MISSING ARAGONITIC BIOTA AND THE DIAGENETIC EVOLUTION OF HETEROZOAN CARBONATES: A CASE STUDY FROM THE OLIGO-MIOCENE OF THE CENTRAL MEDITERRANEAN

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ABSTRACT: Several previous studies have addressed the diagenetic evolution of heterozoan carbonate assemblages. Generally it is assumed that early diagenetic processes in heterozoan settings are mainly destructive, including abrasion and dissolution on the sea floor. Constructive diagenesis (cementation) is delayed to later stages in the burial environment, with pressure solution of calcitic grains acting as a cement source. This paper presents a study of Oligo-Miocene inner- to outer-ramp heterozoan carbonates from the Central Mediterranean (Maltese Islands and Sicily) indicating that early diagenetic processes are more important than previously assumed.

Four to five different cement types, including fibrous, two types of epitaxial, bladed and blocky cement, are distinguished based on transmitted light microscopy. Cathodoluminescence microscopy allowed a differentiation between primary high-Mg calcitic (fibrous and epitaxial cement I) and primary low-Mg calcitic (epitaxial cement II, bladed and blocky) cements. Stable-isotope data indicate cement precipitation from marine, marine-derived, and meteoric waters. Trace-element analyses point to cementation in an open system (Maltese Islands) and a closed system (Sicily).

Our investigations show that the majority of constructive diagenetic processes in these rocks occurs rather early in the shallow, marine burial environment, which is transitional between the marine seafloor and the deep-burial diagenetic environment. The main cement source in this environment is assumed to be aragonite. We suggest careful consideration of the importance of aragonitic components in fossil heterozoan settings, which seem to be more abundant than previously assumed and can act as a major early cement source. Due to the low preservation potential of these components, detailed geochemical studies are necessary to detect aragonite as the cement source. Our findings also have implications when considering the reservoir qualities of these rocks, because primary porosity can be occluded early and secondary porosity is not preserved.

INTRODUCTION

Numerous studies have dealt with the diagenetic evolution of photozoan carbonates (*sensu* James 1997) deposited in shallow-water environments. Photozoan carbonates consist predominantly of light-dependent biota such as hermatypic corals, calcareous green algae, larger benthic foraminifers, and marine precipitates such as ooids. The occurrence of this assemblage is controlled mainly by warm sea-water temperature ($> 22°C$) and low nutrient content (James 1997; Mutti and Hallock 2003). In these settings, the importance of early seafloor and meteoric processes in modifying and stabilizing the sediment is undoubted (James and Choquette 1990a, 1990b; Dodd and Nelson 1998). Inorganic cement precipitation as aragonite or high-Mg calcite (HMC, > 4 mol% $MgCO_3$) on the seafloor occurs commonly as the result of the high carbonate saturation state in equatorial regions (Opdyke and Wilkinson 1990). Moreover, typical photozoan carbonate sediments consist mainly of metastable aragonitic and HMC skeletons, giving the sediment a high early diagenetic potential. When entering the meteoric environment these components are dissolved or neomorphosed in solutions undersaturated with respect to aragonite and HMC, but supersaturated with respect to low-Mg calcite (LMC, < 4 mol% $MgCO_3$). This leads, on the one hand, to the development of secondary moldic porosity and, on the other hand, to extensive cementation by LMC. When entering the burial environment these rocks are mineralogically stable and well lithified, have lost much of

their primary porosity, but also have gained a high amount of secondary porosity.

The diagenetic evolution of heterozoan carbonates (*sensu* James 1997) has recently become of interest (e.g., James and Bone 1989; Hood and Nelson 1996; Nicolaides and Wallace 1997b; Brachert and Dullo 2000; Haywick 2004). The heterozoan association, in contrast to the photozoan, consists mainly of light-independent biota such as mollusks, bryozoans, and echinoderms, with only minor amounts of light-dependent taxa such as calcareous red algae. This association develops in cooler waters ($< 22°C$) and/or at elevated nutrient contents in shallow-water environments, or at greater water depths below the photic zone as the deeper-water continuation of the photozoan association. Generally it has been assumed that the potential for constructive early diagenetic processes in heterozoan carbonates is low. Instead, early marine diagenesis is thought to be mainly destructive, including abrasion, bioerosion, maceration, and dissolution in cool seawaters with a low $CaCO_3$ saturation state (Smith and Nelson 2003). This low saturation state is also thought to be the cause of the scarcity of marine cements in heterozoan carbonates, inasmuch as it is unfavorable for inorganic cement precipitation (Morse and Mackenzie 1990). Also in the meteoric environment, the diagenetic potential of heterozoan carbonates is limited because of their common LMC-dominated primary mineralogy (Nelson et al. 1988b), and only minor dissolution of metastable biota and precipitation of stable cements

takes place. Hence, when entering the burial environment, much of the primary porosity of heterozoan carbonates is preserved and little secondary porosity develops. Not until the stage of burial diagenesis does the main phase of cementation of heterozoan carbonates take place—with pressure solution of LMC grains providing a principal cement source (Nelson et al. 1988a; Nicolaides and Wallace 1997a; Dodd and Nelson 1998).

When dealing with carbonate diagenesis the amount of aragonite in the sediment is important because it strongly determines its diagenetic potential. Heterozoan carbonate assemblages are generally described as being aragonite-poor (James and Bone 1989; Smith and Nelson 1994; Nicolaides 1995; James et al. 1999), even if several examples of moderately to very aragonite-rich heterozoan sediments and rocks are reported (Nelson et al. 1982; Nelson and Hancock 1984; James et al. 2005). The generally assumed scarcity of aragonite might not represent the primary sediment composition, inasmuch as several authors describe that aragonite in these settings is removed early from the sediment record by dissolution, often without leaving a trace (Brachert and Dullo 2000; Cherns and Wright 2000; Smith and Nelson 2003; Wright et al. 2003; Wright and Cherns 2004). This potentially leads to a misinterpretation of the primary biotic assemblage and the sedimentation rate and an underestimation of the importance of aragonite as an early diagenetic cement source.

This paper deals with the diagenetic evolution recorded in Oligo-Miocene shallow-water, heterozoan limestones present in the Central Mediterranean (Maltese Islands and Sicily). In these two locations, inner-ramp to outer-ramp deposits composed of different types of heterozoan biotic assemblages are well exposed, and although they were never deeply buried they are very well lithified and form, in the case of the Maltese Islands, steep coastal cliffs. This raises the question of the source and the timing of cementation and lithification. The sections on the Maltese Islands and Sicily were chosen for comparison because during deposition both islands were part of the same, extensive carbonate-ramp system, but they represent different depositional environments. The carbonates on the Maltese Islands were deposited on a low-energy ramp with low sedimentation rates comprising a well developed rhodolithic carbonate factory in the mid-ramp interval. The carbonates in Sicily were deposited on a high-energy ramp with high sedimentation rates comprising a small mid-ramp interval dominated by redeposited echinoid-rich grainstones. Therefore by comparing these two settings it is possible to discern the influence of different energy levels, sedimentation rates, and facies types on the diagenetic evolution of heterozoan carbonates.

GEOLOGICAL SETTING AND STRATIGRAPHY

The studied sedimentary succession comprises upper Oligocene to lower Miocene shallow-water carbonates cropping out on the Maltese Islands and southwest Sicily (Fig. 1 A, B). During this time interval, both locations were part of a geographically extensive carbonate-ramp system located on the more distal part of the passive African continental margin (Pedley 1998). The sedimentary rocks deposited on the Sicilian Ragusa Platform form the northern margin of a high which extends 80 km south to the Maltese Islands (Pedley 1981). Paleogeographic reconstructions (Fig. 2A, B) locate both areas between 28° and 30°N during the early Chattian moving northward to about 29° and 31°N by the early Burdigalian (Dercourt et al. 2000). Accumulation took place in an open marine environment since the separation of the proto-Mediterranean–Atlantic region from the proto-Indo–West Pacific region was completed only in the late Burdigalian (Harzhauser et al. 2002).

Climatologically, sedimentation took place at the boundary between the subtropical and the temperate climate zone. Calculated water temperatures for the study area range between 17 und 23°C (Knoerich and Mutti 2003) and are consistent with the described larger benthic foraminifer association (*Lepidocyclina, Heterostegina, Amphistegina*)
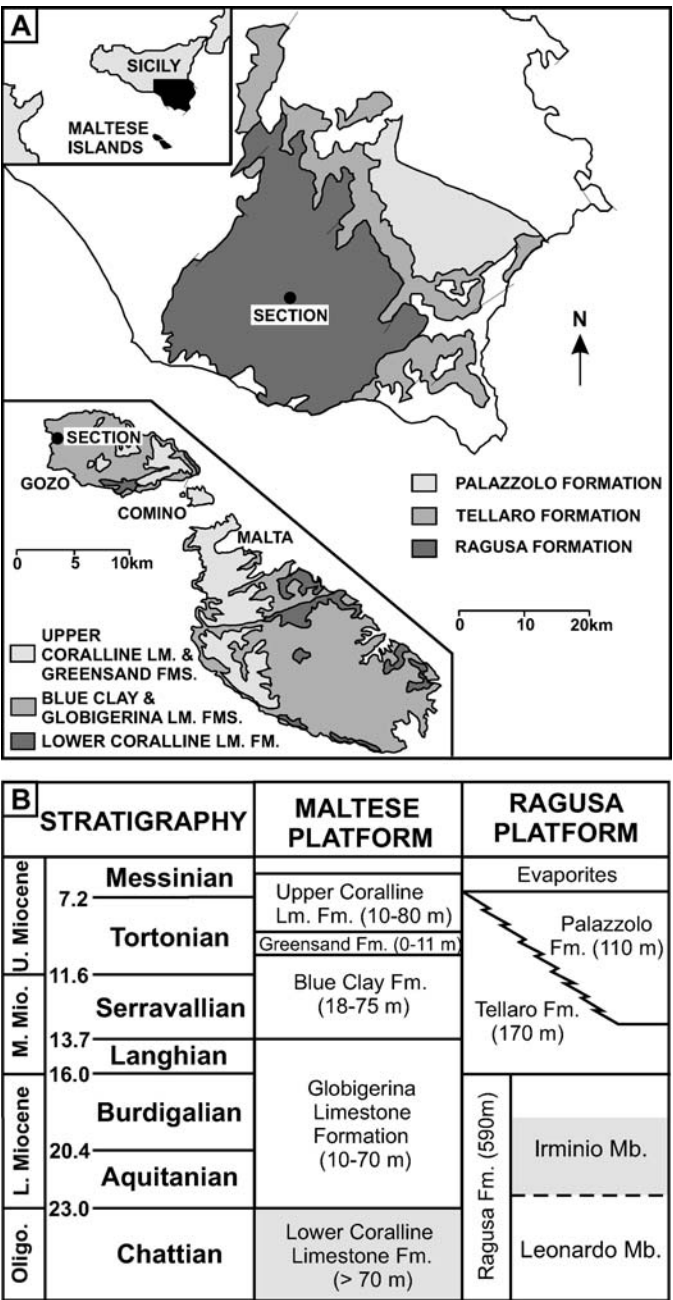

Fig. 1.— **A)** Geological map of the Maltese Islands and SE Sicily. For the Maltese Islands the lithostratigraphic units are displayed according to Felix (1973) and Carbone et al. (1987), for Sicily according to Pedley (1981) and Grasso and Lentini (1982). The location of the measured sections Dweira on Gozo and Ragusa on Sicily is indicated. LM Limestone; FM(S) Formation(s) (modified after Knoerich and Mutti 2003). **B)** Comparison of the studied lithostratigraphy and the age of the units. The focus of this study is indicated by gray shading. FM, Formation; MB, Member (modified after Knoerich and Mutti 2003).

(Pedley 1978, 1998), which is indicative of minimum water temperatures between 14° and 18°C (Betzler et al. 1997).

Maltese Islands

The Oligo-Miocene sedimentary succession of the Maltese Islands comprises four major lithostratigraphic units (Fig. 1A, B) and records
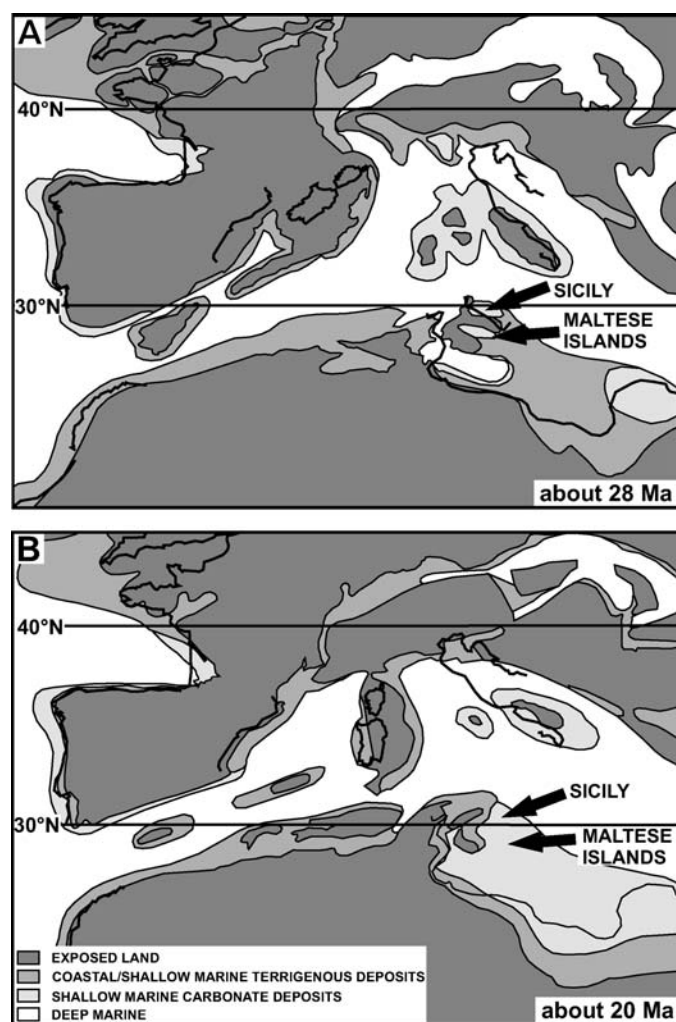
FIG. 2.— Paleogeographic reconstruction of the Mediterranean region. The studied locations are indicated by black arrows. **A)** Early Chattian times (~ 28 Ma). **B)** Early Burdigalian times (20.5–19 Ma) (after Dercourt et al. 2000).

a stepwise evolution as described by several stratigraphic and sedimentological studies (Pedley 1978; DiGeronimo et al. 1981; Carbone et al. 1987; Jacobs et al. 1996; Pedley 1998; John et al. 2003).

The focus of this study is the Lower Coralline Limestone Formation (Chattian, Fig. 1B). It was deposited in an inner-ramp to outer-ramp setting and consists of rudstones and packstones comprising skeletal fragments typical of heterozoan carbonates (Fig. 3). This unit progressively drowned, as recorded by the increasing amount of planktonic foraminifers and the development of several phosphatic hardgrounds (Jacobs et al. 1996; John et al. 2003), and is overlain by hemipelagic wackestones rich in planktonic foraminifers (Globigerina Limestone Formation, Aquitanian–Langhian). The deepening trend continued throughout the Serravallian and early Tortonian when hemipelagic claystones to marlstones were deposited (Blue Clay Formation). The trend is reversed with the Blue Clay Formation unconformably overlain by shallow-water carbonates of the Upper Coralline Limestone Formation (Tortonian to Messinian), which are composed of photozoan associations (*sensu* James 1997).

Sicily

The Oligo-Miocene sedimentary succession of the Ragusa Platform (Fig. 1A, B) starts with the Ragusa Formation (Chattian to Upper Burdigalian; Grasso and Lentini 1982), which is subdivided into a lower, more marly part (Leonardo Member) and an upper more micritic (Irminio Member) part. The Leonardo Member is about 400 m thick, consists predominantly of planktonic foraminiferal debris and micrite, and was deposited in an outer-shelf environment (Pedley 1981). The Irminio Member, the focus of this study, is about 190 m thick and consists of shallow-water deposits comprising small and larger benthic foraminifers, echinoids, coralline algal fragments, and subordinate planktonic foraminifers (Grasso and Lentini 1982) (Fig. 4). It includes small- to medium-size submarine channels (Pedley 1981).

Hemipelagic marls and marly limestones (Tellaro Formation, Langhian to Upper Tortonian) and marls and biomicrites (Palazzolo Formation, Serravallian to Tortonian) conformably overlie the Ragusa Formation and are indicative of a deepening trend (Pedley 1981). Similarly to the Maltese setting, the Palazzolo Formation shallows upwards and is overlain by Messinian evaporites (Grasso and Lentini 1982).

ANALYTICAL METHODS

Two sections were logged in both locations (Dweira on Gozo, Maltese Islands; Ragusa on Sicily), sampled, and described in detail with respect to the sediment structures and the biotic assemblages. Diagenetic and compositional characteristics of the investigated lithologies are based on transmitted-light microscopy on 43 thin sections from the Maltese Islands and 15 thin sections from Sicily. Quantification of the biotic components and diagenetic features (cement types, secondary porosity) was performed by point counting (400 points per thin section) on all thin sections. All thin sections were examined under cathodoluminescence (CL) microscopy. Operating conditions for the Citl Cold Cathode Luminescence (model 8200 mk 3) at the University of Stuttgart (Germany) were maintained at approximately 22 kV beam energy and 0.8 mA beam current.

Eight thin sections from Malta and 6 from Sicily were polished and coated with carbon for electron microprobe analysis, using a Cameca SX 100 at the University of Stuttgart. Conditions were 15 kV accelerating potential, 8 nA beam current on a Faraday cup, and 60 seconds counting time on peaks. Measurements were carried out with a defocused beam (diameter 20 μm) in order to avoid vaporization of the carbonate minerals. Standards were acquired under the same beam conditions (periclase for Mg; wollastonite for Ca; hematite for Fe; rhodonite for Mn; celestite for Sr). Detection limits were 350 ppm for Fe and Mn, 400 ppm for Mg and Sr, and 670 ppm for Ca.

Mineralogical and geochemical analyses were performed on powdered bulk-rock samples. X-ray diffraction analyses and quantification of the inorganic carbon content were performed on 43 samples from Malta and 12 samples from Sicily. XRD measurements were carried out with a Bruker D8 Advance with 2 Theta = 25°–35° at the University of Stuttgart. The increment used was 0.005 degree; the scan speed was 1 degree/minute. For each measurement four scans were stacked to minimize the error. The inorganic-carbon content was determined on 40 to 60 mg of sample material with a Coulometer 5020 from UIC Inc. attached to a CM TIC Auto sampler from Orbis bv at the University of Stuttgart. Results were converted to calcite values by multiplying the TIC (total inorganic carbon) result by the appropriate factor (8.33).

Forty-two powdered bulk-rock microsamples from Malta ranging in size from 0.5 to 4 mg were analyzed for carbon and oxygen stable isotopes at the Laboratory for Stable Isotope Geochemistry at the University of Southern California. Analyses were performed according to on-line common acid bath method using a PRISM mass spectrometer. The analytical precision of the mass spectrometer is ± 0.1‰ or better for δ^{13}C and ± 0.2‰ or better for δ^{18}O. The δ^{18}O values have been corrected for calcite phosphoric acid fractionation. The δ^{13}C and δ^{18}O values are given in the PDB notation.
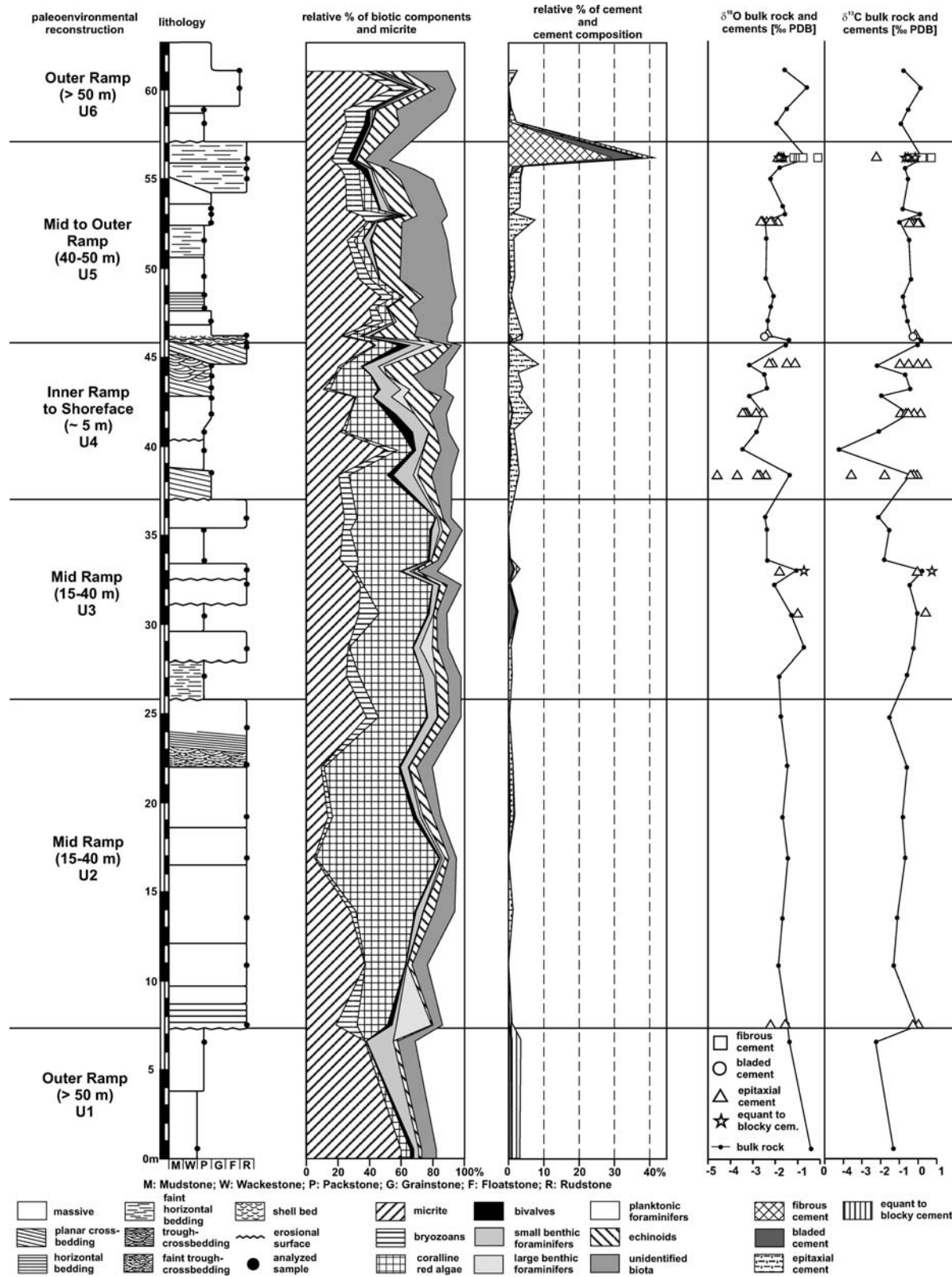
Fɪɢ. 3.—Measured section ("Dweira") of the Lower Coralline Limestone Formation on Gozo. The lithology, including sedimentary textures, the amount of biotic components and micrite and the cement amount and composition based on point counting are displayed to the right of the stratigraphic profile. The stable-isotope composition (δ^{18}O and δ^{13}C) of the bulk rock and the various cement types is presented. The inferred depositional environments are shown to the left of the stratigraphic profile (modified after Knoerich and Mutti 2003).
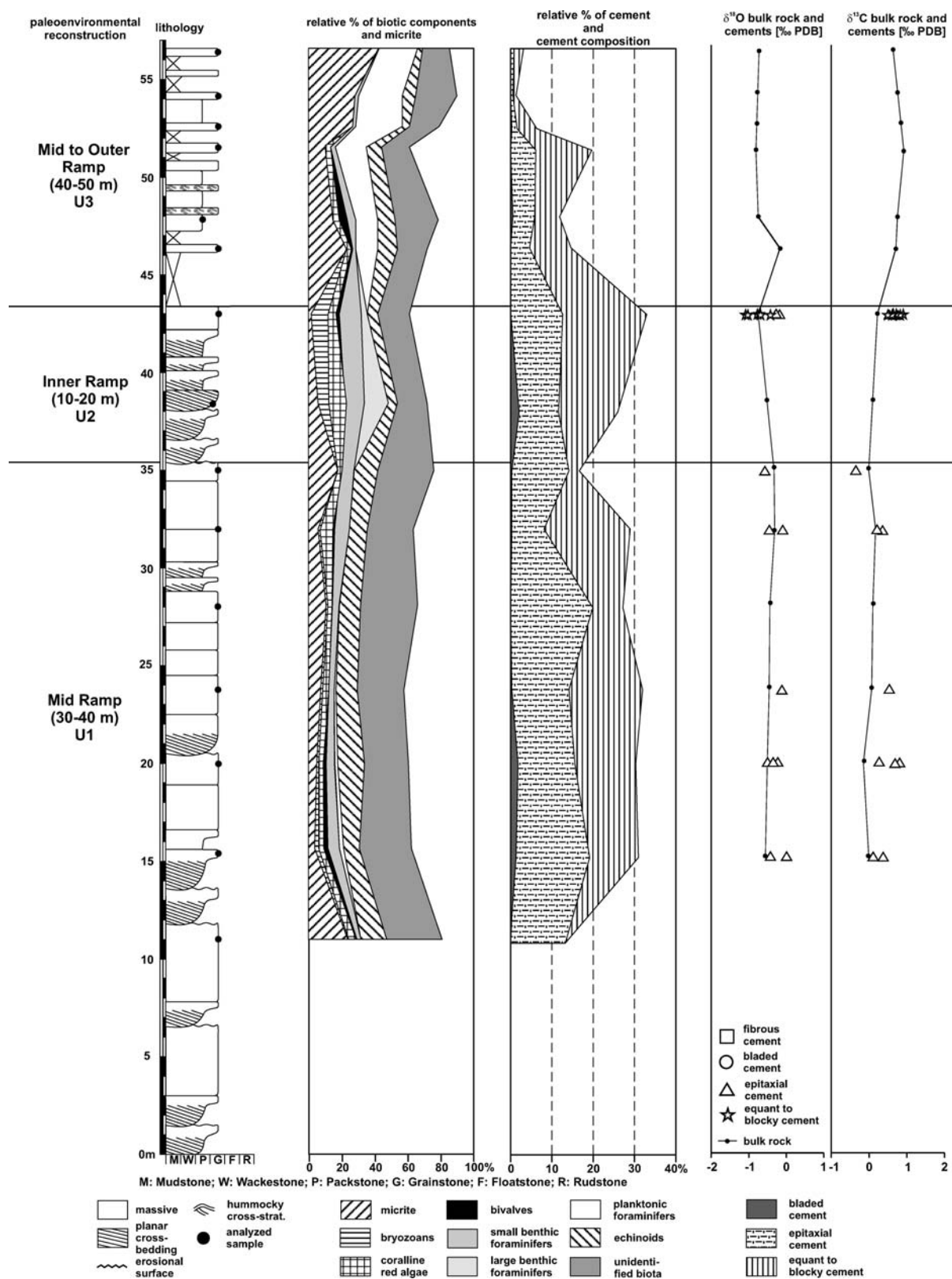
FIG. 4.— Measured section ("Ragusa") of the Ragusa Formation on Sicily. The lithology, including sedimentary textures, the amount of biotic components and micrite, and the amount and composition of cement based on point counting are displayed to the right of the stratigraphic profile. The stable-isotope composition (δ^{18}O and δ^{13}C) of the bulk rock and the various cement types is presented. The inferred depositional environments are shown to the left of the stratigraphic profile.

Sample ID	Section height [m]	$^{87}Sr/^{86}Sr$	Analytical error	Age [Ma]
Dweira 12	30.6	0.708173	0.000003	25.5 ± 0.7
Dweira 30	58.1	0.708224	0.000003	24.4 ± 0.7
Ragusa 9593	15.2	0.708200	0.000003	24.9 ± 0.7
Ragusa 9598	35.0	0.708328	0.000003	22.5 ± 0.7
Ragusa 95101	46.4	0.708370	0.000003	21.4 ± 0.7

Eleven powdered bulk rock samples from Sicily and 56 microdrilled cement samples from Malta and Sicily were analyzed for their stable carbon and oxygen isotopic composition at the University of Bremen (Germany). The samples for the single cement analyses were acquired by microdrilling of thick sections ($\sim$ 300 μm) under the binocular microscope to ensure that only single cement types were sampled. The powder was dissolved in phosphoric acid and the resulting CO_2 gas was measured with a FINNIGAN MAT 251 mass spectrometer. An internal standard calibrated against the international NBS 19 standard was used. Standard deviation was 0.07‰. Oxygen and carbon isotope results are expressed in the PDB notation.

Sr-isotope composition was measured on five bulk rock samples, two from the Dweira section and three from the Ragusa section, with the Re-double filament technique on a Finnigan MAT 262 RPQ$^+$ in static mode at the Geomar Kiel (Germany). Usually 100 to 200 $^{87}Sr/^{86}Sr$ ratios were collected for each measurement, which resulted in an internal precision better than 10 ppm (2SE) of the $^{87}Sr/^{86}Sr$ value for most of the samples. The Sr blank was less than 500 pg. However, sample sizes are sufficiently large (approx. 1 μg of Sr) that blank corrections are negligible. A mean $^{87}Sr/^{86}Sr$ value of 0.710236 ± 34 (2 standard deviations) for the NIST 987 standard solution was calculated from 12 analyses, covering the whole measurement period of the data set. It represents the minimum uncertainty assigned to any individual sample measurement.

SEDIMENT COMPOSITION AND DEPOSITIONAL FACIES

The data presented here were acquired at two sections (Chattian to Aquitanian, Table 1). The Dweira section was measured on the Island of Gozo (Maltese Islands) and comprises the most complete succession of the Lower Coralline Limestone Formation (LCLF). The basal part of the formation is not exposed, while at the top the LCLF grades into the overlying Globigerina Limestone Formation. The exposed 63 m can be subdivided into six units, based on changes in sediment composition, depositional facies, bedding, and sediment structures (Fig. 3).

The Ragusa section was sampled on the island of Sicily and comprises a part of the Irmino Member, which belongs to the Ragusa Formation. The outcrop investigated is located in a deeply incised valley, where only the upper part is accessible. The measured 57 m can be subdivided into three units (Fig. 4).

Dweira (Gozo, Maltese Islands)

Facies Description.—The lowermost unit (U1) consists of massive wackestones to packstones (Fig. 3). The rocks are rich in small and larger benthic foraminifers (*Lepidocyclina*) and biodetrital micrite. This micrite was formed by physical abrasion and biological erosion of larger biogenic skeletons and was not precipitated as cement, as indicated by small biogenic fragments floating in the micritic matrix. The base of the unit is not exposed, and at the top it is bounded by an erosional unconformity.

The overlying unit 2 comprises massive rudstones at the base which grade into planar- or trough-cross-bedded rudstones. Unit 2 is erosionally overlain by alternating packstone and rudstone beds of unit 3, which sometimes display horizontal bedding. Both units consist predominantly of calcareous red algae. Unit 2 is dominated by rhodolites, whereas in unit 3, rhodolites gradually disappear and the content of articulated calcareous red algae and biodetrital micrite increases. Unit 4, consisting of well sorted, planar- and trough-cross-bedded grainstones and rudstones, is rich in small and larger benthic foraminifers and echinoid fragments and displays at least one subaerial exposure surface (40.5 m section height), recognizable by dissolution features. Unit 5 follows above an erosional unconformity and consists of faintly horizontally bedded grainstones to packstones and rudstones. The base of the unit is composed of two shell beds of pectinids and echinoids (*Scuttella*), with overlying deposits dominated by echinoid fragments and unidentifiable broken biota with biodetrital micrite. The topmost unit 6 was deposited above an erosional unconformity and consists of massive wackestones to floatstones. Biodetrital micrite and unidentifiable biota are the main rock components. Planktonic foraminifers start to appear, indicating the transition to the overlying Globigerina Limestone Formation.

Interpretation.—Unit 1 was probably deposited in an outer-ramp setting below storm wave base, as indicated by the abundant biodetrital micrite. Units 2 and 3 were deposited in a mid-ramp environment, with unit 2 representing a rhodolitic carbonate factory (rhodolite pavements of James 1997) with some wave agitation and unit 3 indicating an increase in water energy resulting in strongly reworked grains. Unit 4 accumulated in a well agitated inner-ramp setting, as indicated by good sorting, cross-stratification, and reworking. This was followed by the development of syndepositional solution subsidence structures associated with subaerial exposure. The overlying unit 5 was deposited in a deeper, less agitated mid-ramp setting, as indicated by the abundant micrite. The abundance of abraded material and the enrichment in echinoid fragments points to reworking in agitated waters. These beds could be interpreted as storm deposits washed into the deeper ramp. Unit 6 was deposited in poorly agitated waters of the outer ramp, as indicated by the high amount of biodetrital micrite and the occurrence of planktonic foraminifers.

Overall the Dweira section displays a shallowing-upward trend from an outer-ramp to an inner-ramp setting (U1–U4), which is reversed after a pronounced erosional unconformity and followed by drowning of the succession (Fig. 3). For a detailed description of the biotic assemblages and depositional environments, the reader is referred to Knoerich and Mutti (2003).

Cement Types and Distribution.—On average the Dweira section contains about 3 vol. % cement (Fig. 3). The dominant cement phase (up to 8%) is represented by epitaxial calcite cement precipitated around echinoid fragments. This cement type becomes especially abundant in the inner-ramp and upper mid-ramp unit (U4 and U5) and occurs predominantly in well-sorted, well-bedded, grain-supported strata. One especially well cemented horizon, containing over 40% of cement, occurs along a drowning surface.

Ragusa (SE Sicily)

Facies Description.—The lowermost unit (U1) consists of packstones to grainstones. Intercalated channel-like structures (1.5 to 4 m high) with planar cross-bedding and erosional bases are common features (Fig. 4). Echinoid fragments and unidentifiable biogenic components are the fossil constituents. Except for the channel-like structures the bedding is massive. The overlying unit 2 is composed of packstones to

grainstones and contains abundant planar cross-beds in channel-like bodies with erosional bases (1 to 1.5 m high). The biodetrital micrite content of the rocks diminishes towards the top, whereas smaller and larger benthic foraminifera become abundant. Unit 3 is composed of intercalated massive packstones and grainstones, frequently exhibiting hummocky cross stratification. Towards the top of the unit the biodetrital micrite and planktonic foraminifer content increases while the amount of larger benthic foraminifers and echinoid fragments decreases.

Interpretation.—Unit 1 was probably deposited in a high-energy mid-ramp setting, as evident by the frequent occurrence of erosional, channel-like structures and the scarcity of micrite. Unit 2 also accumulated in highly agitated waters, as suggested by channel-like structures, but the abundant occurrence of larger benthic foraminifers point to deposition in the photic, inner-ramp environment. Unit 3 was deposited below fair-weather wave base but above storm wave base, as indicated by hummocky cross-stratification. The increasing amount of biodetrital micrite, and planktonic foraminifers, points to gradual drowning.

Overall the depositional sequence in the Ragusa section is similar to that of the Dweira section, with shallowing upward from a mid-ramp to an inner-ramp setting (U1 + U2), followed by a deepening trend towards the top of the section (U3). In contrast to Dweira, shallow-water sedimentation in Ragusa occurred on a more open, high-energy shelf. Sediment deposition was episodic, most probably associated with storm events, which must have resulted in temporary high accumulation rates. In contrast to Dweira, Ragusa did not experience a syndepositional subaerial exposure but was rapidly drowned.

Cement Content and Distribution.—The Ragusa section contains on average about 20% cements, consisting of equal amounts of epitaxial and equant to blocky calcite cement (Fig. 4). In the uppermost unit, which experienced drowning (U3), the cement content decreases. Overall the cement content in this section is inversely correlated with the biodetrital micrite content; the highest cement content where micrite is least abundant.

CEMENT PETROGRAPHY AND PARAGENESIS

Based on detailed petrographic observations and cement quantification, several diagenetic features can be distinguished (Figs. 5–7), including different episodes of cementation, dissolution, neomorphism, and compaction. In both localities the diagenetic history is similar. Therefore, they are combined in the following description according to their paragenetic succession from oldest to youngest.

Fibrous to fibrous-bladed cements (Fi) occur in significant quantities only in one sample from Dweira (Fig. 3), where this cement type forms about 30% of the total rock volume (Fig. 5A–C). In all other samples from Dweira and Ragusa, fibrous to fibrous-bladed cement is volumetrically unimportant (1–2%), and mainly lines intraparticle pore spaces such as bryozoan or foraminifera chambers.

Fibrous to fibrous-bladed cements nucleate on grain surfaces and line them in isopachous rinds. The single crystals are elongated along the *c* axis and nucleate in a picket-fence style on the substrate. Their average length is 10 to 20 μm. In the Ragusa samples, this cement type is less elongated and has a more bladed appearance (Fig. 7A). The crystals are inclusion-rich, which causes a patchy pattern under CL (Fig. 5B). The inclusions indicate that this cement type was recrystallized.

Epitaxial cement I (Ep I) precipitated in optical continuity with single-crystal grains. It commonly grows as the first precipitate around echinoid fragments, where it forms thin rims (10–20 μm) (Figs. 5D, E, 6A, 7A, C). This cement type constitutes only 1–2% of the rock volume at Dweira and Ragusa. The crystals are inclusion-rich, grew perpendicular to the substrate, and have prismatic terminations. Under CL, they display a patchy pattern, indicating recrystallization (Figs. 5F, 6B, 7D). In some places the outermost rim of the epitaxial cement I is dissolved and forms moldic porosity (Figs. 5E, F, 7A).

Fabric selective dissolution I affected aragonitic biota (gastropods, bivalves, and foliose bryozoans) and created open moldic porosity in the Dweira samples (up to 9%), which was later not filled with cement (Figs. 5C, 6C). At Ragusa, no biomoldic porosity was observed, but large interparticle pore spaces in some areas (Fig. 7B, see arrows) and ghost structures later filled with cement suggest prior selective dissolution of grains.

Epitaxial cement II (Ep II) precipitated in optical continuity with the substrate, usually around echinoid fragments but occasionally also around benthic foraminifers. This second phase of epitaxial cementation postdates epitaxial cement I (Figs. 5D, E, 6C, 7A–C, E). Epitaxial cement II consists of inclusion-free calcite (average crystal size of 200 μm) with zonations clearly visible under CL (Figs. 5F, 7D), often forming a poikilotopic texture. This is the volumetrically most important cement phase, contributing up to 8% of the rock volume at Dweira and between 10 and 20% at Ragusa.

Fabric selective dissolution II preferentially affected epitaxial cement I and HMC echinoid fragments and coralline red algae. EP I was dissolved in some places, leading to the formation of open moldic porosity (Figs. 5E, F, 7A), both at Dweira and at Ragusa. Echinoids were partly dissolved along the crystal cleavage planes, whereas coralline red algae display a more patchy dissolution.

Neomorphism occurred most probably at the same time as fabric-selective dissolution II. It led to the alteration of HMC cement phases (fibrous and epitaxial cement I) and biota (calcareous red algae and echinoids) to LMC, as indicated by their patchy appearance under CL (Fig. 5B).

Bladed-prismatic cement (Bla) consists of elongated scalenohedral crystals with prismatic terminations. Crystals are on average 20 to 90 μm long and 5 to 10 μm across and form rims of uneven thickness in intraparticle and interparticle pore spaces (Fig. 5A, C). The original crystal fabric of this cement type is well preserved, with clearly recognizable zonations under CL (Fig. 5B). In the Dweira samples, this cement type constitutes on average 1 to 2% of the rock total volume but can reach up to 8%. This cement phase was not observed at Ragusa.

Blocky sparry calcite cement (Blo) comprises plane-sided, equant crystals with an average width of 40 to 60 μm. The original crystal fabric is well preserved with clearly visible zonations under CL. This cement phase either grew poreward, postdating fibrous and/or bladed cements (Fig. 5C), or directly on the substrate, where the other cement phases are missing. In the Dweira samples, blocky cement on average contributes 1 to 2% to the total rock volume. At Ragusa, this cement phase is equally important as epitaxial cement II and forms between 10 and 20% of the rock volume (Fig. 7B–E).

Compaction includes mechanical and chemical compaction (pressure solution). Mechanical compaction can be very intense on fragile components such as larger foraminifers (Fig. 6D). Chemical compaction includes grain-to-grain contacts, concavo-convex contacts, and even sutured contacts (Fig. 7F). No microstylolites were found. In both study locations chemical compaction postdates precipitation of the volumetrically important epitaxial cement II (Figs. 6A, B, 7C, D).

Non-fabric-selective dissolution forms small vugs and channels, crosscutting biota, matrix, and cements. These vugs and channels either stay open or sometimes are filled with blocky calcite cements. Vugs also postdate compaction features, suggesting that this phase of dissolution occurred quite late in the diagenetic sequence. At Dweira up to 13% of vuggy porosity was observed, whereas at Ragusa the vuggy porosity represents only 1%.
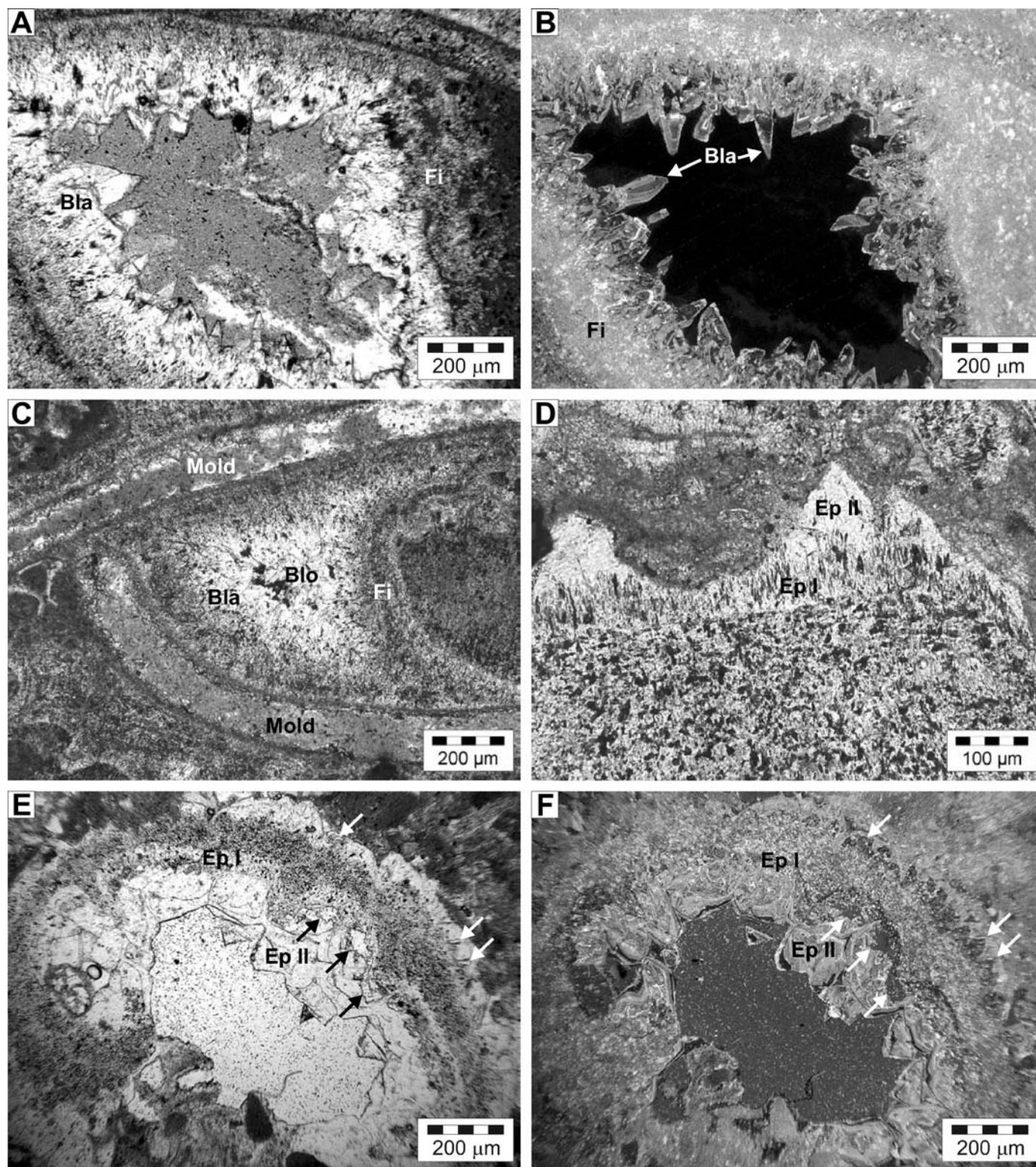
FIG. 5.—Photomicrographs of cements and dissolution features observed in the samples from Dweira (Maltese Islands). **A)** Note fibrous, inclusion-rich cement (Fi) followed by inclusion-free bladed-prismatic cement (Bla). **B)** As Part A in CL. Note the patchy luminescence in the fibrous cement and the clear zonations visible in the bladed-prismatic cement. **C)** Fibrous, inclusion-rich cement (Fi) postdated by inclusion-free bladed-prismatic (Bla) and blocky cements (Blo). Note that the molds after aragonitic biota are totally cement-free and not compacted. **D)** Two phases of epitaxial cements growing on an echinoid fragment. The first (Ep I) is inclusion-rich, the second (Ep II) is inclusion-free. **E)** Part of an echinoid shell overgrown by two phases of epitaxial cement. The first (Ep I) is inclusion-rich and its outermost rim is partly dissolved (see arrows). The second (Ep II) is inclusion-free. **F)** As Part E in CL. Note the patchy luminescence in the epitaxial cement I and the clearly visible zonations in epitaxial cement II.
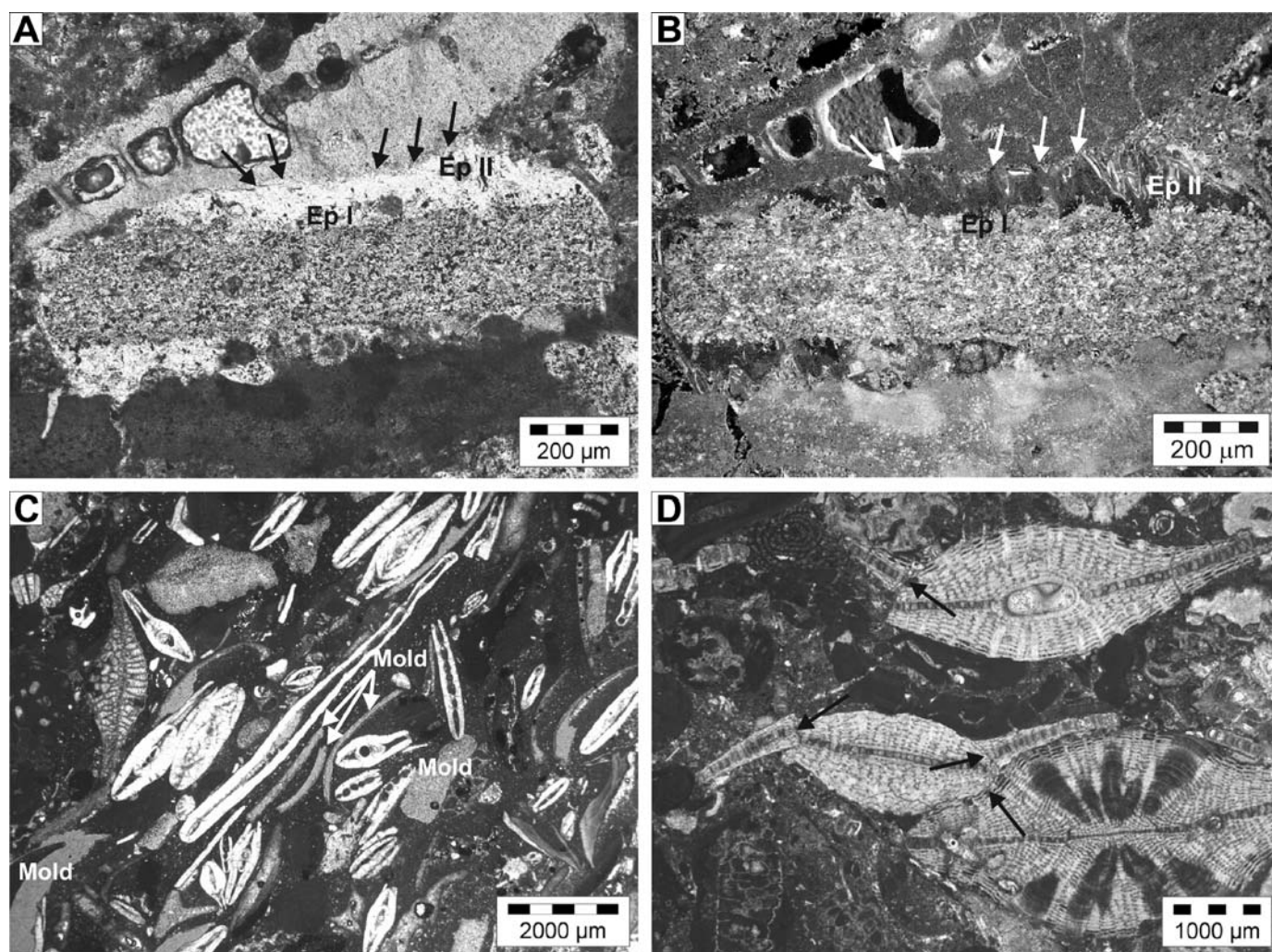
Fig. 6.—Photomicrographs of dissolution and compaction features observed in the samples from Dweira (Maltese Islands). **A)** Note the sutured contact between the foraminifer and the echinoid overgrown by epitaxial calcite cement I and II (Ep I and Ep II). **B)** As Part A in CL. Note how the epitaxial cement II (Ep II) has been partially dissolved by pressure solution (arrows), indicating that precipitation of epitaxial cement II predated compaction. **C)** Note the abundance of molds after aragonitic biota and that all molds are cement-free and not compacted. **D)** Note the occurrence of mechanical (breaking) and chemical (concavo-convex and sutured contacts) compaction in the foraminiferal packstone.

MINERALOGICAL AND GEOCHEMICAL ANALYSES

XRD and Coulometer Analyses

Results from XRD analysis indicate that only LMC is preserved at both locations; no traces of aragonite or HMC were measured. Both sections consist of very pure carbonate (> 90%).

In both areas, the noncarbonate content, consisting mainly of clay minerals, is facies dependent. The mid-ramp and inner-ramp settings contain less noncarbonate material (Dweira, 0.4 to 2.5%, mean 1.2%; Ragusa, 1.0 to 2.7%, mean 2%) than the outer-ramp environments (Dweira, 0.9 to 5.8%, mean 2.3%; Ragusa, 2.8 to 8.4%, mean 4.5%).

Stable Isotopes

Dweira.—Stable-isotope values of the bulk-rock and the different cement types are plotted against stratigraphic thickness (Fig. 3) and in a cross-plot (Fig. 8A). The bulk rock values range from −0.5 to −3.3‰ for $\delta^{18}O$ and from +0.3 to −4.2‰ for $\delta^{13}C$. Generally, bulk and single-cement values are similar; the main exception is the inner-ramp unit (U4), where the bulk rock is up to 4‰ higher, both for $\delta^{18}O$ and $\delta^{13}C$, than the single-cement values.

The stable-isotope values for the different cement types at Dweira generally vary between −0.1‰ and −4.6‰ for $\delta^{18}O$ and between +0.8‰ and −3.6‰ for $\delta^{13}C$ (Figs. 3, 8A). Fibrous cements display the highest values, both for $\delta^{18}O$ and $\delta^{13}C$, ranging between +0.1 and −1.2‰ for $\delta^{18}O$ and between +0.7 and −0.2‰ for $\delta^{13}C$.

Since epitaxial cement I and II could not be sampled separately due to the small size of the cement rims, the isotopic values for this cement type represent a mixture of both epitaxial cement phases, but more likely represent the volumetrically more important epitaxial cement II. This cement type shows a wide range of $\delta^{18}O$ and $\delta^{13}C$ values, varying between −1.0 and −4.6‰ for $\delta^{18}O$ and between +0.5 and −3.6‰ for $\delta^{13}C$. The lowest values, found in the inner-ramp unit (U4) ($\delta^{18}O$, −3.3 to −4.6‰; $\delta^{13}C$, −0.7 to −3.6‰) occur directly below the subaerial exposure surface, which is located at a section height of 40.5 m.

Bladed-prismatic and blocky cements display intermediate isotopic compositions, ranging from −1.0 to −1.9‰ for $\delta^{18}O$ and from −0.2 to −2.2‰ for $\delta^{13}C$.
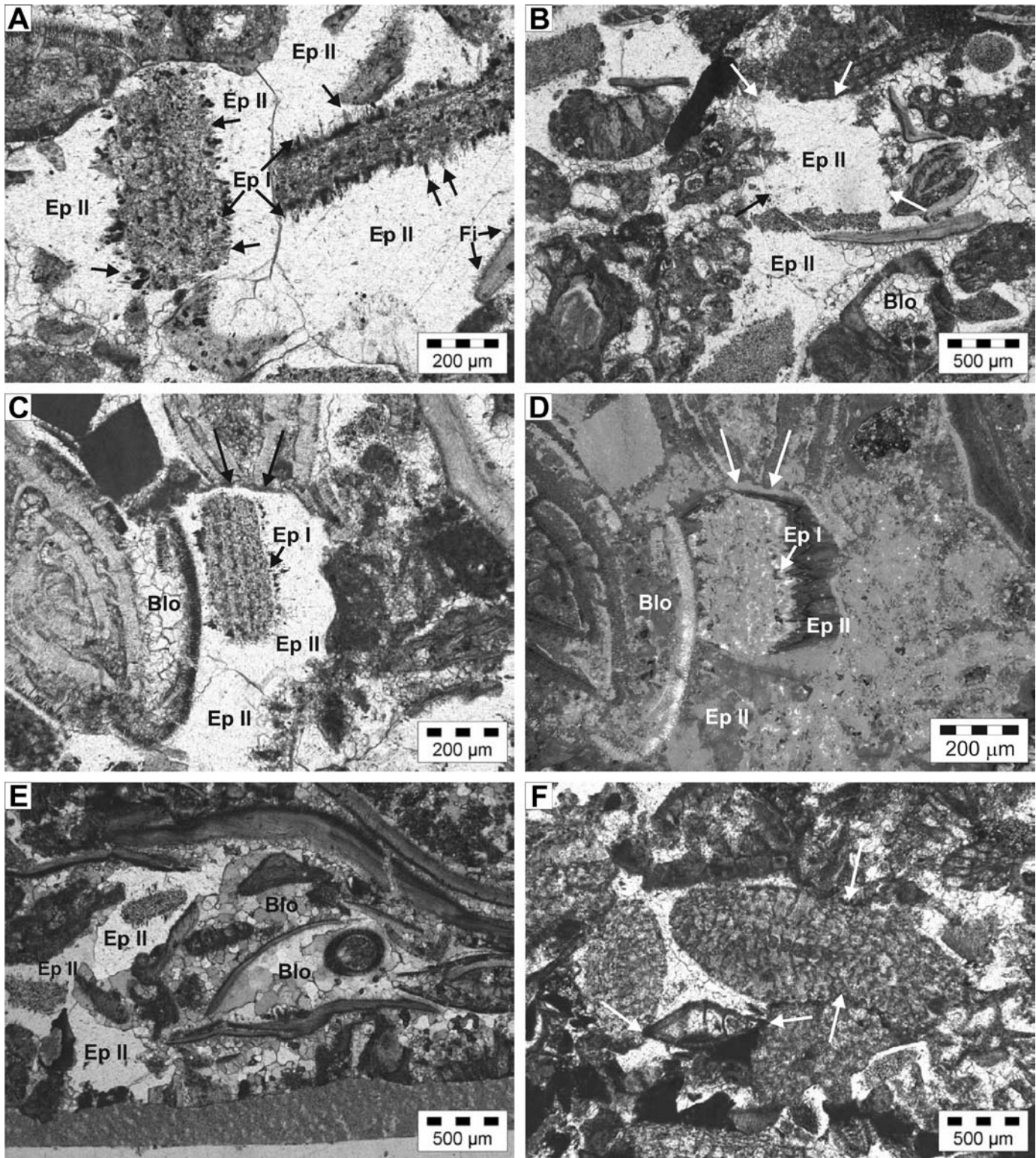
Fig. 7.—Photomicrographs of cements and other diagenetic features observed in the samples from Ragusa (Sicily). **A)** Note the paragenesis of the diagenetic features. Depending on the substrate either fibrous-bladed (Fi) or epitaxial cement I (Ep I) precipitated first, both phases are inclusion-rich. Epitaxial cement I is partly dissolved (see arrows). Both cement phases are then overgrown by a second epitaxial cement phase (Ep II), inclusion-free and in parts poikilotopic. **B)** Note the large, oversized pore spaces (see arrows) filled by inclusion-free epitaxial cement II (Ep II). **C)** Visible is an echinoid fragment overgrown by two phases of epitaxial cement (Ep I and Ep II). Inclusion-free blocky cement (Blo) precipitated inside a foraminifer chamber. Note the sutured contact of the foraminifer and the epitaxial cement II (arrows). **D)** As Part C in CL. Note how the foraminifer has been partly dissolved by pressure solution against the epitaxial cement II (arrows), indicating that pressure solution postdated precipitation of this cement phase. **E)** Overview of the distribution and abundance of the volumetrically important epitaxial cement II (Ep II) and blocky cement (Blo). Picture taken with crossed nicols. **F)** Note the concavo-convex and the sutured contacts between the foraminifers (arrows), indicating chemical compaction by pressure solution.
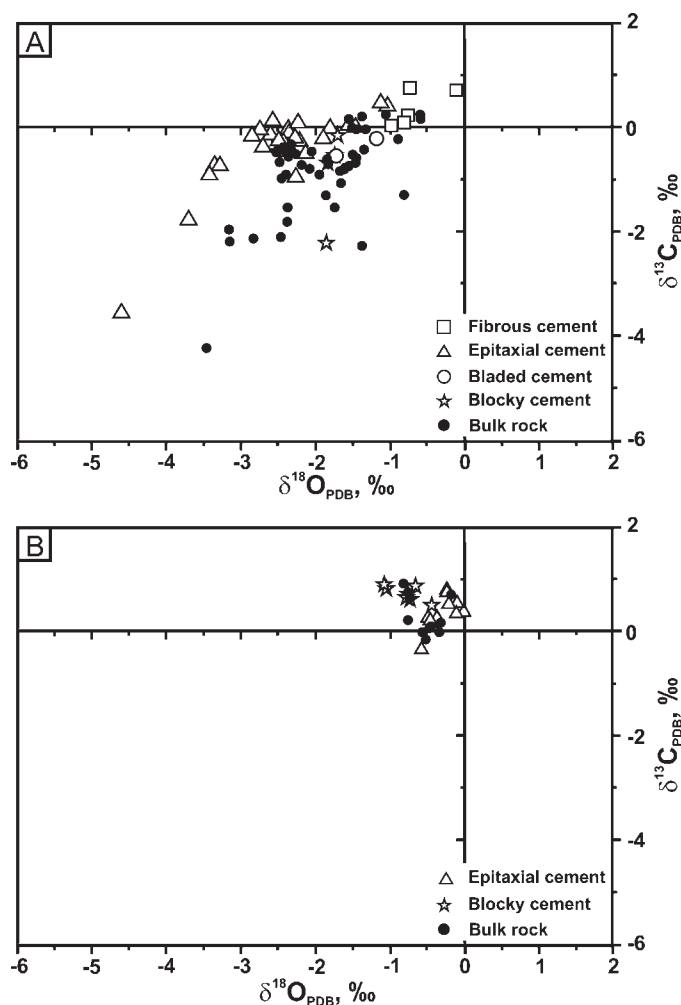
FIG. 8.—Cross-plot of the carbon and oxygen isotopic composition of the bulk rock and the different cement types. **A)** Data for the Dweira section on Gozo. **B)** Results for the Ragusa section on Sicily.

Ragusa.—The stable-isotope values of the bulk rock and different cement types in Ragusa are quite similar and do not show significant variation throughout the section (Figs. 4, 8B). The bulk-rock samples range from -0.2 to -0.8‰ for $\delta^{18}O$ and from $+0.9$ to -0.2‰ for $\delta^{13}C$. The various cement types in Ragusa vary from 0.0‰ to -1.1‰ for $\delta^{18}O$

and from $+0.9$ to -0.4‰ for $\delta^{13}C$ (Figs. 4, 8B). The values for the combined epitaxial cement phases I–II, with predominance of epitaxial cement II, show slightly higher $\delta^{18}O$ (0.0‰ to -0.6‰) and slightly lower $\delta^{13}C$ ($+0.8$‰ to -0.4‰) values than the blocky cements ($\delta^{18}O$: -0.2‰ to -1.1‰; $\delta^{13}C$: $+0.9$‰ to $+0.5$‰).

In comparison, the cements from Dweira display lower ^{18}O and ^{13}C values than the cements from Ragusa and scatter over a much wider range.

Major-Element and Trace-Element Cement Composition

The major-element and trace-element composition (Ca, Mg, Sr, Fe, and Mn) of the various cement types from Dweira and Ragusa was determined by measuring transects through the cement rims (Figs. 9, 10) to document variations within single cement zones. The mean and maximum values as well as the standard deviation of the major and trace elements are displayed in Tables 2 and 3.

Dweira.—The Mg content of the various cement types ranges from 2500 to 3500 ppm, which corresponds to 1 to 1.4 mol % $MgCO_3$, indicating that all cement types are LMC. The Sr, Fe, and Mn values of the single cement types differ significantly from one another, with fibrous-bladed cements having the lowest Sr and the highest Fe and Mn (Sr, 87 ppm; Fe, 429 ppm; Mn, 102 ppm) contents and epitaxial cement II having the highest Sr and lowest Fe and Mn (Sr, 317 ppm; Fe, 101 ppm; Mn, 37 ppm) values. (Note that the detection limit for Fe and Mn is 350 ppm and for Sr it is 400 ppm.)

Ragusa.—The Mg content in the cement samples ranges from 1600 to 3100 ppm (0.7 to 1.3 mol % $MgCO_3$), indicating a LMC composition. Of the two analyzed cement types, epitaxial cement II has lower Sr and higher Fe and Mn (Sr, 1190 ppm; Fe, 205 ppm; Mn, 119 ppm) values than the blocky cement type (Sr, 2658 ppm; Fe, 139 ppm; Mn, 63 ppm). (Note that the detection limit for Fe and Mn is 350 ppm and for Sr it is 400 ppm.)

In comparison, the various cement types from Dweira contain less Sr and more Fe and Mn than the cements from Ragusa. In particular, the relatively high Sr value of the cements from Ragusa (up to 5900 ppm) is rather remarkable. Moreover, the measured transects show that the trace-element composition varies not only between the different cement types (Fig. 9), but also within single cements related to CL zones internally (Fig. 10).

DISCUSSION

Environments of Cement Precipitation and Alteration

The following discussion aims to constrain the diagenetic environments of cement precipitation and alteration using detailed petrographic
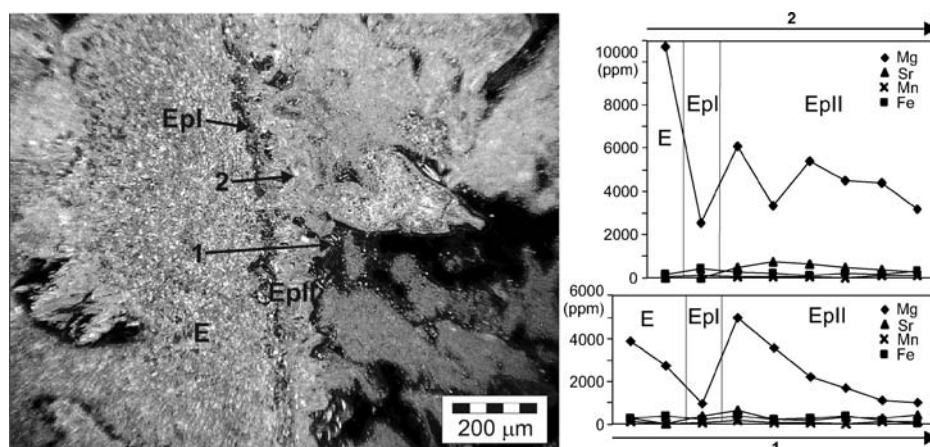

FIG. 9.—Cathodoluminescence photo of an inner-ramp sample from section Dweira (Maltese Islands) showing the two phases of epitaxial cement growing on top of an echinoid fragment. Lines (1 and 2) show the course of microprobe transects. Profiles of Mg, Sr, Mn, and Fe are plotted alongside, with zones labeled as on photo: E, echinoid fragment; Ep I, epitaxial cement I, Ep II, epitaxial cement II.
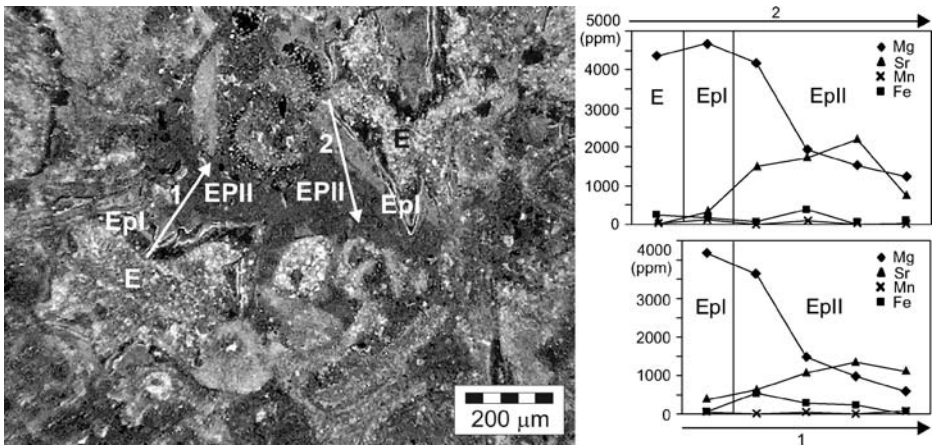
FIG. 10.—Cathodoluminescence photo of a mid-ramp sample from section Ragusa (Sicily) showing the two phases of epitaxial cement growing on top of echinoid fragments. Lines (1 and 2) show the course of microprobe transects. Profiles of Mg, Sr, Mn, and Fe are plotted alongside, with zones labeled as on photo: E, echinoid fragment; EpI, epitaxial cement I, EpII, epitaxial cement II.

observations combined with stable-isotope and trace-element measurements on the single cement phases.

Fibrous, Fibrous-Bladed, and Epitaxial Cement I.—Fibrous, fibrous-bladed, and epitaxial cement I are grouped together because they all grew in direct contact with their respective substrate and represent the first phase of cement precipitation. Under CL, all three cement types display a bright patchy luminescence, indicating neomorphism in Mn enriched waters (Tucker 1990). This, together with the elongated crystal shape of the fibrous and fibrous-bladed cements, suggests an original HMC mineralogy (Given and Wilkinson 1985). Geochemical data available for Dweira support this assumption. The elevated Fe contents (Table 2), especially of the fibrous cements, would also be consistent with a former HMC mineralogy (Richter and Füchtbauer 1978). HMC cement precipitation most probably occurred early, in the marine environment.

During later stages of diagenesis the metastable HMC was recrystallized to LMC. This is suggested by the low Sr contents (Table 2, Fig. 11A), inasmuch as Sr is preferentially lost to the fluid because of its distribution coefficient of > 1 (Veizer 1983). Therefore, 1200–1500 ppm Sr in an original HMC can be depleted to many tens or a few hundred ppm in the diagenetic LMC (dLMC) (Tucker 1990). The stable-isotope composition (Fig. 8A) suggests that recrystallization to LMC occurred in marine or marine-derived waters, inasmuch as fibrous cements display marine $\delta^{18}O$ and $\delta^{13}C$ values (+0.1 to $-1.2‰$ for $\delta^{18}O$; +0.7 to $-0.2‰$ for $\delta^{13}C$).

As suggested by this marine-derived isotopic composition, neomorphism of HMC to dLMC most probably occurred in the marine-burial diagenetic environment similar to that described by Melim et al. (1995). The marine burial environment represents the transition between the near-surface marine and the deeper-burial diagenetic environment (Melim et al. 1995). The dissolution–precipitation reactions taking place in this environment might start at very shallow subsurface depth and continue into the deeper-burial environment until the supply of metastable minerals becomes exhausted (Melim et al. 2002). In the marine-burial

environment diagenesis takes place in marine pore fluids and mimics many petrographic aspects of meteoric diagenesis (blocky spar precipitation, neomorphism, moldic porosity). Therefore stable-isotope measurement is the crucial method to distinguish the marine-burial diagenetic environment from the meteoric diagenetic environment. In general, aragonite dissolution, triggered by bacterial oxidation of organic matter elevating the CO_2 content, is considered to be the cement source in this environment (Hendry 1993).

A prominent phenomenon at both locations is the partial dissolution of the outermost rim of epitaxial cement I and the formation of moldic porosity. The inner part of the cement, in direct contact with the original substrate, is only altered to LMC. The formation of moldic porosity by dissolution of HMC is rather unusual since it generally dissolves incongruently, releasing its Mg without significant $CaCO_3$ transport (Morse and Mackenzie 1990; Moore 2001). Budd (1992) described similar partial to total dissolution of HMC grains (foraminifers) and attributed this atypical reaction pathway to the sudden influx of CO_2. This led on the one hand to an exponential increase in the dissolution rate of HMC and on the other hand precluded LMC precipitation. Once equilibrium with respect to LMC was reached, stabilization continued as incongruent dissolution. Budd (1992) suggested meteoric influence associated with subaerial exposure surfaces and water-table sources of such high CO_2 influx.

An increase in CO_2 could also explain the formation of HMC molds in this study. In contrast to the example described by Budd (1992) we suggest that the CO_2 increase was not caused by meteoric influences, for which no evidence can be found in the stable-isotope data, but could have been generated by the decay of organic matter in the marine-burial environment. Although the organic matter content in the gain-supported fabrics most probably was low, its decay might have been sufficient to cause undersaturation, as described for other neritic systems (Sanders 2003). This undersaturation might have favored the congruent dissolution behavior of HMC.

TABLE 2.—*Trace-element composition (ppm) of the various cement types measured in the samples from Dweira (Maltese Islands); n, number of measurements; SD, standard deviation; Max, maximum value.*

| Element	Fibrous cement				Epitaxial cement I				Epitaxial cement II				Bladed cement				Blocky cement			
	n	Mean	SD	Max	n	Mean	SD	Max	n	Mean	SD	Max	n	Mean	SD	Max	n	Mean	SD	Max
Ca	21	384991	3337	387990	56	393571	7166	402113	110	395452	6041	403266	44	389830	3806	399849	29	391516	4354	398530
Mg	21	3291	550	4002	56	2558	1333	6120	110	2858	1267	6466	44	3509	1493	6557	29	3216	1568	6476
Sr	21	87	159	542	56	251	204	871	110	317	193	1087	44	259	209	729	29	323	240	842
Fe	21	429	664	2093	56	176	203	1035	110	101	156	1135	44	313	374	1803	29	312	415	1789
Mn	21	102	72	226	56	74	155	625	110	37	87	685	44	82	143	662	29	78	104	475
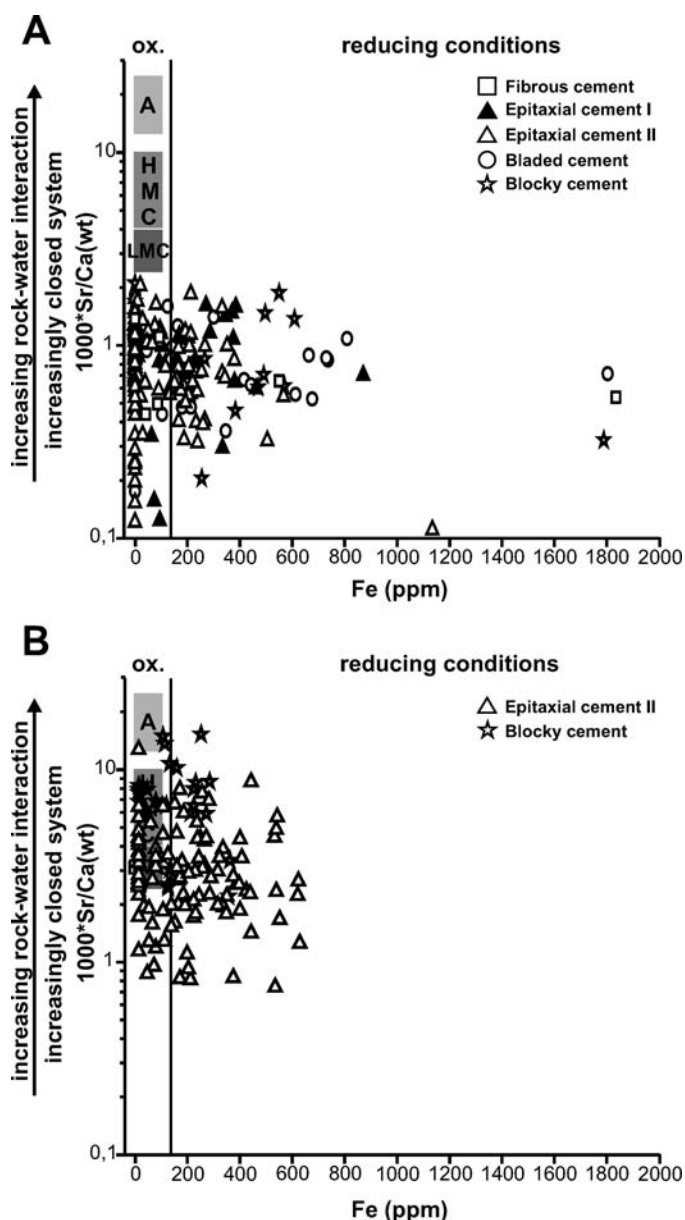
Fig. 11.— Cross-plot of the Fe and Sr content (normalized to Ca) of the different cement types and the possible precursor mineralogy. A, aragonite; HMC, high-Mg calcite; LMC, low-Mg calcite (plot by Brand and Veizer 1980, modified by Lohmann 1988). The Sr value shown on the vertical axis indicates the amount of rock–water interaction and is a measure of the openness of the system. The Fe content displayed on the horizontal axis points to the redox conditions of the system during cement precipitation (availability of Fe assumed). Note that the detection limit for Fe is 400 ppm. **A)** Data for the Dweira section (Maltese Islands); **B)** Results for the Ragusa section (Sicily).

The only partial dissolution of epitaxial cement I could also have been caused or favored by different amounts of Mg incorporated in the HMC. As evident from the transects measured trough Ep I and Ep II (Figs. 9, 10), the Mg content can vary significantly within a single cement, depending on the composition of the fluid from which it precipitated. Because the stability of HMC decreases with increasing Mg^{2+} content (Tucker 1990), a high amount of Mg incorporated in the calcite lattice might have caused or favored the partially congruent dissolution behavior of the epitaxial cement I.

Epitaxial Cement II, Bladed, and Blocky Cement.—Epitaxial cement II and bladed and blocky cements are grouped together because they all show well preserved internal zonations under CL and were therefore most probably precipitated as mineralogically stable LMC. This conclusion is also supported by the more equant crystal shapes, indicating minor incorporation of Mg in the calcite lattice (Given and Wilkinson 1985). The isotopic and trace-element compositions reflect the composition of the diagenetic fluid from which these cements precipitated and were not considerably altered during later stages of diagenesis.

The Dweira and Ragusa samples will be discussed separately because their geochemical results differ significantly. The LMC cements from Dweira (epitaxial cement II, bladed and blocky cement) display two different isotopic trends (Fig. 8A). The first is characterized by a covarying trend of low $\delta^{18}O$ (down to $-$ 5.0‰) and low $\delta^{13}C$ (down to -4.0‰) values, and the second is delineated by low $\delta^{18}O$ values (-1.0 to -3.0‰) with $\delta^{13}C$ values varying around zero.

The first trend suggests cement precipitation from meteorically derived fluids in an open system with little rock–water interaction (Lohmann 1988). Here the $\delta^{18}O$ values mirror the composition of the fluid and the $\delta^{13}C$ composition is not well buffered by limestone-derived CO_2, but rather displays the low $\delta^{13}C$ values of the soil-derived CO_2 (Allen and Matthews 1982). Cement precipitation in an open system is also supported by the trace-element data, which display low Sr and Mg values pointing to low rock–water interaction (Veizer 1983; Lohmann 1988). The elevated Fe values (Fig. 11) suggest cement precipitation under slightly reducing conditions (Veizer 1983), which would be consistent with the dysoxic meteoric environment. The meteoric trend is consistent with field data and depositional environments. All samples occur in the inner-ramp unit (U4) of the Dweira section, where at least one subaerial exposure surface (section height 40.5 m) was observed. This suggests that the meteoric influence occurred early in the diagenetic history.

The second trend, evident by low $\delta^{18}O$ values and $\delta^{13}C$ values varying around zero, could be caused either by meteorically derived fluids or by elevated temperatures during cement precipitation. If meteoric fluids were responsible for this trend, cement precipitation should have occurred in a more closed system with a high amount of rock–water interaction buffering the low $\delta^{13}C$ values of soil-derived CO_2 and increasing the Mg and Sr content (Lohmann 1988). Because both the Mg and the Sr contents are rather low (Table 2), cement precipitation most likely took place in a more open system with low rock–water interaction (Fig. 11A) unable to buffer low $\delta^{13}C$ values (Veizer 1983). Therefore, it is suggested that cement precipitation took place at elevated temperatures, which typically results in lower $\delta^{18}O$ values because the $\delta^{18}O$ fractionation is temperature-dependent (Moore 2001), but fractionation does not alter the $\delta^{13}C$ values. Because only a trend towards slightly lower $\delta^{18}O$ values is observed, a moderately elevated temperature (around 30°C; calculated using the equation by Epstein et al. 1953), corresponding to shallow burial in the marine burial diagenetic environment, is estimated (Choquette and James 1990).

The LMC cements from Ragusa (epitaxial cement II and blocky cement) show a different geochemical signal. The stable-isotope composition suggests precipitation from marine or marine-derived fluids (Fig. 8B) even if the values ($\delta^{18}O$: 0.0 to -1.0‰) are slightly lower than modern marine values. Nevertheless, they are consistent with $\delta^{18}O$ values of unaltered neritic skeletal material of Oligocene age ranging between $+2$ and -1‰ (Nelson et al. 1988a). Also the $\delta^{13}C$ values, ranging between $+0.9$ and -0.3‰, are in agreement with precipitation from marine or marine-derived fluids, taking place most probably in the marine-burial environment (*sensu* Melim et al. 1995).

The elevated Sr contents (Table 3) point to cement precipitation in a nearly closed to closed system with high rock–water interaction (Fig. 11B) as described by Veizer (1983). In such a system the diagenetic fluid is not removed but constantly changed due to the ongoing

TABLE 3.—*Trace-element composition (ppm) of the various cement types measured in the samples of the Ragusa section (Sicily); n, number of measurements; SD, standard deviation; Max, maximum value.*

Element	Epitaxial cement II				Blocky cement			
	n	Mean	SD	Max	n	Mean	SD	Max
Ca	105	386782	6264	394420	28	389823	1897	392447
Mg	105	3167	1514	6161	28	1615	538	3652
Sr	105	1190	781	4898	28	2658	1477	5971
Fe	105	205	174	618	28	139	129	511
Mn	105	119	106	567	28	63	62	204

dissolution–precipitation processes. Depending on the distribution coefficient (D), major and trace elements will be enriched or depleted in the diagenetic fluid. Sr and Mg will be enriched (D < 1); Fe and Mn will be depleted (D > 1) in the fluid. Therefore, later forming cements will precipitate from altered, e.g., Sr-enriched, fluids and will, with a constant D assumed, incorporate more Sr than the earlier-precipitated cements (Dickson 1990). Such a pattern is clearly visible in the cement transects measured (Fig. 10), where the Sr content increases steadily from the grain to the pore center.

Therefore, three diagenetic environments of cement precipitation can be depicted for the studied rocks. In the early marine environment generally small amounts of HMC fibrous, fibrous-bladed, and epitaxial cement I precipitated (1–2%, one exception with 30%). In the meteoric environment (inner-ramp part of the Dweira section) some cementation with LMC epitaxial cement II (2–8%) took place. The main cementation phase with LMC epitaxial cement II, bladed and blocky cement (up to 8% in Dweira, up to 30% in Ragusa) took place at both locations in the marine burial environment. The contrasting geochemical signals of the two locations are not caused by different diagenetic environments but are related to the degree of openness of the diagenetic system.

Cement Sources

In heterozoan carbonate assemblages (*sensu* James 1997), it has generally been assumed that cementation is delayed to later stages of diagenesis, because the low saturation state of the seawater and the predominantly stable primary mineralogy of the grains are not able to source large amounts of early cementation. Only in the burial environment (deeper than 400–500 m) pressure solution, acting as a principal cement source, promotes cement precipitation (Nicolaides and Wallace 1997a; Dodd and Nelson 1998).

Results from this study suggest that minor cementation occurred in the early marine and meteoric environments and most cementation took place in the marine burial environment. Pressure solution of calcitic grains can be excluded as a cement source because in both areas cementation of the volumetrically important epitaxial cement II clearly predates compaction (Figs. 6A, B, 7C, D). In addition, oxygen isotope data indicate cementation at only slightly elevated temperatures (around 30°C), not at higher temperatures as would be expected in the deeper-burial environment (Choquette and James 1990). This raises the question of cement source(s) in the different diagenetic environments.

Early Marine Environment.—In the early marine environment, primary HMC cements (fibrous, fibrous-bladed, and epitaxial cement I) in both settings precipitated directly out of marine pore waters. Even in waters with a low $CaCO_3$ saturation state biochemical activities can induce precipitation, especially in intraparticle pore spaces of biota such as foraminifers or bryozoans, or along the outer rims of echinoid fragments (Smith and Nelson 2003). Due to the high Mg/Ca ratio in seawater this

leads, depending on the substrate, to the precipitation of fibrous or epitaxial HMC cement (Given and Wilkinson 1985). The low abundance of these cements (about 3% of the rock volume) can be explained satisfactorily by this process.

The large amount of fibrous cement in the single Dweira sample (30%) also precipitated directly out of marine waters. However, to explain the abundance of early marine cement in a heterozoan setting another process is required. Nelson and James (2000) suggest that large amounts of early marine HMC cements can precipitate in heterozoan carbonate settings under special conditions including stacked, cross-bedded sand bodies, meter-scale, subtidal carbonate cycles, or in association with certain unconformities. In these settings cement precipitation is promoted by either high permeability or prolonged time lying at the sediment–water interface. The Dweira sample is associated with an unconformity, most probably with a condensed section (Knoerich and Mutti 2003), so the cement is likely due to prolonged exposure at the sediment–water interface.

Meteoric Environment.—Some epitaxial cement of the inner-ramp setting at Dweira precipitated in the meteoric environment (6–8%). The source for these cements was probably dissolution of aragonitic biota, as suggested by the occurrence of biomoldic porosity after aragonite shells (Fig. 6C). Since cement precipitation at Dweira occurred in an open system, no further hints of an aragonitic cement source, such as elevated Sr contents, are observed.

Marine-Burial Environment.—In both study areas, most of the cementation (primary LMC epitaxial cement II, bladed and blocky cement) occurred in the marine-burial environment. The source of these cements at Dweira is unknown because cement precipitation took place in an open system and the geochemical signature of the cement source was lost.

In contrast, at the Ragusa section cement precipitation took place in a nearly closed to closed system, retaining the geochemical signature of the cement source (Lasemi and Sandberg 1993). Therefore, the high Sr content in the LMC cements is suggestive of an aragonitic cement source. Since Sr can be depleted only during a dissolution–precipitation process because of its D < 1, the high Sr contents (up to 5900 ppm) measured in the LMC cements from Ragusa point to a cement source with a higher Sr content. Of the common sedimentary carbonate minerals only aragonite has such high Sr contents (up to 9400 ppm).

An aragonitic cement source could also explain the blocky morphology of the Ragusa cements. Generally, this morphology is interpreted as indicative of meteoric environments and is caused by the low Mg/Ca ratio in fresh water (James and Choquette 1990a). In our case a meteorically influenced cement source can be excluded because stable-isotope measurements display marine values. However, the shift towards low Mg/Ca ratio can also be caused by an aragonitic cement source (Melim et al. 2002). Because aragonite can incorporate less Mg in its lattice than calcite, a diagenetic fluid supersaturated with respect to calcite by the dissolution of aragonite would also have a low Mg/Ca ratio, favoring a more blocky cement morphology.

Origin of the Aragonite

The origin of the aragonite that acts as a cement source in the meteoric and marine burial environment is not well understood. In the Dweira section, open molds after aragonitic fossils (especially bryozoans and bivalves) indicate an aragonite cement source. In the Ragusa section this it not the case. Because precipitation in Ragusa took place in a more or less closed system, an internal source is needed. Yet, the observed biotic assemblage lacks primary aragonitic fossils, and no direct evidence for the existence of aragonitic grains is noted. However, oversized pore
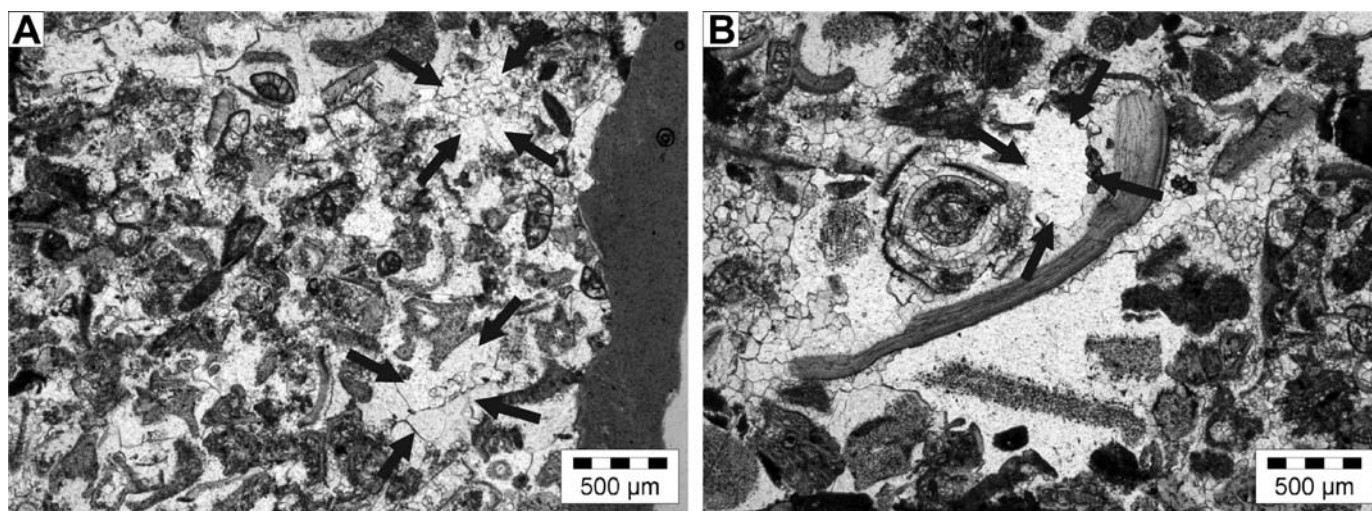
Fig. 12.—Photomicrographs of oversized pore spaces and filled "bio" molds in the samples from Ragusa (Sicily). **A)** Oversized pore spaces (arrows) with respect to the mean grain size of the sediment. **B)** Hardly visible "bio" mold (arrows) now filled with LMC epitaxial cement II.

spaces and molds, both now filled with blocky cement, are observed (Fig. 12). These oversized pores likely formed by dissolution of aragonitic grains but did not preserve the specific shape of the grain. This suggests that the current biotic assemblage is strongly biased towards calcitic fossils and that the aragonitic constituents were totally removed by dissolution.

Inasmuch as no molds after aragonitic fossils were observed, their dissolution must have occurred early, as described by Brachert and Dullo (2000) and Smith and Nelson (2003) for other heterozoan carbonate settings. This process has also been reported from other predominantly calcitic settings in the Ordovician, Silurian, and Jurassic (Cherns and Wright 2000; Wright et al. 2003; Palmer and Wilson 2004). In these studies the depletion of aragonitic biota took place either on the seafloor or during shallow burial, before any calcitic replacement of shells. The lack of molds is explained by rapid dissolution while the sediment was unconsolidated and lacked sufficient matrix strength to preserve the mold space against collapse (Cherns and Wright 2000). This could explain why molds and oversized pore spaces in the Ragusa samples are observed only in grain-supported lithologies, where the grain-supported fabric was able to stabilize the molds.

Another explanation why direct evidence of aragonitic components is scarce might be their small size. This could result either from intense reworking of grains or be a primary feature. Tunicate spicules, for example, are abundant sediment constituents in some recent heterozoan settings (James et al. 2005; Betzler et al. 2005), forming up to 36% of the sediment volume. Tunicates possess small aragonitic spicules in their tissues, which disintegrate upon death. Because these spicules are most abundant in the silt and finer grain-size fraction (Saxena and Betzler 2003) they are very prone to dissolution and leave no visible molds. Some or much LMC cementation in Ragusa could be explained by this aragonite source.

Controls on Aragonite-Sourced Cementation

We interpret that the main part of early cementation in the study area was sourced by aragonite dissolution. This raises the question why the amount of aragonite-sourced cementation is so different at the two locations (Dweira, mean 3%, Ragusa, mean 20%).

One possible explanation could be that the amount of aragonitic fossils present in the primary sediment was very different, therefore sourcing very different cement volumes at the two locations. The other possibility

would be that the primary amount of aragonite was the same in both locations but that it was preserved differently.

A low preservation potential for aragonite exists in lower-energy settings, where the accumulation of fine-grained organic matter is more likely as a result of less winnowing, and decay of this organic matter leads to an increase in CO_2 and to aragonite dissolution (Wright et al. 2003; Wright and Cherns 2004). Also, low sedimentation rates, typical of heterozoan carbonate settings, are unfavorable for aragonite preservation because dissolution at the sediment–water interface takes its effect (Smith and Nelson 2003).

In Dweira, sedimentation preferentially occurred under lower-energy conditions, as evident by the overall high amount of micrite, favoring the deposition of organic matter. The decay of this organic matter could have caused early aragonite dissolution directly on the sea floor, as described by Wright and Cherns (2004), Palmer and Wilson (2004) and James et al. (2005). This aragonite would have been lost as a later cement source.

In Ragusa, on the contrary, sedimentation took place under higher-energy conditions and sediment accumulation occurred more episodically (storm-related), with high sedimentation rates. So aragonitic components could have been buried rapidly and therefore could have been protected from early dissolution on the seafloor (Alexandersson 1979; Young and Nelson 1988; Smith and Nelson 2003), later acting as a cement source.

Diagenetic History

By combining detailed petrographic descriptions of the cements with the interpretation of the diagenetic environments, based on geochemical data, it is possible to establish diagenetic histories for the two study locations (Figs. 13, 14). Despite the different amounts of cement, the pathways appear quite similar.

In both areas, the diagenetic evolution starts with the early marine precipitation of HMC fibrous and epitaxial cement I directly from seawater. The shallowest-water environments of the Dweira section were exposed to meteoric fluids, which caused fabric-selective dissolution of aragonitic biota and neomorphism of HMC to LMC. Precipitation of LMC epitaxial cement II was sourced by the aragonite dissolution in this environment.

In the marine-burial environment, the main phase of dissolution, neomorphism, and cement precipitation took place. The process started with the fabric-selective dissolution of aragonitic biota. In Dweira, biomolds of aragonitic biota are preserved; for Ragusa this process is
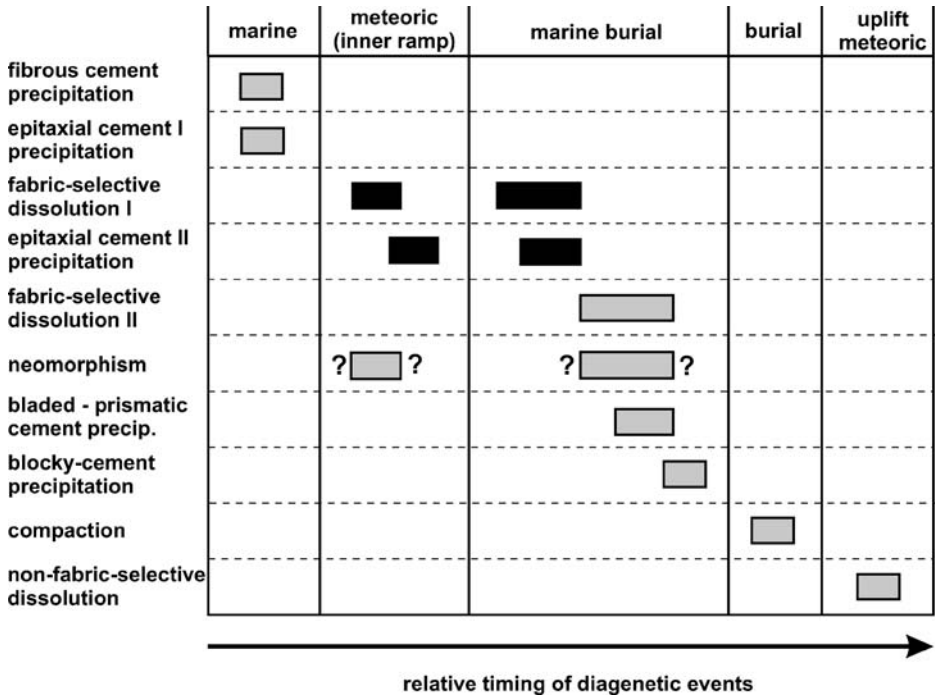
FIG. 13.—Paragenetic pathway of the diagenetic events recorded in the heterozoan limestones of the Dweira section (Maltese Islands). Note that the meteoric environment only refers to the paleobathymetrically shallowest part of the section (modified after Knoerich and Mutti 2003). Black bars = major processes, gray bars = minor processes, question marks indicate uncertain timing.

inferred, as discussed above. This phase of aragonite dissolution is the main source for the precipitation of LMC epitaxial cement II. This was followed by another phase of fabric-selective dissolution and neomorphism, preferentially affecting the HMC epitaxial cement I and HMC biota (Figs. 13, 14). The next phase of cementation included LMC bladed-prismatic and blocky cement precipitation, most probably also sourced by aragonite and HMC dissolution.

Upon entering the deeper-burial environment, mild chemical compaction, causing concavo-convex and some sutured contacts, occurred.

Subsequently, both areas were tectonically uplifted and reentered the meteoric environment, which caused some late, non-fabric-selective dissolution.

CONCLUSIONS

Our investigations highlight early cementation directly out of the seawater in the marine environment in heterozoan carbonates. This generally leads to the precipitation of minor amounts of HMC fibrous
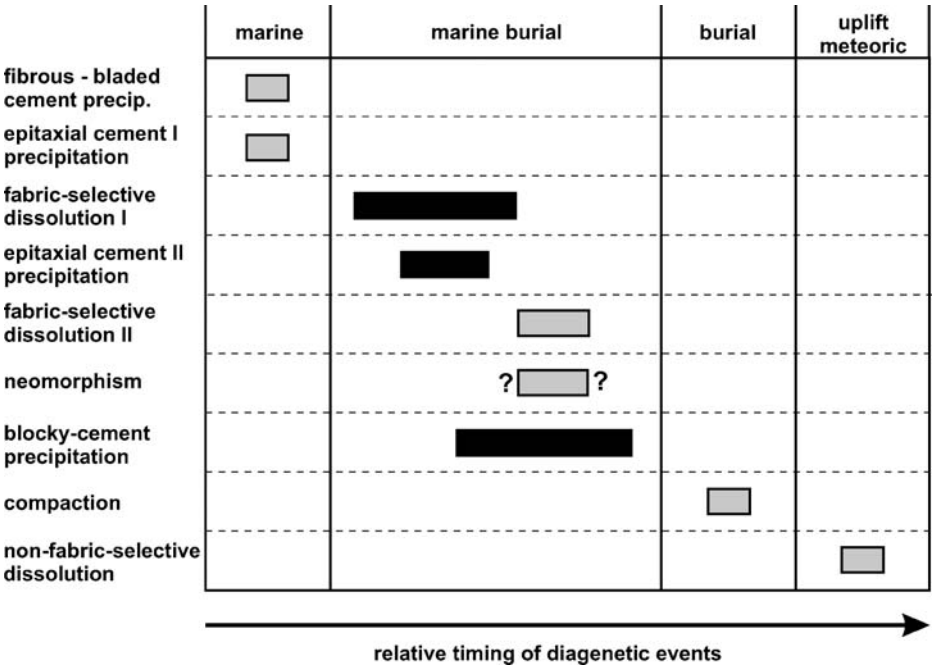

FIG. 14.—Paragenetic pathway of the diagenetic events recorded in the heterozoan limestones of the Ragusa section (Sicily). Black bars = major processes, gray bars = minor processes, question marks indicate uncertain timing.

and epitaxial cement, as observed in both locations. At specific stratigraphic positions (condensed sections?) these early marine cements can reach volumes of up to 30%, resulting in an early lithification of the rocks.

Meteorically influenced cementation with LMC (epitaxial cement II) can be observed depending on the aragonite content and the paleoenvironmental position of the rocks. As noted in the Dweira section, only the shallowest, inner-ramp unit was affected by meteoric fluids. This cementation in the meteoric environment is rather unusual for heterozoan carbonates since it is generally assumed that they are unaffected by meteoric alteration because of their stable, dominantly LMC mineralogy.

The main phase of mineralogical stabilization, dissolution, and cementation by LMC takes place near the seafloor in the shallow-marine burial environment. The source for the LMC cements most probably was the dissolution and neomorphism of early marine precipitated HMC cements (epitaxial cement I) and of aragonitic and HMC biota. This is especially distinct at Ragusa, where cement precipitation occurred in a nearly closed system, which allowed the preservation of the high Sr content of the precursor aragonite. In contrast to many previous studies, little cement precipitation took place in the deep-burial environment and pressure solution was unimportant as a cement source for the studied heterozoan carbonates.

These results have three implications: (1) The importance of early, shallow burial diagenesis for the chemical stabilization and cementation of heterozoan carbonates must be carefully considered. The marine-burial environment seems to particularly play an important role in this process. (2) This study underlines the importance of aragonitic components in heterozoan settings. By means of trace-element analysis it was possible to conclude that cementation was sourced by the dissolution of aragonitic components, even in rocks that show no petrographic evidence of former aragonite. It is suggested to carefully consider aragonite as a possible early cement source when dealing with heterozoan rocks. (3) In heterozoan limestones, secondary porosity formed by dissolution of aragonitic skeletons need not be preserved. In contrast to photozoan carbonates well lithified in the marine environment by early cementation, heterozoan carbonates lack this stabilization. Therefore, molds formed by dissolution often collapse, leaving no secondary porosity. Oversized pore spaces or filled molds can be preserved only in grain-supported textures, giving hints to formerly existing aragonitic components.

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