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The Formation of Micritic Limestones and the Development of Limestone-Marl Alternations in the Silurian of Gotland, Sweden

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KEYWORDS: LIMESTONE-MARL ALTERNATIONS – MICRITIC LIMESTONES – ARAGONITE – DIAGENESIS – MICRITE – MICROSPAR – GOTLAND – SILURIAN

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SUMMARY

Micritic limestone-marl alternations make up the major part of the Silurian strata on Gotland (Sweden). Their position on the stable Baltic Shield protected them from deep burial and tectonic stress and allowed the preservation of early stages of burial diagenesis, including lithification. In the micritic limestones certain characteristics have been preserved (e.g., pitted microspar crystals, sharp boundaries between microspar and components, lack of deformation phenomena) that offer insights into their formation. We suppose the formation of these micritic limestones and limestone-marl alternations to be based on a rhythmic diagenesis within an aragonite solution zone (ASZ) close below the sediment surface. The micritic limestones are the product of a poikilotopic cementation of carbonate muds which consisted of varying portions of aragonitic, calcitic and terrigenous matter. Their microspar crystals show the primary size and shape of the cements lithifying the original carbonate mud. Dissolution of aragonite in the marls provided the carbonate for the lithification of the

limestones. By cementation, the limestone beds evaded further compaction. The marls, which already underwent a volume decrease by aragonite depletion, lacked cement and became more and more compacted due to increasing sedimentary overburden. Although field observations show that primary differences in material influence the development of limestone-marl alternations they are not required for their formation.

1 INTRODUCTION

When investigating carbonate rocks, a basic differentiation between micritic and sparitic matrix is made (FOLK 1959, DUNHAM 1962). On a worldwide scale limestones with a fine-grained matrix and carbonate muds as their modern equivalents make up the largest part of carbonate rocks. Nevertheless, attention usually does not focus on the micritic matrix since on the one hand larger components are easier to assess, on the other hand many important questions regarding the formation and diagenesis of micritic limestones are not fully understood. Moreover, an actualistic approach to the question, why many micritic carbonates of ancient shallow water depositional environments occur as limestone-marl alternations is difficult because there are no modern analogues for such alternations.

1.1 Micrite

Research on fine-grained carbonates commenced with the work of SORBY (1879) who assumed them to be a product of the decay of calcareous skeletons of organisms into micrometer-sized components. Further investigations on fine-grained carbonates were hampered methodologically by the thickness of thin sections because everything smaller than 25-30 μm ('matrix') evaded examination. These rocks were called 'calcite mudstones', 'calclutites', 'Pelite', or 'Kalkpelite'. Still today, most publications dealing with fine-grained carbonates refer to the investigations of FOLK (1959,

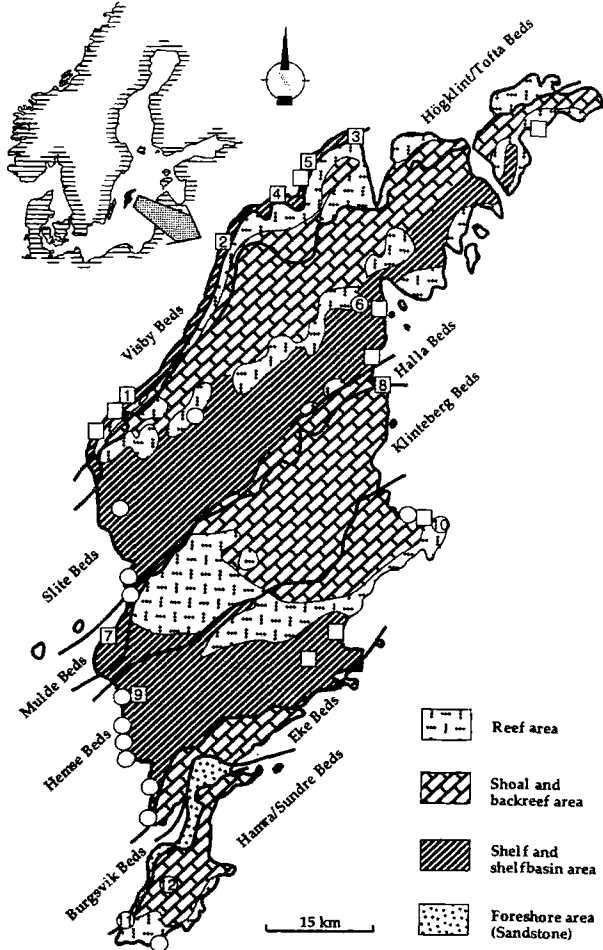


Fig. 1. Map of the Silurian of Gotland. Topostratigraphy and facies distribution modified after HEDE (1921, 1960), LAUFELD & JEPSSON (1976), WATTS (1981), SUNDQUIST (1982) and FRYKMAN (1989), and sample localities

(squares = detailed profiles, circles = additionally investigated sample series; numbered localities are mentioned in the text: 1 = North of Höglint, 2 = Rönklint, 3 = Hallshuk, 4 = Irevik, 5 = North of Häftingsklint, 6 = Klinteklnt west of Slite, 7 = Djauviks Fiskeläge, 8 = Gothemhammar, 9 = Snoder, 10 = Herrvik fishing harbour, 11 = Hoburgen, 12 = Northwest of Vamlingbo).

1965, 1974), who introduced the term 'micrite' for 'microcrystalline calcite'. FOLK distinguished 'micrite', having grain sizes around $2\mu\text{m}$ from 'microspar' with mean grain sizes between $5\text{--}15\mu\text{m}$. He presumed microspar to be formed out of micrite by aggrading neomorphism. The introduction of the scanning electron microscope (SEM) into carbonate sedimentology and petrology in the 1960's for the first time offered the opportunity to approach the fine fractions of modern and ancient carbonate sediments. Numerous investigations dealt with the problems of micrite formation (e.g., FOLK 1965, 1974, FISCHER et al. 1967, FLÜGEL 1967, HONJO 1969).

Investigations on Tertiary and Quaternary carbonates led to the assumption that the gap in grain size between micrite (about $2\mu\text{m}$) and microspar (about $5\text{--}15\mu\text{m}$) is caused by the mineralogical composition of the original

sediment (LASEMI & SANDBERG 1983, 1984, 1993): while originally aragonite-dominated sediments are altered to form microspar (ADP-'micrite'; aragonite-dominated precursor), calcite-dominated muds form real micrites (CDP-micrite; calcite-dominated precursor). ADP-'micrites' are characterized not only by their grain size (microspar) but also by a higher content in strontium and **typically pitted crystals**. Sometimes they show inclusions of primary aragonite needles in their original orientation. That shows that the transformation can proceed without breaking down the primary fabric (STEINEN 1978, 1982, LASEMI & SANDBERG 1984, 1993). Partly the engulfed needles are converted to calcite, partly dissolved, leaving empty pits in the calcite crystals. During further diagenesis these pits rapidly disappear. Primary aragonite in calcite as well as a pitted structure of microspar crystals are regarded as indications of a primary aragonitic composition. In sediments of pre-Tertiary age, however, usually there is no evidence of such hints (FISCHER et al. 1967).

The principal difficulties in reconstructing the formation of micritic limestones are summarized by MOSHIER (1989 p. 194): "Since lithification involves dramatic reorganization of material and pore space, the problem is to reconstruct how, when, where, and why lime muds become the products we see today."

1.2 Limestone-marl alternations

Limestone-marl alternations are common forms of carbonate rocks, including the entire spectrum from nodular alternations to well-bedded sequences (EINSELE et al. 1991). Limestones in these alternations normally show a symmetrical structure with highest carbonate content in the central part and no or only very low degrees of compaction, whereas marls display a significant reduction in thickness (KENT 1936, ILLIES 1949, SEIBOLD 1962, NOBLE & HOWELLS 1974, BATHURST 1980, WALTHER 1982, 1983, RICKEN 1986, 1992, MÖLLER & KVINGAN 1988). Lack of compaction phenomena in limestones points to an early lithification (KENT 1936, WELLER 1959, PRAY 1960, SEIBOLD 1962, HENNINGSMOEN 1974, NOBLE & HOWELLS 1974, JONES et al. 1979, EDER 1982, LASEMI et al. 1990).

It is tempting to regard limestones or marl beds in limestone-marl alternations as results of sedimentation events triggered by external processes like oscillations in orbital parameters (Milankovitch-cycles). Other causes could include irregularly varying sedimentation rates of carbonate or clay. The opinion that primary differences in sedimentary composition are necessary for the development of limestone-marl alternations is widely accepted by, e.g., WEPFER (1926), SEIBOLD (1952), GRÜNDEL & RÖSLER (1963), WEBER (1969), EINSELE (1982a,b), RICKEN (1986), EINSELE & RICKEN (1991), and ORTOLEVA et al. (1993). These authors presume that primary differences are enhanced by diagenesis.

Other researchers suppose that rhythmic limestone-marl alternations could as well be the product of diagenetical

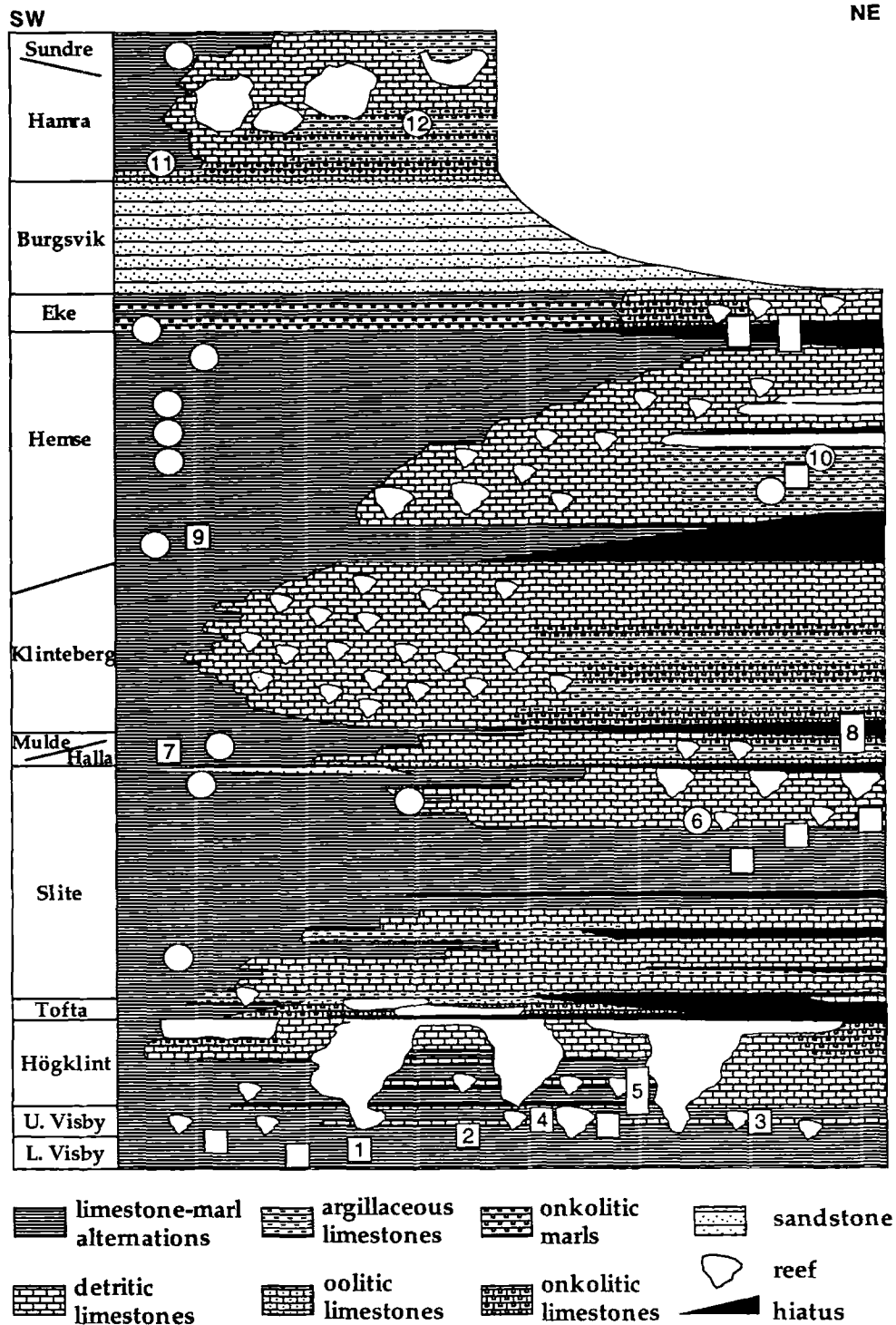


Fig. 2. Lithostratigraphic sequence modified after HEDE (1921, 1960), LAUFELD & JEPSSON (1976), WATTS (1981), SUNDQUIST (1982) and FRYKMAN (1989). Topostratigraphic frame after HEDE (1921); sample localities cp. figure 1.

alterations of relatively homogeneous sediments. Consequently, they argue that primary differences in material are not necessarily the cause for the alternations (SEMPER 1917, KENT 1936, ILLIES 1949, SUJKOWSKI 1958, HALLAM 1964, 1986, NOBLE & HOWELLS 1974, BYERS & STASKO 1978, JONES et al. 1979, EDER 1982, WALTHER 1982, 1983, BEIERSDORF & KNITTER 1986). These authors are of the opinion that sedimentary structures affect diagenesis but are not required for the formation of limestone-marl alternations.

1.3 Open questions

Many questions mentioned in previous publications, concerning the formation and diagenesis of micritic limestones and the development of limestone-marl alternations, are still not answered:

a) While modern carbonate muds on carbonate platforms and shelf areas of low latitudes mostly consist of aragonite and high-Mg-calcite (HMC), most ancient micritic

limestones are composed predominantly of low-Mg-cal-cite (LMC) and dolomite (e.g., FOLK 1965, BATHURST 1970). How does the transformation of the instable phases of the unconsolidated sediments into the stable phases of the solid rocks proceed? Comparison to diagenetic processes in young sediments is only of limited use, as the Pleistocene regressions led to exposure and hence to fresh water diagenesis of most of the young shallow water carbonates (STEHLI & HOWER 1961). However, fresh-water diagenesis can hardly be regarded as the normal case of carbonate-diagenesis (including micrite-diagenesis) (BATHURST 1993).

b) A large part of modern carbonate muds consists of debris of calcareous nannoplankton, especially in pelagic areas. It is supposed, however, that calcareous plankton developed during the Mesozoic (TAPPAN & LOEBLICH 1973). Which groups of organisms played a role in the accumulation of *Palaeozoic* carbonate muds (GARTNER 1977, CONIGLIO & JAMES 1985)?

c) Do limestone-marl alternations always result from primary rhythms in sedimentation, or could they develop from homogeneous sediments, too? If they are of sedimentary origin, it is still not clear *which* primary differences in sediment composition are responsible for their formation and why they have not been obscured by bioturbation (GINSBURG 1957, BATHURST 1987, 1991). If limestone-marl alternations are of diagenetical origin, which processes of separation led to their development (SEMPER 1917, ILLIES 1949, HALLAM 1964, WALTHER 1983)?

d) Although experiments have shown that clays and carbonate muds exhibit a similar behaviour when compacted (TERZAGHI 1940, EBHARDT 1968, SHINN et al. 1977) numerous micritic carbonates obviously have been lithified before compaction has affected them (WELLER 1959, PRAY 1960, ZANKL 1969, BATHURST 1970). What is the source of the carbonate required for cementation, how is it transported and precipitated (WELLER 1959, BATHURST 1970, 1975, FRIEDMAN 1975, RAISWELL 1988b, RICKEN & EDER 1991)? Solution of aragonite within the limestones would make carbonate available, but by far not enough to fill the primary pores (BATHURST 1970). Many authors consider interbedded marls, which are always reduced in thickness, to be the source area for cement carbonate (e.g., EDER 1982, WALTHER 1982, RICKEN 1986). But which diagenetic processes affect the redistribution? Pressure solution cannot be the only factor, because the limestones usually are lithified prior to the commencement of pressure solution in the marls (BATHURST 1980, 1987). And what is the role of biologically mediated carbonate precipitation during microbial sulfate reduction and methane production as a source for early cementation (RAISWELL 1988a)?

e) What are the diagenetic environments in which lithification takes place - the seafloor, shallow subsurface, deep burial, and/or under influence of fresh water (BATHURST 1975, 1993, FRIEDMAN 1975, SCHOLLE & HALLEY

Profile (m)	Gotland Stratigr. (-Beds)	Chronozones	Stages	Series	Age (m.y.)	
425	Sundre				411	
375	Hamra	Whitcliffian	Ludfordian	Ludlow	415	
	Burgsvik					
328	Eke	Leintwardinian	Gorstian			
314	Hemse					Bringewoodian
						Eltonian
224	Kilnteberg	Gleedonian	Homerian			424
160	Halla	Whitwellian	Wenlock	425		
145	Mulde			426		
	Site	Sheinwoodian				
55	Tofia					
47	Högkint					
12	Upper Visby	Telychian	Llandov.	430		
0	Lower Visby					

Fig. 3. Stratigraphic frame of the Silurian of Gotland after HEDE (1921), MARTINSSON (1967), LAUFELD & JEPSSON (1976), FRYKMAN (1989), and JEPSSON et al. (1994); ages according to HARLAND et al. (1990).

1985, HALLEY 1987, DIX & MULLINS 1988, MOSHIER 1989, RICKEN & EDER 1991)?

Purpose of our study is an attempt to answer these questions by macroscopical and microscopical analysis of structural phenomena in limestones and marls from the Silurian of Gotland.

2 GEOLOGIC SETTING

The Silurian of Gotland (upper Llandovery to upper Ludlow), Sweden, consists of about 450m of almost horizontally lying carbonates deposited in a tropical, epicontinental sea at the border between a carbonate platform complex and an open shelf (LAUFELD & BASSETT 1981). While on the west side of the island the whole series is developed as alternations of micritic limestones and marls (with the exception of the Burgsvik Sandstone), the eastern side is dominated by shallow water deposits, reefs, tidal and lagoonal sediments with numerous breaks in deposition (Fig. 1, 2). Terrestrial influx was low; indications of a nearby coastline are lacking.

A number of different limestone-marl alternations are observed: wavy bedded to nodular, marl-dominated series accumulated below wavebase in a shelfbasin, well bedded

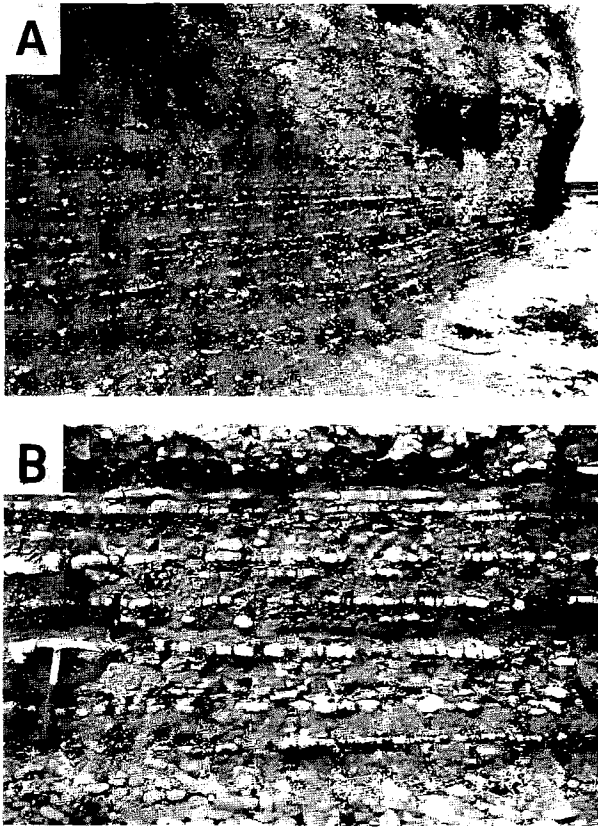


Fig. 4. A) Regular alternation of marl and nodular to wavy bedded micritic limestones, shelfbasin facies, Lower Visby Beds (upper Llandoverly) near Fridhem (north of Höglint; locality 1). B) Closer view to the upper part of the Lower Visby Beds (locality as A, hammer for scale).

Although the carbonates on Gotland are more than 400 million years old (Fig. 3), they are in an exceptionally good state of preservation because they have not experienced deep burial and strong tectonic stress due to their position on the stable Baltic Shield. Furthermore, they have not been subject to dolomitization. This favourable geological setting was the reason for our investigation of the facies-dependant formation and diagenesis of micritic limestones.

3 METHODS

At different stratigraphic levels and in various facies of the carbonate sequence, 21 profiles were mapped in a scale 1:5 and sampled in detail (Fig. 1, 2). One of them will be shown as an example in the following chapter. Additional sample series were taken from other locations (Fig. 1). More than 250 samples were polished, etched for 20 seconds with 0.1 M hydrochloric acid and investigated with the SEM. Chemical identifications of components were made with an EDX-analyser. Geochemical analysis (Sr, Mg, CaCO₃) have been carried out in order to determine the composition of limestones and marls in different facies as well as to determine the variations of carbonate content within single beds. But an interpretation of the development of limestone-marl alternations by means of geochemical analyses is difficult because the primary composition of the sediment is altered by diagenetical

limestone-marl alternations of inter-reef-facies, and shallow marine to lagoonal, wavy bedded, limestone-dominated series (Fig. 1).

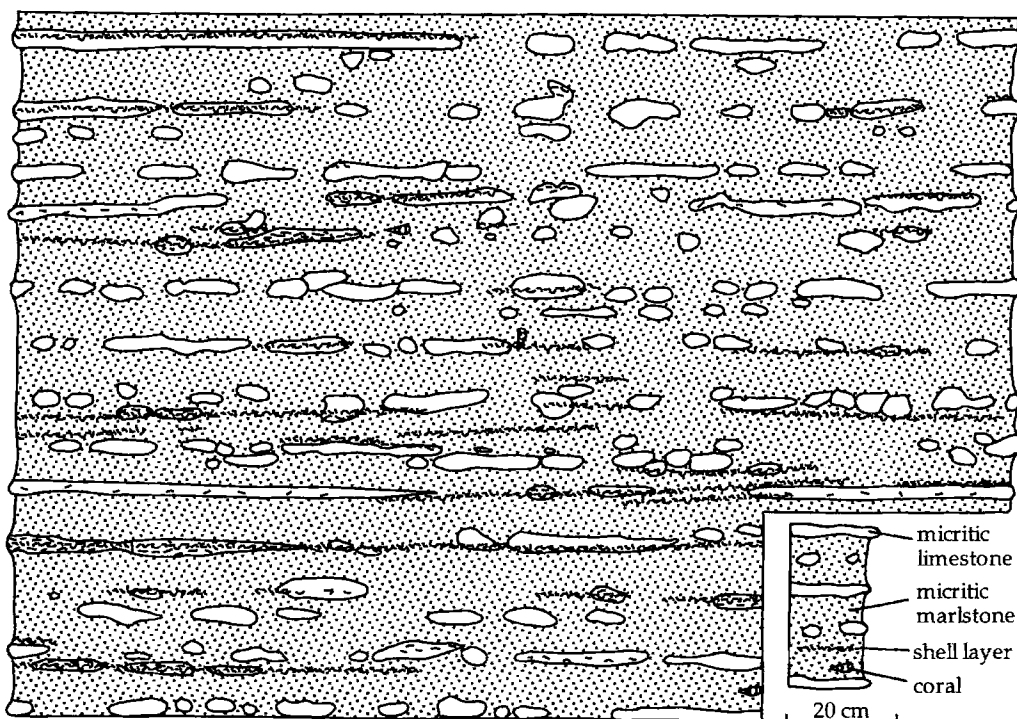


Fig. 5. Nodular to wavy bedded micritic limestone-marl alternation in the Lower Visby Beds (upper Llandoverly) near Fridhem (north of Höglint; number 1 in Fig. 1).

process. An analysis of faunal content is equivocal due to different diagenetic behaviour (solution and/or compaction) of fossils in limestones and marls (EINSELE & RICKEN 1991). Therefore, in this paper preponderantly observations regarding the macroscopic and microscopic structure of limestones and marls are presented. An attempt is made to investigate differences in sedimentary and diagenetical structures as well as the preservation of different components in micritic limestones and marls on Gotland. These observations are the basis for a hypothesis on the formation of micritic limestones and the development of limestone-marl alternations.

In the following, the terms 'limestone' and 'marl' do not imply specific ratios of clay to carbonate, which can be rather different in various series (Tab. 1). They refer to field observations, where beds that are more resistant to weathering are termed as 'limestones' and the intercalated softer ones as 'marls'.

4 OBSERVATIONS

4.1 Limestone-marl alternations

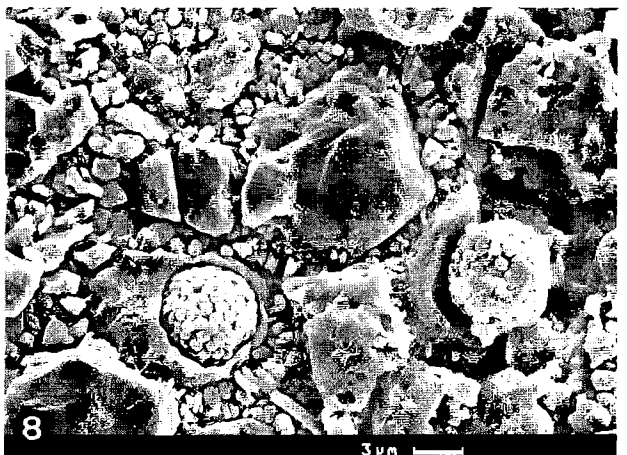
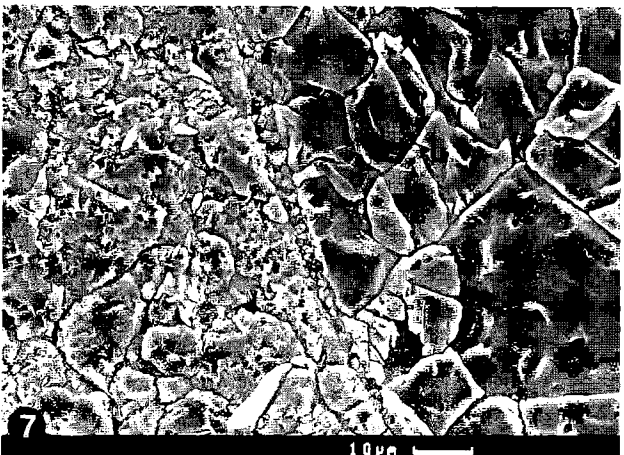
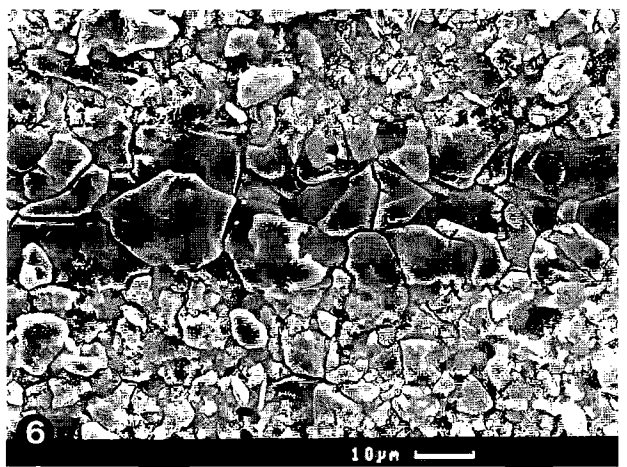
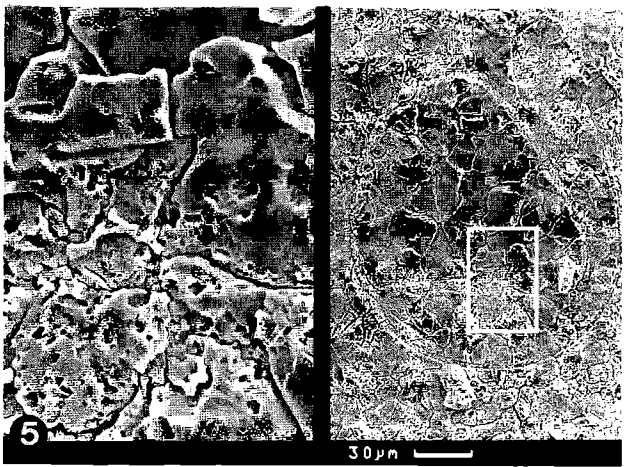
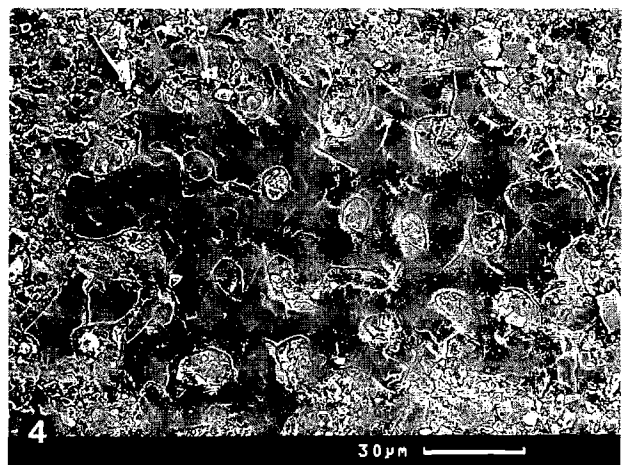
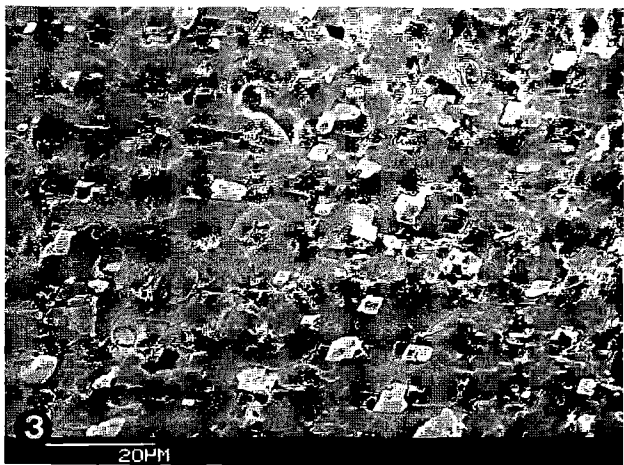
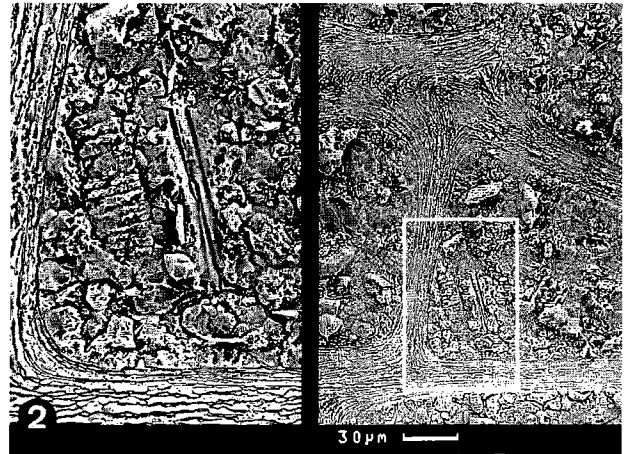
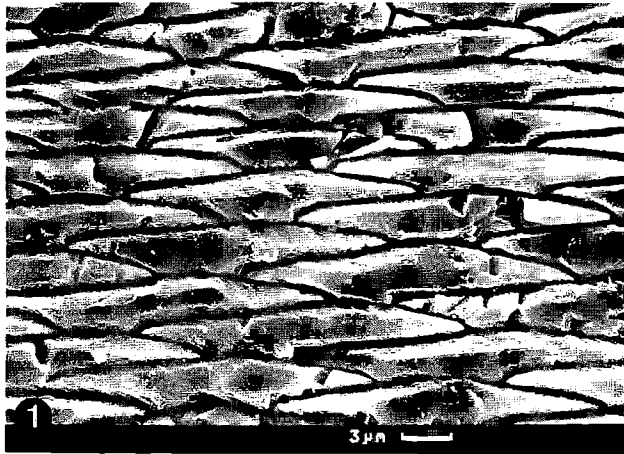
The typical characteristics in micritic limestones and marls (see below), leading to a hypothesis of their formation, are observed in every investigated sample series from Gotland (Fig. 1). Therefore, the results of our study are regarded as valid for limestone-marl alternations in the Silurian of Gotland, independent of their depositional environment or stratigraphic position.

The differences in limestone-marl alternations comprise carbonate content (in limestones and marls, Tab. 1), limestone/marl-ratio, bedding features, and the regularity of the alternating sequences. Regular limestone-marl alternations can be developed as well bedded series or as regularly arranged layers of micritic limestone nodules within marl sequences (Fig. 4), with intermediate stages between the extremes. Like in other sequences (e.g., SEIBOLD 1962, RICKEN 1986, BATHURST 1987), also on Gotland the carbonate content of limestone beds (or nodules) is highest in their central parts. The formation of limestone-marl alternations is influenced by primary sediment features: in regular alternations visible depositional structures are rare or missing due to intense bioturbation and/or lack of hydrodynamic sorting. Irregular alternations show structures and material differences which can be traced back to the primary sediment. If, for example, the wave base sometimes reached the bottom, the resulting alternation is much more irregular, with a preferential cementation of the washed, coarse sediments.

As an example one profile in the Lower Visby Beds (upper Llandovery) north of Högklint (locality 1) shows marls and nodular to wavy bedded micritic limestones alternating on a dm-scale (Fig. 5). Both types of rocks are predominantly mudstones with wacke- to packstones in some horizons due to irregularly intercalated thin lenses of brachiopod shells and bryozoans. The position of the shell layers is not directly correlated with the distribution of limestones and marls, because some of the layers are

Plate 42 Micritic limestones and the development of limestone-marl alternations in the Silurian of Gotland. SEM-Photomicrographs of limestones (localities cp. Fig. 1)

- Fig. 1. Ultrastructure of a brachiopod test with excellently preserved prisms (limestone, Upper Visby Beds, Rönnskint, locality 2).
- Fig 2. Bryozoan skeleton with sharp boundaries between skeleton and microspar. Note the two bioclasts embedded in microspar (scale bar indicates the magnification of the right picture; limestone, Lower Visby Beds, Rönnskint, locality 2).
- Fig. 3. Microdolomite inclusions within a recrystallized crinoid (limestone, Hamra/Sundre Beds, Hoburgen, locality 11).
- Fig. 4. Clast of a crinoid (limestone, Halla Beds, Gothemhammar, locality 8).
- Fig. 5. Geopetally filled ostracod with sharp boundaries of the test and a gradual transition between micritic infilling (now microspar with pitted crystals) and precipitated sparite, indicating that sparite and microspar are different products of the same lithification process (cp. Fig. 6b-c and compare with Pl. 42/6) (scale bar indicates the magnification of the right picture; limestone, Högklint Beds, Häftingskint, locality 5).
- Fig. 6. Primarily aragonitic bioclast replaced by neomorphic calcite which shows no traces of the original structure. This indicates that the original shell has been dissolved and refilled with calcite. The neomorphic crystals of the micritic matrix acted as templates for the growth of the mold filling cements, indicating that the refilling of the mold with calcite must have taken place *after* lithification, in contrast to void-filling sparite (see Pl. 42/5), which obviously precipitated *during* lithification (limestone, Högklint Beds, Irevik, locality 4).
- Fig. 7. Ostracod with sharp boundaries against microspar (left) and sparitic filling (right). Note that the precipitation of sparite starts with grainsizes similar to the microspar (limestone, Slite Beds, Klinteklint west of Slite, locality 6).
- Fig. 8. Framboidal pyrite and unpitted calcite within nannospheres (limestone, Högklint Beds, Häftingskint, locality 5).



completely within the marls, others completely in a limestone bed, sometimes crossing the boundaries between limestones and marls. Limestones containing coarse material (wacke-/packstones) are less nodular than pure mudstones. The thickness of the shell layers is reduced in the marl sections whereas they show no signs of compaction in the limestones. Sometimes a slight angle discordance is observed between the limestone-marl alternations and the interbedded shell layers (Fig. 5).

4.2 Limestones

Most constituents in micritic limestones from limestone-marl alternations on Gotland are exceptionally well preserved. Usually they are not recrystallized and show no or only little diagenetic interlocking with the surrounding matrix. The micritic limestones contain the following constituents:

a) **Primarily low-Mg-calcite components.** Debris of organisms with calcareous skeletons can be identified down to the submicroscopic scale. Especially debris of organisms with a skeleton of primary LMC, e.g., brachiopods, ostracods, trilobites, some bryozoans, clearly exhibit their primary ultrastructure (Pl. 42/1-2). A variety of microfossils down to few micrometers in size is observed in the micritic limestones (Pls. 42/8; 43/1, 3, 5). Spheres of 10 to 25 μm in size, which show tests with radially arranged crystallites, are very common (Pls. 42/8; 43/1). For these fossils we introduce the term 'nannospheres'. Analogous to macrofossils, microfossils with well preserved skeletal ultrastructures are supposed to consist originally of LMC.

b) **Primarily high-Mg-calcite components.** Components which consist primarily of HMC, like crinoids or rugosan corals, often show small inclusions of dolomite

(Pl. 42/3). Normally, crinoids are easily discernible by their structure (Pl. 42/4) while corals are generally slightly recrystallized.

c) **Primarily aragonitic components.** Bioclasts of originally aragonitic composition, like molluscs, are calcitized, without traces of the original structure; that is, they are dissolved and refilled with calcite (Pl. 42/6).

d) **Microspar (Folk 1959).** Microspar crystals form the dominant part of most of the fine-grained limestones. Their sizes range from 5 to 20 μm (in fillings up to 40 μm ; Pl. 44/5), they show a mosaic-like fabric, and they are conspicuously pitted (Pls. 42/2, 5, 7; 43/1, 7). On the other hand, void-filling calcite (e.g. in nannospheres) is unpitted (Pls. 42/7; 43/1, 3, 5). Boundaries between neomorphic crystals and components are distinct, whereas transitions to sparite are fluent. This phenomenon can clearly be observed in geopetal fillings (Pl. 42/5).

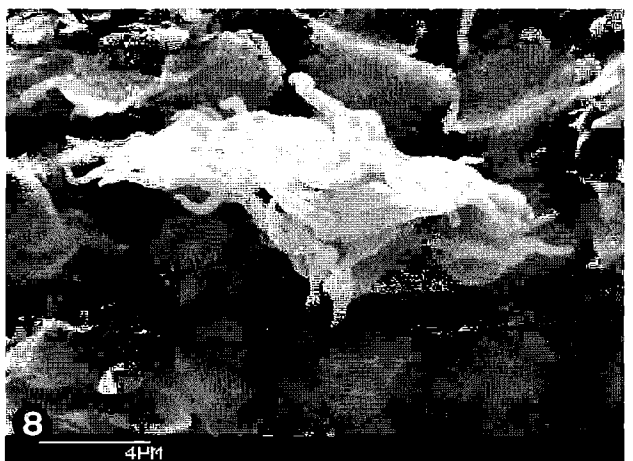
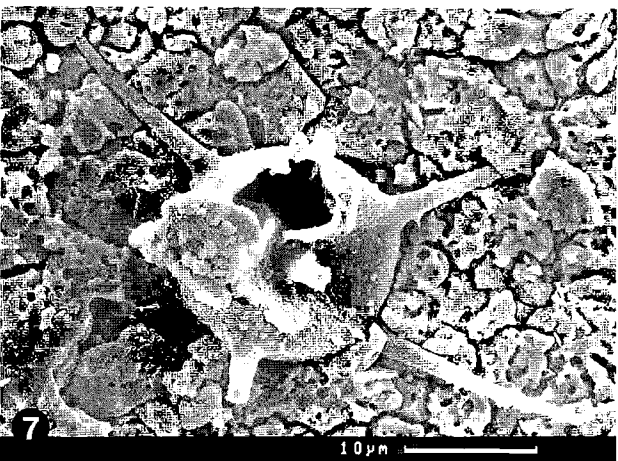
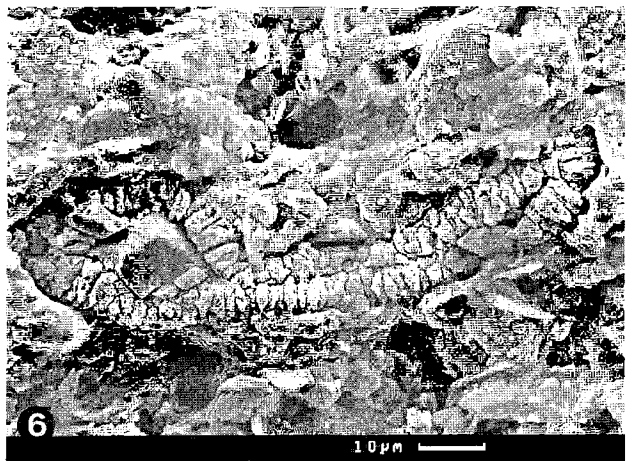
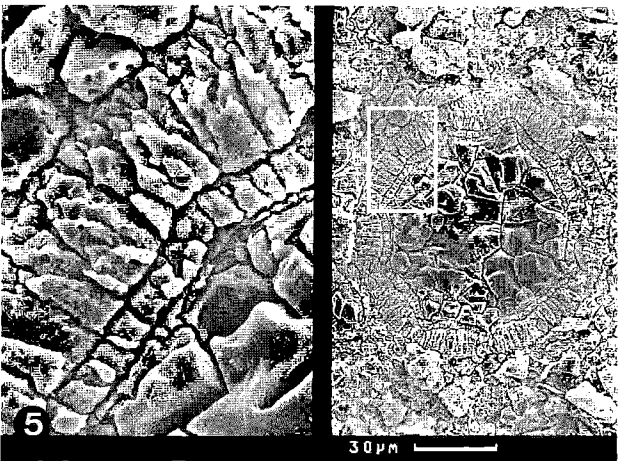
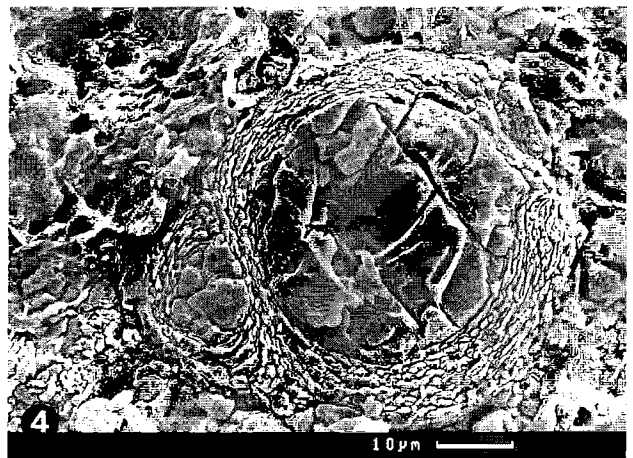
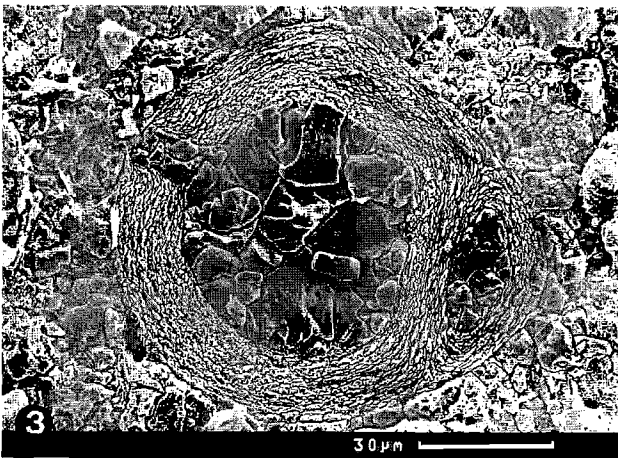
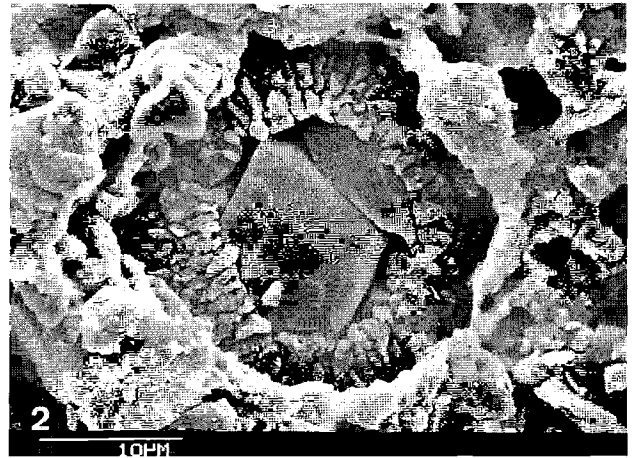
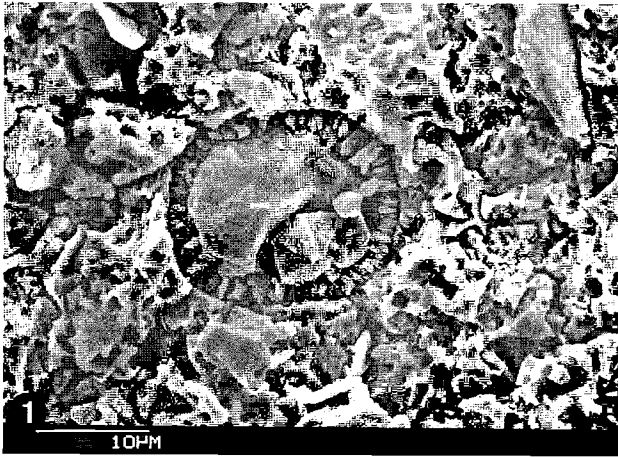
e) **Micrite (Folk 1959).** Often, a considerable portion of the micritic limestones consists of irregularly shaped calcite crystallites with an average size of 2 μm (micrite). These crystallites are normally distributed in patches and surrounded by microspar crystals (Pl. 44/3-4). Occasionally, their origin can be seen, for example, from the decay of nannospheres (Pl. 44/3).

f) **Other particles.** Other constituents of the micritic limestones are framboidal pyrite, dolomite, organic microfossils (e.g., acritarchs, chitinozoans), and terrigenous detritus, ranging in size from clay to silt. Organic microfossils show a very good preservation with no signs of compaction (Pl. 43/7; cf. MUNNECKE & SERVAIS subm.). The framboidal pyrite is located in primary pores, e.g., cavities in fossils (Pl. 42/8).

Plate 43

Micritic limestones and the development of limestone-marl alternations in the Silurian of Gotland. SEM-micrographs with typical preservation of components in limestones (left) and marls (right) (localities cp. Fig. 1).

- Fig. 1. Nannosphere with massive calcite crystals inside embedded in microspar with pitted crystal structures (cp. Fig. 6 c) (limestone, Mulde Beds, Djauviks Fiskeläge, locality 7).
- Fig. 2. Nannosphere with massive calcite crystals inside embedded in predominantly terrigenous material (marl, Högklint Beds, Irevik, locality 4).
- Fig. 3. Two-chambered microfossil with a bryozoan-like skeletal ultrastructure and massive calcite crystals inside (limestone, Högklint Beds, Häftingsklint, locality 5).
- Fig. 4. Two-chambered microfossil with massive calcite crystals within the test (marl, Högklint Beds, Irevik, locality 4).
- Fig. 5. Calcisphere with well preserved skeletal ultrastructure, sparitic filling (with increasing grainsizes towards the center) and sharp boundaries (scale bar indicates the magnification of the right picture; limestone, Högklint Beds, Häftingsklint, locality 5).
- Fig. 6. Compacted calcisphere with early (before compaction) precipitated calcite crystal inside (marl, Högklint Beds, Häftingsklint, locality 5).
- Fig. 7. 'Acanthomorphic' acritarch specimen (*Baltisphaeridium?*) showing no signs of compaction embedded in pitted microspar (limestone, Lower Visby Beds, Rönklint, locality 2).
- Fig. 8. Undeterminable specimen of an 'acanthomorphic' acritarch flattened by mechanical compaction embedded in predominantly terrigenous material (marl, Hemse Beds, Herrvik, locality 10).



4.3 Marls

In contrast to the limestones, marls within a limestone-marl alternation show a significant reduction in thickness which can be recognized by deformed ichnofossils. The marls contain the same constituents as the limestones, except for primarily aragonitic *bioclasts* and microspar (Tab. 1). Whole primarily aragonitic fossils, such as bivalves, are preserved as steinkerns. Organic microfossils with thin tests are flattened (Pl. 43/8, cp. MUNNECKE & SERVAIS subm.), while calcitic microfossils are often uncompact and filled with cement (Pl. 43/4). Even nannospheres can be found in the marls, with no traces of dissolution (Pl. 43/2). Although some calcitic microfossils show indications of mechanical compaction (Pls. 43/6; 44/1) signs of pressure solution between bioclasts are rare (Pl. 44/2). Cements occur exclusively as fillings in tests (Pl. 43/2, 4).

5 RESULTS AND DISCUSSION

5.1 Formation of micritic limestones

Most Silurian micritic limestones on Gotland consist predominantly of microspar. It has been known for long that the interlocking, mosaic-like fabric of microspar crystals cannot be of sedimentary origin, but must be a product of diagenesis (FOLK 1965). However, some observations in fine-grained limestones from Gotland exclude a formation by 'aggrading neomorphism' as proposed by FOLK (1959, 1965, 1974) to be the normal way of microspar formation:

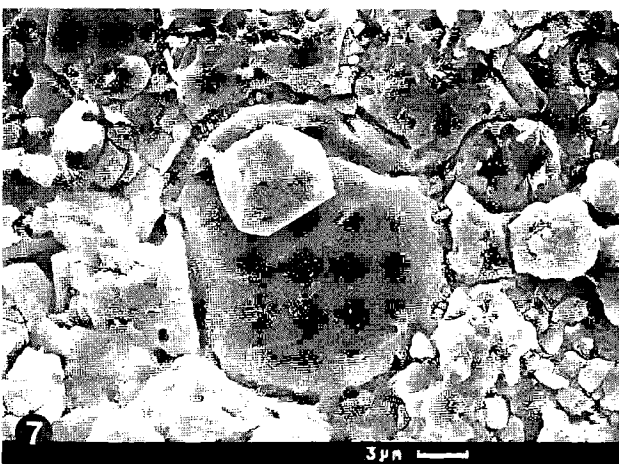
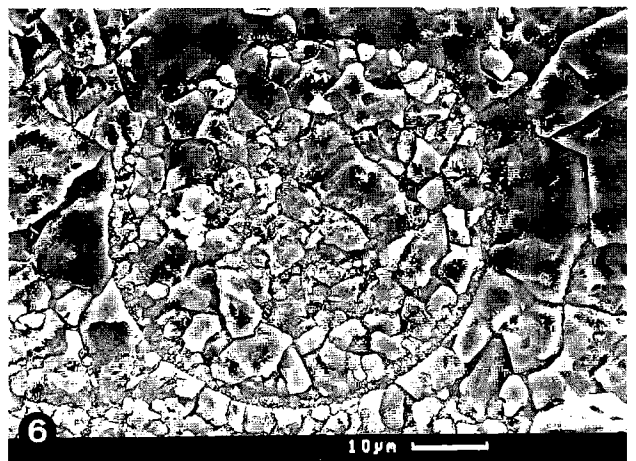
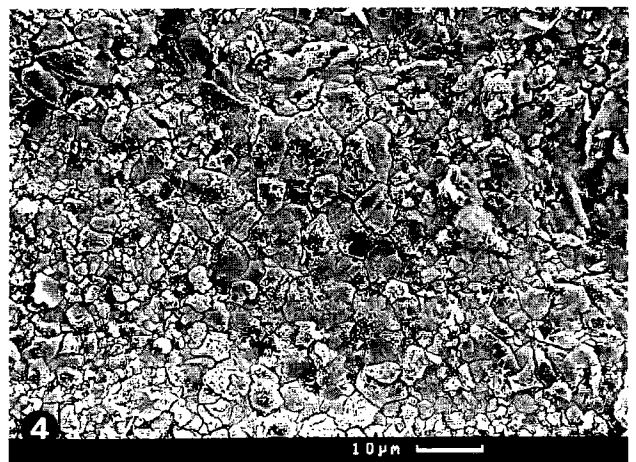
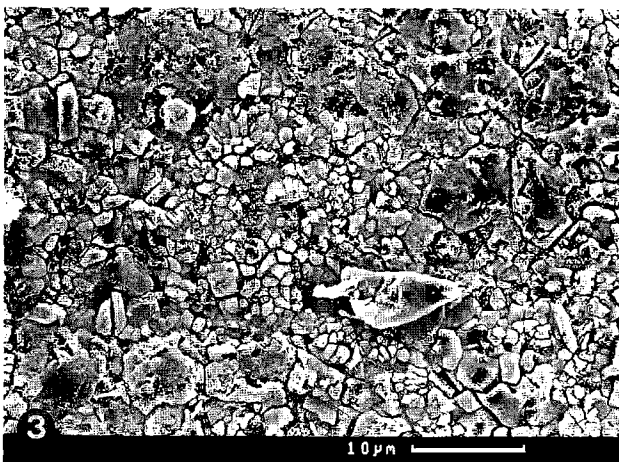
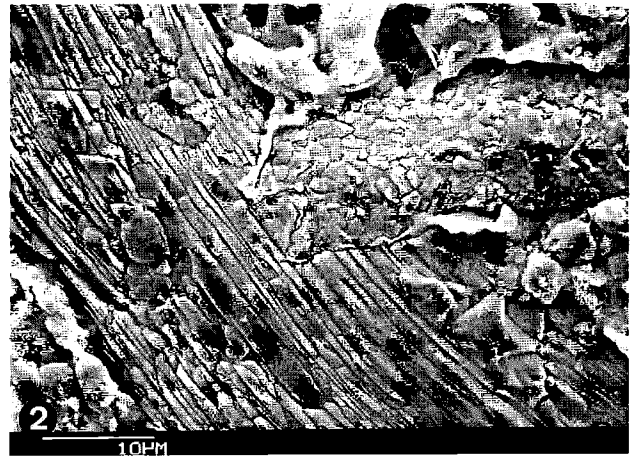
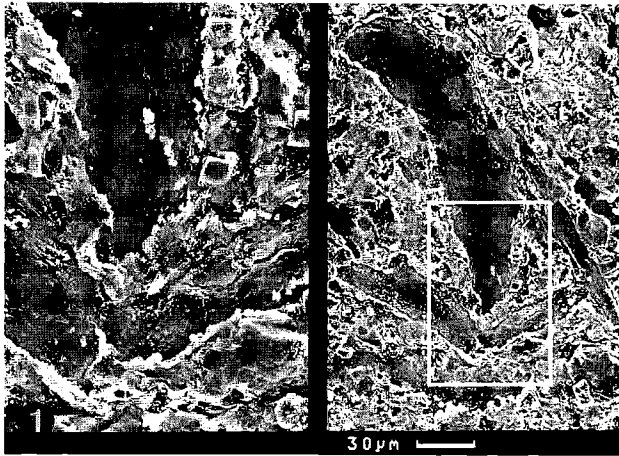
a) Most constituents of the micritic limestones have clear boundaries, even in the smallest fractions (Pl. 42/2, 5, 7; 43/1, 3, 5). Especially the boundaries between fossils or bioclasts and microspar are not in accordance with a recrystallization process of such profundity as FOLK's 'aggrading neomorphism'. Formation of neomorphic calcite crystals by this process should have led to interlocked, uneven boundaries between microspar and calcitic components.

b) Microspar as well as micrite can be found in almost every micritic limestone. It appears unlikely that only one part of the micrite crystals has grown to microspar, while the rest retained its original size (BATHURST 1970). Moreover, according to the theory of FOLK (1959, 1965, 1974), test-crystallites of originally calcitic organisms embedded in microspar should have grown as well because their size-ranges are at the same level as micrite.

These observations and the pitted structure of the microspar crystals suggest that the microspar precipitated as *cement* from a carbonate-supersaturated solution as it has been described for Pliocene/Pleistocene carbonates by STEINEN (1982) and LASEMI & SANDBERG (1984); the empty-pit-fabric (Pls. 42/2, 5, 7; 43/1, 7; 44/4-5) points to an aragonite-dominated composition of the primary sediment. Aragonite needles of only few micrometers size have been engulfed poikilotopically by neomorphic calcite crystals (microspar) during cementation of the soft sediment (Fig. 6). Calcitic and aragonitic components were embedded in the mosaic of neomorphic calcites, maintaining a sharp contact (Fig. 6). Within small voids, where no aragonite needles were present (e.g., in nannospheres), the precipitated microspar crystals are massive (Pls. 42/8; 43/1, 3, 5; Fig. 6). The cementation occurred after the formation of framboidal pyrite, because the pyrite has been precipitated in primary voids which did not exist after cementation (at least the small ones, Pl. 42/8). The engulfed aragonite needles in the microspar crystals were dissolved later, leaving pit-like structures (Pls. 42/2, 5, 7; 43/1, 7; Fig. 6). Larger aragonitic components were dissolved, too; the empty moulds, however, have been refilled with calcite (Pl. 42/6), in contrast to the voids within the neomorphic calcite crystals. Calcitic components, especially those consisting primarily of LMC, retained more or less their original skeletal ultrastructure (Pls. 42/1, 2, 5, 7; 43/3, 5; Fig. 6). Consequently, the micritic limestones are the product of a poikilotopic cementation of carbonate muds consisting of calcitic, aragonitic, and terrigenous matter in varying fractions.

Plate 44 Micritic limestones and the development of limestone-marl alternations in the Silurian of Gotland. SEM-micrographs of limestones and marls (localities cp. Fig. 1)

- Fig. 1. Breakage of a bioclast due to mechanical compaction (scale bar indicates the magnification of the right picture; marl, Halla Beds, Gothemhammar, locality 8).
- Fig. 2. Pressure solution contact between two calcitic bioclasts (marl, Upper Visby Beds, Hallshuk, locality 3).
- Fig. 3. Micrite (resulting from the decay of nannospheres) and microspar (limestone, Klinteberg Beds, Gothemhammar, locality 8).
- Fig. 4. Micrite and pitted microspar. In contrast to Pl. 44/3 the origin of the micrite cannot be determined (limestone, Klinteberg Beds, Gothemhammar, locality 8).
- Fig. 5. Coarse microspar with pitted crystal structures (limestone, filling of an ostracod test, Högklint Beds, Irevik, locality 4).
- Fig. 6. Calcisphere with partly disappeared test due to growth of larger adjacent crystals (cp. Fig. 6d) (limestone, Hemse Beds, Snoder, locality 9).
- Figs. 7. - 8. Two examples of nannospheres with nearly disappeared tests due to growth of surrounding crystals (cp. Fig. 6d) (Fig. 7: limestone, Högklint Beds, Häftingsklint, locality 5; Fig. 8: limestone, Hamra/Sundre Beds, Vamlingbo, locality 12).



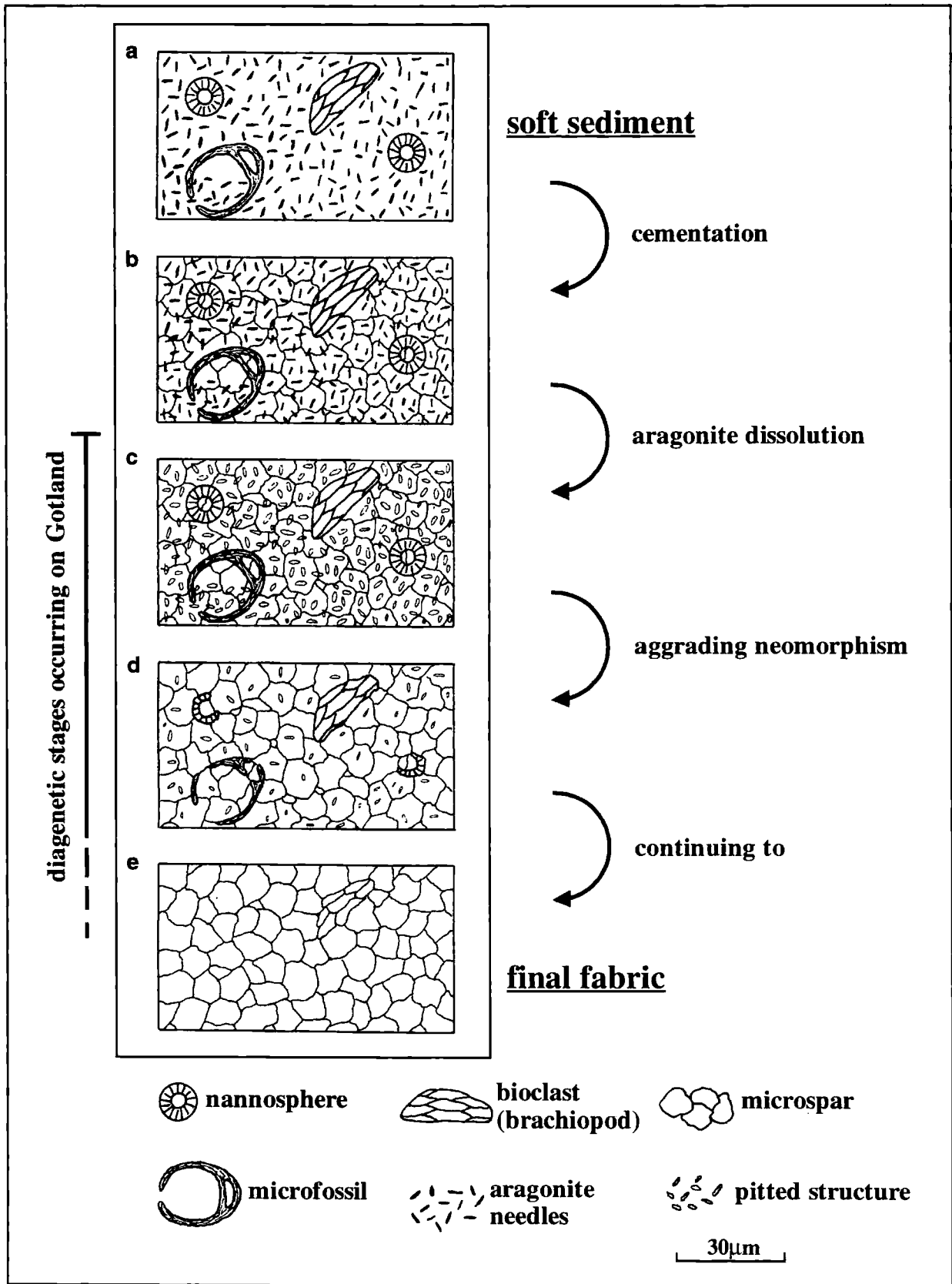


Fig. 6. Diagenetical processes during burial and limestone lithification; based on observations in the Silurian of Gotland ('cementation' as described from Pliocene/Pleistocene micritic carbonates of Florida and the Bahamas by STEINEN (1982) and LASEMI & SANDBERG (1984)). Note that size and shape of microspar crystals are largely determined in the first stage of diagenesis (cementation).

	LIMESTONES	MARLS
primarily LMC-components	X	X
primarily HMC-components	X	X
primarily aragonitic components	X	(only Steinkerns)
organic microfossils	X (uncompacted)	X (compacted)
ichnofossils	X (uncompacted)	X (compacted)
micrite	X	X
microspar	X	
sparite	X (as matrix in grainstones and as filling in fossils)	X (within fossils)
terrigenous matter	X	X
dolomite	X	X
pyrite	X	X
compaction phenomena		X
pressure solution		X (rare)
carbonate content (%) all samples (n=476) example Lower Visby Beds (n=63) example Högklint Beds (n=168)	62 - 97 (Ø 78) 62 - 91 (Ø 72) 64 - 95 (Ø 78)	14 - 74 (Ø 39) 16 - 25 (Ø 22) 37 - 74 (Ø 51)

Tab. 1. Compositions and diagenetic characteristics of limestones and marls in the Silurian of Gotland.

This means that the original carbonate muds on Gotland did *not* first alter to micrite and later to microspar by aggrading neomorphism. The formation of microspar crystals obviously is a one-step process: they show the **primary grain size and shape of the cements** lithifying the original aragonite-dominated carbonate mud. The 'SANDBERG-curve' (SANDBERG 1983), distinguishing between calcitic and aragonitic episodes in the Phanerozoic (using results of investigations on ooids and cements), does not seem to be applicable to the mineralogical composition of carbonate *muds*. According to the 'SANDBERG-curve' the Silurian lies near the Paleozoic calcite maximum.

During subsequent stages of diagenesis the micrite crystals, fragile microfossils, nanofossils, as well as thin shells recrystallize into larger crystals by slight aggrading neomorphism of the larger microspar crystals (Pl. 44/6-8; Fig. 6). This neomorphism blurs the clues to primary structures and early diagenetic processes. Due to modest overburden pressure and absence of tectonic stress, the limestone diagenesis on Gotland stopped shortly after lithification, so the early stages of diagenesis are preserved (Fig. 6).

5.2 Development of limestone-marl alternations

What happens to the the marls during the lithification of the limestones? On the one hand they show a significant reduction in thickness but on the other hand almost no signs of pressure solution (Tab. 1). Also, the calcitic components down to the size of nanospheres lack disso-

lution (Pl. 43/2). Cements do not occur in the marls with the exception of fillings in some fossils. Because no primary aragonitic bioclasts, for example molluscs, are found in the marls (Tab. 1), it is assumed, that they represent those parts of the original sediment in which aragonite has been removed. The pitted structure of the microspar crystals of the *limestones* points to a primary aragonite-dominated mud. It is presumed that the precursor sediment of the *marls* consisted to a large degree of aragonite, too, because sedimentary structures sometimes cross the boundaries between limestones and marls (Fig. 5), indicating that the same sediment can alter to limestone as well as to marl. It is likely that solution of aragonite in the marls made carbonate available for the cementation of the limestones.

Generally, HMC with more than 12% mol Mg-content is more soluble than aragonite (WALTER 1985). However, as observed in younger sediments (Tertiary-Quaternary), *aragonite* is the most unstable calcium carbonate mineral in diagenesis because HMC-components usually release their magnesium prior to the solution of aragonite, thus decreasing their solubility (FRIEDMAN 1964, WINLAND 1968, TOWE & HEMLEBEN 1976). Because on Gotland components which are composed primarily of HMC (e.g., crinoids) are found in limestones and marls (Tab. 1), it is assumed that *aragonite* was the only carbonate mineral involved in the processes of early diagenetic carbonate redistribution.

Field observations on Gotland show that the same precursor sediment can alter to limestone or marl (see above). Although obviously coarser layers are preferentially cemented it cannot be supposed that diagenesis principally enhanced primary differences of sediment

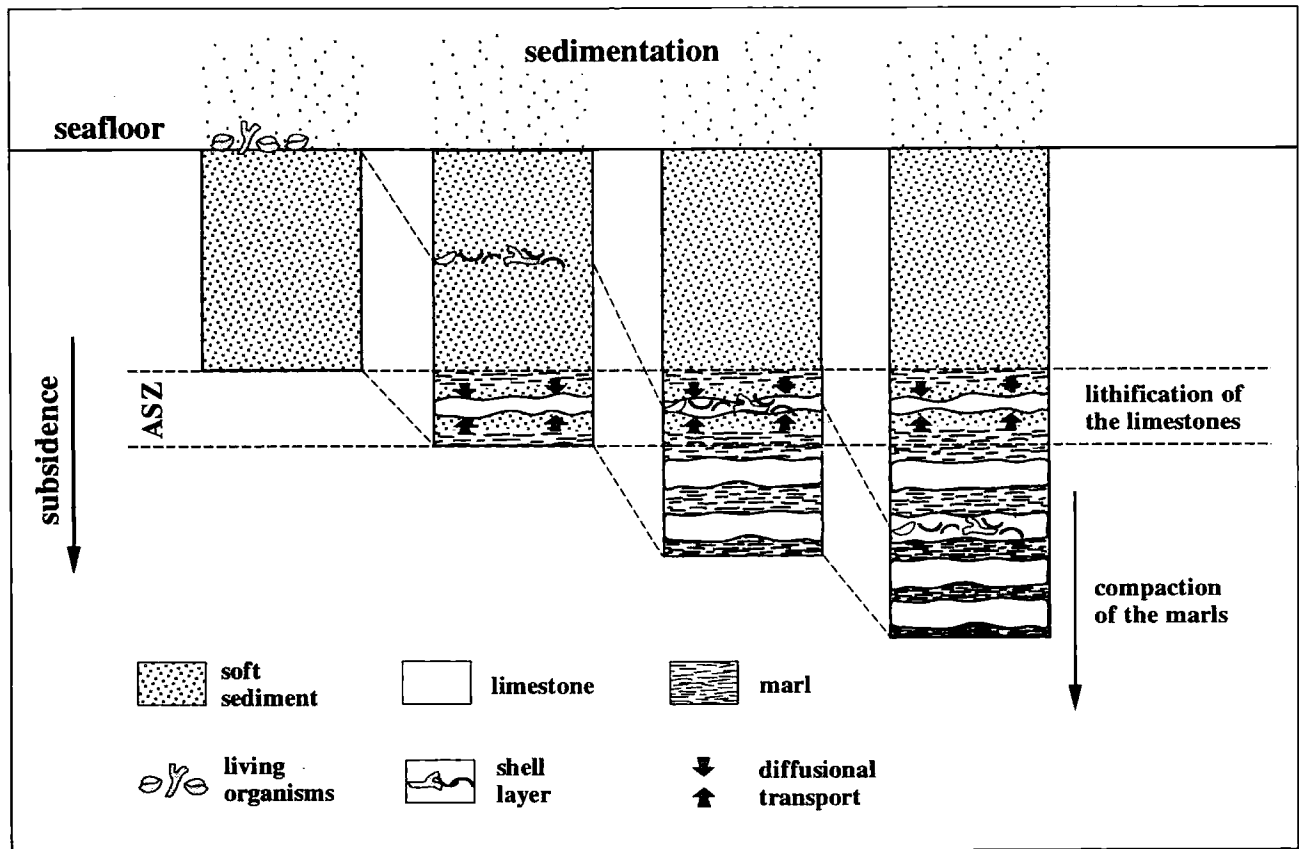


Fig. 7. Development of rhythmic limestone-marl alternations caused by aragonite dissolution and calcite precipitation within the ASZ (aragonite solution zone) and further compaction of the marlstones due to continuous sedimentary overburden. The dissolved carbonate is transported by diffusion. Occasionally, regularly arranged layers of limestone nodules replace well bedded limestones.

material because not every shell layer is cemented (Fig. 5). In addition, the slight angle discordance between shell layer and limestone beds (Fig. 5) can only be explained by a diagenetic origin of the alternation. Obviously, the diagenetical redistribution of carbonate is the dominant process in the development of limestone-marl alternations. This process is *only influenced* by primary differences in material.

Based on our observations, we propose the following hypothesis for the development of limestone-marl alternations in the Silurian of Gotland: in the original soft muds vertical changes in pore water chemistry, mainly produced by microbial decomposition of organic material, are presumed, resulting in vertical geochemical gradients. Decreasing carbonate saturation within the sediment column caused aragonite to be unstable and to be dissolved at a certain depth, producing a dissolution-zone roughly parallel to the seafloor, in the following called 'ASZ' (aragonite solution zone). Within this zone aragonite was dissolved, transported, and precipitated as calcite (microspar) in regions where aragonite was still stable (Fig. 7). As the amount of pore water is by far not large enough for the transportation of the dissolved carbonate by pore water flow (ENOS & SAWATSKY 1981), the transport mechanism must be a diffusion process, as it is assumed for Lower Jurassic deposits in Great Britain by RAISWELL (1988b). Because of the symmetrical structure of limestone beds or

nodules with highest carbonate contents in their central parts, the transport of the carbonate ions by diffusion must have been in upward as well as in downward direction (Fig. 7). The parts of the sediment depleted in aragonite became marls, those with precipitation of calcite were formed to limestones. Although primary differences in sediment composition (e.g., carbonate content, bioclasts, organic matter, grain size, porosity) can serve as centers of cementation, they are not essential for the development of limestone-marl alternations: the processes of aragonite-dissolution and calcite-precipitation must have been effective in homogeneous sediments, too, because the maximal distance of a diffusional transport is limited by geochemical gradients (RAISWELL 1987, 1988b). Our assumption is that the regularity of the resulting limestone-marl alternation is substantially influenced by the homogeneity of the primary sediment: homogeneous sediments led to regular alternations. Notwithstanding the fact that in numerous shallow marine sequences on Gotland irregularly distributed sediment materials are common which strongly influenced the diagenetical processes and resulted in more irregular limestone-marl alternations. By cementation, the limestone beds evaded further compaction; the dissolution of their aragonitic components presumably took place in a later stage, this caused the empty-pit-fabric of the microspar crystals. The diagenesis of the micritic limestones was nearly completed just below the ASZ. The marls, on the

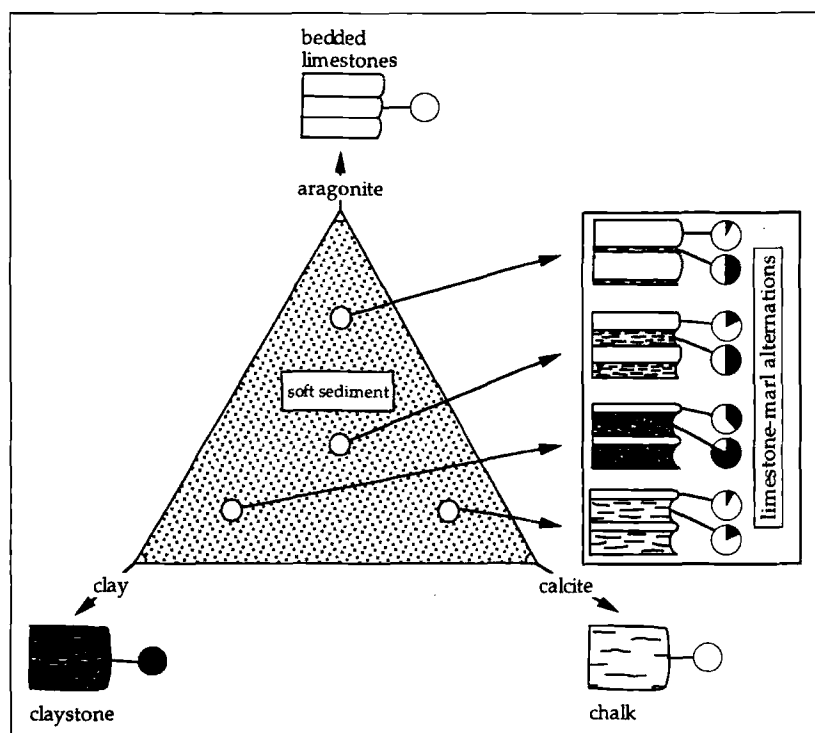


Fig. 8. Schematic presentation of the transformation of homogeneous soft sediments (porosity about 50%) with different portions of aragonite, calcite, and clay into alternating sequences of solid carbonate rocks. The carbonate content of the rocks is outlined in circles (white = lime, black = clay/terrigenous material).

other hand, which already underwent a volume decrease by aragonite depletion, lacked cement and became more and more compacted due to increasing sedimentary overburden (Fig. 7). They represent a compacted residual sediment depleted in aragonite. While the *relative* thicknesses of alternating limestone and marl beds depend mostly on the primary aragonite content (see below), the *absolute* thicknesses probably are influenced or even determined by the geochemical gradients.

The nature of the presumed geochemical gradients is not clear. In recent sediments the strongest changes in pore water chemistry are observed within the uppermost 2 m of the sedimentary column. A sudden increase in carbonate saturation is found in the zone of anaerobic methane oxidation (CANFIELD & RAISWELL 1991). Above and below this zone carbonate saturation of pore water is considerably lower, sometimes it is even undersaturated with respect to aragonite (CANFIELD & RAISWELL 1991). However, an explanation of fossil limestone-marl alternations by comparison with recent geochemical pore water processes is questionable because there are no modern analogues for such alternations. Nevertheless, rhythmic precipitations produced at a moving reaction front (Liesegang band formation) are common phenomena in recent geochemical processes (DEE 1986, JACOB et al. 1994, ORTOLEVA 1994).

The question concerning the sediment depth of the ASZ arises. Our observations point to a position just below the seafloor:

a) Easy to compact organic microfossils, such as acritarchs, generally are flattened in the marls (Pl. 43/8), whereas they are excellently preserved in limestones (Pl. 43/7, cf. MUNNECKE & SERVAIS *subm.*). If lithification of the limestones would have taken place several hundred meters

below the seafloor their organic microfossils should show signs of compaction.

b) Fragile calcitic components in the limestones (e.g., thin-branched bryozoans) do not exhibit any signs of bending, breaking, or pressure solution, likewise pointing to an early lithification.

c) Micritic nodules overgrown by stromatoporoids, which are observed occasionally, and numerous hardgrounds in the Silurian of Gotland indicate exposition of the lithification front to seawater. Also in similar sequences of other regions incrustation of micritic nodules by sessile organisms or hardgrounds are observed and taken as indication of an exhumation of early lithified sediment (VOIGT 1968, KENNEDY & KLINGER 1972, BAIRD 1976, JONES et al. 1979, LINDSTRÖM 1979, MÖLLER & KVINGAN 1988). This could hardly be possible with a deep positioning of the lithification front.

Our observations indicate that the aragonite content of the soft sediment is the most important factor for the development of limestone-marl alternations. The more aragonite is available in the precursor sediment the higher must be the amount of limestone related to marl in the resulting alternation (Fig. 8). The carbonate content of the *marl* is determined by the ratio of calcitic to terrigenous material in the primary mud because aragonite is completely removed. Consequently, pure aragonitic muds alter to bedded limestones because those parts of the primary sediment in which aragonite dissolution took place are entirely dissolved, leaving a bedding plane (Fig. 8). The carbonate content of the *limestones* depends as well on the amount of carbonate minerals in the soft sediment as on its porosity (Fig. 8).

6 CONCLUSIONS

The Silurian carbonates on Gotland represent an extraordinary case among Palaeozoic carbonates because of their exceptionally good state of preservation. 'Extraordinary case' does not refer to sedimentary facies, but to the diagenesis that came to an end shortly after early lithification. Early diagenetic characteristics well preserved on Gotland have been altered by later phases of diagenesis in most carbonates of other regions, even if they are considerably younger. Our observations allow interpretations concerning the formation of micritic limestones and the development of limestone-marl alternations:

a) Micritic limestones originated as carbonate mud consisting of varying portions of calcitic, aragonitic and terrigenous material. The calcitic part consists of microspar, micrite, bioclasts, microfossils and spheroidal nanno-organisms ('nannospheres'), presumably tests of planktonic algae. Disintegrated nannospheres obviously produced a major part of the fine-grained material (micrite). The pitted crystals of microspar are inferred to be cemented aragonite needle mud; they show the primary size and shape of the cements, which poikilotopically lithified the original mud.

b) The differentiation of aragonite-containing sediments into limestones and marls occurred during early burial diagenesis, that is, without heavy overburden. The most important factor is not *pressure* solution but *aragonite* dissolution and calcite precipitation within a diagenetical zone close below the seafloor. These early diagenetical processes result in more or less regular limestone-marl alternations. The higher the content of aragonite in the primary sediment, the higher is the limestone/marl-ratio in the resulting limestone-marl alternation.

c) Regular limestone-marl alternations can result from relatively homogeneous carbonate muds. Primary depositional differences are not necessary for their development, but have an influence on the formation of the sequences. At least for a part of the limestone-marl alternations on Gotland a diagenetic origin is concluded, although in most alternations a positive proof is hardly possible because of the parallelism of sedimentary and diagenetic structures.

d) Although the question about the source of the aragonitic needles - organic or inorganic - cannot be answered yet, it is concluded that in the Silurian aragonite-containing muds played an important role in formation and diagenesis of marine carbonates. The 'SANDBERG-CURVE' (SANDBERG 1983) does not seem to be applicable to mineralogical composition of carbonate muds. According to the 'SANDBERG-CURVE' the Silurian lies near the Palaeozoic calcite maximum.

Although the processes of aragonite dissolution and calcite precipitation seem to be general in early diagenesis of carbonate muds, we do not conclude that limestone-

marl alternations are generally produced solely by diagenesis. But one should be able to exclude the possibility of a diagenetic formation before interpreting such sequences as results of externally controlled sedimentary processes and claim, for example, climatic (Milankovitch-) cycles as possible causes.

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