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MICROTEXTURES IN RECENT AND FOSSIL CALICHE

BY

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ABSTRACT

Microtextures of calcite in recent caliche are similar to those of authigenic calcite in Upper Carboniferous, Permian and Lower Triassic continental sandstones and mudstones in the South-Central Pyrenees, Spain. Except for one profile in the Permian, no complete caliche profiles containing calcrete occur in the ancient deposits. It is suggested that the fossil authigenic calcite crystallized in early stages of diagenesis under climatic conditions favourable to the development of caliche.

INTRODUCTION

The presence of fossil caliche in continental deposits provides important paleoclimatological information, since caliche mainly forms in regions with high temperatures and moderate to low, seasonal precipitation. A complete caliche profile showing well-differentiated horizons commonly consists of an uppermost zone of lenses of indurated fine-grained limestone (caprock, calcrete, 'croûte zonaire' or 'Kruste'), variously underlaid with large, bolster-like masses, small nodules, irregular encrustation zones and even, semi-lithified zones in which the calcite acts as a cement (Bretz & Horberg, 1949; Durand, 1953, 1954; Brown, 1956; Gaucher, 1957; Rutte, 1958; Blank & Tynes, 1965; Mosely, 1965).

Where such complete profiles occur in ancient deposits, there is little difficulty in recognizing their true nature. Matters are less certain, however, where the ancient caliche consists only of zones containing replacement and cementing calcite, or levels of nodules, or levels containing encrusting calcite.

Such levels may have formed where, under lithologic and climatic conditions conducive to the formation of caliche, a high rate of clastic deposition prevented the development of complete and thick profiles. As the distinctive calcrete is the last to form (Gaucher, 1957), it may be expected to be infrequent or even altogether absent in rapidly accumulated clastic deposits.

The importance of the rate of deposition for the thickness of caliche has recently been demonstrated by Ruhe (1967) for the caliche in southern New Mexico, U.S.A. He records that the thickness of caliche increases significantly with the age of the geomorphic surface on which the caliche occurs.

Besides the rate of deposition, many other factors, such as the availability of calcium in the substratum, the amount of aerially supplied carbonate-carrying dust, the relative elevation of the site of formation, ground water movements and, of course, the climate proper, may affect the characteristics and thickness of caliche profiles (Brown, 1956; Rutte, 1958; Ruhe, 1967).

Since so many factors exist in the formation of caliche, the term caliche in this paper is taken to apply indiscriminately to calcite precipitated from descending solutions in soil profiles, from run-off water, and from ascending groundwaters.

It is the aim of the present paper to show that the assumedly characteristic association of calcite microtextures in recent caliche may assist in recognizing fossil, incompletely developed caliche.

RECENT CALICHE, MODE OF OCCURRENCE

Recent caliche deposits were studied along the mediterranean coast of Spain, in the area situated between Tarragona and the Ebro delta (exposures 1—5, Fig. 1). The area forms part of a larger region of

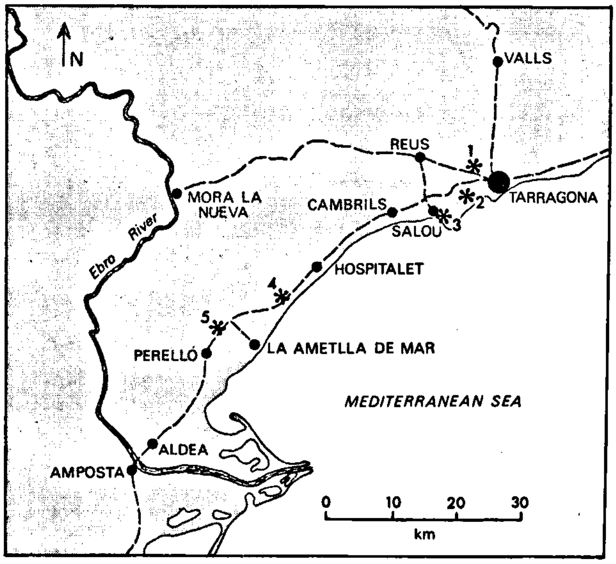

Fig. 1. Topographic map showing localities in which recent caliche deposits were studied. Localities are numbered 1 to 5

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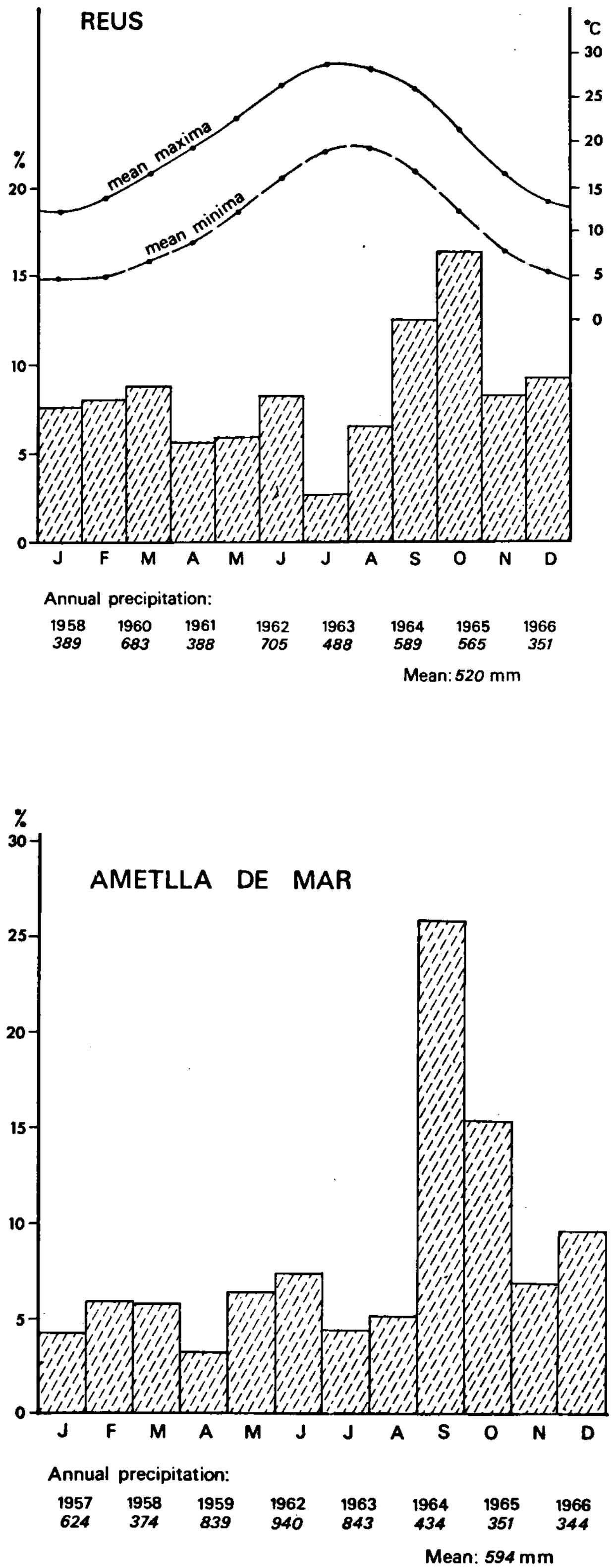


Fig. 2. Precipitation diagrams for Reus and Ametlla de Mar, and temperature curves for Reus. Data supplied by the Comisaria de Aguas del Pirineo Oriental (Ministerio de Obras Publicas, Dirección General de Obras Hydraulicas), Barcelona, Spain.

caliche deposits, which has previously been investigated by Rutte (1958).

The exposures visited, with the exception of nr. 3 (Fig. 1), show differentiated horizons within the caliche. Lenses, usually less than 30 cm thick, of light brown, pinkish-grey, or occasionally almost white, dense aphanitic limestone occurring 10—100 cm below the surface overlie zones of nodules, masses of encrusting calcite, irregular calcite veins running in apparent random directions and zones of partial, but evenly distributed cementation with calcite. The caliche profiles reach thicknesses of up to 10 m. Exposure 3 shows thick masses of encrusting calcite in a small sea cliff.

The differentiated caliche profiles which have been studied closely follow the present-day relief. The ties of the caliche with the relief evidence that the caliche cannot be very old. On the other hand, it can be observed in many places that the caliche is now being eroded, for instance at the sea cliff of exposure 3 and wherever small, narrow gorges (dry in summer) traverse the caliche.

Some data on the climate of the last 10-year period are given in Fig. 2. The mean annual precipitation for this period (520 mm for Reus, 594 mm for Ametlla de Mar) is somewhat higher than the 400—500 mm value shown on pluviometric maps (Masachs, 1958; Lautensach, 1964). However, there is a considerable variability in annual precipitation. The maximum temperature recorded during the period was 36.0° C (August, 1964, Reus); the minimum temperature was — 0.4° C (Febr., 1963, Reus).

According to Rutte (1958) the optimal conditions for the generation of calcrete at the Spanish east coast occur at precipitation values between 150 and 250 mm. In his opinion, mainly the lower levels in caliche profiles, the calcite nodules, develop at values above 500 mm.

It is very likely that the investigated caliche profiles resulted from the accumulated effects of various climatic periods. On the basis of the current data it is impossible to decide in which way present-day conditions affect the profiles: whether they remain constant, or whether addition or substraction takes place. This could be solved for example, by using direct methods such as radiocarbon dating.

The caliche developed on a variety of substrata, which all are rich in carbonates. At exposure 1 it occurs on shallow-marine, Quaternary deposits; in exposure 2 it occurs on Quaternary eolian sands (former coastal dunes) and at exposures 3, 4 and 5 it occurs on Quaternary alluvial fan deposits extending seaward from the Catalan Coastal Mountain Range (Solé Sabaris *et al.*, 1966). The caliche in the former coastal dunes may have had a complex history; as suggested by Schwegler (1948) for similar cemented dunes in Egypt, cementation may have started early by means of $CaCO_3$ mobilized by dew and sea-spray solutions.

FOSSIL CALICHE, MODE OF OCCURRENCE

Calcite considered to be of a caliche origin has been studied in three, stratigraphically successive, continental formations in the South-Central Pyrenees, Spain (Fig. 3).
The sedimentology and diagenesis of these deposits, called the Malpas Formation (Stephanian), the Peranera Formation (Permian red beds) and the Bunter Formation (Lower Triassic red beds), are treated in detail elsewhere (Nagtegaal, 1969).

In the Malpas Formation one member, near the top of the formation, contains calcite attributable to caliche. The remainder consists largely of coal and

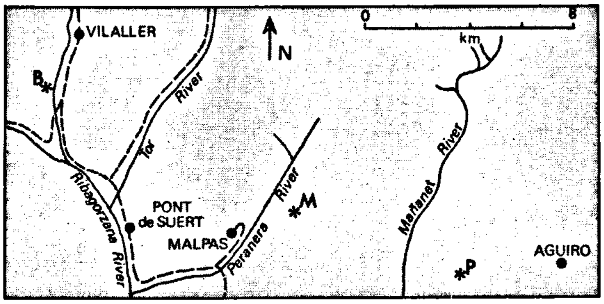


Fig. 3. Topographic map of part of the South-Central Pyrenees showing localities in which ancient caliche deposits were studied. Localities are indicated by B = Bunter Formation (Lower Triassic), M = Malpas Formation (Stephanian), and P = Peranera Formation (Permian).

ironstone-bearing mudstones and shales, dark-grey to black in colour, and of sandstone sheets commonly showing point-bar sequences. These deposits, with a thickness of approx. 400 m, are interpreted as fluvial systems with meandering streams bordered by flooded back-swamp areas. Authigenic parageneses include kaolinite, quartz, siderite, marcasite and pyrite; calcite is almost absent, both as clasts and as an authigenic product. Intercalated near the top of the sequence, however, there occurs a 75 m-thick unit of conglomerates and coarse sandstones, probably deposited by braided streams. The difference in sedimentary environment has its counterpart in a difference in authigenesis: in this level the cement is largely calcite, considered to be of a caliche origin. No complete caliche profile with differentiated horizons, however, is present in the deposits.

The Peranera Formation is a sequence more than 700 m thick, largely consisting of an alternation of greyish-red mudstones and conglomeratic sandstone channel-fills. The environment of deposition is thought to correspond to steppes bordering alluvial fans. Texturally and mineralogically the deposits are immature, the sandstones consisting of moderately sorted quartzite grains, phyllite debris, limestone fragments, and angular to subangular quartz. The main cement in the formation is calcite. Levels of calcite nodules, some containing vertically oriented nodules, are very common in the mudstones. The maximum thickness of the nodules-carrying mudstone layers is approx. 30 m. One level was found which may correspond to a complete caliche profile; it shows lenses 10—20 cm thick, of dense, almost white limestone (calcrete) underlaid with calcite nodules. All calcite nodules, and much of the replacement calcite and calcite cement, are considered to be of a caliche origin, as will be shown when the micro-textures are considered.

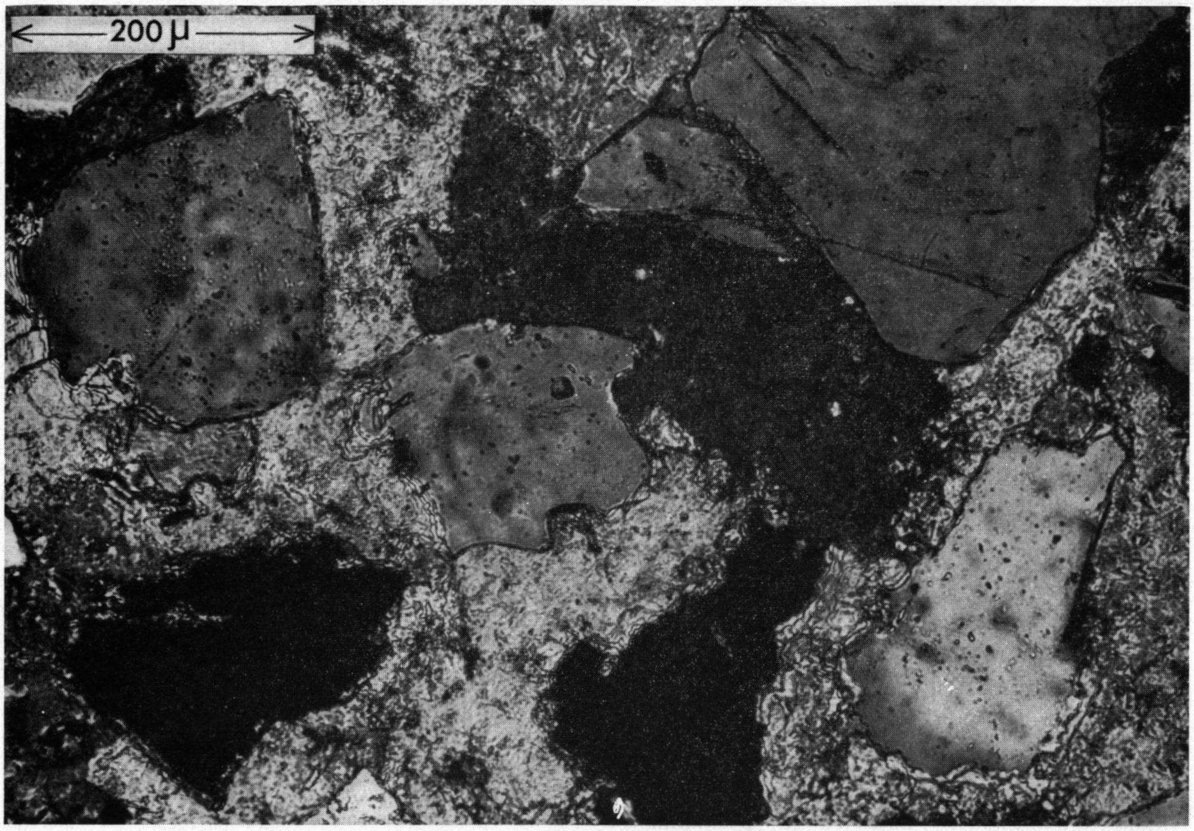
The Bunter Formation consists of greyish-red mudstones intercalated with many sandstone channel-fills, and some conglomerate sheets in a sequence varying in thickness between 50 and 300 m.
Mineralogically the sandstones are mature; they consist of detritogeneous quartz, quartzite, chert, muscovite and rare phyllite and altered feldspar grains, while there are no limestone fragments. The cement is largely quartz. The deposits, which are of a fluvial facies, accumulated on wide pediment surfaces in a tropical, savannah-like environment. Authigenic calcite occurs in some mudstone levels in the form of numerous, densely packed spherulites, most of which were dolomitized at a later stage. The spherulites are thought to be of a caliche origin.

MICROTEXTURES IN RECENT AND FOSSIL CALICHE

A wide variety of microtextures, both with respect to crystal habit and crystal size, occurs in caliche. Although the recent and ancient caliche probably both are polygenetic (Ruhe, 1967), similar calcite microtextures can be distinguished. Only those which in the present author's view are the most common and most significant, have been singled out for discussion.

Replacement and cementing calcite

These types of calcite are by far the most abundant volumetrically, both in the ancient rocks and in the recent caliche. In fully lithified rock the two types of calcite, i.e. the replacement calcite and the calcite which indurates the rock, cannot possibly be distinguished since the calcite has both functions simultaneously. As the two types commonly occur together, they are treated under one heading.
The replacement and cementing calcite normally exhibits crystalline equigranular (mosaic) textures. There is a wide range in coarseness. In mudstones the calcite is of the finest grain, although a considerable range in grain-size exists (of less than 4 micron up to 100 micron, with a maximum in the 5—30 micron range); in sandstones the textures are coarser and show a still wider range (of less than 4 micron up to 2 mm, with maxima varying from sample to sample). Apparently there is a relation between the grain-size of the rock, and the coarseness of the replacement and cementing calcite formed in it.
There is also a complete range in degrees of replacement, both in ancient and recent items. In fully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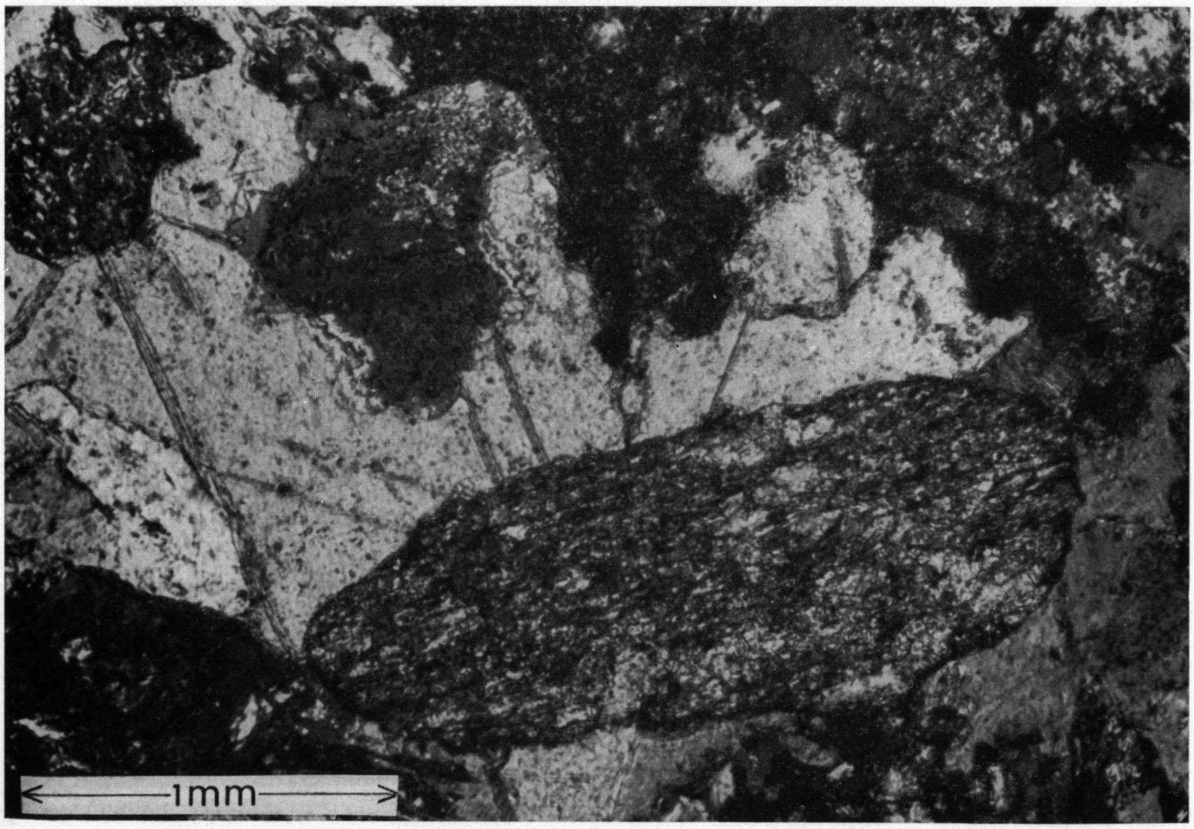
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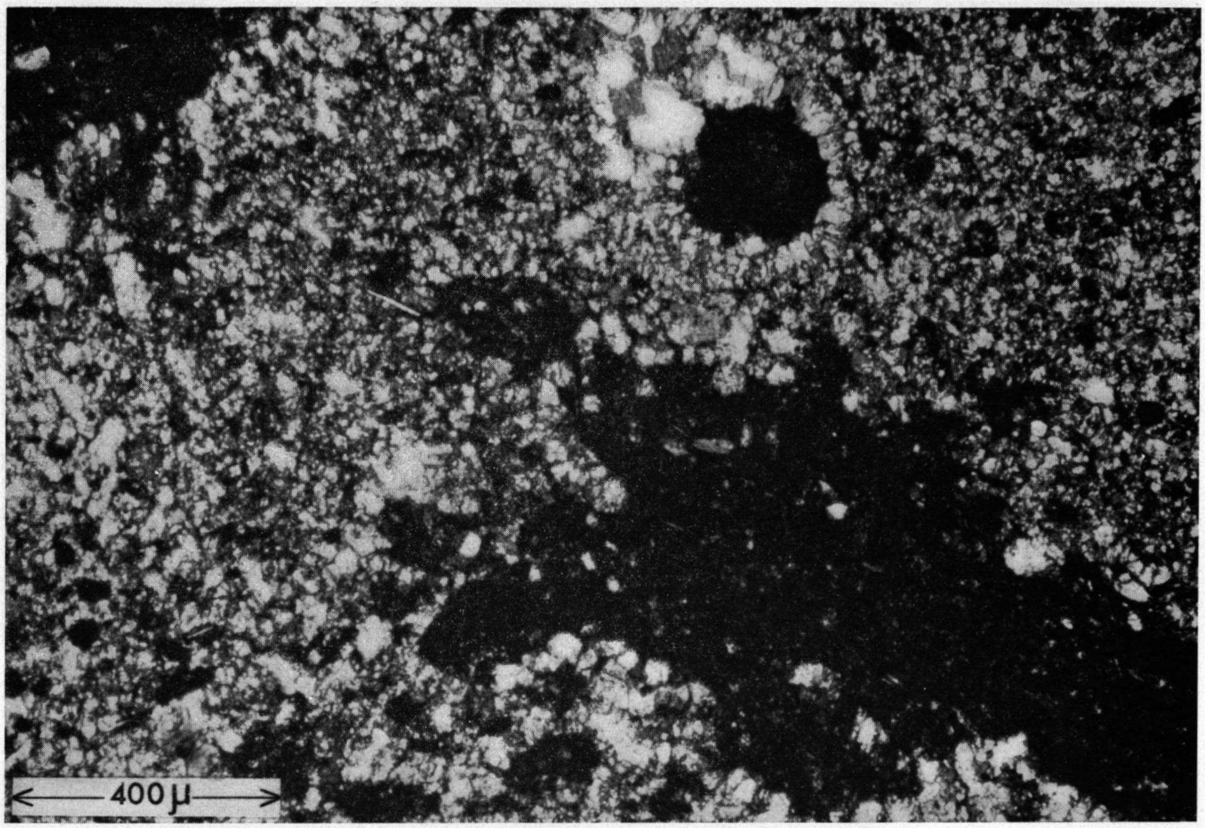


Fig. 6. Fossil caliche; replacement and cementing calcite in Permian ferritic mudstone (Peranera Formation). In this sample, approx. 80 % (volume) of the mudstone has been replaced by fine-grained calcite. Only irregular relics of the mudstone (darkened by ferric oxides) are preserved. A spherical, isolated relic of mudstone is enveloped by a calcite crust. Sample 1296, locality P in Fig. 3. Plain light.

indurated samples the degree of replacement by calcite may vary from almost nil (calcite mainly occurring as an inert cement), *via* stages where detritogeneous, strongly corroded silicate grains "float" in aggregates of calcite, to stages where almost nothing is left of the original constituents of the sandstone. The intermediate stage of silicate grains "floating" in calcite, which Brown (1956) considered to be characteristic of caliche, is illustrated in Figs. 4 and 5.

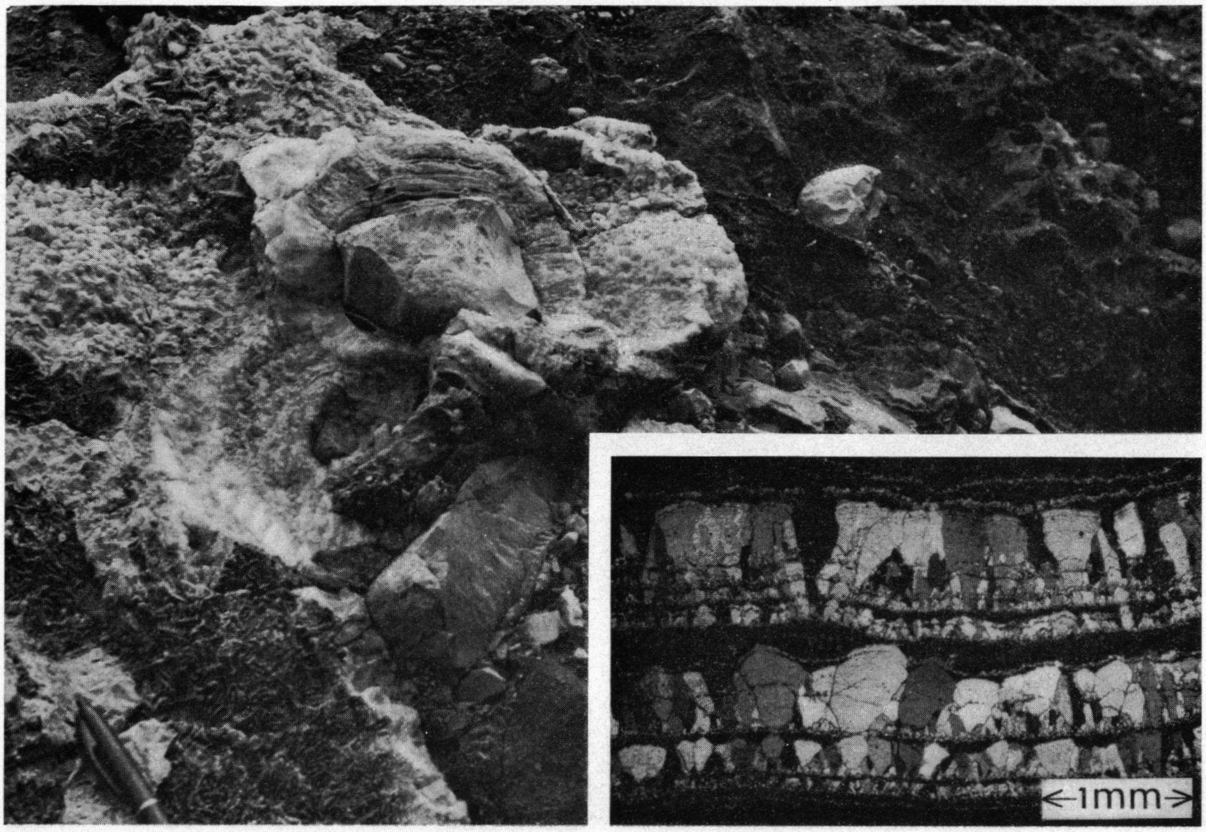
Fig. 4. Recent caliche; replacement and cementing calcite. The sample is of a caliche which formed in Quaternary, marine sands. The grains of quartz are all strongly corroded and replaced by calcite, which crystallized in crystals approximately of the size of the sand grains. Traces of craggy replacement fronts are preserved in the calcite crystals. The two lower quartz grains are fully extinguished; large crystal of calcite is almost extinguished. Secondary calcite occupies 56 % (volume) of the rock. Sample 2510, locality 1 in Fig. 1. Nicols crossed.

Fig. 5. Fossil caliche; replacement and cementing calcite in Permian phyllitic sandstone (Peranera Formation). Calcite, in crystals approximately of the size of sand grains, extensively replaces phyllite and quartz. The calcite crystal at the extreme right is almost extinguished. The phyllite grains are rich in ferric oxides, which probably formed before the grains were included in the deposit. Secondary calcite occupies 52 % (volume) of the rock. Sample 1302, locality P in Fig. 3. Nicols crossed.

A common property of the calcite crystals forming part of the mosaic pattern in these cases is that they join along irregular, craggy boundaries instead of essentially straight planes, as is normal in void and pore-filling calcite (Fig. 4). The feature probably results from the replacement history of the calcite crystals. Also, in recently formed calcite, craggy replacement fronts are to a large extent preserved *within* the crystals (Fig. 4). This is also a feature of older crystals, though it appears to have been preserved less frequently (Figs. 5 and 8). In nodules in mudstones bulk replacement may proceed along highly irregular fronts, which leads to a rock composed of shard-like relics of mudstone suspended in fine-grained crystalline granular calcite (Fig. 6). Similar replacement relations were noted in the nodules of recent caliche.

Encrusting calcite

Abundant crusts or rinds enveloping single fragments or aggregates of sediment probably are a highly significant feature in the recognition of calcite of a caliche origin. Most calcite crusts vary in thickness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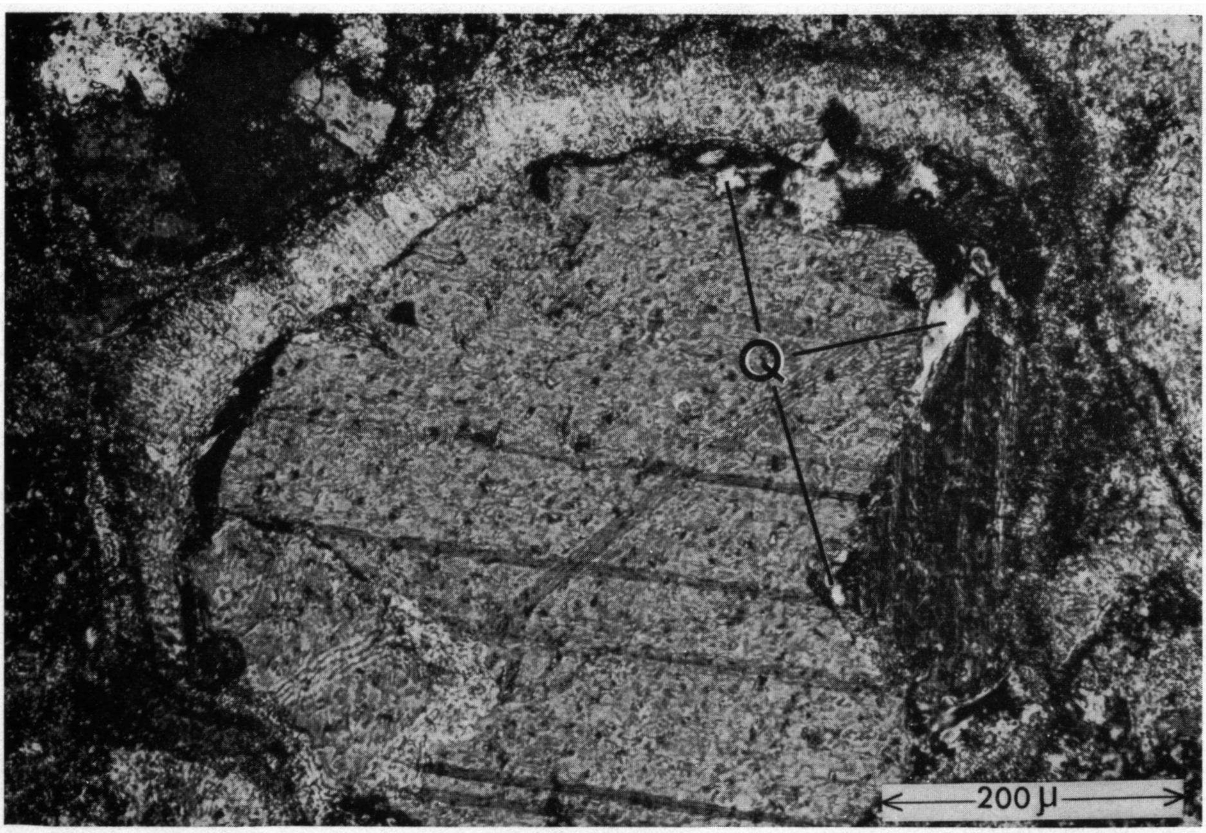
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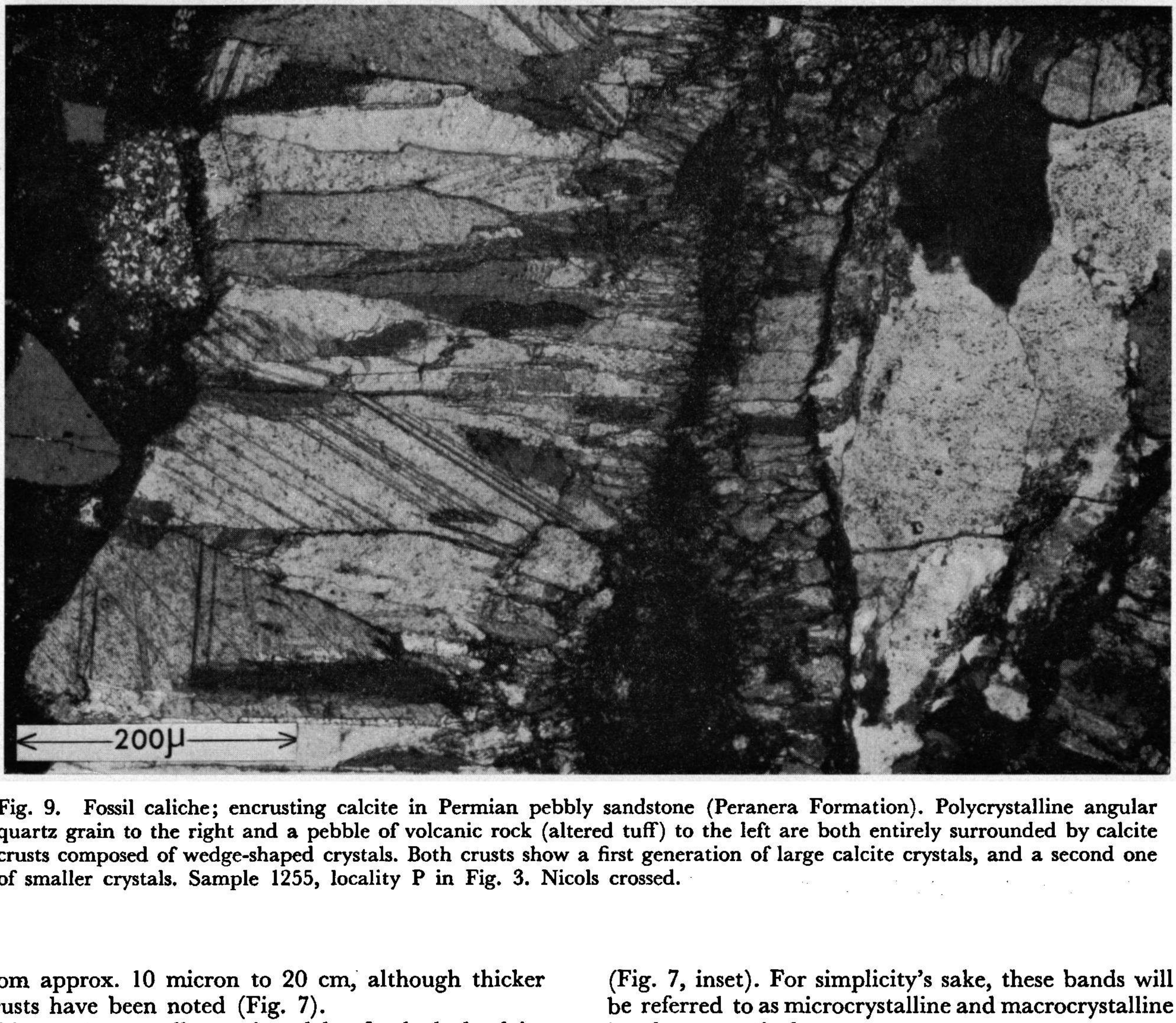


Fig. 9. Fossil caliche; encrusting calcite in Permian pebbly sandstone (Peranera Formation). Polycrystalline angular quartz grain to the right and a pebble of volcanic rock (altered tuff) to the left are both entirely surrounded by calcite crusts composed of wedge-shaped crystals. Both crusts show a first generation of large calcite crystals, and a second one of smaller crystals. Sample 1255, locality P in Fig. 3. Nicols crossed.

from approx. 10 micron to 20 cm, although thicker crusts have been noted (Fig. 7).

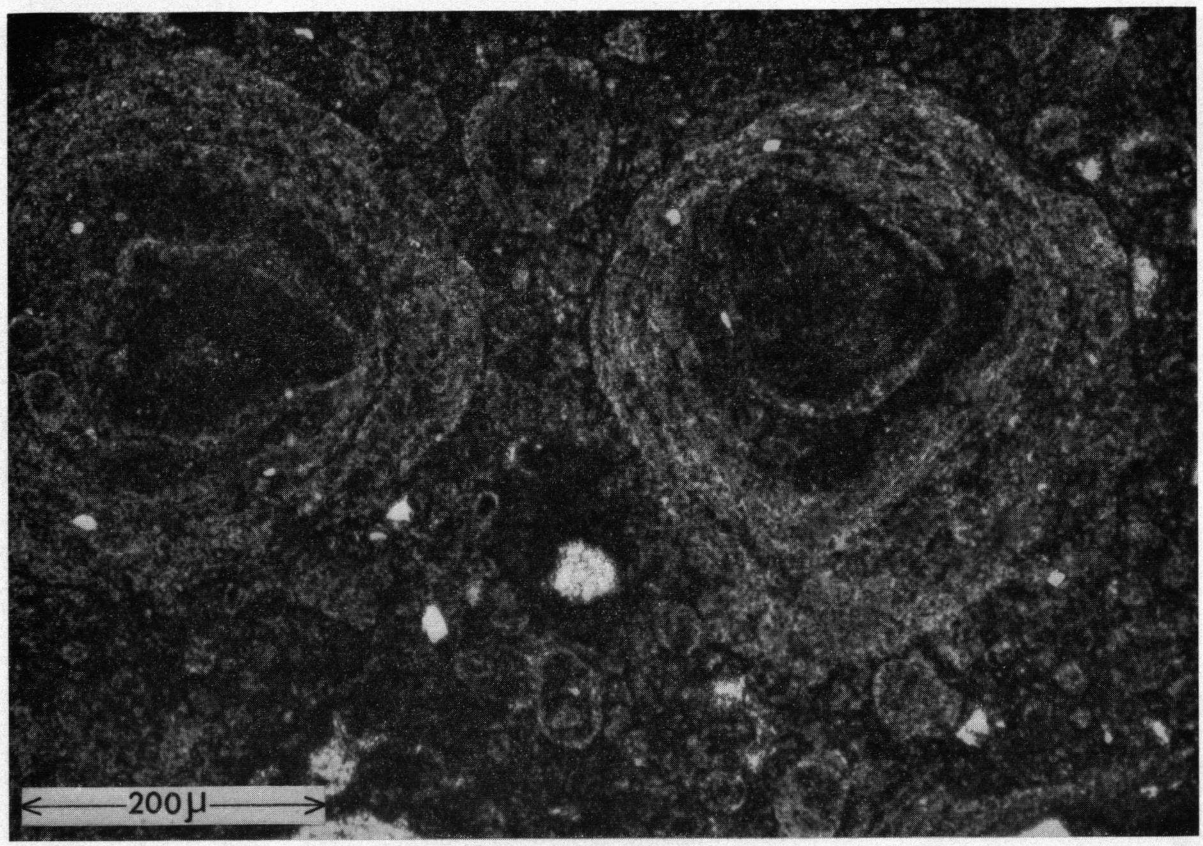
Thin crusts normally consist solely of anhedral calcite crystals, many of which occur throughout the thickness of the crust; thicker crusts are composite, consisting of alternating bands of extremely fine-grained calcite and bands composed of large anhedral crystals (Fig. 7, inset). For simplicity's sake, these bands will be referred to as microcrystalline and macrocrystalline bands, respectively.

Fig. 7. Recent caliche; encrusting calcite. Limestone boulder in Quaternary alluvial fan deposit is entirely surrounded by a calcite crust, which is thickest at the upper surface of the boulder. Inset shows alternation of macrocrystalline and microcrystalline calcite bands in a sample taken from the crust shown in the larger photograph. Sample 2503, locality 3 in Fig. 1. Nicols crossed (inset).

Fig. 8. Fossil caliche; replacement and encrusting calcite in Stephanian sandstone (Malpas Formation, braided river facies). Silicate sand grain almost fully replaced by calcite; small relics of quartz are still preserved (Q). Fibrous calcite crust envelopes the grain; it probably formed after the grain had been replaced by calcite. All calcite is ferroan. In the same sample numerous crusts of columnar, wedge-shaped calcite crystals occur as well. Sample 1158, locality M in Fig. 3. Nicols crossed.

The maximum thickness of macrocrystalline bands in the fossil and recent instances is the same: approx. 3 cm. In the macrocrystalline bands calcite displays two habits: (a) columnar and wedge-shaped interlocking crystals, and (b) fibrous calcite crystals or whiskers. The latter habit was noted only in one (fossil) case (Fig. 8). Virtually all calcite crystals are arranged perpendicularly to the surfaces on which the encrustation took place. In many cases the surface on which encrustation occurred is beset with a first generation of small crystals, presently succeeded by much larger crystals, though reverse relations are found as well (Fig. 9).

Oval or spherical calcite aggregates, up to a few cm in size, in which the crystals have the wedge-shaped habit as in encrusting calcite and radiate outward, were noted in various samples of the Peranera Formation. These aggregates, classed as encrusting calcite, may have formed on very small nuclei, or on nuclei which now have disappeared through replacement by calcite.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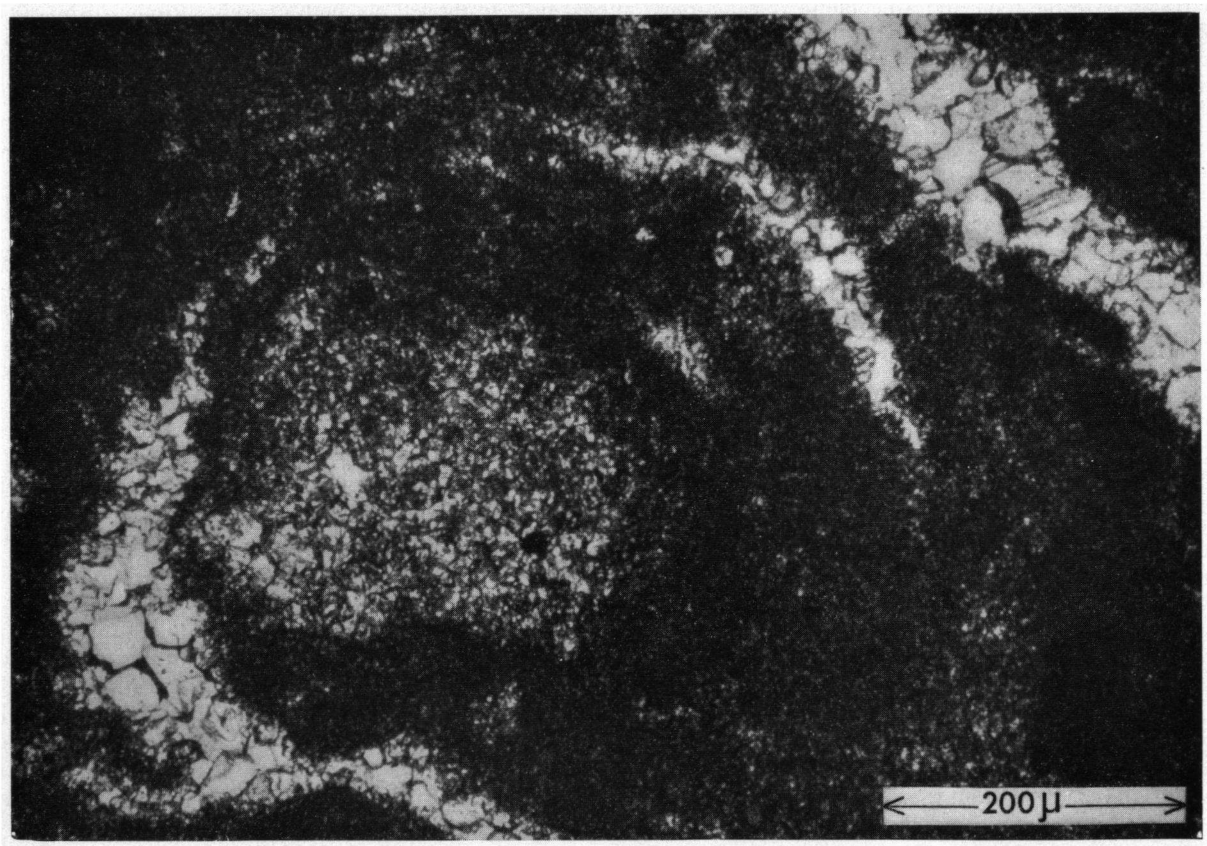
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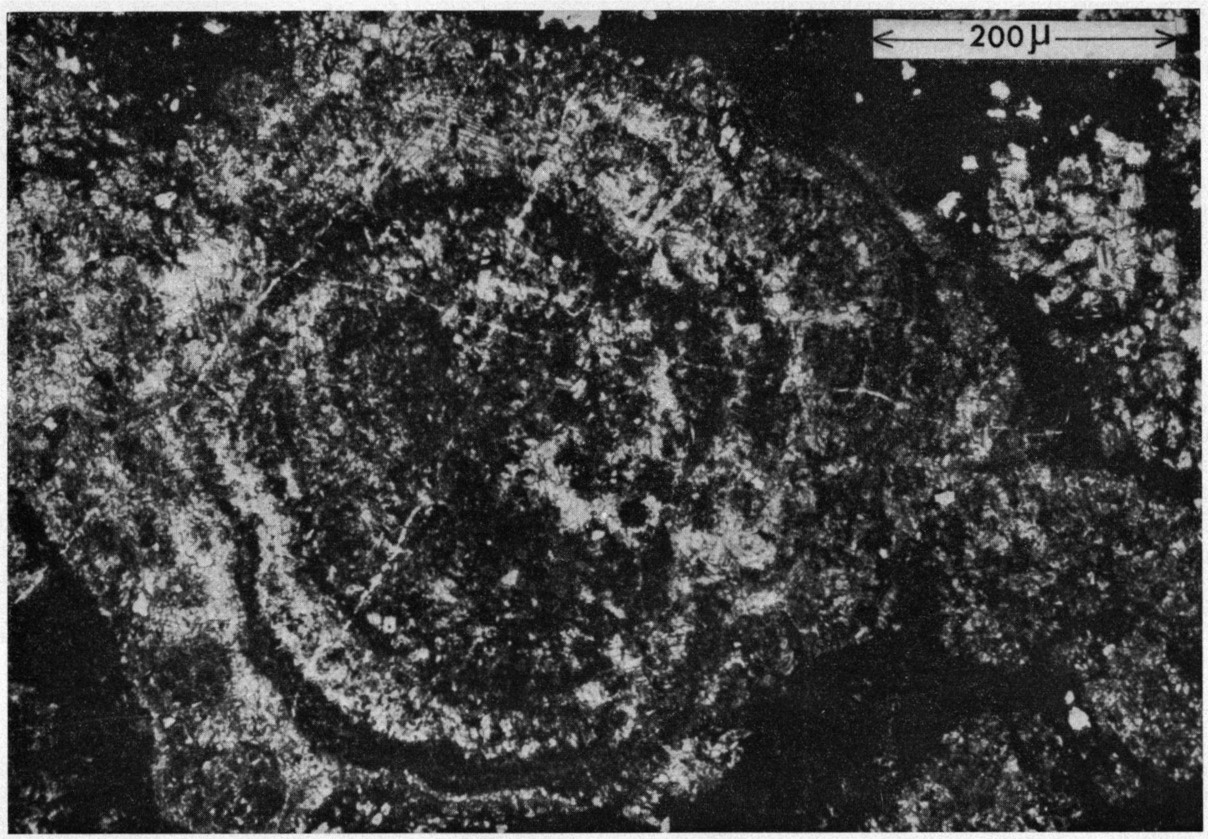

Fig. 12. Fossil caliche; calcite/dolomite spherulite in mudstone of Bunter Formation (Lower Triassic). The mudstone is densely packed with similar spherulites. Though in size and concentric structure the spherulites are similar to those in Permian calcrete (Fig. 11) and recent calcrete (Fig. 10), the carbonate is coarser-grained, probably due to dolomitization. Also, the content of ferric oxide (hematite, black) is higher; precipitation of calcite and of ferric oxides appear to have been alternating processes. Small spherulites are incorporated in the large one; two distinct items are visible in the lower left of the microphotograph. Sample 1464, locality B in Fig. 3. Plain light.

A common feature in macrocrystalline bands is the occurrence of rows of inclusions of calcite of the microcrystalline size range. The rows are arranged parallel to each other and to the encrustation surface; they are continuous through large calcite crystals and crystal boundaries alike. The rows probably record cessations in the growth of the macrocrystalline bands; subsequently, the growth of the large calcite crystals continued syntaxially. The maximum thickness of the rows is approx. 120 micron; with greater thicknesses further growth of the macrocrystalline calcite is no longer syntaxial, and the microcrystalline calcite behaves as a distinct band (Fig. 7, inset).

Distinct microcrystalline bands have not been found in the fossil caliche: there all encrusting calcite is macrocrystalline, although parallel rows of inclusions do occur.

In handspecimens the microcrystalline calcite is delicately stratified, showing alternations of laminae differing slightly in lightness. The thickness of microcrystalline bands ranges in many cases between the minimum thickness mentioned (120 micron) and a few cm. The laminated structure of the bands is caused by slight variations in included ferric oxides and differences in the coarseness of the crystallinity; most laminae range in thickness between 250 and

Fig. 10. Recent caliche; calcite spherulites in calcrete. All spherulites shown have nuclei of fine-grained calcite. Both large spherulites engulfed, in the course of their growth, smaller spherulites; two incorporated small spherulites are clearly seen at the extreme left in the micro-photograph. Just below the centre and at the lower left occur two light patches of void-filling calcite. In the same sample, many voids are still only partly filled or even open. Sample 2509, locality 1 in Fig. 1. Plain light.

Fig. 11. Fossil caliche; calcite spherulite in Permian calcrete (Peranera Formation). The nucleus of the spherulite consists of fine-grained calcite, probably somewhat recrystallized. The three light patches of macrocrystalline calcite are filled-in voids, similar in size and form to those in recent calcrete. Though of the same habit as in recent spherulites (Fig. 10), the concentric structure is not as distinct. Many even less distinct spherulites occur in the same sample. Sample 1270a, locality P in Fig. 3. Plain light.

1000 micron. The microtexture of the calcite is crystalline granular, with crystal sizes ranging between approx. 4 and 50 micron, most crystals lying in the 10—20 micron range. Many laminae show a gradation in crystallinity, slightly coarsening outwards.

A peculiarity of the encrusting calcite is that an estimated majority of the crusts is thickest in their stratigraphic upper parts. To the author's knowledge, this feature has only been described of caliche (Rutte, 1958) and may consequently be a distinctive one. The variation in thickness occurs on a macroscopic as well as a microscopic scale; it was noted both in the recent and in the ancient caliche (Fig. 7).

The origin of the alternating macrocrystalline and microcrystalline bands is not quite clear. Considering its much larger crystals, the calcite in the macrocrystalline bands may have crystallized at a slower rate than the calcite in the microcrystalline bands.

Spherulites

The term 'spherulite' will be used for spheroidal concretionary aggregates of calcite, which have a radial internal structure, or a concentric internal structure, or a combination of both. The meaning of spherulite is therefore taken in a different sense from that of oolites and pisolites, terms which are generally applied to spheroidal aggregates of calcite formed in agitated waters.

Calcite spherulites are commonly associated with caliche. According to Rutte (1958; 'Pseudooids'), they form in loosely packed, porous soil material in which the temperature may fluctuate strongly due to insolation. In the recent profiles studied, the spherulites are restricted to the uppermost zone in caliche (calcrete).

The fossil occurrences of spherulites were found to occur in a thick terrestrial tuff bed in the Malpas Formation, in calcrete in the Peranera formation, and in mudstones in the Bunter Formation. The spherulites vary in size from 20 micron to approx. 3 mm, both in the recent and in the ancient samples. The majority of the spherulites contain a nucleus which may be composed of a single quartz grain, a rock fragment or an aggregate of fine-grained calcite or clastic sediment. If the nucleus is small in relation to the spherulite, or if no nucleus is present, the concentric internal structure of the spherulites is more or less radial-symmetric. Occasionally also large grains or particles (up to 1.5 cm), as if by a vagary of nature, served as nuclei. In these cases the spherulite is no more than a thin rind, its outward shape reflecting that of the nucleus. Such 'spherulites' are transitional types to encrustations (p. 135).

In the recent spherulites, most calcite is extremely fine-grained, having grain-sizes of less than 4 micron. The calcite is arranged in a vaguely-defined, fully concentric pattern (Fig. 10). Staining with alizarine red S brings out the structure somewhat more clearly. In many recent spherulites microcrystalline quartz is present; in some, the quartz-content may reach an estimated 10 % by volume. Ferric oxides are usually present as a vague light-brown stain.

The internal structure of the spherulites looks like that of onkolites; where few, and less perfect, spherulites occur, they are apt to be confused with onkolites. Similarly, the finely laminated structure also common in calcrete (to which no further reference is made here) can assume the appearance of algal mats (Multer & Hoffmeister, 1968).

The variation in the characteristics of the ancient spherulites is somewhat wider than in the recent ones studied. The calcrete in the Peranera Formation shows, apart from parallel laminations, a vaguely globular texture. Presumably, it partially consists of calcite spherulites, the internal structure of which has almost disappeared (Fig. 11). On the other hand, spherulites in the Bunter, which occur densely packed in mudstone (no true calcrete being present), show a much clearer internal structure as a result of the occurrence of distinct rings rich in ferric oxides. Nearly all Bunter spherulites, moreover, show dolomitization in varying degrees (Fig. 12). Recrystallization and dolomitization usually led to a radial structure, although this is imperfectly developed.

ADDITIONAL REMARKS

Apart from the differences in microtexture of the calcite in recent and ancient caliche as mentioned above, there are noteworthy compositional differences. In the encrustations-carrying sandstone of the Malpas Formation, for instance, nearly all calcite is ferroan, while dolomite locally occurs as a last pore-fill (determinations with the combined carbonate staining technique, Dickson 1966). In the recent caliche, however, no authigenic ferroan calcite was noted in any of the 21 samples collected. Some authigenic dolomite occurs in the caliche formed in the Quaternary alluvial fan deposits (exposures 4 and 5, Fig. 1). The alluvial fan deposits carry dolomite rock fragments in abundance; it is likely that the Mg was derived from this source. Likewise, in the case of the Malpas sandstones, the Fe and Mg may have been derived *in situ* by replacement of the volcanic components which abound in the sandstones. As is clear from Fig. 8, replacement by calcite occurred on an extensive scale before the encrusting calcite formed; the Fe included in the encrusting calcite and the Mg necessary for the formation of dolomite could have been mobilized by this process.

Both in the samples of recent and of ancient caliche, calcite either is the first precipitate, or it comes second after colloform ferric oxides. In some cases the precipitation of calcite and ferric oxides did alternate (Fig. 12). Calcite commonly replaces the ferric oxides. Where microcrystalline, authigenic quartz is present, it appears to have formed simultaneously with the calcite. In the recent spherulites microcrystalline quartz occurs intergrown with the fine-grained calcite; in some Peranera sandstones chalcedonic quartz occurs intergrown with replacement and cementing calcite.

To summarize, it has been shown that the microtextures of calcite in recent caliche and of calcite in Stephanian, Permian and Bunter continental deposits in the South-Central Pyrenees are very similar. This fact lends support to the assumption that the fossil calcite microtextures did originate during early stages in diagenesis, under climatic conditions favourable to the formation of caliche.

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