000 and 3000 bars. Best results were obtained in experiments lasting 2 to 4 weeks at temperatures of about 150°C and pressures of 2000 to 3000 bars. Under these conditions, silica crystallizes relatively rapidly and thermal alteration of the included algae is slight [approximating the "thermal alteration index 2" of Staplin (*13*)]. At lower temperatures or pressures, several months are required for crystallization (*7*); at higher temperatures the algae are severely altered and may volatilize (*14*).

In order for the "artificial fossils" produced in these experiments to be considered close facsimiles of naturally occurring microfossils, it must be demonstrated that hydrothermal treatment has resulted in crystallization of the amorphous silica gel and that the embedded algae are structurally intact and are completely permeated by the siliceous matrix. The crystallinity of the hydrothermal products is readily observable by optical or scanning electron microscopy; x-ray diffraction patterns of this material (Fig. 1A) exhibit all characteristic peaks of alpha quartz and occasionally include some peaks of residual cristobalite, a naturally occurring crystalline intermediate between amorphous silica and quartz. In order to determine the degree of silica permeation of the included algae, we measured the distribution of silicon and carbon in transverse sections of *Lyngbya* filaments embedded in solid, amorphous silica with an electron probe x-ray microanalyzer. As is shown in Fig. 1B, although relatively less silicon is present in highly organic portions of such filaments, the algae are thoroughly permeated by silica in a manner quite comparable to that which occurs in nature. And, as is evident in Figs. 1B and 2A, the algae remain structurally intact during silica infiltration.

Two forms of quartz have been synthesized in these experiments. The most common product is in the form of small spheres, less than 20 μm in diameter and composed of euhedral to subhedral quartz crystallites; these spheres occur both singly (Fig. 2C) and aggregated in sheets (Fig. 2B). Natural opals are composed of a rather similar aggregation of closely packed silica spheroids (*15*). Fine-grained cherty material is also synthesized; as is shown in Fig. 2, D and E, this second form of synthetic quartz occurs as an interlocking mosaic of subhedral to anhedral crystals that is essentially indistinguishable in texture and mineralogy from natural chert.

Artificially fossilized algae have been detected in both forms of synthetic quartz. Figure 2F shows an algal filament contained in a fragment of the cherty material. Figure 2, G to I, shows similar filaments embedded in quartz predominantly of the small spheroidal form; the organic filaments appear to have influenced crystallization, presumably by serving as sites for crystallite nucleation. The predominance of small quartz grains in highly fossiliferous areas of both the Gunflint (*3*) and Bitter Springs cherts (*2*) may reflect a similar phenomenon.

In Fig. 2, J to L, are shown algal filaments that were freed from their synthetic quartz matrix by maceration in 5 percent HF. Although individual cells are not preserved in these "artificial fossils," strands of brownish cellular material are clearly visible within the amber organic sheaths. This degree of organic preservation is widespread in nature and commonly coexists with cellularly preserved microorganisms; one example of such an occurrence is illustrated in Fig. 2M, which shows filamentous blue-green algae freed by acid maceration from an Ordovician chert (*16*).

Unlike previous attempts to artificially silicify modern plant material (*17*), the technique described here results in structurally intact, permineralized microorganisms that are thoroughly embedded in a crystalline silica matrix. These "artificial fossils" are comparable in morphology, color, and mineralogic setting to their naturally occurring counterparts and are produced by a series of stages that seem analogous to processes involved in the deposition of natural bedded cherts. Experiments in progress suggest that this new technique will provide useful information regarding ultrastructural and organic geochemical changes that occur during "carbonization" of permineralized fossils and, as we suggested earlier (*18*), may eventually provide a basis for interpretation of the micromorphology and chemistry of organically preserved microorganisms.

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