Syndepositional and Late Diagenetic Alteration of Primary Gypsum to Anhydrite

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ABSTRACT

Because primary gypsum is made over to anhydrite upon burial, as a consequence of rising temperature, it is necessary to establish criteria that can distinguish between anhydrite which formed by syndepositional alteration at the surface from that which results from burial diaggnesis. It is suggested that syndepositional alteration results in an anhydrite fabric resembling that seen in the supratidal areas of the Persian Gulf whilst burial diagenesis results in a different set of textural fabrics. In those instances where the amounts of impurities in the gypsum beds are large, conversion to anhydrite may still preserve the outlines of the original crystal morphology. In those beds where only very small amounts of impurity is present, massive featureless beds of coursely crystalline to spherulitic anhydrite results. If the escape of the water of crystallization is impeded for a time and then released, partial fluidization of portions of the beds may result, and may also be recorded in the rock record.

INTRODUCTION

Although gypsum is common in Recent evaporitic sediments and primary gypsum is preserved in some Neogene evaporite formations that have been buried to only relatively shallow depths, it is characteristically not found in borehole cores taken from evaporites at depths greater than approximately 1000 metres where anhydrite is normally the only calcium sulphate mineral present. It is therefore generally believed that gypsum becomes unstable in consequence of the increase in rock temperature which accompanies burial and that at some critical depth, determined by the local geothermal gradient and the salinity of the connate waters, it is made over into anhydrite. Predicted depths lie in the order of 1000 metres; Murray (1964) and Holser (1979), and are thus in accord with the observed distribution. However anhydrite is also known to occur in comparative abundance in certain present day and Neogene evaporites, where some of it demonstrably formed by syndepositional alteration of gypsum. Thus a question that arises in the study of borehole cores from evaporite successions is that of criteria which can be used to distinguish between anhydrite which formed by syndepositional alteration of gypsum from that which was produced by later replacement of gypsum as a result of burial diagenesis. It is with those criteria that this paper is concerned.

In simplest terms some anhydrite rocks are nodular, others are not; and the nodular varieties stand apart from the rest both in their appearance and in their crystal fabrics. There are lines of evidence which suggest that in the nodular anhydrite rocks the anhydrite was mostly syndepositional in origin, and that in many instances it formed by syndepositional alteration of gypsum. The evidence for that will be dealt with in the first part of the paper, by reference to Recent and Neogene occurrences. The non-nodular anhydrite rocks are a somewhat diverse group, and they did not all have the same origin. However if the arguments set out in this paper are correct, the group should include anhydrite rocks which were produced by burial diagenesis of gypsum. In order to attempt to identify these latter, a brief review will be made of the conditions thought likely to prevail during burial diagenesis of gypsum and the ways in which those conditions might influence the structure and crystal fabric of the ultimate anhydrite rock. Attention will then be directed to certain anhydrite rocks that have the appropriate features.

NODULAR ANHYDRITE ROCKS

The term nodular anhydrite is here used in the broad sense to include: isolated nodules, masses of coalesced nodules and enterolithic nodular structures. Evidence that these various nodular structures characterise syndepositional anhydrite is discussed, first by dealing with the mode of formation of anhydrite nodules in Recent sediments, and then by reference to the way in which various lithofacies of primary gypsum were made over into nodular anhydrite by syndepositional alteration of primary gypsum in superficially buried Neogene evaporites.

Nodular Anhydrite Formed by Syndepositional Alteration of Gypsum in Present Day Sabkhas

Anhydrite is forming at the present day on a regional scale in the coastal sabkhas of Abu Dhabi, UAE, and the review by Butler *et al.*, (1982) provides a resume of the setting in which it occurs. The anhydrite is characteristically nodular and the nodules grow displacively, mostly in the sediments of the supratidal facies. Some of the nodules clearly arose by alteration of earlier formed gypsum, but others evidently formed *de novo* without gypsum precursors. Further, anhydrite nodules have been seen to hydrate to gypsum as a result of dilution of the sabkha brines following heavy rainstorms, but those are uncommon events and the gypsum is made back into nodular anhydrite within a few months as high salinities become reestablished. Irrespective of their origin all of the nodules that have been examined to date have essentially similar crystal fabrics.

In situ, the nodules are soft and easily deformed so that it is difficult to collect and section them without disturbing their crystal fabrics. However nodules near the surface are sometimes cemented by halite and provide suitable material for petrographic studies. As seen in thin section (Figure 1) the nodules comprise loose aggregates of small platelets of anhydrite, many of which appear to be bent and broken. The arrangement of the platelets commonly varies from place to place in the one nodule, ranging from decussate to aligned, but there is a strong tendency for those in the outer parts to lie subparallel to the margin.

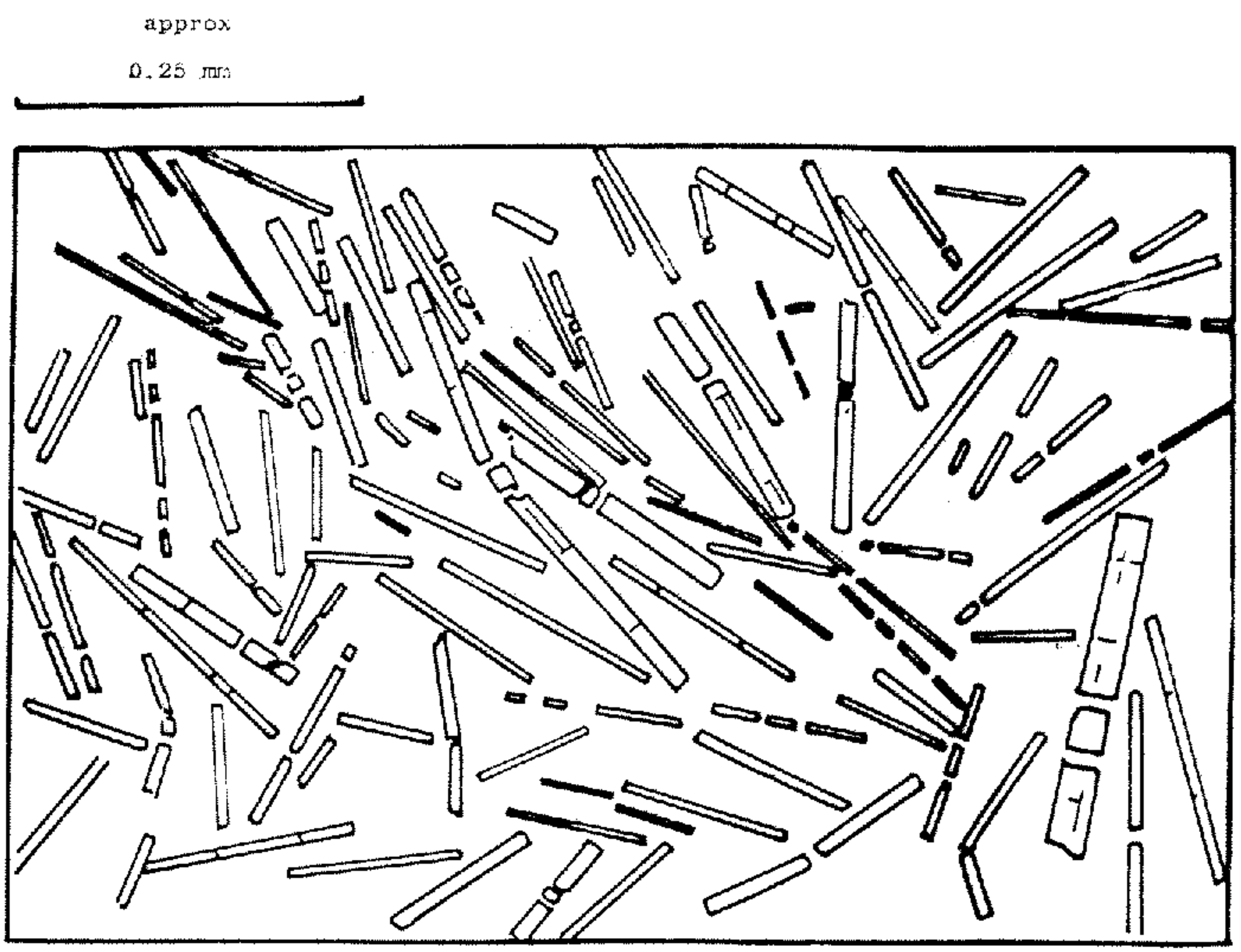


Figure 1. Schematic representation of the crystal fabric of a recent anhydrite nodule. For simplicity approximately only half of the crystals have been drawn in.

The nodules fall apart easily in water and many of the individual crystals are seen to be very thin rectilinear platelets, the shapes of which are controlled by the three orthorhombic pinakoidal cleavages. These are, in fact, cleavage flakes. Unbroken crystals do occur: they too are very thin but they are lanceolate in outline. The platy nature of the crystals is not readily apparent in thin sections where they mostly appear lath-like. Many of the 'laths' have oblique extinction, but that optical effect arises naturally in orthorhombic minerals with certain orientations of the thin section relative to the traces of the outline of the crystal and its optic axial plane.

Gypsum crystals in the sabkhas are mostly lenticular in habit and grew displacively. Where they commence to undergo alteration to anhydrite, cavernous hollows develop in them which are occupied by loose accumulations of tiny anhydrite crystals. The corroded appearance of the hollows leaves little doubt that the change from one mineral to the other was a dissolution/precipitation process. As alteration proceeds (Figure 2) each gypsum crystal is made over into a loose mass of anhydrite crystals that first roughly pseudomorph it, but as more and more anhydrite crystals are addded the pseudomorph swells out into a nodular mass that ultimately loses all resemblance to its parent. Some nodules that evidently had gypsum precursors have grown to such an extent that, even allowing for the loose packing of the anhydrite crystals, much more calcium sulphate must now be present than could have been provided by the original gypsum crystal. The additional calcium and sulphate ions can only have been supplied from the sabkha brines. Thus it seems that there is a self-perpetuating mechanism built into anhydrite nodules, so that once initiated a nodule will continue to grow, provided that there is a continuing supply of ions. The nodules appear to grow by nucleation of new crystals within the framework of earlier formed ones, and as the new crystals grow they push the others aside and those in turn displace the surrounding host sediment. The presence of broken crystals and cleavage fragments in the nodules is fair testimony to the extent of the internal movements that accompany the expansive growth. It is inevitable that the internal movements must from time to time cause the anhydrite crystals to scratch one another, and collision between crystals is known to be a potent mechanism for generating nuclei.

The anhydrite nodules are remarkably pure, and that purity is a measure of the efficacy of the platy shapes of the crystals in pushing the surrounding sediment aside. Where anhydrite nodules grow in close proximity to one another and coalesce, the displaced sediment comes to form partitions between them and gives rise to the familiar mosaic or chicken wire appearance. Continued growth of

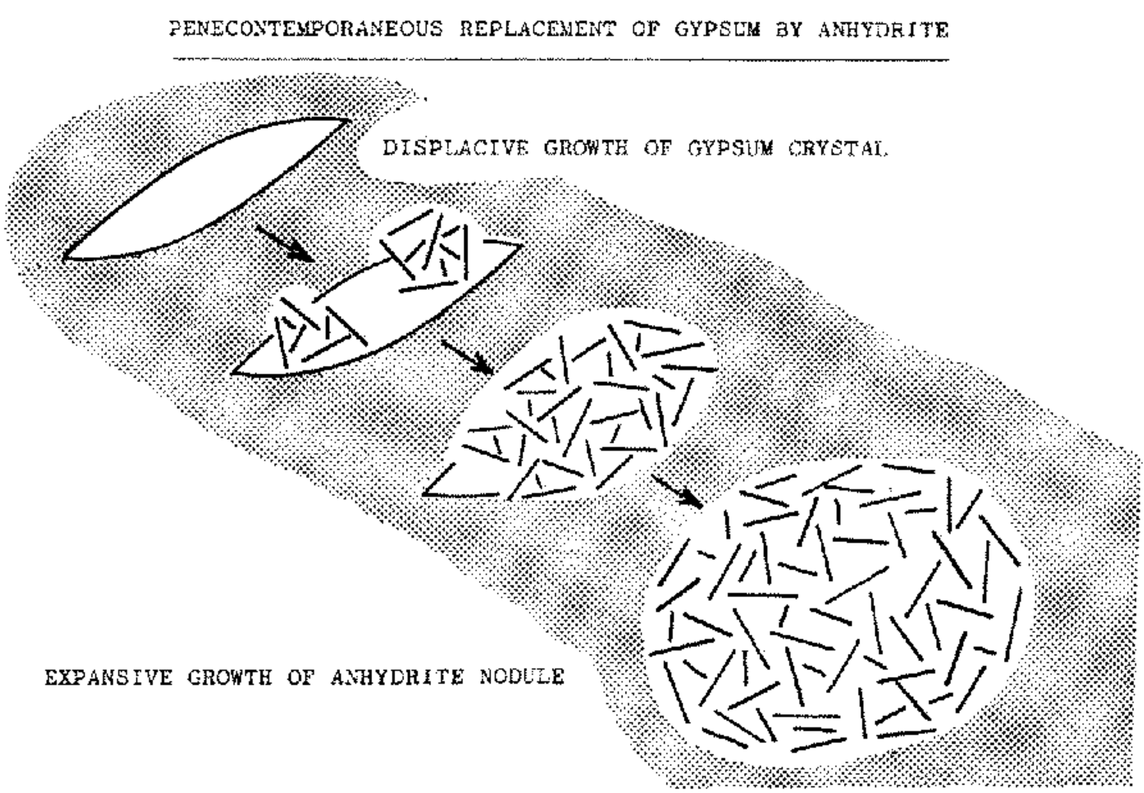


Figure 2. Stages in the transformation of a crystal of primary gypsum to nodular anhydrite by syndepositional diagenesis in the recent sediments of the Abu Dhabi sabkhas.

coalesced nodules causes the mass to distort, and enterolithic structures develop. The movements necessary to produce the enterolithic structures are no doubt facilitated by the natural moisture of the nodules and the platy shapes of the crystals which allow easy slip between them.

Rough anhydrite pseudomorphs after gypsum with crystal fabrics similar to those described above are known from sabkha facies evaporites in various parts of the stratigraphic record (Holliday, 1973); and the crystal fabrics of anhydrite nodules seen in borehole cores from sabkha cycles of the past match those of the Recent. However in these older rocks, the fabrics are often modified by compaction which has tightened the packing and caused further breakage of the crystals.

Nodular Anhydrite Formed by Syndepositional Alteration of Primary Gypsum in Neogene Evaporites

Many of the Neogene evaporites exposed at outcrop in countries around the Mediterranean include beds of unaltered primary gypsum. They range from thick massive beds of competitively grown gypsum crystals, through current-bedded gypsum sands to very fine-grained delicately bedded gypsum laminites, all of which were clearly subaqueous in origin. There are also some current-bedded gypsum sands that may have been aeolian. Th field relationships of these formations indicate that at no time had they been buried to depths of more than a few hundred metres and those depths were evidently insufficient to carry them out of the stability field of gypsum. In some places the beds of primary gypsum enclose nodules of secondary gypsum, but the secondary gypsum carries tiny corroded relics of anhydrite crystals and those demonstrate that the nodules were originally nodules of anhydrite. The hydration of the anhydrite to gypsum was a late effect caused by introduction of meteoric water during exhumation of the rock. The relationships of the former anhydrite nodules to the primary gypsum which encloses them leaves no doubt that the nodules formed by alteration of the primary gypsum. The frequency of the nodules varies from bed to bed. They are absent from many beds, sparsely scattered in others where they are usually developed at particular levels; but they are so abundant elsewhere that only mere vestiges of the original gypsum remain. The extent of alteration varies randomly from bed to bed in any stratigraphic succession, and the change clearly took place on a *lit par lit* basis. The only reasonable conclusion is that the change from gypsum to nodular anhydrite was the result of changes in the chemistry of the interstitial brines penecontemporaneously with deposition.

Three examples will serve to illustrate the ways in which syndepositional alteration to nodular anhydrite affected various lithofacies of primary gypsum in the Neogene.

Syndepositional Alteration of Displacively Grown Crystals of Primary Gypsum in the Miocene of Turkey

Part of the Miocene succession seen at outcrop in the Cankari-Corum Basin of Turkey comprises calcium sulphate bearing carbonates, in which the individual beds range in thickness from a few centimetres up to several metres (Ergun, 1977). Some of the beds enclose scattered lenticular crystals of primary gypsum which grew displacively in the carbonate host. In many beds, however, each gypsum 'crystal' is in part unaltered primary gypsum and in part a swollen mass of secondary gypsum (Figure 3). In other beds primary gypsum is wholly absent, but there are abundant nodules of secondary gypsum, many of which are obviously pseudomorphous after the lenticular habit of primary gypsum.

The disposition of the original gypsum crystals indicates that they grew displacively in the carbonates, and

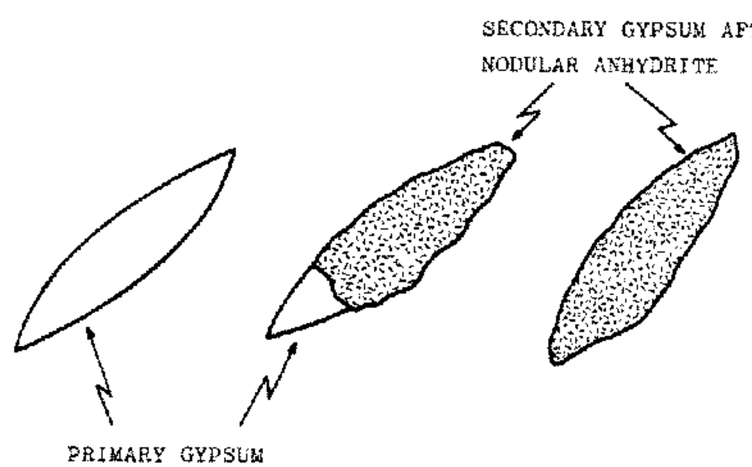


Figure 3. Syndepositional alteration of primary gypsum to nodular anhydrite (now secondary gypsum): Miocene, Turkey.

they must, therefore, have crystallized from the interstitial brines. The extent to which the primary gypsum was made over into nodular anhydrite (now nodular secondary gypsum) varies randomly throughout the succession, and in any one bed the change must have occurred before deposition of the overlying one. The mode of alteration was essentially similar to that which takes place at the present day in the coastal sabkhas of Abu Dhabi.

Nodular Anhydrite Formed by Alteration of Primary Gypsum in Beds of Competitively Grown Crystals: Messinian S E Spain

Beds of competitively grown crystals of primary gypsum are locally well exposed in the Messinian evaporites of S E Spain, Sicily, Cyprus and elsewhere in the Mediterranean region. The beds range in thickness from a few centimetres up to several metres, and in some of the thicker beds individual crystals are very large indeed: five metres or more in length. In some exposures in Sicily and Spain the gypsum crystals were partly replaced by masses of nodular anhydrite, now nodular secondary gypsum (Schreiber, Roth & Helman, 1982., and Rouchy, 1980), while in other places the replacement went almost to completion.

Rouchy (1980) illustrated one of the partly altered crystals of primary gypsum, and the example is of particular interest because it throws light on some of the factors that influence the shapes and patterns of nodules. The gypsum (Figure 4) is a large twinned crystal of a type commonly seen in the Messinian evaporites. Each side of the twin comprises a succession of descrete arms that are reminiscent of the multiplicity of the arms of Siva. Each arm carries two sets of zones of included sediment. One set lies parallel to the prism {120}, and the other, which has irrational curved traces, is more sediment rich and delimits the arms (Orti-Cabo and Shearman, 1977). Where alteration to nodular anhydrite took place, there was a tendency for some nodules to become elongate along the length of an arm, as though guided by it, but other nodules spread across the sediment boundaries between successive arms and were in no way confined. Where adjacent nodules grew against one another, they are separated by thin partitions of sediment, and that sediment could only have been derived by redistribution of the material originally present in the sediment zones of the now altered parts of the gypsum. Factors which influenced the shapes and arrangements of the nodules clearly included: the spacing of the centres about which alteration was initiated, the distribution of included sediment in the parent gypsum, and the extent to which the discontinuities between successive arms of the twin were able to confine growth of the nodules.

Ways in which the factors might operate are illustrated schematically in Figure 5, which illustrates a Siva twin of gypsum in which there is some included sediment zoned parallel to {120} and a relative abundance along the discontinuities between successive arms. On the right hand side of the twin it is assumed that alteration to anhydrite is initiated at only one or two centres in each arm. As the nodules expand they push the sediment in the {120} zones aside, and that becomes concentrated around their margins. If the discontinuities between successive arms are sufficiently profound, the growing nodules do not easily cross them. But the nodules are free to extend along the length of the arms, and in consequence the shapes and arrangements of the nodules ultimately come to mimic the array of arms of the former twin. On the left hand side of the twin it is assumed that alteration sets in about more closely spaced centres, several to each arm of the twin. Expansion and coalescence of these closely spaced nodules will produce a nodular mosaic that carries little hint of the morphology of the parent crystal. Schreiber, Roth & Helman (1982) and Loucks & Longman (1982) were quick to appreciate the importance of Rouchy's observations, and they used the elongation of anhydrite nodules seen in some borehole cores as evidence for an origin by alteration of primary gypsum; although the examples which they cited may have arisen from groups of single rather than twinned crystals.

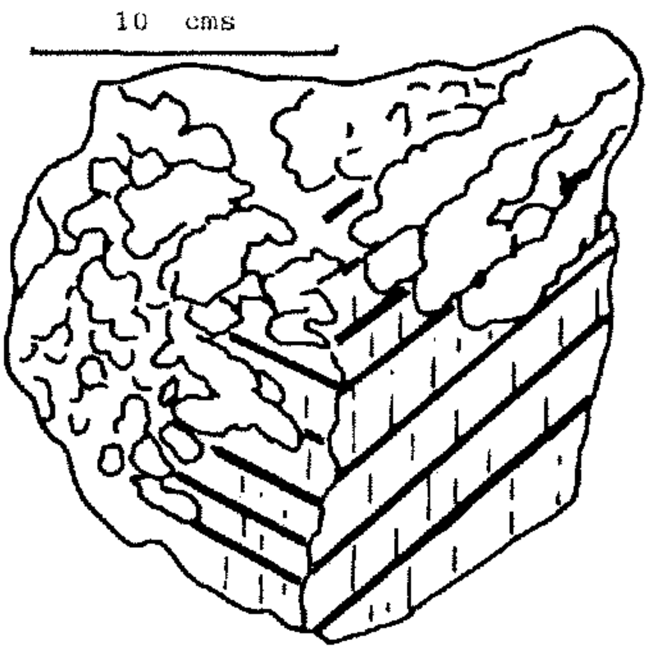


Figure 4. Siva twin of primary gypsum in part made over into nodular anhydrite (now secondary gypsum). Messinian, SE Spain Drawing from Rouchy (1980) Plate 43.

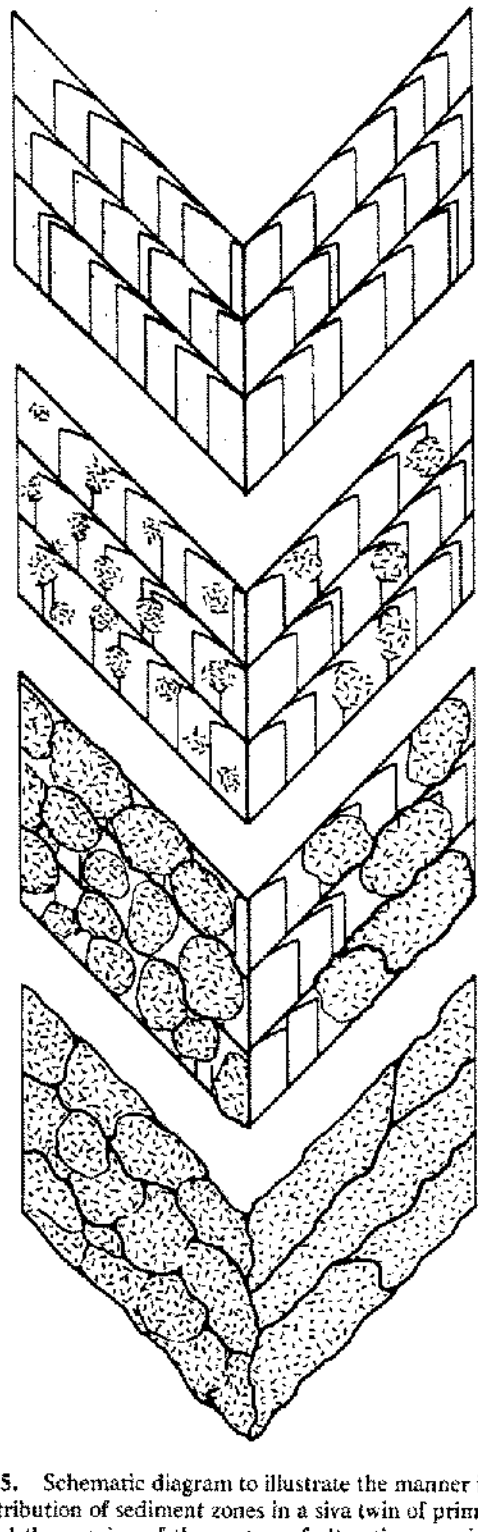


Figure 5. Schematic diagram to illustrate the manner in which the distribution of sediment zones in a siva twin of primary gypsum and the spacing of the centres of alteration may influence the shapes of anhydrite nodules formed by syndepositional alteration of the gypsum.

Nodular Anhydrite Rocks Formed by Alteration of Primary Gypsum Laminites in the Messinian Evaporites of Cyprus

Very fine-grained, delicately-bedded gypsum laminites of Messinian age are locally well exposed in parts of Cyprus. Some comprise very tiny prismatic to short acicular crystals of gypsum that lie with their long axes in the plane of bedding, but randomly arrayed in that plane. The laminae are in part defined by slight differences in crystal size, in part by occasional laminae of carbonate, but mostly by what appear to have been thin films of organic matter. The gypsum crystals were clearly primary and were presumably deposited subaqueously. In most places the laminae are continuous and undisturbed, but there are some exposures where at particular levels, the continuity of the laminae was interrupted by growth of lenticular masses of nodular anhydrite, now nodular secondary gypsum (Figure 6). The laminae terminate abruptly against the sides of the nodules which obviously replaced them, but there is no hint at all of the former laminae anywhere in the nodules. In some of the clusters the individual nodules are not all clearly defined because the partitions of displaced sediment that separate them are thin and impersistent. That vagueness of the partitions is an expression of the general paucity of sediment impurities in the primary laminites, and it follows that if the primary gypsum had been completely pure the alteration would have produced an apparently homogeneous mass of anhydrite without obvious nodular structure.

Review of Syndepositional Nodular Anydrite

All of the occurrences of nodular anhydrites from the Neogene that have been described in the preceding sections of the paper are from rocks now exposed at outcrop, and most of the anhydrite has been hydrated to secondary gypsum. The crystal fabrics of the earlier anhydrite have been largely obliterated, but where the secondary gypsum is porphyroblastic, the porphyroblasts commonly enclose abundant corroded relics of the former anhydrite crystals. These tiny relics are often slightly elongate and they show random optical orientations no matter how closely spaced they are; and that combination of features is reminiscent of the crystal fabrics of Recent anhydrite nodules and of nodular anhydrite found in borehole cores.

The intimate association of nodular anhydrite (now secondary gypsum) with unaltered primary gypsum in Neogene evaporites, together with the *lit par lit* mode of replacement of the primary gypsum by the anhydrite, testifies to the alteration having been a syndepositional process. The change from gypsum to anhydrite appears to have been a dissolution/reprecipitation process, so enlargement of any nodule required dissolution of the gypsum immediately surrounding it. Two possibilities arise with respect to the way in which the nodules increased in size.

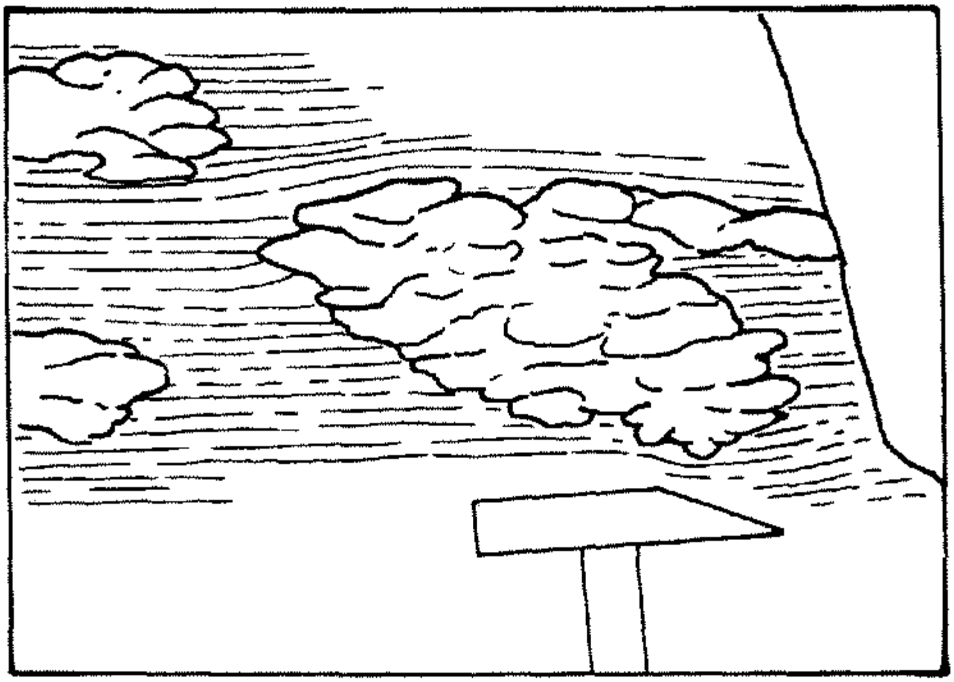


Figure 6. Syndepositional nodular anhydrite (now secondary gypsum) formed by alteration of a primary gypsum laminite. Miocene, Cyprus.

One is that the nodules grew by addition of new crystals around the margins of the nodules in the spaces left by dissolution of the gypsum. If that had been the case, the probability is that a large proportion of the impurities in the original gypsum would have been retained in the nodules. The extreme purity of the nodules suggests the other possibility, that is, that the nodules grew by nucleation of new crystals inside the framework of the nodules, so that they expanded like an inflated balloon. With such a mode of growth, the anhydrite crystals in the outer parts of the nodules would have been shoved mechanically outward into the spaces left by dissolution of the surrounding gypsum, where because of their platy shapes they would have pushed impurities ahead of them.

The occurrences in the Neogene show that all lithofacies of primary gypsum are alike made over into nodular anhydrite by syndepositional alteration. The pattern of nodules that result are determined not so much by the lithofacies, as by the spacing of the centres of alteration and the distribution of sediment impurities in the gypsum. In some instances where the concentration of impurities is sufficiently profound, they may exert some guidance on the shapes of the nodules, but in many instances little hint remains of the nature of the lithofacies of the original gypsum.

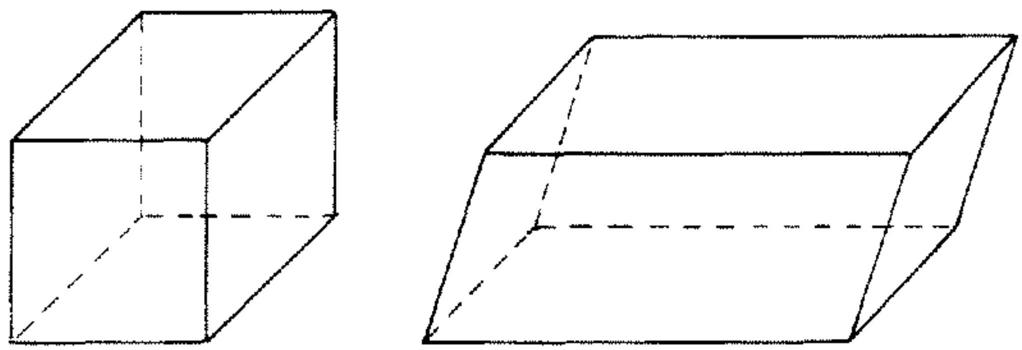
It is thus the writer's impression that syndepositional anhydrite is characteristically nodular, irrespective of whether it was generated by alteration of gypsum or arose *de novo*. However, it must not be forgotten that in the Messinian evaporites of Tuscany and Sicily, Schreiber, Roth and Helman (1982) found that primary gypsum had been converted to massive nodular anhydrite in the hinge of a fold at one place, and immediately adjacent to a fault at another. The tectonic setting suggested to Schreiber et al. that in those instances the change had been promoted by frictional heating.

BURIAL DIAGENESIS OF GYPSUM

The late change from gypsum to anhydrite which should take place during burial would be mainly a response to the rise in rock temperature that accompanies increasing depth of burial. That increase in temperature would be expected to affect any group of beds uniformly, so the diagenetic effects should be pervasive.

Thought must be given to the possible effects of the volume changes that should result from the mineral alteration. If the system is closed so that calcium and sulphate ions are neither lost nor added, the change should result in a decrease in volume of the solid phase of approximately 38 percent (Figure 7). Where the change from gypsum to anhydrite takes place syndepositionally, the reduction in volume is largely compensated for by the very loose packing of the anhydrite crystals in the resultant nodules. However, at the depths of burial where the late diagenetic change is believed to occur the system should be virtually

COMPARISON OF UNIT CELL VOLUMES



ANHYDRITE		GYPSUM	
$CaSO_4$		$CaSO_4 . 2H_2O$	
ORTHORHOMBIC		MONOCLINIC	
Density	2.963	Density	2.317
Unit cell volume	305.09	Unit cell volume	493.5
Z	4	Z	4
molecular volume	45.94	molecular volume	74.3

Figure 7a. Comparison of unit cell volumes.

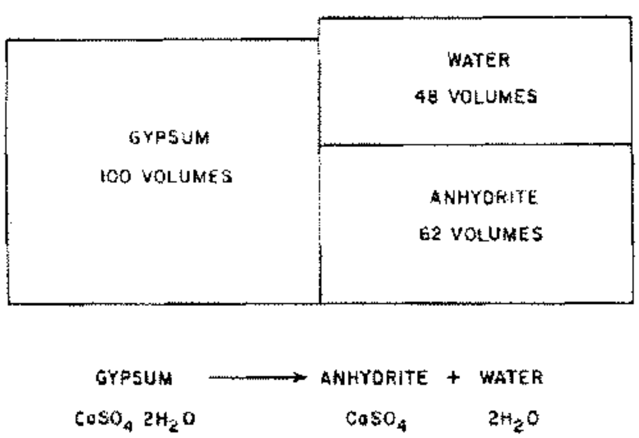


Figure 7b. Theoretical changes in volume involved in the alteration of gypsum to anhydrite.

closed with respect to calcium and sulphate ions. Under the overburden load which would prevail, the effects of the 38 percent reduction in bed thickness should be expressed in the detail of the structure of the anhydrite bed. Further, account must be taken of the manner in which the water released by the reaction escapes, because that too could affect the structure of the anhydrite rock.

One can only speculate on the way in which the diagenetic change takes place, but it is reasonable to suppose that during the early stages sufficient unaltered gypsum would be present to provide a rigid framework that would support the overburden load. That framework would progressively deteriorate, and the bed would ultimately be made over into a loosely packed mush of anhydrite crystals and water. As the water escapes, the mush would compact into a tighter packing, and because of the overburden load some breakage of the anhydrite crystals would be expected. However, there would be a limit to the extent of the mechanical compaction, and substantial water-filled porosity should remain. The sustained overburden load and further burial would lead to more slowly progressing compaction and expulsion of water by pressure dissolution. If the change does take place via a crystal mush, then the expulsion of water which would accompany the first phase of compaction might tend to fluidize the bed. That fluidization would not only involve the anhydrite crystals but also redistribute the sediment impurities formerly present in the gypsum so that many of the primary features of the original gypsum rock would be obliterated. In thick beds, the fluidization could also induce water-escape structures in the upper parts.

There are many anhydrite rocks which in borehole cores show no obvious nodular structure, but which appear either relatively featureless or show vague uncertain structures, some of which could be palimpsest, while others could have been induced. The beds come in all thicknesses from a few centimetres to several metres. They are more coarsely crystalline than nodular anhydrite rocks and have markedly different crystal fabrics.

In thin sections of some of these rocks, most of the anhydrite crystals have sub-rectangular outlines and they are tightly impressed against and into one another in a manner reminiscent of pressure dissolution. Here and there in the thin sections there are other anhydrite crystals that are much more elongate and which tend to split and fan slightly in one direction along their length (Figure 8). The likelihood is that all of the crystals are, in fact, similarly elongate and split, the differences in shapes of outlines being simply the result of orientation of the crystals relative to the plane of the thin section. This probability is supported by the fact that the elongate sections consistently display the lowest order interference colours, usually grey, and so contrast markedly with the bright higher order colours of the others. Where the arms of the elongate sections split they are commonly broken, and the broken parts are displaced to a greater or less extent. The fractures, which are now healed, took place along cleavage planes, and the displacement is clearly revealed under crossed polars by abrupt changes in extinction direction. There is little evidence of breakage in the other cross sections through the anhydrite crystals, but that again could be the result of their orientations and the position of the thin section across them. Thus, interpreting the thin sections in three dimensions, the likelihood is that there is much more breakage of crystals than appears at first sight. The fracturing of the crystals was clearly mechanical, and it implies that at one time the rock was a loosely packed uncemented aggregate. Sediment impurities are present as inclusions in some of the crystals and also as small pockets between crystals here and there in the thin sections, but there appears to be no regular pattern in their distribution.

There are other anhydrite rocks, which are of similar

Figure 8. Coarsely crystalline anhydrite rock: Permian, U.K. Schematic. Note the fracture and displacement at the extremity of the anhydrite crystal with the elongate cross section.

ilk, in which the anhydrite crystals are present as more strongly divergent fans or as spherulites. In these, too, there is evidence of breakage of crystals by an episode of compaction. It is perhaps less obvious in the spherulitic varieties, as it is usually only seen where former points of contact between the spherulites lie in the plane of the thin section.

Anhydrite rocks with these various types of crystal fabrics are well known, and they have usually been described as 'recrystallized.' Holliday (1973), for example, considered them to be the result of recrystallization of earlier aphanitic anhydrite rocks. The suggestion in this paper is that these apparently recrystallized rocks are the products of late diagenetic transformaton of beds of primary gypsum. The mechanical breakage of the anhydrite crystals was caused by compaction during the first phase of reduction in bed thickness which accompanied the mineralogical change. The final tightening up of the crystal fabric was by later pressure dissolution consequent on sustained overburden load and further burial.

The state of affairs seen in the Messinian evaporites, where only parts of a bed of primary gypsum were made over into nodular anhydrite by syndepositional diagenesis, leaving a large proportion of the gypsum unaltered, is unlikely to have been unique in the stratigraphic record. It is predictable, therefore, that there should be anhydrite rocks amongst older, more deeply buried evaporites where nodules of syndepositional anhydrite float in a background of anhydrite that has a totally different structure and crystal fabric.

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