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PRE-CENOZOIC NODULAR CHERTS: EVIDENCE FOR OPAL-CT PRECURSORS AND DIRECT QUARTZ REPLACEMENT

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ABSTRACT. Deep-sea nodular and bedded cherts recovered by the Deep Sea Drilling Project form by a maturation process whereby chert evolves from originally dispersed biogenic opal-A, through an intermediate opal-CT porcellanite stage, to quartz porcellanite or chert. Quartz-replaced opal-CT lepispheres and rim cements are evidence that many Mesozoic and Paleozoic shelf nodular cherts also had an opal-CT precursor. Nodular chert formation by the same maturation process is supported by the similar microcrystalline quartz mineralogy of many cherts. Chert formation by direct replacement of limestone by quartz also occurs. These cherts may be distinguished by the absence of quartz-replaced lepispheres and a coarser (>10 μm) crystal size. Small crystal sizes of cherts with opal-CT precursors are predicted by application of the Freundlich-Ostwald equation relating quartz solubility and surface area.

INTRODUCTION

The discovery of a great variety of Cenozoic and late Mesozoic cherts in various stages of development during the Deep Sea Drilling Project has greatly increased our knowledge of the formative processes of chert. In the deep sea, both nodular and bedded cherts appear to form by a maturation process, whereby chert evolves from original biogenic opal-A, through an intermediate opal-CT composition (terminology of Jones and Segnit, 1971), to quartz (Heath and Moberly, 1971; Heath, 1973; Von Rad and Rösch, 1974; Wise and Weaver, 1974; Keene, 1975; Von Rad, Reich, and Rösch, 1978; Hein, Vallier, and Allan, 1981; and others).

The free growth forms of opal-CT are lepispheres, which are 5 to 20 μm spherulitic aggregates of blade-shaped opal-CT crystals (Wise and Kelts, 1972), and less commonly, non-spherulitic blades (Hein and others, 1978), rim cements (Von Rad, Reich, and Rösch, 1978), and overgrowths (Hein and others, 1978). Opal-CT also occurs as a massive cement (Hein and others, 1978). Quartz-replaced lepispheres are the only textural evidence for an opal-CT precursor in deep-sea quartz cherts. These replaced lepispheres occur as either massive spheres or spheres composed of radiating quartz crystals (Von Rad, Reich, and Rösch, 1978; Hein, Vallier, and Allan, 1981). No evidence of the bladed ultrastructure is preserved. Similar quartz-replaced lepispheres in pre-Cenozoic nodular cherts would be evidence for this maturation from opal-CT to quartz. Rim cements may also be preserved with textures reminiscent of opal-CT.

Although the maturation theory has been proposed for some Paleozoic cherts (for example, DeCelles and Gutschick, 1983; Selleck, 1985), possible quartz-replaced lepispheres have been described only from the Mississippian Lake Valley Formation (Meyers, 1977) and Arkansas Novaculite (Jones and Knauth, 1979). In this paper we: (1) present the evidences of opal-CT precursors found in some Mesozoic and Paleozoic cherts; (2) evaluate the maturation theory for the origin of pre-Cenozoic nodular cherts; and (3) apply Ostwald-ripening theory to crystal sizes of replacement cherts.

OPAL-CT RELICS IN MESOZOIC AND PALEOZOIC CHERTS

The following chert-bearing formations were examined for possible opal-CT relict fabrics: (1) Upper Chalk (Upper Cretaceous) of southern England, a deep water pelagic deposit (Hancock, 1975); (2) Portland Limestone (Upper Jurassic) of southern England, a shallowing upward shelf sequence (Townson, 1975); and (3) Onondaga Limestone (Lower Devonian) of eastern New York, a deepening upward shelf limestone (Oliver, 1956; Lindholm, 1967). Both quartz-replaced and possibly persistent opal-CT lepispheres have previously been described from the Upper Cretaceous chalks of Europe (for example, Buurnam and Van der Plas, 1971; Scholle, 1974; Pomerol and Aubry, 1977; Clayton, 1982).

In addition to the above formations, single chert samples from 10 other Paleozoic formations were examined by optical and/or scanning electron microscopy. Possible opal-CT relics were found in half the samples. Of these additional Paleozoic cherts, only those relics from the Markgraf Member of the Joliet Formation (Middle Silurian) of northern Illinois (Mikulic and others, 1985, stop 2) are discussed in this paper.

Upper Chalk.—Chert in the Upper Cretaceous Upper Chalk of southern England occurs predominantly as burrow flints, which are chert nodules that nucleated within thalassinidean burrows (Bromley, 1967). Burrow flints have a core of black vitreous chert and commonly a thin outer rim of white porcellanous chert. This texture is similar to that of some deep-sea nodular cherts (for example, Lancelot, 1973; Wise and Weaver, 1974; Keene, 1975), except that in the Upper Chalk the rim is now composed of quartz rather than the opal-CT of deep sea cherts.

It is within the outer porcellanous rim that quartz-replaced opal-CT lepispheres are best preserved. The replaced lepispheres are 10 to 15 μm in diameter and consist of radiating quartz crystals (fig. 1A). No evidence of the bladed lepisphere ultrastructure is preserved. Possible ghosts of lepispheres are poorly visible by optical microscopy in the black chert core, which consists of relatively pure microcrystalline quartz. The lepispheric microstructure of the burrow flint core can be seen more clearly with SEM on hydrofluoric acid etched polished surfaces of some nodules (fig. 1B). The darker spheres and patches in figure 1B are similar in size, shape, and distribution to the quartz-replaced lepispheres in the porcellanous rim. The lighter appearing

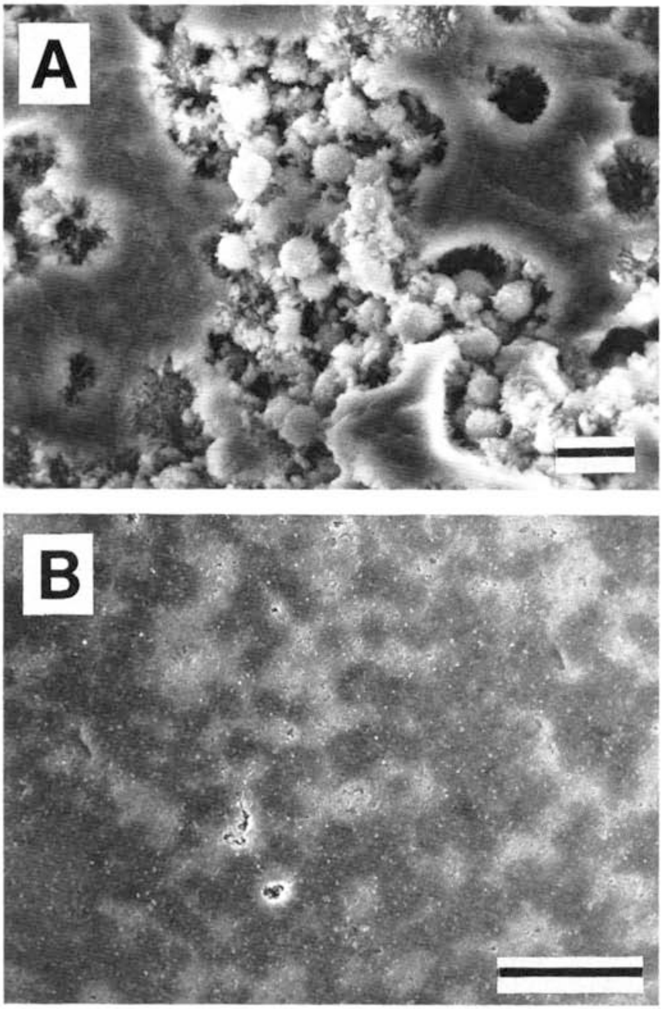

Fig. 1. Upper Chalk. (A) Quartz-replaced lepispheres in porcellanous rim of flint nodule. SEM. Bar scale 20 μm. (B) Etched polished surface of core of flint nodule. Ghosts of lepispheres appear as dark areas. Note that ghosts of lepispheres in the core of the flint nodule have the same size and distribution as quartz-replaced lepispheres in the nodule rim. SEM. Bar scale 100 μm.

interstitial silica was more strongly etched than the quartz-replaced lepispheres. Clayton (1982) indicated that this interstitial silica is a structurally disordered chalcedony. The lepispheric microstructure occurs throughout the flint nodules.

Portland Limestone.—A wide variety of presumable opal-CT relict textures was found in Portland Limestone chert nodules. All textural types are now microcystalline quartz or chalcedony. Most common are 20 to 30 μm microspheroids, which line pores and replace intergranular and geopetal micrite. The microspheroids are either homogenous or are differentiated into an inner core of quartz with abundant water or air vaculoes and an outer rim of clear quartz about 10 μm thick (fig. 2A). In some microspheroids the core is empty or filled with calcite. The microspheroid quartz is either length-fast chalcedony, with crystals radiating outward from the core, or microcrystalline quartz. These microspheroids were found in all silicified carbonate sands.

Some intergranular pores and solution voids of spicules of the sponge *Rhaxella* are lined with a layer of relatively clear quartz similar in its thickness, mineralogy, and appearance to the outer rim of the microspheroids. These layers have a broad hemispherically lobate morphology and may be continuous with the outer rim of some microspheroids (fig. 2B). The remaining pore space is filled predominantly with centripetally oriented sheaves of length-fast chalcedony. In one sample both the pore-lining silica layers and micropherules were replaced by calcite, whereas the adjacent pore-filling length-fast chalcedony was not replaced. This difference in susceptibility to replacement by calcite strongly suggests that at the time of replacement the microspheroids and pore-lining silica layers had a different mineralogy than the pore-filling chalcedony.

The pore-lining silica layers observed in the Portland Limestone are morphologically similar to opal-CT rim cements in Miocene deep sea conglomerates (Von Rad, Reich, and Rösch, 1978) and a Cretaceous deep sea breccia (Hein, Vallier, and Allan, 1981). In these deep-sea occurrences, Opal-CT occurs as rims or hemispheres, up to 80 μm thick, lining cavities. Opal-CT cementation was followed by quartz precipitation, which filled the remaining intergranular porosity in the conglomerate. Some of the opal-CT rim cements were later replaced by chalcedonic quartz.

Onondaga Limestone.—Possible former opal-CT lepispheres, now replaced by quartz, were found lining pore walls and as a replacement of matrix in Onondaga Limestone chert nodules (fig. 3A). The pore lining microspheroids range in diameter from 20 to 25 μm and are composed of microcrystalline quartz. The remaining intergranular porosity was typically filled centripetally with inward oriented sheaves of length-fast chalcedony and granular megaquartz. In one sample from the Moorehouse Member, quartz microspheroids were found lining a pore that was unfilled (fig. 3B). These spheres, which are approx 25 μm in diameter, are similar in size and morphology to pore lining opal-CT spheres in the Cretaceous deep-sea chert breccia. The opal-CT crystals in the deep-sea spheres are apparently randomly oriented and thus lack the normal lepisphere ultrastructure (Hein, Vallier, and Allan, 1981). Possible ghosts of lepispheres were observed by both optical microscopy and SEM in etched polished samples of Seneca and Moorehouse Member chert nodules. The circular zones revealed by etching are similar in size

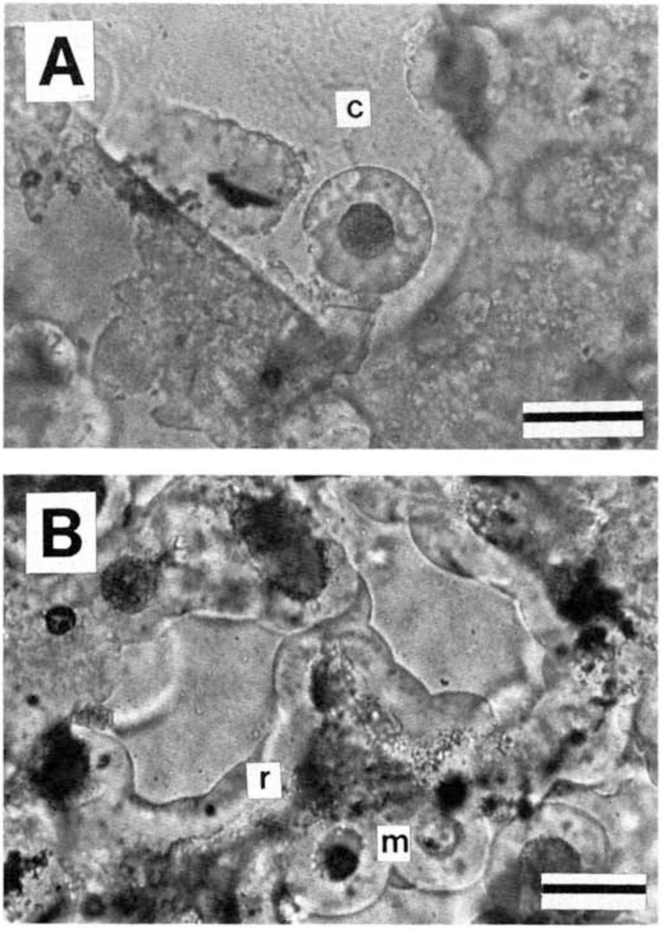

Fig. 2. Portland Limestone. (A) Microspheroid differentiated into an inclusion-rich core and clear rim. Microspheroid occurs along wall of pore that was later filled with length-fast chalcedony (c). Plane polarized light. Bar scale 30 μm. (B) Quartz-replaced opal-CT rim cement (r) lining solution voids of *Rhaxella* spicules. Length-fast chalcedony filled rest of void. Note similarity of outer rim of microspheroids (m) and rim cement. Plane polarized light. Bar scale 30 μm.

and manner of preservation to those found in Upper Chalk flint nodules.

Joliet Formation.—Quartz microspheroids, 20 to 30 μm in diameter, are extremely abundant in a chert nodule from the Markgraf Member of the Joliet Formation. The microspheroids, which are composed of microcrystalline quartz, line intergranular pores and replace the matrix of this chertified fossiliferous wackestone (fig. 4A). Length-fast chalcedony and to a lesser degree granular megaquartz filled the remaining intergranular porosity. Interstices between microspheroids in the silici-

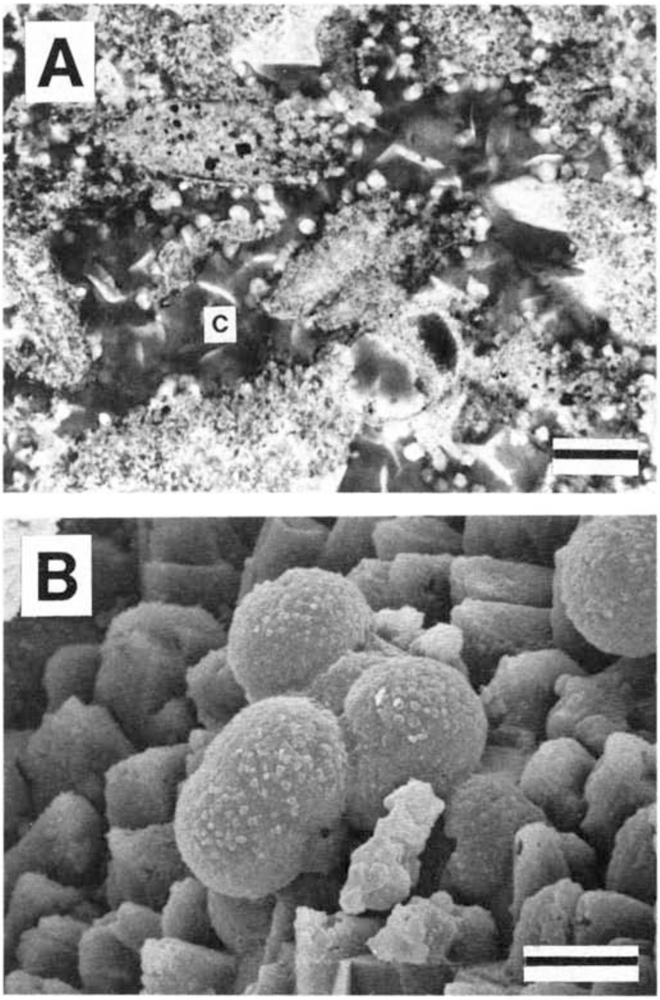

Fig. 3. Onondaga Limestone. (A) Microspheroids lining intergranular pores in a silicified grainstone nodule. Remaining porosity was filled with length-fast chalcedony (c). Plane polarized light. Bar scale 100 μm. (B) Microspheroids lining pore wall. No evidence of lepispheric ultrastructure is preserved. SEM. Bar scale 20 μm.

fied matrix are filled with length-fast chalcedony or are empty. Fusion of microspheroids occurs in the matrix (fig. 4B), but the original spheroidal habit is still discernible. The microspheroids are present throughout the silicified matrix of the chert nodule.

MATURATION SEQUENCES

The microspheroids and rim cements found in this study are similar in size, shape, and distribution to opal-CT and quartz-replaced lepispheres and rim cements found in Cenozoic and Cretaceous deep-sea cherts. This suggests that many pre-Cenozoic deep and shallow water

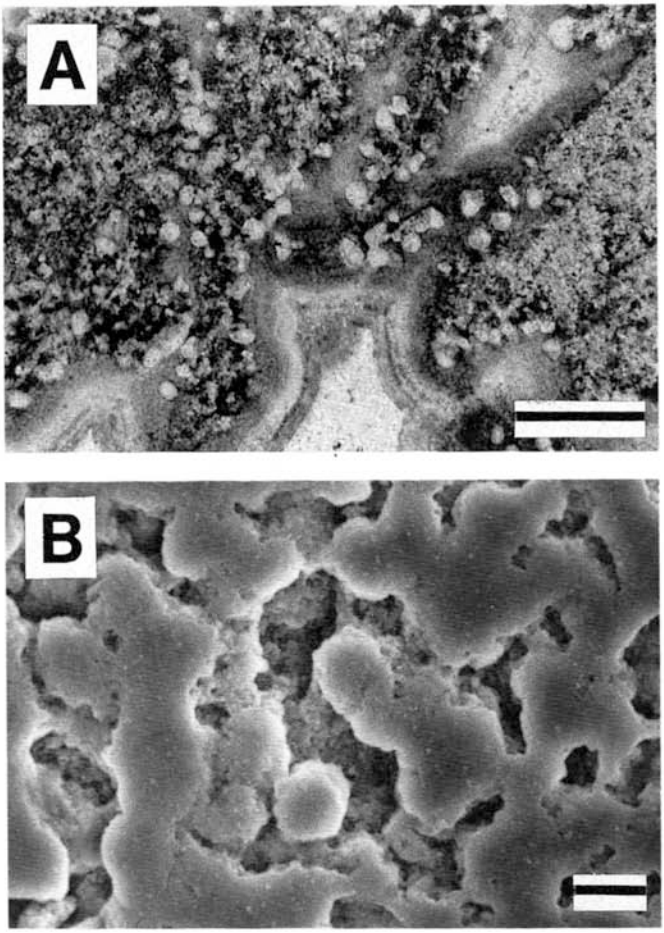

Fig. 4. Joliet Formation. (A) Microspheroids lining pores that were later filled with length-fast chalcedony and granular megaquartz. Similar microspheroids are abundant in silicified matrix. Plane polarized light. Bar scale 100 μm. (B) Etched polished surface of chert nodule. Fusion of microspheroids is common, but 20 μm spheres are still discernable. SEM. Bar scale is 20 μm.

nodule cherts very likely formed by a maturation process similar to that of Cenozoic deep-sea cherts. From our observations on a variety of Phanerozoic nodular cherts and the studies of deep-sea cherts by Heath and Moberly (1971), Keene (1975), and Hein, Vallier, and Allan (1981) we can recognize the following 7 stage maturation replacement sequence:

1. Blades of opal-CT are precipitated in the interstices of the carbonate sediment and within intragranular pore spaces (for example, foraminiferal chambers).

2. The opal-CT blades join and grow to form lepispheres and, less commonly, rim cements.
3. Carbonate matrix is replaced by massive opal-CT.
4. Chalcedony and to a lesser degree granular megaquartz is precipitated within remaining intra- and intergranular porosity.
5. Fossils and other carbonate grains are replaced either by opal-CT or quartz.
6. Opal-CT is converted to quartz.
7. Further quartz crystal growth and recrystallization combine to form dense chert.

The 7 stages of the above sequence occur simultaneously in some growing chert nodules. Opal-CT may be precipitating and replacing carbonate at the boundaries of the nodules (stages 1–3), while opal-CT is being recrystallized into quartz in the nodule interior (stages 4–7). This results in nodules with cores of chert and thin porcellanous outer rims of opal-CT. The relative timing of pore-filling quartz precipitation (stage 4) and the replacement of fossils (stage 5) are variable.

Foraminiferal tests in deep-sea cherts can be replaced directly by chalcedony (Heath and Moberly, 1971; Keene, 1975). Replacement of individual fossils in shelf limestone matrices by quartz euhedra and granular megaquartz crystals with some euhedral terminations appears to have occurred with no opal-CT precursor (Maliva and Siever, 1988). Opal-CT, which has a completely different crystal habit and form than quartz, would not have replaced calcite with a hexagonal form having quartz rational faces. We find no firm evidence of an opal-CT precursor to replacements by microcrystalline quartz or chalcedony. If densely silicified grains had an opal-CT precursor, the opal-CT would have been in a massive rather than lepispheric or balded form leaving no traces surviving recrystallization to quartz.

CRYSTAL SIZE AND MATURATION

Quartz crystal size is another criterion, albeit not by itself unequivocal, that can be used in distinguishing between cherts that formed by a maturation process and primary quartz cherts. Crystal size is a function of the rates of nucleation and crystal growth. If the rate of quartz crystal nucleation from opal-CT is very rapid relative to the crystal growth rate then coarsely crystalline chert could not form by the recrystallization of an opal-CT precursor. Deep-sea cherts with opal-CT precursors typically consist of cryptocrystalline or microcrystalline quartz or very finely crystalline chalcedony. Similarly, all the pre-Cenozoic shelf chert containing ghosts of lepispheres now consists of either microcrystalline quartz or very finely crystalline chalcedony. These observations lead us to conclude that the rate of quartz crystal nucleation during opal-CT recrystallization is typically rapid enough to limit quartz crystal size in cherts that formed by a maturation process to a maximum of about 10 μm.

Another argument predicts the small crystal size in cherts derived from opal-CT. Williams, Parks, and Crerar (1985) proposed that in chert, quartz crystal size increases by Ostwald-ripening from an earlier chalcedony or cryptocrystalline quartz stage to a later microcrystalline quartz stage though they did not calculate specific crystal sizes. The driving force of Ostwald-ripening, whereby larger crystals grow at the expense of smaller ones, is the solubility difference resulting from excess Gibbs free energy associated with the solid-liquid interface. As crystal size increases, and therefore specific surface area decreases, the excess free energy and hence solubility of the crystals decrease. Ostwald-ripening would continue at a decreasing rate until a crystal size is reached where the excess free energy associated with the crystal surface becomes negligible. We may calculate the maximum size of crystals produced by this process.

The relationship between quartz solubility and specific surface area can be expressed by the Freundlich-Ostwald equation (Enüstün and Turkevitch, 1960; Parks, 1984; Williams, Parks, and Crerar, 1985):

$$\mathrm{Log}\,\frac{S_a}{S_o} = \frac{\gamma M}{2.3RT}\left(\frac{2A}{3}\right)$$

where S_a = solubility of quartz with specific surface area A, S_o = solubility of quartz particles with an infinite radius, R = gas constant, T = absolute temperature, γ = surface free energy at the solid-water interface, M = molecular weight, and A = specific surface area (square meters per gram). This equation is strictly valid only for populations of geometrically similar particles with a uniform crystal size. The relationship between the index of excess solubility (S_a/S_o) and quartz particle diameter is illustrated in figure 5 for particles with different degrees of roughness. Quartz crystals with diameters of less than 0.1 μm have solubilities significantly increased over those of larger grains. As crystal diameter exceeds 1 μm the excess solubility due to surface energy becomes vanishingly small. Cherts in which quartz crystal growth occurred by Ostwald-ripening would, therefore, have an upper limit to crystal size of between 1 and 10 μm, the crystal size range that encompasses most Mesozoic and Paleozoic nodular and bedded cherts. Quartz crystal size may increase to a greater degree, however, as a result of metamorphism (for example, Jones and Knauth, 1979).

Petrographic evidence indicates that replacement quartz and cements that precipitated directly as quartz, and are not the product of recrystallization of opal-CT, do not have such an upper limit to crystal size and may, therefore, grow to much coarser sizes. Brachiopod and oyster shells in limestone matrices, for example, were observed to be replaced by granular megaquartz crystals that were in excess of 1 mm in maximum dimension (Maliva and Siever, 1988). Euhedral quartz terminations at some quartz-shell contacts are evidence that these crystals precipitated directly as quartz.

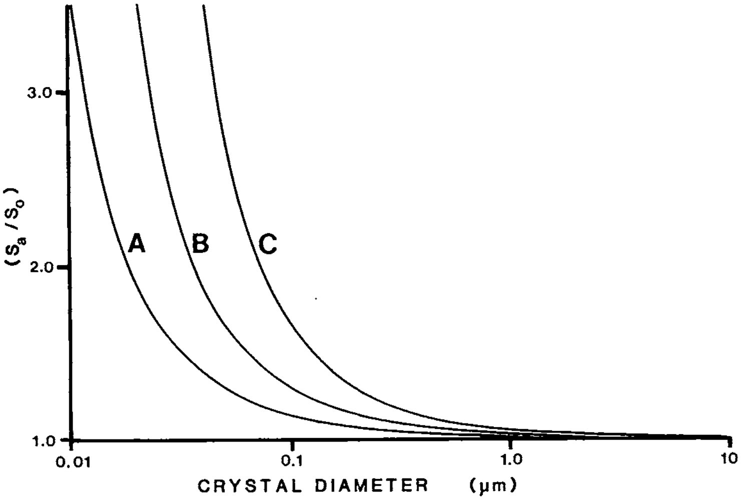

Fig. 5. Graph of index of excess solubility (S_a/S_o) versus quartz crystal diameter. Curve (A) represents smooth spherical crystals and curves (B) and (C) rough crystals with surface areas 2 and 4 times, respectively, that of smooth spherical crystals of the same diameter. Interfacial free energy of 350 mj m^{-2} (Parks, 1984, from data of Stöebber, 1967) and temperature of 25°C were used to calculate quartz solubility.

Direct replacement by quartz would be expected to occur in pore waters undersaturated with respect to opal-CT but supersaturated with respect to quartz (Kastner, Keene, and Gieskes, 1977). As the rate of crystal nucleation is directly proportional to the degree of supersaturation, quartz that precipitated in pore waters with low degrees of supersaturation would have relatively low nucleation rates and coarse crystal sizes. Chert formation by direct quartz replacement is indicated, therefore, by an absence of quartz-replaced opal-CT lepispheres and rim cements and a crystal size of greater than 10 μm. In chert containing both micro- and macrocrystalline quartz, we infer an earlier stage of opal-CT precipitation from pore waters supersaturated with respect to opal-CT. After cessation of opal-CT precipitation, pore water silica concentration would drop to or below opal-CT saturation and precipitation of quartz would begin.

CONCLUSION

Quartz microspheroids and rim cements found in some pre-Cenozoic cherts have sizes, morphologies, and positions similar to opal-CT lepispheres and rim cements found in deep-sea cherts and porcellanites. These quartz-replaced opal-CT lepispheres and cements are evidence that many Mesozoic and Paleozoic cherts formed by the same kind of maturation process that formed Cenozoic deep-sea cherts.

The similarity of crystal sizes, textures, and mineralogies of deep-sea and shelf cherts of various ages is further evidence of maturation.

Some cherts formed by the direct replacement by quartz, presumably in pore waters undersaturated with respect to opal-CT but supersaturated with respect to quartz. These cherts may be distinguished by their coarser crystal size (>10 μm) and absence of quartz-replaced lepispheres and rim cements.

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