Zoned silicate nodules in brucite marbel Santa Olalla, Western Sierra Morena, Spain

Zoned silicate Nodules in Brucite Marble Santa Olalla, Western Sierra Morena, Spain

by *E. Puga** and *J. M. Fontboté***

Abstract

A Block of dolomite (1 km² in outcrop) enclosed in a tonalite pluton of Hercynian age has been converted by contact metamorphism to brucite marble (brucite believed to be pseudomorphous after periclase). Within the block are zoned nodules, mostly 10-30 cm in diameter, in which a diopside core is enclosed in a rhythmically banded shell of calcite and forsterite (partly serpentinized).

The pressure of metamorphism, estimated from the thickness of cover, was about 1 Kb. The maximum temperature, based on Jaeger's cooling models, cannot have exceeded 700°C. A prograde stage in the nodule reaching about 540°C, $X_{CO_2} \simeq 1$, was followed by a retrograde stage of hydration (in forsterite bands of the shell) at 475°C, $X_{H_2O} \simeq 1$. In the dolomite envelope a prograde phase (later than that recorded in the nodule) at maximum temperature above 610°C and high X_{H_2O}, was followed early in the cooling phase by retrograde hydration of periclase to brucite at 610°C, X_{H_2O} remaining high.

A quantitative model has been developed to explain the formation of the nodules by interaction between primary chert and a dolomite envelope of finite extent. Together, these constituted a system closed to everything except to CO_2 and H_2O.

Resumen

Un bloque de dolomía (de 1 Km² de afloramiento) incluido en un plutón tonalítico, de edad hercínica, se ha convertido por metamorfismo de contacto en mármol brucítico (la brucita se considera pseudomorfa de periclasa). Dentro del bloque hay nódulos zonados, en su mayoría de 10 a 30 cm de diámetro, en los que un núcleo de diópsido está rodeado por una capa con bandeado rítmico de calcita y forsterita parcialmente serpentinizada.

La presión del metamorfismo, estimada a partir del espesor de cobertera, debió ser del orden de 1 Kb. La temperatura máxima no debió exceder los 700°C de acuerdo con los modelos de enfriamiento de Jaeger. En los nódulos se registran un estadio de metamorfismo prógrado, que alcanzó aproximadamente 540°C, para $X_{CO_2} \simeq 1$, seguido de otro retrógrado, con hidratación en las bandas forsteríticas, a unos 475°C y $X_{H_2O} \simeq 1$. En la dolomía envolvente una fase prógrada (posterior a la registrada en los nódulos) alcanzó una temperatura superior a 610°C, para X_{H_2O} alta, que fué seguida durante el enfriamiento por hidratación de la periclasa a brucita, a unos 610°C, permaneciendo alta la X_{H_2O}.

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Un modelo cuantitativo ha sido desarrollado para explicar la formación de los nódulos por interacción entre el chert primario y la dolomía envolvente. Ambos constituyen un sistema cerrado a todos los componentes con excepción del CO_2 y del H_2O.

GEOLOGICAL SETTING

The Santa Olalla batholith is one of several granitoid plutons located along the core of the Zafra anticlinorium in the western Sierra Morena. The batholith has an outcrop area of 50 km^2 and was emplaced in, essentially unmetamorphosed, uppermost Precambrian and Cambrian formations during the Hercynian orogeny (Fig. 1). Stratigraphic and structural evidence indicates that the present erosion level lay at a depth of not more than 4 km at the time of emplacement. Cambrian (Georgian) rocks outcrop widely near the pluton, and are conspicuously affected by contact metamorphism adjacent to it. Between the villages of Real de la Jara and Santa Olalla, there is a large inclusion (0,5 km x 2 km) of brucite marble (derived from Cambrian dolomite) within the batholith
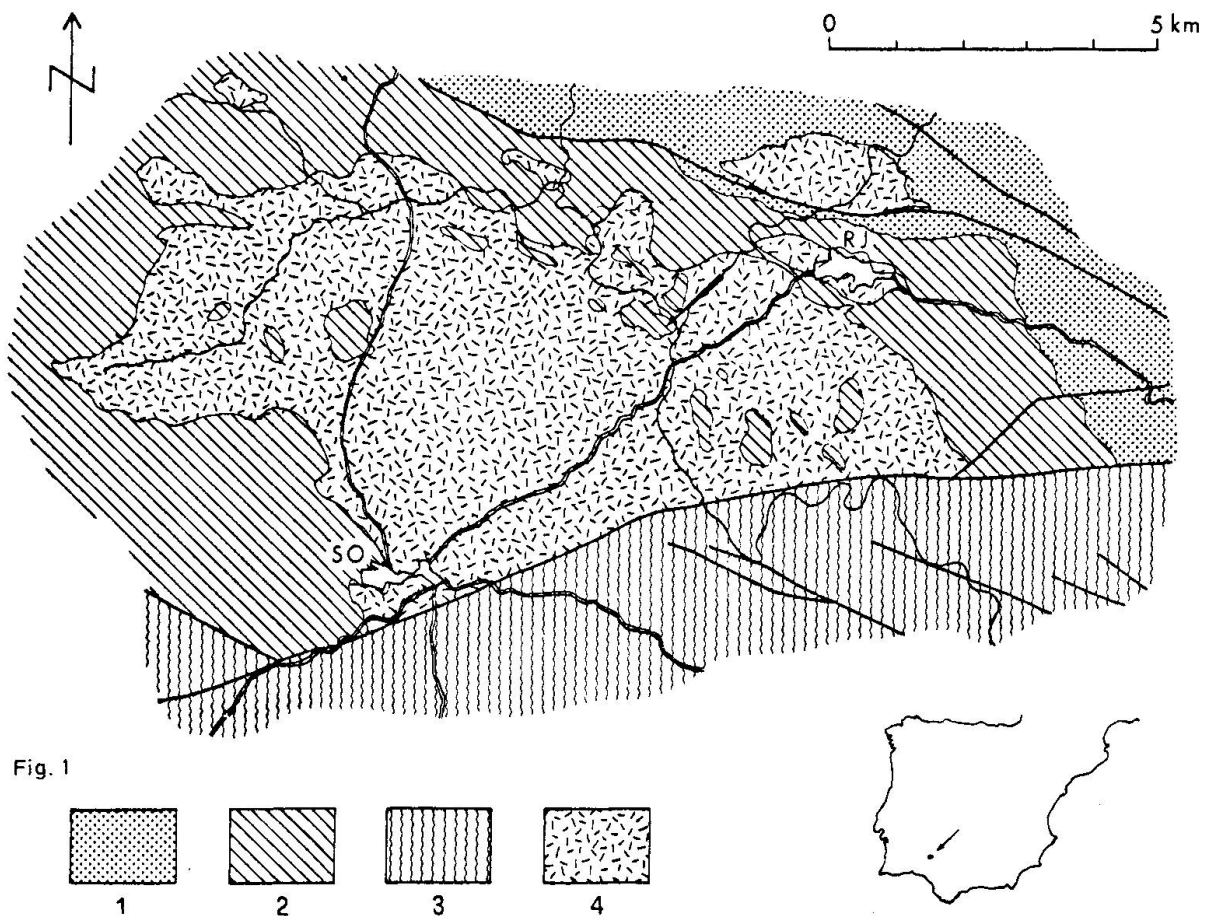

Fig. 1

Fig. 1   Location sketch map: 1-Upper Proterozoic, 2-Cambrian, 3-Devonian, 4-granitoid plutons. (Date from geological maps: INSTITUTO GEOLÓGICO Y MINERO DE ESPAÑA, 1970 and 1974; VELASCO, 1977). SO, Santa Olalla; R.J. Real de la Jara. Between these two villages, an arrow indicates the surveyed inclusion.

and close to its southern border. This marble contains numerous ellipsoidal silicate nodules, commonly 20-30 cm, and rarely as much as 1 m, in diameter. These nodules are the main subject of this paper.

We have seen similar nodules in other inclusions, both in this and in other batholiths, such as Burgillos del Cerro, roughly 60 km NNW of Santa Olalla. These may well have been formed in the same way as the ones discussed here.

PETROLOGY

The host rock of the silicate nodules is a brucite marble with roughly equal amounts of brucite and equant twinned calcite grains (up to 1 mm in diameter). The brucite forms fibrous aggregates, 1-3 mm across, with regular outlines suggesting that they are pseudomorphous after periclase, no remnants of which survive.

Scattered within the marble are nodules, macroscopically zoned, with cores of dark diopside rimmed by concentric alternating shells of white calcite, and dark partially serpentinized forsterite containing minor amounts of spinel (photos 1, 2).

The mineralogical and textural characteristics of these rocks, indicate that the following parageneses formed during prograde metamorphism:

10 cm
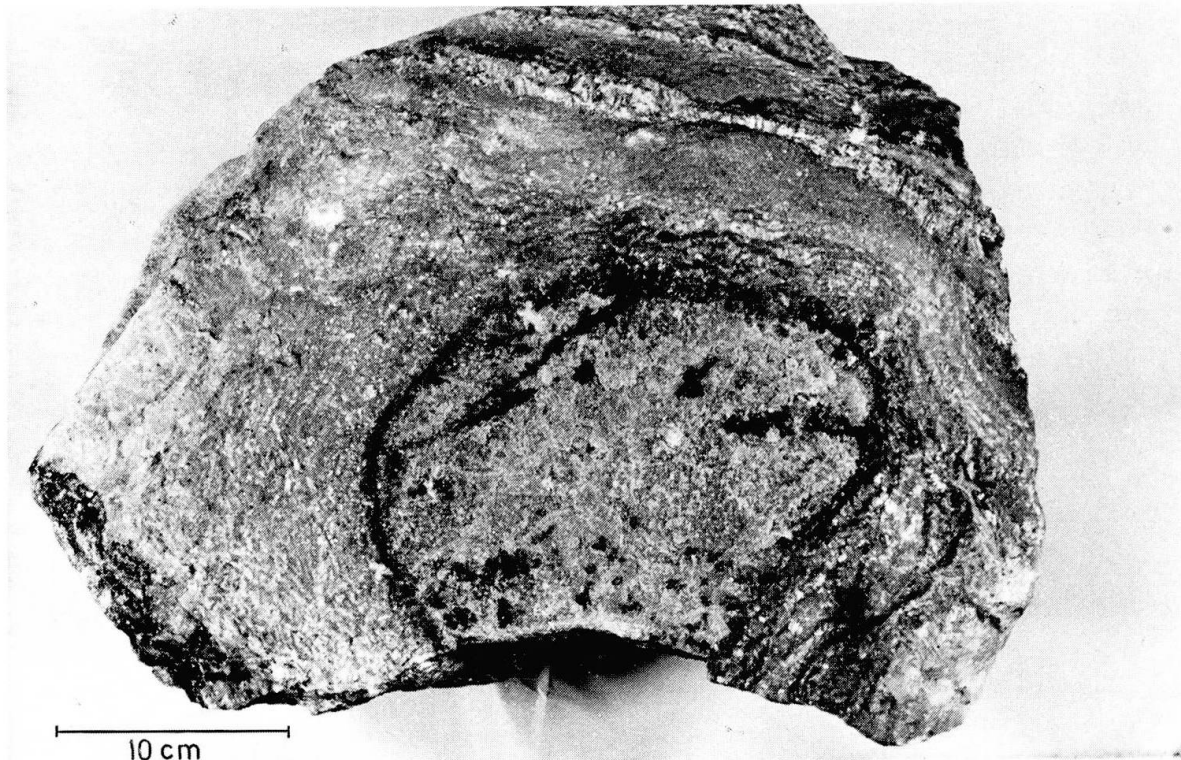
Photo 1   Spheroidal Ca-Mg silicate nodule from la Garranchosa Quarry (Santa Olalla). Relatively dark core of diopsidite surrounded by rhythmically striped shell composed of calcite and (partly serpentinized) forsterite.
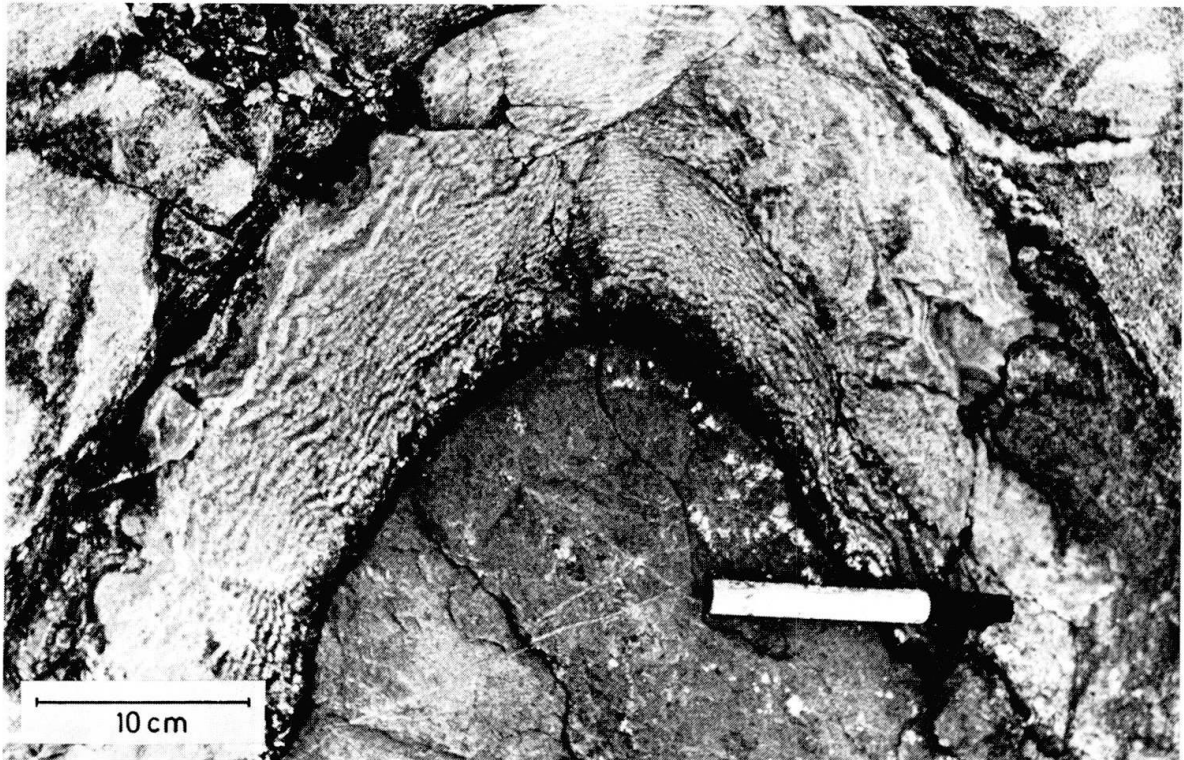
Photo 2 More detailed view of another nodule. The dark strip a few millimetre thick shell boundary is composed of serpentine, iron oxide, and phlogopite. These minerals formed in several stages of retrograde metamorphism. The dark strip in the nodule surface is largely composed of serpentine.

calcite + periclase ± forsterite ± spinel, in what is now brucite marble;
calcite + forsterite + spinel, in the shells of the nodules;
diopside, in the core of the nodules.

As we show below these parageneses formed at different temperatures.

During retrograde metamorphism, the brucite replaced periclase, and forsterite was partly altered to serpentine and minor amounts of ore, magnesite and brucite. Phlogopite appears locally at the boundary between the core and the shell.

Most of these phases can be related by equilibria in the $CaO-MgO-SiO_2-H_2O-CO_2$ system; spinel and phlogopite must be excluded because they require Al, Fe and K. Available experimental and thermodynamic data for this system, critically summarized by HELGESON et al. (1978), are represented in fig. 2.

Skarn parageneses are locally present filling fissures in marble as well as in nodules. We consider these skarn minerals to result from metasomatism linked to the circulation of fluids bearing silica, alumina, potassium and volatiles. These processes postdate the retrograde metamorphism, and will not be considered here.

We think that the silicate nodules are derived from chert, which occurs as scattered bodies of similar size in unmetamorphosed dolomite outside the aureole. This has been proposed for similar nodules elsewhere by, for example,

JOESTEN (1974) in the Christmas Mountains (Big Bend region, Texas), and by MOORE and KERRICK (1976) in the Alta aureole (Utah). On the other hand, VELASCO (1977) suggested that the Santa Olalla nodules are derived from boudins of pelitic layers. Our field observations, the geometry of the nodules, and textural and mineralogical considerations, are compatible only with the hypothesis that the nodules formed from chert.

PETROGENESIS

*Prograde stage* - The lowest temperature mineral belonging to this phase is diopside, which forms the core of the silicate nodules. It shows no textural indication of having replaced another older mineral. It probably formed by two-way diffusion between the chert and the surrounding dolomite. The implied reaction would be:

$$1 \text{ dolomite} + 2 \text{ quartz} = 1 \text{ diopside} + 2 \text{ CO}_2$$
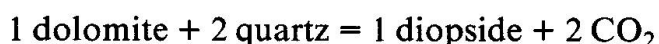
Thermodynamic conditions of this reaction are approximately represented by the curve 1, fig. 2. For $P_1 = P_f = 1$ kb, and $X_{CO_2} \geqq 0.95$, this reaction starts at $442° \pm 15°C$ (SLAUGHTER et al., 1975), $470° \pm 30°C$ (HELGESON et al., 1978), $480°C$ (SKIPPEN, 1974) or $494°C$ (METZ and TROMMSDORFF, 1968). Once that one of the reactants has been used up, silica in this case, as no quartz remains in the nodules, the heat supplied to the system causes the temperature to rise. Eventually, dolomite starts to react with diopside:

$$1 \text{ diopside} + 3 \text{ dolomite} = 2 \text{ forsterite} + 4 \text{ calcite} + 2 \text{ CO}_2$$
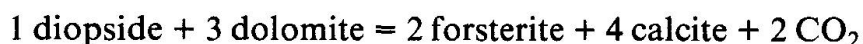
This leads to the growth of concentric shells around the diopside cores. Thermodynamic conditions of this reaction are approximately those of curve 2, fig. 2. At a lithostatic load of 1 Kb, and $X_{CO_2} = 0.95$, this reaction starts at $539°C$ (METZ and TROMMSDORFF, 1968), $540°C$ (SKIPPEN, 1971) or $548°C \pm 10°C$ (METZ, 1977).

If the temperature rises further, periclase forms within the marble envelope by the reaction:

$$1 \text{ dolomite} = 1 \text{ calcite} + 1 \text{ periclase} + 1 \text{ CO}_2$$
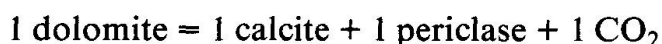
This reaction can occur between $610°C$ and $820°C$, depending on X_{CO_2}, for $P_1 = 1$ kb (METZ, 1968) as shown in curve 3, fig. 2.

In the Santa Olalla marbles, this reaction probably took place between $610°C$ and $700°C$, if the fluid pressure remained at 1 kb. It requires $X_{H_2O} \geqq 0.8$

in the fluid phase. The lower temperature limit is constrained by the lack of primary brucite (its typical tabular habit has not been recorded anywhere) which could eventually develop according to the reaction:

$$1 \text{ dolomite} + 1 \text{ } H_2O = 1 \text{ brucite} + 1 \text{ calcite} + 1 \text{ } CO_2$$
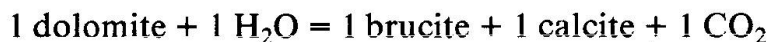
The upper limit is given by the composition of the magma surrounding the carbonate inclusion. The application of the calcite-dolomite geothermometer does not permit estimate of the temperature during the metamorphic climax. $Mg \text{ } CO_3$ contents from 15 samples of calcite have been compared to the calcite-dolomite solvus (GOLDSCHMIDT and NEWTON, 1969) using the equation for the solvus devised by SHEPPARD and SCHWARCZ (1970). This indicates a temperature below 485 °C, suggesting that a reequilibration occurred. This conclusion is supported by exsolution textures in some of the samples.

Retrograde stage – As the temperature dropped after the metamorphic climax hydration of periclase became possible; and no periclase now remains in the marble inclusion. Forsterite is also extensively serpentinized. These reactions require that $X_{H_2O} > 0.90$.

For this fluid composition, at $P_1 = P_f = 1$ kb, periclase is transformed into brucite between 620 °C and 610 °C (BARNES and ERNST, 1963; TROMMSDORFF and SCHWANDER, 1969; HEMMLEY et al., 1977; and HELGESON et al., 1978). (See curve 4, fig. 2).

With a further fall in temperature, serpentinization of olivine would start according to the reaction:

$$2 \text{ forsterite} + 2 \text{ } H_2O + 1 \text{ } CO_2 = 1 \text{ serpentine} + 1 \text{ magnesite}$$
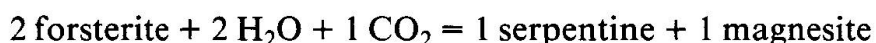
below 475 °C for $X_{H_2O} > 0.92$ and $P_1 = P_f = 1$ kb (JOHANNES, 1969),(see curve 5, fig. 2). Magnesite occurs mainly around the pre-existing serpentinized olivine crystals as well as in veinlets with serpentine, defining the mesh texture.

Serpentinization would progress according to the reaction:

$$2 \text{ forsterite} + 3 \text{ } H_2O = 1 \text{ serpentine} + 1 \text{ brucite}$$
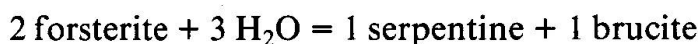
at about 360 °C, for $X_{H_2O} > 0.99$ and $P_1 = P_f = 1$ kb (JOHANNES, 1968),(see curve 6, fig. 2).

Serpentinization is accompanied by liberation of iron oxide that remains closely mixed with serpentine building the mesh structure. The amount of iron oxide is more conspicuous along both inner and outer boundaries of the nodule shell. It suggests that the olivine was richer in iron near these boundaries. Iron enrichment along the inner shell boundary could be explained by diffusion of iron out of the original chert nodules, during their transformation to diopside,
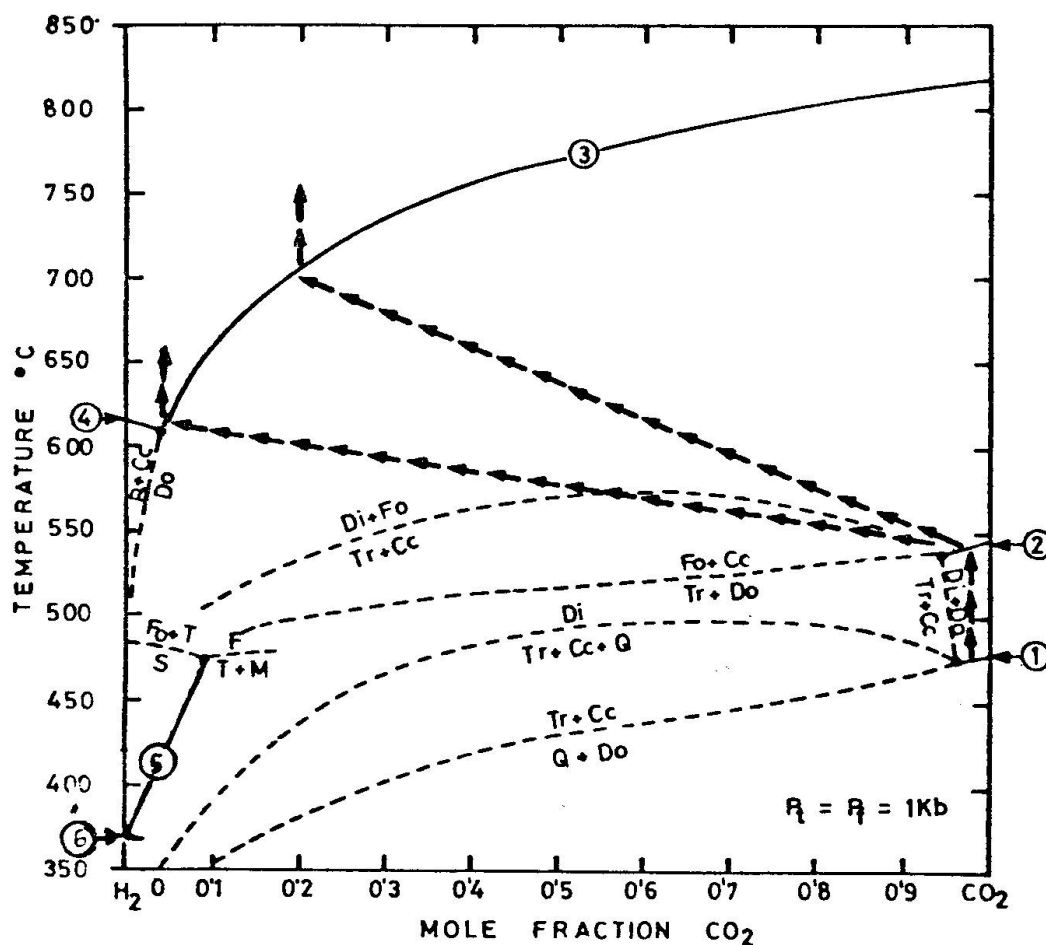
Fig. 2 T-X_{CO_2} diagram at P_f = 1 kb illustrating the prograde and the retrograde metamorphism of brucite marble and included nodules from Santa Olalla.

① 1 Do + 2 Q = 1 Di + 2 CO_2 (METZ and TROMMSDORFF, 1968; SKIPPEN, 1974; SLAUGHTER et al., 1975; HELGESON et al., 1978).

② 1 Di + 3 Do = 2 Fo + 4 Cc + 2 CO_2 (METZ and TROMMSDORFF, 1968; SKIPPEN, 1971; METZ, 1977).

③ Do = Cc + P + CO_2 (METZ, 1968).

④ P + H_2O = B (BARNES and ERNST, 1963; TROMMSDORFF and SCHWANDER, 1969; HEMLEY et al., 1977; HELGESON et al., 1978).

⑤ 2 Fo + 2 H_2O + 1 CO_2 = 1 S + 1 M (JOHANNES, 1969).

⑥ 2 Fo + 3 H_2O = 1 S + 1 B (JOHANNES, 1968).

Abbreviations for minerals in figure: B = brucite; Cc = calcite; Di = diopside; Do = dolomite; Fo = forsterite; M = magnesite; P = periclase; Q = quartz; S = serpentine; T = talc; Tr = tremolite.

which can only incorporates a small amount of iron. Chert occurring in unmetamorphosed dolomite around the thermal aureole contains abundant small hematite crystals.

In the bigger nodules there is a strip , one to several millimeters wide, of interleaved phologopite poikiloblasts along the boundary between core and shell. The texture suggests that it formed from olivine and diopside at high X_{H_2O}, after serpentinization of olivine had started. Al_2O_3 and K_2O may have been derived from clay impurities of the chert that were expelled from the nodule during diopside formation.

EVOLUTION OF METAMORPHIC CONDITIONS AND FLUID PHASE

Emplacement of tonalite magma (initially undersaturated in water) at a liquidus temperature of about 800-850 °C, possibly 900 °C, and at shallow depth places limits on the maximum temperatures attained within a dolomite block 1 km wide. Applying the cooling models of JAEGER (1957, 1968) and taking into account a possible supply of heat by convection towards the marble inclusion, temperatures can scarcely have exceeded 700 °C.

Regional pressure may have been as high as 1 kb, but could have been perhaps as low as 0,5 kb. Fluid pressure is assumed to have been rougly equal to the load pressure.

The gas phase generated in the nodule from the early stages of metamorphism must have been almost pure CO_2. Otherwise (even at X_{H_2O} as low as 0,05) tremolite would have formed and remained stable up to 475 °C or more (see fig. 2) and there is no indication of this. Under such conditions the system must have been open to CO_2. Otherwise decarbonation reactions could not have proceeded. In fact, assuming T = 470 °C, the CO_2 yielded by the formation of only a mole of diopside from dolomite, would generate a pressure of about 15 kb if gas was not removed. Extremely high P_{CO_2} would also result from forsterite formation. Although fluid must have left the system, the fluid pressure probably remained close to load pressure. Mineralogical evidence is lacking, but it is possible that P_f dropped below P_1 while the temperature was between that for curves 2 and 3, because fluid would continue to leave the system, while no more CO_2 was being evolved.

After forsterite formation was complete, P_f fell with respect to P_1, or the fluid was diluted with water to $X_{H_2O} > 0.8$, or both, so that periclase could form at temperatures below 700 °C according to curve 3, fig. 2.

The retrograde stage, both in the nodule and its envelope, involved hydration of periclase and forsterite. This implies X_{H_2O} close to 1, the water being of external (plutonic) origin. Minor carbonation occurred in the nodule shell.

CONCLUDING REMARKS AND PROPOSED MODEL

Zoned silicate nodules, developed from primary chert carbonate rocks, are well known from several contact metamorphism aureoles (TILLEY, 1951; KENNEDY, 1959; JOESTEN, 1974; MOORE and KERRICK, 1976; SUZUKI, 1977).

The formation of these nodules requires transfer of matter between the primary chert and the enclosing carbonate rock, along several centimetres. This transfer is effected by local diffusion metasomatism only (KORZHINSKII, 1968) as no material supply from the igneous body can be detected. We are therefore dealing simply with contact metamorphism, although the mineral assemblages are similar to those of some skarns. In this way material exchange between siliceous bodies and enclosing carbonate rock gives rise to concentric zones of different composition. These zones constitute «local equilibrium» systems (THOMPSON, 1959).

Differentiation of zones within the nodules results from a radial gradient of composition. The $SiO_2/(Ca-Mg)O$ ratio decreases from the core to the margin of the nodule. The gradient of chemical potentials are the driving forces for diffusion and redistribution of elements, and will depend on the original contrast in composition between the nodule and its envelope (VIDALE, 1969). The rate of diffusion processes is also dependant on the availability of interstitial fluid. During the quoted reactions giving rise to diopside and forsterite + calcite, large amounts of CO_2 were evolved and were available to assist diffusion.

In fig. 3 growth sequences of Ca-Mg silicate nodules from Santa Olalla are shown. An attempt to quantify their development process is presented for a nodule of the size of that shown in photo 1, as follows: For an approximately
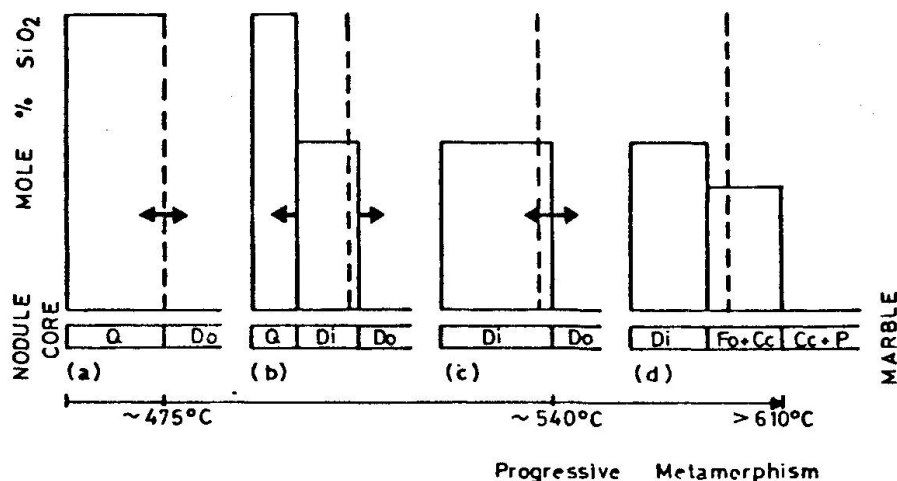

Fig. 3 Growth sequences of reaction zones building up the Ca-Mg silicate nodules. Arrows indicate the direction of movement of the boundary of each zone. Vertical dash line marks initial location of the quartz dolomite interface, which lies in the zoned shell after the process is complete. Approximate T starting for the reactions leading to each stage are shown below.

spherical nodule, the volume of primary chert can be calculated from the thickness of the core and shell. The growth history of the diopside core can then be calculated from volumetric reactions leading to the differentiation of the shell. In this way we calculate: 1°.- The present volumes of the diopside core plus the concentrically banded shell (stage d, fig. 3). 2°.- The volume of diopside consumed by the reaction: 1 Di + 3 Do = 2 Fo + 4 Cc + 2 CO$_2$, that gave rise to the banded shell; the sum of 1° and 2° then gives: 3°.- The total volume of diopside before formation of the shell (stage c, fig. 3). 4°.- The volume of chert consumed by the reaction: 1 Do + 2 Q = 1 Di + 2 CO$_2$ giving rise to volume 3° of diopside. The nodule in photo 1 has a diopside core 10 cm in radius, and a 10 cm thick shell. This gives a radius of 14,4 cm for the diopside nodule at the intermediate stage c; the radius of the primary chert nodule (stage a, fig. 3) was 12,7 cm. The thickness changes of the different reaction zones during the metamorphic process are represented in fig. 3.

The small volume increase due to serpentinization of forsterite has not been taken into account. Molar volumes used in these calculations have been taken from ROBIE et al. (1967). These data are for surface conditions; dilation processes occurring under metamorphic conditions are not taken into account, and real volumes at stage c were certainly greater than those estimated. On the other hand, minor-amounts of other minerals present in the nodules have not been considered. In spite of these limitations, we think the estimates are accurate enough to give a useful picture of the process.

An interesting question remains. In the proposed model diopside continued to form from quartz and dolomite until the quartz was completely consumed. No remnant of chert has been detected within the nodules. By contrast, the later reaction between diopside and dolomite to give forsterite + calcite in the shell did not completely consume the diopside. Completion of the first reaction is understandable as the large amount of CO$_2$ yielded would have greatly assisted diffusion of the solid components. A comparable availability of fluids also existed during the second reaction, however, it did not go to completion. We suggest that this can be explained by the fact that the chemical potential gradient between diopside and dolomite is lower than that between quartz and dolomite. The rate of diffusion would therefore have been slower in the second case. Diffusion would have been slowed still further, as the mineralogically stable forsterite + calcite shell grew thicker, separating the diopside and dolomite (THOMPSON, 1959). As the diffusion velocity of one or some of the reactant components decreases, a threshold will be reached, at which the reaction will stop. It seems likely that this threshold was reached in the case of the diopside + dolomite reaction giving rise to the banded shell of the nodules.

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