INTRODUCTION

Ooids, mainly calcareous, spherical to subspherical coated grains consisting of one to several nearly concentric cortices encrusting a nucleus, are ubiquitous features of (sub)tropical oceans throughout Earth's history (Flügel, 2004; Tucker, 2011). Although there is no commonly accepted elucidation for ooid genesis, processes such as accretion of fine particles around a nucleus while agitating on a soft substrate (Mei and Gao, 2012), abiotic precipitation from an ambient supersaturated water around a nucleus (Duguid et al., 2010) and organomineralization of a surface biofilm (Diaz et al., 2014; 2017; Li et al., 2017; Batchelor et al., 2018) have been diversely put forward. Ancient ooids are
valuable palaeoenvironmental proxies for water energy, temperature, salinity, seawater geochemistry and bathymetry (after Sandberg, 1975; 1983; Plee et al., 2008). Oolites, that is carbonate rocks formed predominantly by ooidal facies, form on marine carbonate platforms and ramps and make up a significant part of the world's carbonate hydrocarbon reservoirs (Flügel, 2004). The mineralogy of oolites and ooids is extremely diverse. Previous works describe low-Mg to high-Mg calcitic, aragonitic, dolomitic and chamositic (Van Houten and Purucker, 1984) ooids, as well as gypsum (gypsolites; Tekin et al., 2008) and halite ooids (Radwanski and Birkenmeyer, 1977; Kimberley, 1983). Even collophanitic ooids made solely of fine-grained phosphatic material have been reported (Barr, 2007).

Previous workers have used the mineralogy of ooids as a proxy for secular variations in ancient seawater chemistry and, in particular, the reconstruction of what was referred to as oscillating trends in calcite versus aragonite seas has largely been based on ooidal cortex mineralologies (Sandberg, 1983; Wilkinson et al., 1984; 1985; Opdyke and Wilkinson, 1990; Tucker and Bathurst, 1990a; MacKenzie and Morse, 1992; Cantrell, 2006). The present understanding is that high-Mg calcitic (>4 mol.% MgCO3; Stanley and Hardie, 1998) and aragonitic ooids form when Mg/Ca molar ratios in seawater are above a threshold limit of 2 (Stanley and Hardie, 1998; 1999; Stanley, 2006; Ries, 2010). Conversely, calcite ooids are mainly precipitated during periods of lower seawater Mg/Ca ratios. In this paper, the term ‘calcite’ is used for low-Mg calcite (<4 mol.% MgCO3, according to Stanley and Hardie, 1998).

Other works document the relevance of different compositions of marine ooid cortices in calcite versus aragonite seas to changes in global $\rho$CO2 (MacKenzie and Pigott, 1981; Sandberg, 1983; Algeo and Watson, 1995; Corsetti et al., 2006) during the Phanerozoic. Stanley and Hardie (1998) (supported by Broecker, 2014; Kimmig and Holmden, 2017), however, indicated that $\rho$CO2 is a viable control only over a narrow range and, within this, values are all greater than that of the present. For seawater of modern composition, these $\rho$CO2 values lead to supersaturation with respect to calcite while remaining undersaturated with respect to aragonite. For present-day seawater at 25°C, raising $\rho$CO2 from a low value of $10^{-15}$ to $10^{-1}$ atm lowers the MgCO3 content of precipitated high-Mg calcite from 12 to 8 mol.%, but does not cause precipitation of calcite instead of aragonite (Burton and Walter, 1991). Mineralogical variation in modern ooids is thought to be controlled mainly by short-term changes in the Mg/Ca ratio of the precipitating fluid (Kimmig and Holmden, 2017). Large fluctuations in atmospheric $\rho$CO2 and changes in the mineralogy of tropical marine carbonate sediments are, however, related to Pleistocene glacial/interglacial cycles (Algeo and Watson, 1995). The driving factor that links seawater chemistry and ooid mineralogy is mainly thermodynamics (Gibbs free energy; Heydari and Moore, 1994; Morse et al., 1997; Riding and Liang, 2005). Under elevated fluid Mg content, thermodynamics predicts that increasingly abundant Mg is incorporated into the calcite crystal lattice. Under more fluid Mg contents (above 150 mEq/L; Berner, 1975; Stanley, 2006), aragonite mineralogies are the more stable carbonate phase (Bischoff, 1968; Berner, 1975).

The focus of this paper is on coated grains from Middle Triassic deposits of the Island of Hydra, Greece (Muttoni et al., 1994; Richter, 1999; Wignall et al., 2012). In this locality, grain flows shed basins from unknown Triassic shoal-water settings, which are characterized by an uncommonly complex variety of coated grains, including carbonate ooids, oncoids, carbonate bioclasts and other allochemical particles (e.g. peloids, intraclasts). Among these, two specific types are of interest: (a) half-moon ooids and (b) micritic to microsparitic ooids. Revealed through micropetrography, half-moon ooids combine a former aragonitic cortex and a high-Mg calcitic nucleus (Carozzi, 1963). In contrast, an accretionary, concentric pattern in the cortical layers of micritic to microsparitic ooids is primarily made up of aragonite and high-Mg calcite (Tucker, 1984). In this study, these fabrics are explored as examples of synsedimentary changes in carbonate mineralogy, with the petrographic and diagenetic modifications of formerly bimineralic, ancient ooids documented and discussed.

The goals of this study are threefold: (a) to describe the shape and inner fabric of half-moon ooids and micritic/microsparitic ooids by integrating petrographic and cathodoluminescence (CL) evidence. This was done by describing the different types of coated grains present in thin sections from Hydra; (b) to construe the original composition of bimineralic coated grains based on the degree of preservation of the primary cortex and the subsequent cementation pattern; and (c) to investigate the significance of bimineralic, non-skeletal coated grains for ambient seawater geochemistry. Focus was placed on the environmental parameters that drive changes in ooid mineralogy. The results shown here are of general significance for those concerned with the use of ancient carbonate deposits as archives of past seawater properties.

2 GEOTECTONIC SETTING AND STRATIGRAPHY OF HYDRA ISLAND

Hydra, a Saronic Island of Greece, is a large, mainly carbonate province between the Saronic Gulf and the Argolic Gulf (65 km south-southwest of Athens; Figure 1). The sedimentary succession of Hydra, spanning from the Early Permian to the Late Jurassic, is situated within the Subpelagonian Zone of Eastern Greece (Angiolini et al., 1992; Muttoni et al., 2004). The Subpelagonian Zone is the NNW-SSE trending, non-metamorphic structural zone of the Central Hellenides that forms the south-western margin of the Pelagonian Platform boundary to the Pindos (Figure 1). The Central
Hellenides is a multiphase deformed branch of the broader Alpine orogenic belt in Laurasia (Adamantios, 2018). The Subpelagonian Zone is characterized by a Triassic–Jurassic carbonate platform overlying a volcano-sedimentary Permo–Triassic complex and underlying an Upper Jurassic ophiolite mélangé formation (Papanikolaou and Papanikolaou, 2007). Hydra’s succession, at the southern end of the Subpelagonian Zone, forms a >2,500 m thick lithostratigraphic series arranged in four main thrust sheets and dissected by three NW-SE transcurrent regional faults (Figure 2). The northward homoclinal dipping of the succession caused the outcropping of the Permian Series along the southern side of the island and the Jurassic along the northern. The formation of multiphase solids with internal breccias and mass flows is associated with strike-slip faults activated during the Hellenic rifting phase. The stratigraphic sequence documents the build-up, fracture and subsidence of three carbonate platforms (Richter, 1999). Permian shallow-marine deposits, the first carbonate platform, are overlain by Scythian gravity flows and deep-water limestones of the lowermost Anisian substage (Figure 2). Reef limestones characterize the formation of a second carbonate platform of Middle to Upper Anisian age. Massive tectonic activity at the second carbonate platform is demonstrated by the formation of tuffs and ammonoid-bearing (Trinodosus Zone; Brack and Rieber, 1993) red limestones following Illyrian-aged mass flows. This event caused strong facies differentiation, with basinal (often fine-grained clastic) deposits on the southern thrust sheet of Hydra and shallow-water carbonates on the northern. During the evolution of a third carbonate platform of Upper Triassic age, deposition of laminated cherty limestones suggests deep-water sedimentation in the south, while reef limestones document the growth of a carbonate platform on the northern thrust sheet. The transitional area of this reef basin complex is represented by well-bedded fore-slope limestones containing gravity flows with shallow-water bioclasts.
2.1 Study section

The Middle Triassic oolitic limestones of the NW–SE trending Agios Nikolaos profile situated along the southern coast of Hydra between Kap Rigas (to the east) and Klimaki (to the west), south of the solitary Miras homestead (Figure 1), were investigated. This section is a major subset of the second carbonate platform in Hydra which incorporates several series deposited during the Early to Middle Triassic (Figure 2). Stratigraphically, the section is located within the tectonically suppressed Upper Mass-flow Series of the Early to Middle Anisian age (Angiolini et al., 1992) and is close to the boundary of Late Anisian limestones and base limited by the Lower Mass-flow Series which is known to span the Permian–Triassic in other localities on Hydra (Shen and Clapham, 2009). The abundance of grain-supported, mud-poor limestones suggests the prevalence of high-energy conditions across much of the broad carbonate platform accommodating the study section (after Cantrell, 2006).

The lithified, ooidal facies (Agios Nikolaos section) of (Upper Spathian ?) Early to Middle Anisian age (Angiolini et al., 1992) is organized into two parts separated by a fracture zone (Figure 3). The first part of the section, from the coastline to the beginning of the fracture zone, shows bed thicknesses ranging from medium-bedded (15–30 cm) to heavily weathered, massive, grey oolitic limestones (>100 cm). This part of the section accommodates well-developed karst features, including limestone pavement and dissolution vugs.
FIGURE 3  Stratigraphic column of the study section, Agios Nikolaos, west of Kap Rigas, measured within the ooid grain flow. The section is divided into eight major facies units, represented by colour codes (refer to Table 1 for details of these facies). A short description is given in front of each facies. AN-1 to AN-15 mark the samples taken from the profile. AN, Agios Nikolaos
The second part of the section, above the fracture zone, is mostly characterized by fractured, massive, cream limestones; however, the first few metres exhibit alternating well-bedded limestone (10–20 cm thick) and brown marl. The study section is divided into eight major facies (Table 1; Figure 3) on the basis of observed sedimentary structures and textural features, including the proportion of carbonate matrix and larger components (skeletal and non-skeletal grains).

Based on an assessment of the depositional structures (especially the occurrence of breccia), bed thickness and the presence of mud, Richter (1999) concluded that the Middle Triassic oolite outcrop in Hydra represents a grain flow that formed during a laminar, highly concentrated, cohesion-less flow event. This grain flow has carbonate coated grains formed syndepositionally, indurated in an unknown source area outside the Island of Hydra together with detrital grains, and transported basinwards to Hydra (Richter, 1983; 1999). Angiolini et al.'s (1992) field observations show that only the conspicuous micrite bands within the grain flow are autochthonous (Figure 3). These micrite bands were re-arranged after the interruption and erosion of the micritic series from the Permian–Triassic transition by the grain-flow deposit rich in allochthonous coated grains (Angiolini et al., 1992).

### METHODOLOGY

The results of this study are based on the petrographic investigation of 30 polished thin sections. Petrographic arguments were emphasized because the small size of the different coated grains and their complex internal structure inhibited an in-depth geochemical analysis. Additionally, alteration occurring from the Middle Triassic to the present seems to have been thorough enough to remove the signatures of the original mineralogy from the coated grains. Hence, this study focused on texture and mineralogy using optical microscopy (OM) and CL. One set of thin sections (15 thin sections) stained with alizarin red-s and potassium ferricyanide and one unstained set (15 thin sections) were studied. All 30 thin sections came from slabs cut perpendicular to the bedding.

#### Thin-section analysis

The shape and internal fabric of coated grains were analysed by OM. In particular, the cortical specification and intragranular cementation patterns were considered key for characterizing the formerly bimineralic ooids. A polarizing Zeiss Microscope, model 47 30 34-9902, was used for the

### TABLE 1 Description of major carbonate facies, Agios Nikolaos study outcrop

| No. | Lithofacies type               | Structure                          | Textural characteristics                                                                 |
|-----|--------------------------------|------------------------------------|------------------------------------------------------------------------------------------|
| 1   | Grey ooid grainstone           | Medium to thick-bedded             | Different sand-sized carbonate ooids, minor fractions of peloids, micritic/microsparitic clasts and oolitic intraclasts |
|     |                                | Karstic pavement                   | Both grains and calcite cement cut through by stylolites                                  |
|     |                                | Stylolitic                          |                                            |
| 2   | Yellowish grey ooid grainstone | Thick-bedded to massive            | Sand-sized carbonate ooids, minor micritic/microsparitic clasts, mollusc bioclasts and oolitic intraclasts |
|     |                                | Heavily weathered                   |                                            |
|     |                                | Vuggy                              |                                            |
| 3   | Yellowish dark grey ooid grainstone | Medium to thick-bedded           | Carbonate ooids, smaller fractions of mollusc bioclasts                                  |
|     |                                | Bioclastic                          |                                            |
|     |                                | Dissolution vugs                    |                                            |
| 4   | Purplish grey ooid grainstone (to packstone) | Thick-bedded to massive    | Carbonate ooids, smaller fractions of oncoids, minor mollusc bioclasts, micritic clasts and peloids |
|     |                                | Karstic pavement                   |                                            |
|     |                                | Dissolution vugs                    |                                            |
| 5   | Reddish pink ooid packstone    | Thick-bedded                        | Deformed carbonate ooids, limestone breccia fragments, minor mollusc bioclasts and oncoids |
|     |                                | Breciated                           |                                            |
| 6   | Brownish grey ooid grainstone (to packstone) | Medium to thick-bedded       | Carbonate ooids, minor detrital quartz and peloids                                        |
|     |                                | With well-bedded marl intercalations | Deformed carbonate ooids                   |
| 7   | Dark cream ooid grainstone (to packstone) | Medium-bedded to massive         | Carbonate ooids, minor fractions of mollusc bioclasts, peloids and oncoids                |
|     |                                | Moderately fractured                | Recrystallized carbonate groundmass        |
| 8   | Light cream ooid grainstone (to packstone) | Massive                            | Carbonate ooids, minor bioclasts, micritic/microsparitic clasts and peloids               |
|     |                                | Heavily fractured                   | Both grains and cement (and/or matrix) cut through by stylolites                           |
|     |                                | Moderately weathered               | Spar-filled microfractures developed throughout the unit                                   |

*Note: The abbreviation represents: No. = the identification number of facies colour codes presented in Figure 3.*
thin-section studies. Photomicrographs were taken using a Canon EOS 60D digital reflex camera attached to a Leica DM 4500 P microscope.

3.2 | Cathodoluminescence

A hot-cathode CL microscope (HC1-LM) was used to investigate the diagenetic modification and cementation of coated grains. Polished, unstained thin sections were sputter coated with a very thin gold layer to make their surfaces electrically conductive and to prevent charging of the samples. Operation conditions were as follows: 14 kV gun potential, unfocused beam diameter of 4 mm, \(10^{-5}\) mbar vacuum and 0.1 mA beam current. The CL spectra were obtained using an EG&G triple-grating spectrograph with a liquid-N\(_2\) cooled CCD-detector (Götze et al., 1999), which allows the documentation of very short-lived and dull luminescence. A Kappa DX30C video camera linked to the CL microscope recorded the digital images.

4 | DATA PRESENTATION

4.1 | Petrography

The investigated oolite outcrop displays a varying assemblage of ancient, calcitized coated grains which are often poorly sorted in the framework of grain-supported (oid backstone and grainstone) fabrics (Figure 4). Under OM and CL, there is a diversity of well-developed ooid morphologies, ranging in size from 0.1 to 2 mm (Table 2), for the grain-flow mass at the Agios Nikolaos section. These concentric features are commonly circular or elliptical (some with unregulated structures), containing well-preserved multiple fine cortical laminations encrusting a small carbonate or clastic nucleus (Figure 4). In other ooidal morphologies, the concentric lamellae have been mostly dissolved and recrystallized (half-moon ooids and oomoulds in Figure 4A,C through O). The cement is composed of anhedral to subhedral crystals, ranging in size from 0.1 to 1 mm. In interparticle spaces, it represents the pore-occluding stage of cementation occurring after Type A cements form. Blocky spar is also present in fractures and veins, and cross-cuts earlier cement phases.

Type B: This cement is a blocky calcite spar. It is the dominant component that fills oomoulds, mollusc bioclasts, some oncoids and the upper coarse part (spar-filled) of the half-moon ooids (Figure 4). The cement is composed of well-developed anhedral to subhedral crystals, ranging in size from 0.1 to 1 mm. In interparticle spaces, it represents the pore-occluding stage of cementation occurring after Type A cements form. Blocky spar is also present in fractures and veins, and cross-cuts earlier cement phases.

Type C: This typical microsparry cement forms the finer-grained lower part of the studied half-moon ooids (Figure 4A,C through O), and records the first stage of cementation inside this coated grain. It also forms the microsparry laminae in the studied micritic to microsparitic ooids (Figure 4A,D through O). The cement is composed of anhedral crystals that display a mosaic microfabric.

The studied ooidal microfacies also accommodate other diagenetic phases, including mechanical grain compaction/deformation (e.g. Figure 4F,G,L,M) and grain-to-grain pressure solution features such as microstylolites (Figure 4D,E,N,O) and grain distortion (e.g. Figure 4N,O). With respect to the ooidal coated grains present in thin sections from Hydra, the morphological features and the inner architecture of the cortex were used to identify different types of ooids. In addition, oncoidal and mixed oncoidal–ooidal features have been found, although the focus here is on two specific types, half-moon ooids and micritic to microsparitic ooids.

4.1.1 | Coated grains – Description

Half-moon ooids

Half-moon ooids from this study are circular to oval-shaped coated grains exhibiting an exotic, internal geopetal facet consisting of porosity-occluding, intragranular blocky calcite cement and the residual parts of largely destroyed cortices and nuclei (Figure 5). This bi-partite structure is encrusted with a thin, micritic shell (envelope) with an average thickness of 0.02 mm for the analysed samples. Judging from quantitative estimates of two-dimensional thin sections, half-moon ooids constitute about 8.5% of the analysed coated grains. The regular external shell coating half-moon ooids continues unaffected by the impact of the penetration of the interstitial groundmass. Rare examples of ooids with discontinuous outer shells, parts of which have obviously ruptured and pushed inside the oomouldic cavity (Figure 5K,L), are the only exception. A concentrically zoned outer shell (closely spaced) coating the bi-partite structure was observed on some occasions, exhibiting a few microcrystalline rings of variable
are highly variable (0.15–2 mm; Table 2). The growth index
of (sometimes with a detrital nucleus) while ooid dimensions
ble thickness. The nucleus is subspherical and generally peloi-
that consists of nearly concentric, regular laminae with varia-
tic to microsparitic ooids show a complex, zonal cortex
boundary between zones 5 and 6 (Figure 6F).
These luminescent zones are regular, except for an irregular
zone and (f) a very dull zone. The crystal boundaries between
succession of zonations (Machel, 1985), including (a) a dark
The sequence exhibits six subtle leading phases based on a
paragenetic sequence filling the intragranular cavity is a clas-
pattern observed in the half-moon ooids. The luminescent
calcite cement fills between the coated grains (CL) (Sample AN-9). (G) Same view of F
under PPL (Sample AN-9). (H) An assemblage of various coated grains, including micritic to microsparitic ooids and their broken parts, micritic ooids, half-moon ooids, ooids with detrital nuclei, ovoidal fragments and bioclasts in a grainstone (CL) (Sample AN-9). (I) Same view of H
under PPL (Sample AN-9). (J) Micritic to microsparitic ooids, half-moon ooids, oomoulds, ooids with a quartz nucleus, micritic ooids, bivalve fragments and micritic/microsparitic clasts in a packstone (CL) (Sample AN-12). (K) Same view of J under PPL (Sample AN-12). (L) Micritic to microsparitic ooids, their fragments, and deformed types, half-moon ooids, cemented oomoulds, ooids with a quartz nucleus, mollusc shells and micritic/microsparitic clasts in a grainstone bound by crystals of sparry calcite cement (CL) (Sample AN-14). (M) Same view of L under PPL
(Sample AN-14). (N) Oolite grainstone with micritic/microsparitic ooids and their distorted examples, half-moon ooids, micritic ooids, oomoulds, micritic/microsparitic clasts and bioclasts. Also, stylolites cut through upper left part of the section (CL) (Sample AN-14). (O) Same view of N under PPL (Sample AN-14). Arrows: a, l, s, v, x) half-moon ooids. b, q, t) micritic ooids. c, j, u) cemented oomoulds. d, k, m, w) micritic to microsparitic ooids. e, f, g) asymmetrical, circular and elliptical oncoids, respectively. h, o) ooids with a quartz nucleus. i, n) mollusc fragments. p, r) broken micritic/microsparitic ooid and oncoid, respectively
Based on CL observations, the blocky calcite spar in the
g eo petal ooids shows the thorough zonation of a cementa-
tion sequence. The concentric zonation (Figure 6), that is
 compositional changes parallel to the growth morphology of
blocky calcite, makes up the most widespread cementation
pattern observed in the half-moon ooids. The luminescent
paragenetic sequence filling the intragranular cavity is a clas-
micrite/microsparite hardly recognizable by OM
(Figure 7E,G). However, the micron-scale laminae of mi-
critized grains were recognized to some extent under CL
(Figure 7F,H). Very small superficial ooids encrusted by a
single thin micritic/microsparitic envelope are observed in
rare examples (Figure 7I). In such cases, the cortical lamina-
tion cannot be distinguished from the nucleus and the central
lamination immediately around it. In particular, the radius of
this pseudocore is commonly far more than the thickness of
the outer shell (growth index between 1.9 and 7.7).

**Micritic to microsparitic ooids**

As seen in Figure 7A through D, the round to oval-shaped
micritic to microsparitic ooids show a complex, zonal cortex
that consists of nearly concentric, regular laminae with variable
thickness. The nucleus is subspherical and generally peloi-
dal (sometimes with a detrital nucleus) while ooid dimensions
are highly variable (0.15–2 mm; Table 2). The growth index
ranges from 1:3 to 1:1, generally representing a thick cortex.
Micritic to microsparitic ooids were the most abundant type
of coated grain (ca 42%) found in the grain flow (Figure 4).

Most of the cortices observed were wholly composed of
concentric laminations, alternating between the far better de-
veloped micritic laminae and subordinate microsparitic rings
(Figure 7A through F). In the thin sections, micritic laminae
appear dark, featureless and microcrystalline, whereas the
microsparitic layers are white to milky-coloured and coarser
grained. In most of the samples analysed, the micritic and
microsparitic lamellae are distinguished as different lumino-

cer dolomite, the outer shell (growth index between 1.9 and 7.7).

**Cemented oomoulds**

As seen in Figure 8A,B, the studied cemented oomoulds are
round to oval features developed from primary ooids whose
cortical layers and nucleus were both dissolved, and the for-
mer fabric was replaced by blocky calcite. In thin section, a
dark, thin layer (mean thickness 0.01 mm) coats an oomould
filled by blocky calcite cement crystals. The diameters of the
investigated oomoulds reach between 0.2 and 1.6 mm, the average being 0.9 mm. However, both oomouldic porosity (nucleus and some cortical laminae dissolved) and oomouldic cores (dissolved core) were observed to occur in ooids with a thick, multi-layered, zonal cortex. Cemented oomoulds make up to 32% of the coated grains in the microfacies analysed.

**In situ calcitized ooids**

The small, spherical, in situ calcitized ooids (Figure 8C,D) are the least abundant of the coated grains studied (Table 2). The cortical fabric includes a concentric arrangement of regular laminae made up of brick-like calcite crystals enclosing a nucleus which is often dissolved and filled with intragranular cement. The nuclei sometimes include undissolved, peloidal particles or intraclasts. The ratios of nucleus diameter to cortical thickness lie between 1:2 and 2:1.

**Micritic ooids**

Micritic ooids exhibit the most variability in grain sphericity among the coated grains studied (Table 2). The diameter of micritic ooids vary between 0.1 and 1.2 mm. As seen in Figure 9A,B, they are micritic throughout and lack an identifiable nucleus. However, in rare cases, a darker micritic nucleus can be distinguished from a lighter cortex consisting of many vaguely visible, very thin, concentric laminae. The $d/t$ ratios range between 1:4 and 1:1. Micritic ooids are particularly noticeable in grainstone microfacies (e.g. Figure 4A,F through I,N,O), and they constitute about 11% of the coated particles studied.

**Oncoids**

The oncolitic nature of these nodular coated grains was recognized based on their irregular cortices (Figures 4B and 9C,D). The studied oncoids are characterized by more or less concentric, partially overlapping, zonal, cortical laminae encrusting a lithoclastic (sometimes bioclastic) nucleus. In thin section, oncoids are more variable in size than any other coated grain (Table 2). The thickness of the accretionary, complex cortex remains more than the diameter of the nucleus (the $d/t$ ratio ranges between 1:4 and 1:2). Most oncoids are spherical, but asymmetrical subtypes are also present. The asymmetrical oncoids range in size from 1.8 to 4.1 mm, exhibit downward and upward thickening as well as lateral elongations, and their laminae vanish within the surrounding sediment. Oncoids constitute up to 4.5% of the coated grains studied.

### TABLE 2

Various, ancient, calcitized ooidal and oncoidal coated grains observed in the analysed samples—cortical fabric and microscopic parameters

| Coated-grain type            | Cortical microfabric            | Diameter (mm) | $n^b$ |
|------------------------------|---------------------------------|---------------|-------|
|                              |                                 | Min. | Max. | Mean | $d/t^a$ | Min. | Max. | Mean | Abundance (%) | Sphericity |
| Half-moon ooid               | Regular, outer shell            | 0.35 | 1.9  | 1.13 | –       | 0    | 20   | 5.5  | 8.3           | 0.75–0.93  |
| Micritic to microsparitic ooid| Regular, concentric laminae     | 0.15 | 2    | 1.08 | 0.3–1 | 2    | 55   | 28.2 | 42.1          | 0.67–0.95  |
| Cemented oomould             | Regular, outer envelope         | 0.2  | 1.6  | 0.9  | –      | 1    | 65   | 21.6 | 31.9          | 0.73–0.93  |
| In situ calcitized ooid       | Regular, concentric laminae     | 0.15 | 1.1  | 0.63 | 0.5–2  | 0    | 3    | 0.7  | 0.5           | 0.79–0.95  |
| Micritic ooid                 | Very thin, concentric laminae   | 0.1  | 1.2  | 0.65 | 0.25–1 | 1    | 40   | 7.7  | 10.8          | 0.61–0.94  |
| Oncoid (Vadoid)              | Irregular, concentric laminae   | 0.75 | 4.6  | 2.68 | 0.25–0.5 | 0    | 50   | 2.9  | 4.4           | –          |

$^a$The growth index, i.e. the ratio of the nucleus diameter ($d$) to the thickness of cortex ($t$).

$^b$Number of coated grains counted per thin section using point counting method. The point counting was done on photomicrographs of thin sections using a two-dimensional point grid following Flügel (2004). Every grain (or matrix and cement) which fell under a grid point was counted. The estimated volume of a grain type is proportional to the counted number of grains of that type; 30 thin sections were point counted.

5 | **INTERPRETATION**

5.1 | **Coated grains**

5.1.1 | **Half-moon ooids — Mineralogical remarks**

Petrographic analysis suggests that the half-moon ooids originally consisted of nearly concentric layers of aragonite coating a carbonate core (nucleus and the central concentric rings developed immediately around it) thought to have been made up of high-Mg calcite. The aragonitic composition of the primary ooid cortex is evidenced according to criterion 3 of Sandberg (1983), implying the growth of coarse drusy calcite without aragonite relics inside the oomouldic pore. The existence of a formerly aragonitic cortex may also be suggested by the development of a non-luminescent zone within the sparry cement filling the half-moon ooids. Non-luminescent sparry calcite which replaces aragonite cement in a low water/rock burial system inherits the CL-derived signature of the primary mineralogy, in particular the low Mn and Fe contents (Given and Lohmann, 1985; Major et al., 1988). The luminescence of calcium carbonate minerals depends mainly on the abundance of Mn$^{2+}$ activators, whereas the presence of Fe$^{3+}$ has a fundamental quenching role due to this ions ability to suppress luminescence (Habermann et al., 1998). However, the calcite
cement would not necessarily be wholly related to the chemical composition of the primary mineral if dissolution occurred on a relatively large scale (e.g. destroying primary ooid cortices) (after Johnson et al., 2016). Luminescence of calcite cement, in addition to an original mineralogy, is influenced by processes that produce variations in pore-water chemistry
during burial, including varying redox potential, organic matter degradation and pressure solution (Machel, 1985; Johnson et al., 2016). Therefore, it remains uncertain to what extent the non-luminescence of blocky calcite in the half-moon oolites reflects the chemistry of the fluid that dissolved the cortex-forming primary aragonite. The original high-Mg calcite of the ooid core transformed via recrystallization into the micritic fabric of the calcitic half-moon ooids. This fine-grained calcitic zone appears iron rich (stained purple with alizarin red-s and potassium ferricyanide; Figure 5C). The Fe enrichment (ferroan calcite lamellae) may be indicative of a radial high-Mg calcite precursor transformed to calcite under anoxic conditions during deep burial (Richter and Füchtbauer, 1978; Veizer, 1983; Flügel, 2004).

Examples of formerly bimineralic, ancient, calcitized half-moon ooids with fabrics closely resembling those described here have been reported by a number of workers (Choquette, 1955; Carozzi, 1963; Mazzullo, 1977; Richter, 1983; 1999; Chatalov, 2005). Richter (1983) and Chatalov (2005) in particular described half-moon ooids from the Middle Triassic (Anisian) limestones of Episkopi (Greece) and Western Balkanides (Bulgaria), respectively. Their investigated ooids show a bi-partite fabric developed from primary, concentric laminae of tangential aragonite that coated a high-Mg calcitic core which probably exhibited radial fabric. This presumed primary mineralogy agrees with the distinctive distribution of originally bimineralic ooids in the Anisian strata on a global scale (Wilkinson et al., 1985). A thorough correlation between the original ooid mineralogy, primary microfabric, and the interpreted high-energy environment of formation has been emphasized by these studies. The Late Pleistocene bimineralic ooids from the Florida continental shelf (Major et al., 1988) reveal a concentric fabric consisting of a core of radial high-Mg calcite and cortical layers of tangential aragonite which resemble the original fabrics of ancient ooids described by the above-cited works (Figure 10A,B). The primary fabric in the half-moon ooids from Hydra is therefore assumed similar to those of the un lithified, bimineralic ooids from the Florida Shelf and the primary microfabrics of the Middle Triassic ooids from Episkopi (Richter, 1983) and the Western Balkanides (Chatalov, 2005; Figure 11).

FIGURE 6 Concentric CL zonations of sparry phase, changing from non-luminescent to bright, filling half-moon ooids. A through D: Normal zonation; numbers 1 through 6 mark the location of concentric zones. E: Rhombohedral zonation; note that the non-luminescent zone 1 is missing. F: Irregular crystal boundary between the two dull-luminescent zones 5 and 6; other boundaries of luminescent zones are regular.
5.1.2 | Half-moon ooids — Formation mechanism

Following the protocol outlined by Carozzi (1963), immediately after deposition in Hydra and before the completion of compaction, the aragonitic cortex was predominantly dissolved away by a fluid that would have to be undersaturated with respect to aragonite (no preservation of primary cortical fabric). According to a number of workers (Melim et al., 1995; 2001; Chorns and Wright, 2000; 2009), the dissolution of aragonitic cortices does not require meteoric fluids undersaturated with respect to aragonite, but can occur spontaneously during shallow burial in exclusively marine pore fluids. However, the partial development of karst structures in the studied limestones indicates that meteoric waters likely contributed to the initial diagenesis. Therefore, uncertainty will remain whether the fluid that dissolved the primary cortex was marine. The dissolved cortex left behind the less-soluble residue of a few cortical laminae and the remnants of a micritic high-Mg calcite
core (partly dissolved). The less-dissolved components collapsed in layers, which involved significant irreversible deformation, to the bottom of the mouldic cavity after the diagenetic dissolution of the more-soluble components. The plastic deformation of the remaining cortical layers and the core clearly shows that both were still soft during the onset of mineralogical transformation, whereas the thin outer shell coating the primary ooid was already indurated prior to deposition in Hydra. This interpretation assumes that the outer envelope of the ooid was an ooid laminae possibly recrystallized to calcite before cortical dissolution and plastic deformation. If the internal set of less-soluble rings and the core were indurated at the same time as the outer envelope, traces of primary microfabric would have been preserved in the half-moon ooids. Because the outer shell of the half-moon ooids is presently a micritic calcite with microscopic features resembling those of the fine calcitic part of the geopetal infillings, this situation suggests that the calcitized, microcrystalline calcite was rapidly recrystallized at the end of the oolitization process. Otherwise, the micritic shell would have been plastically deformed at the same time the insoluble cortical rings and core, possessing the same composition, were deformed during early diagenesis. The destruction of cortical laminae, which partly collapsed over the less-soluble part of the micritic carbonaceous core, resulted in an appreciable loss of volume and larger intragranular pore space. This process, together with deformation of the core remnants and collapsed aragonitic lamellae, resulted in an invisible ooid nucleus (i.e. a residual micritic nucleus at the bottom of the oolitic pore), and formation of the fine-grained part of the geopetal fabric. This fine micritic/microsparitic fabric that forms the bottom of the half-moon ooids is demonstrably a product of recrystallization of core components (involving the least interstitial dissolution; Newell et al., 1960). However, it is also partly influenced by interstitial precipitation (partial presence of microsparite crystals within the micritic part of the geopetal structure) during burial. The origin of half-moon ooids with eccentrically displaced, partially dissolved nuclei visible inside the bottom part of the geopetal cavity is attributed to the selective, aggrading recrystallization of the ooid nucleus during a period of burial diagenesis (Mazzullo, 1977).

The empty part of the generated intraoooid void was later filled with interlocking calcite crystals showing the characteristics of cavity-filling cement (for example, Figures 5 and 6). The growth of these cavity-filling sparry crystals occurred as pore fluids saturated with respect to calcium carbonate percolated through the sediment post-lithification or following burial.

5.1.3 | Micritic to microsparitic ooids — Mineralogical remarks

Petrographic examination suggests that the alternating micritic and microsparitic (sometimes pseudospar) ooidal laminae were developed in a marine environment from layers of high-Mg calcite and aragonite, respectively, coating a high-Mg calcite nucleus. The precursor aragonite composition of the microsparitic layers is particularly documented by the growth of microsparite mosaics without aragonite relics (Sandberg, 1983). The precursor high-Mg calcite composition of the micritic calcite laminae is evidenced by the recrystallization of microcrystalline calcite. If the micritic laminae were primarily aragonitic, the relics of precursor aragonite would have likely been retained in the replacing microcrystalline calcite (i.e. micritization of aragonite, Tucker, 1990). The development of micritic/microsparitic ooids from a bimineralic cortex is further justified by an obvious distinction between the luminescent behaviour of micrite and microsparite (Figure 7B). The non-luminescence of microspar crystals reflects low Mn contents which may be inherited from an aragonite precursor, while the bright luminescence of microcrystalline calcite may reflect the high Mn and low Fe levels of a high-Mg calcite precursor (Given and Lohmann, 1985; Major et al., 1988).

The formerly bimineralic nature of micritic to microsparitic ooids has been reported on in a number of papers...
describing ancient ooids with cortical fabrics identical to those investigated here (Wilkinson et al., 1984; Chow and James, 1987; Algeo and Watson, 1995; Richter, 1983; 1999; Chatalov, 2005; Lehrmann et al., 2012). Among these, the works of Richter (1983) and Chatalov (2005) concentrate especially on micritic/microsparitic ooids of Anisian age in Episkopi, Greece and the Western Balkanides, Bulgaria, respectively. These researchers have revealed that micritic/microsparitic ooids were formerly composed of concentric laminae of high-Mg calcite showing radial fabric and/or micritic fabric alternating with tangential aragonitic layers. This interpreted original mineralogical composition is consistent with a marked distribution of formerly bimineralic micritic/microsparitic ooids in the Anisian rocks on a global scale (Wilkinson et al., 1985). Analogous examples from the Holocene of Baffin Bay, Texas (Land et al., 1979) likewise show the coexistence of concentric lamination of radial magnesian calcite and tangential aragonite in ooidal cortices (Figure 10C,D). Based on the analogous cortical fabric and mineralogy of the aforementioned ancient and modern ooids, alternating radially oriented high-Mg calcite and tangential aragonite lamellae is the assumed likely primary cortical fabric for the studied micritic/microsparitic ooids (Figure 11).

5.1.4 | Micritic to microsparitic ooids — Formation mechanism

Previously aragonitic layers were altered to microsparry calcite crystals through a dissolution–precipitation process (intracortical leaching of aragonite and precipitation of calcite with no aragonite relics). Recrystallization of formerly high-Mg calcitic laminae to micritic calcite occurred at the same time. The growth of (micro)spar cement with no traces of the original fabric indicates a mineralogical stabilization that involves thorough dissolution of the precursor aragonite cortex and precipitation of calcite from interstitial fluids under a burial diagenetic realm (after Hashim and Kaczmarek, 2019). Microsparite cement nucleated on the underlying dark lamellae (now transformed to micritic calcite) made up of relatively insoluble material and grew upward until reaching the overlying dark layer. The initiation of microsparitic calcitization of aragonitic layers is due to high microporosity (Richter, 1983). The significance of the microporosity during diagenesis is also reflected by the reaction of different aragonitic particles in the pore fluid. In the studied micritic/microsparitic oolites, the original aragonitic laminations were dissolved and (micro)sparitic calcite was precipitated instead. However, in other aragonitic grains, including a few examples of mollusc shells with
micritized rims, the inner part was mostly replaced with microcrystalline calcite (e.g. Figure 4C) or, in some exceptional ooids, the concentric traces of the primary cortex was preserved as replacive calcite (e.g. Figure 8C).

Formation of micritic calcite with no evidence of crystal growth (e.g. no microsparry crystals in the micritic layers) in place of primary high-Mg calcite laminations indicates an in situ process that involves obliteration of precursor composition through a thin fluid-film transformation with the least interstitial dissolution (Tucker, 1990). The recrystallization of high-Mg calcite layers to microgranular calcite, consisting of small (2–10 μm) crystals, in micritic/microsparitic ooids resulted in the loss of primary microfabrics (Figure 7A through D).

5.1.5 | Cemented oomoulds

Based on petrographic analyses, the dissolved, aragonitic ooids with intraparticle voids formed through the dissolution of both nucleus and aragonitic cortices. Although the assumption that the primary composition of cemented oomoulds is strictly aragonite has been challenged by some workers (Chow and James, 1987), the growth of blocky sparite (without any traces of the former composition) filling the intragranular cavity indicates that the primary cortex dissolved and formed a mould for cement to fill. This is in agreement with Sandberg (1983) who noted that coarse sparite mosaics grow inside oomouldic cavities without preserving relics of the primary mineralogy following the dissolution of an aragonite cortex. Orientation of the original aragonite crystals is unknown. The cemented oomoulds exhibit an intragranular, paragenetic sequence of sparry calcite (Figure 8B) resembling that established in half-moon ooids (Figure 6A through D), suggesting that primary aragonite cortices dissolved and were replaced by calcite. The genetically close relationship between cemented oomoulds and half-moon ooids is delineated by transitional stages (Castellarin and Sartori, 1973). After the dissolution of the cortex, the less-dissolved part of the core was eccentrically displaced, usually to the bottom of the cavity. The oomouldic void was generally filled with blocky calcite growing preferentially from the periphery towards the core (Figure 8A). The outer, micritic envelope coating the oomould was rapidly recrystallized to high-Mg calcite at the last stage of oolitization.
5.1.6 | In situ calcitized ooids

There was only a solution film between the aragonite and the calcite replacement when the primary aragonitic cortex was transformed (after Richter, 1983). It is unknown whether the source of this thin film is organic or inorganic. The metastable aragonitic cement was modified to brickwork calcite through a polymorphic transformation process. The thin-film alteration allowed specific properties of the original, cortical fabrics (including concentric relics; Figure 8C) to be preserved. The aragonitic composition of former cortices is also suggested by the nearly smooth, non-luminescent sparry crystals that form the recrystallized fabric (Figure 8D). The non-luminescent calcite replacing aragonite under burial diagenesis can maintain the CL-derived signature of the original mineralogy, including low Mn levels (Given and Lohmann, 1985; Major et al., 1988). The orientation of the primary aragonite crystals, however, is not maintained. It is therefore inferred that pseudomorphic replacement (paramorphic calcitization) of an aragonite precursor cement with calcite in the ooid cortex is unlikely (Sandberg, 1985). Calcitization is also supported by the penetration of inclusions into the individual calcite crystals (Richter, 1983). The process involves the growth of neomorphic spar which considerably reduces the total porosity of the texture hosting the aragonitic ooids.

5.1.7 | Micritic ooids

Petrographic analysis suggests that these ancient micritic ooids were developed from primary ooids through in situ recrystallization or possibly through the activity of marine microorganisms. A thorough diagenetic micritization of primary ooids made the cortex and nucleus almost indistinguishable from each other, introducing the genetic association of micritic ooids with peloids (Figure 9A). Micritic fabrics can arise either through the addition of equant nannograins in high-Mg calcitic ooids formed originally as micrite or through microendoliths, primarily cyanophytic algae, in calcitic or aragonitic ooids (Wilkinson et al., 1984). Because both processes give rise to nearly identical fabrics, it is difficult to infer the original mineralogy of micritic ooids. Nevertheless, the primary composition of investigated micritic ooids is most likely to be high-Mg calcite because the growth of replacement microsparite mosaics is prevented. Micritization of an aragonite precursor is less likely in this situation because the recrystallization of microcrystalline calcite from aragonite occurs only through an in situ transformation film. This thin-film transformation involves the least dissolution of the primary ooid and can maintain traces of aragonite in the micritic calcite which results. The initiation of micritic calcitization in the primary fabric of ooids here can be attributed to a low microporosity.

5.1.8 | Oncoids

Petrographic analysis suggests that the microsparitic calcitization of initially aragonitic layers of oncoidal cortices occurred after the dissolution of aragonite, at the same time microcalcitization of the originally high-Mg calcite occurred. These mineralogical changes are further demonstrated by an explicit difference between the atypically bright and non-luminescent micrite and microsparite lamellae of the cortex, respectively (Figure 9D). However, it is not clear whether the investigated oncoids were algal (with the exception of red algae), cyanobacteria or bacteria mediated (Peryt, 1983) as the original fabric of the cortical crystals has been entirely destroyed during diagenesis. The irregularity of the oncoid cortex (sometimes discontinuous) is believed to be environmentally controlled (Flügel, 2004). Thus, the concentric, primary laminations of the studied marine oncoids may show that they were originally formed as soft materials in a low-energy, quiet-water setting.

6 | DISCUSSION

6.1 | Diagenetic history of coated-grain facies

According to Richter (1999), the calcitized coated grains of the Middle Triassic grain flow at Hydra formed
syndepositionally in an unknown shallow-water depositional environment and were shed basinwards towards Hydra. The cortical thickness is larger than the size of the nucleus for the majority of the studied ooids (see $d/t$ ratio in Table 2) reflecting the primary depositional environment for this Middle Triassic oolite in a high-energy, agitated setting (Mei and Gao, 2012). Although a non-selective microfabric partially made up of non-oolitic, micritic/microsparitic clasts and oolitic fragments (Figure 4A through C, H through O) in some of the analysed microfabrics represents a before-relocation alteration outside this island, the Middle Triassic grain flow was diagenetically evolved after relocation (Richter, 1999). If diagenetic alteration took place before the coated grains were transported to Hydra, significant fractions of broken coated grains, oolite fragments and intraclasts would be expected in the framework of the grain flow.

Petrographic analysis suggests that the main diagenetic processes involved, based on the order in which they occurred, include dissolution, carbonate cementation, grain deformation and pressure solution (Figure 12). The aragonitic part of the primary coated-grain facies was extensively dissolved away during early pre-compaction diagenesis (possibly long before the completion of compaction) after the re-ordering and deposition of the grain mass flow in Hydra. For this to happen, the fluid dissolving the primary cortex would have had to be undersaturated with respect to aragonite. However, it is unclear whether the dissolution occurred in meteoric fluids (often early in shallow settings) or in undersaturated seawater. According to Bridge and Demicco (2008), the metastable aragonite components of ooids are readily dissolved during early diagenesis. The fabric-selective dissolution of aragonite (Melim et al., 2002) is supported by the common presence of oomouldic porosities later filled by sparry cement (Figure 4). Although the microcrystalline high-Mg calcite coating grains (in micritic ooids and micritic/microsparitic ooids) evolved during early diagenesis before the ooids were relocated to Hydra (following Richter, 1999), the transformation of high-Mg calcite to calcite occurred after the re-arrangement of the grain flow at Hydra. In rare examples, the original high-Mg calcite composition of ancient limestones has been maintained, probably when it originated under the early stages of burial diagenesis, during compaction of carbonates deposited in a shallow-marine setting (Stanienda-Pilecki, 2018). From this, a strong case can be made that the various coated grains present in the investigated grain flow are now calcite as they were formed outside the Island of Hydra and diagenetically altered and cemented once removed to this island. The relationship between carbonate cementation, grain deformation (e.g. Figure 4L, M) and pressure solution events, in particular stylolites (Figure 4D-E, N-O), suggests that the calcite inside and between the coated grains represents a (shallow to deep) burial sequence precipitated during intermediate–late diagenesis after rapid dissolution of aragonite (and the more-soluble parts of high-Mg calcite components) (Carozzi, 1963; Figure 12). Nevertheless, the burial depth over which cement was precipitated cannot be determined from the present data.

There are two possible sources for precipitation of the (shallow to deep) burial calcite cementing the studied limestones: (a) pore solutions derived from the early pre-compaction undersaturated fluid (either fresh water or marine) that dissolved the original minerals and (b) diagenetically derived pore solutions percolating following pressure solution of calcitic grains and calcitic components of coated grains. According to many researchers (Baker et al., 1982; Machel, 2005; Ahm et al., 2018; Lu et al., 2019), the dominant source for precipitation of shallow-burial calcite is the trapped part of the fluid that dissolved the primary carbonates. This solution could be trapped in the sediment pores during deposition, gradually becoming more saturated with respect to calcium carbonate with increasing burial. Despite only having experienced shallow burial, the evidence suggests that mechanical compaction and pressure solution occurred early (i.e. intermediate diagenesis; Zhao et al., 2018), based on a significant decrease in porosity and the replacement of tangential grain contacts with concave–convex margins (Figure 4J through M). Later pressure dissolution during deep burial (equates to late diagenesis), represented by such key features as stylolites delineated by insoluble material (Figure 4D-E) and grain distortion (e.g. Figure 4N, O), is considered to be the last phase of calcite cementation (Zhao et al., 2018). Sediment dissolution can also be a partial source of deep-burial calcite cement, although carbonates are especially susceptible to pressure solution, and stylolites are common.

### 6.2 Cementation sequence

Defining the calcite sequence using CL imaging involves the identification of shallow-burial and deep-burial cements employing cement stratigraphy, pore-fluid chemistry.
and crystallization (Machel, 2005). The CL zonations in the calcite sequence explicitly reflect the differential uptake of Mn$^{2+}$ (Ten Have and Heijnen, 1985) during crystal growth which is caused by changes in the redox potential of the precipitating fluid. The reductive uptake of Mn$^{4+}$ can take place in suboxic waters, and the reduction of Fe$^{3+}$ can occur in waters of even lower redox potential, sometimes close to anoxia (Plunkett, 1997).

The paragenetic sequence established in the investigated half-moon ooids is comparable to that observed in the interparticle blocky calcite (Type B cement) filling the oomoulds and mollusc fragments, and in the interparticle pore-filling drusy calcite cement (Type A cement; Figure 13). Because the number of concentric zones and CL patterns are consistent within all analysed samples from the studied section, the intergranular and intragranular calcite sequences are believed to be correlated in the context of cement stratigraphy (Heydari and Moore, 1993). This correlation provides a tool for deducing the details of the diagenetic setting of the studied grain flow.

Interparticle and intraparticle sparry sequences show a similar pattern of non-luminescent to bright-dull zonation with increasing burial (Meyers, 1978; Grover and Read, 1983; Frykman, 1986). This pattern is thought to relate to the increasingly reducing nature of pore fluids during burial which is a result of the breakdown of organic matter (Tucker and Bathurst, 1990a). Machel (1985) and Curtis (1987) indicated that most water-soluble degradation products of organic matter release Mn, Fe and some other trace metals (e.g. Ag, Cr and Cu) into pore waters during shallow burial. These trace elements could then incorporate into the precipitating carbonate cements and impose further variations in the luminescence of the burial calcite sequence. Among six luminescent concentric zones of calcite, phases 1–4 (Figure 6A through D, F) in both sequences were formed in a shallow-burial setting. The non-luminescent zone 1 was precipitated from oxidizing pore waters that did not contain Mn$^{2+}$ (Heydari and Moore, 1993). The bright zones 2 through 4 were formed from reducing waters that contained Mn$^{2+}$ but not Fe$^{2+}$, still representing shallow burial. The oscillatory zonation of bright-luminescent zones (orange to yellow orange) is presumably explained by a swift chemical re-equilibration of the interstitial fluid precipitating calcite (after Vuillemin et al., 2011). The innermost zones, 5 and 6 in both interparticle

**FIGURE 13** Concentric zonations of sparry cement. Numbers from 1 to 6 mark the location of concentric zones. (A, B) Intergranular sparry calcite. (C) Blocky sparry cement filling an oomould. (D) Blocky sparry calcite cement filling a bivalve shell
and intraparticle sequences, were formed under deep-burial conditions. These dull zones are ferroan calcites precipitated from more negative Eh waters, below the stability field of FeS2. Thus, it is inferred that these zones most likely precipitated during deep burial, commonly after a phase of fracturing (Tucker and Bathurst, 1990a). The influence of iron incorporated into the lattice of calcite could be a cause of dull luminescence (see Figures 5C and 6A through D for comparison). The dark brown luminescence of the youngest zone in the sequence is deemed to represent the phases where Fe is most abundant. Regarding the consistency of this normal, paragenetic sequence in both interparticle and intraparticle cements within the study samples, a local stratigraphic synchronicity in the study coated-grain section is accepted.

6.3 Environmental controls on bimineralic ooid mineralogy

The stratigraphic generalization of coated grains in the studied outcrop follows a trend characterized by commonly bimineralic ooids in the Lower to Middle Anisian, documented by comparable studies of formerly bimineralic Middle Triassic ooids (Richter, 1983; 1999; Chatalov, 2005), and conforms to a significant occurrence of ancient ooids in the Middle Triassic on a global scale (Wilkinson et al., 1985). Similarly, the regional distribution and presumed association of radial high-Mg calcite and tangential aragonite in formerly bimineralic ooids from the study section correlate with their high-energy, shallow-water depositional environment. From this correlation, a strong case can be made that the cortical fabric and mineralogy of formerly bimineralic ooids were strongly controlled by environmental conditions.

The Middle Triassic epoch was related to low-stands (Figure 14A) within the first order (108–109 year) tectono-eustatic cycle specified by slower spreading rates of mid-ocean ridges (MOR) and an increase in the intensity of continental weathering (MacKenzie and Agegian, 1989). The high rates of terrestrial weathering, increased photosynthesis and the decreased levels of CO2 outgassing at subduction zones caused a large-scale reduction in the level of oceanic CO2 (Mackenzie and Pigott, 1981; Sandberg, 1983; Wilkinson et al., 1985; Mackenzie and Morse, 1992). The decreasing pCO2 (other kinetic factors, for instance Mg/Ca ratio, being equal) did lead to a greater seawater carbonate saturation, causing the precipitation of aragonite instead of calcite. Within the saturation domain of aragonite, a lower-grade supersaturation caused similar precipitation rates for both aragonite and high-Mg calcite phases. The dominance of aragonite and high-Mg calcite at low-stands implicates the aqueous pCO2 interacting with the seawater pH as the main control over seawater carbonate supersaturation (Algeo and Watson, 1995).

The occurrence of bimineralic ooids in the Middle Triassic study outcrop and in comparable studies (Richter, 1983; Chatalov, 2005), reflecting aragonite seas, argues against pCO2 as a major control leading to long-term secular switching between calcite and aragonite seas (Lehrmann et al., 2012). The Early to Middle Triassic was a period of extreme global warming at the beginning of the Mesozoic greenhouse episode and still had extremely high CO2 levels associated with extrabasinal volcanic/hydrothermal events (Retallack, 1999; Krull et al., 2004; Payne et al., 2007; Sun

![FIGURE 14](image-url)
et al., 2012; Benton, 2018; Foster et al., 2018), despite photosynthetic reduction of atmospheric and oceanic pCO₂. If pCO₂ were the main cause, then the switch to the Mesozoic calcite sea would have occurred by the Early Triassic instead of being delayed until the Early Jurassic. This conclusion is confirmed by the work of Burton and Walter (1991); Stanley and Hardie (1998); Broecker (2014); Li et al. (2015) who demonstrated that raised pCO₂ could lower the Mg content of modern seawater at 25°C, although this did not lead to supersaturation with respect to the calcite polymorph over aragonite.

The genesis of bimineralic ooids during a Middle Triassic period of rapidly increasing pCO₂ levels (also low SO₄²⁻ levels related to the end-Permian mass extinction, Luo et al., 2010; Bots et al., 2011; Lehmann et al., 2012; Li et al., 2013) indicates that a gradual increase in the Mg/Ca ratio of seawater was the likely mechanism that exerted a primary control on the mineralogy of these carbonate grains. According to a number of papers (Spencer and Hardie, 1990; Hardie, 1996; Stanley and Hardie, 1999; Ries et al., 2008; Ries, 2010), the composition of ancient seawater was a consequence of the mixing of average river water and the MOR hydrothermal brine. Even minor changes in the flux of MOR hydrothermal brine would have been able to change the Mg/Ca ratios enough to influence the original composition of marine carbonates (Spencer and Hardie, 1990).

Baker et al. (1995) and Stanley and Hardie (1998; 1999) suggested that the low spreading rates of MOR (reflecting low fluxes of hydrothermal brines) was a critical factor elevating Mg/Ca ratios in the ancient open-ocean systems above 2 for warm surface seawater. This increase in the secular Mg/Ca ratio of seawater was accompanied by a decline in the concentration of calcium (commonly below 3 mM in the Triassic; Stanley, 2006), leading to supersaturation with respect to aragonite and high-Mg calcite over calcite (Figure 14B). The high saturation of pore fluids with respect to aragonite and high-Mg calcite nucleated instead of calcite in shallow-water nearshore settings (according to Kimmig and Holmden, 2017) and pCO₂ (Lee and Morse, 2010). The effect of salinity on the saturation state of carbonate minerals is minor. According to Zhong and Mucci (1989), even a sharp seawater salinity increase (e.g. from 5 to 45 g/L) has a negligible kinetic effect on the reaction rate of calcite and only a slight decrease in the precipitation rate of aragonite through influencing its stoichiometric solubility. The mineralogical transitions within the structure of bimineralic ooids reflects short-term fluctuations in the chemical and hydrographic setting of ooid formation, represented by fluctuating Mg/Ca ratios in the epicontinental Middle Triassic sea. As the chemistry of marine carbonates varies under relatively wide ranges in seawater temperature (e.g. 18–30°C; Higuchi et al., 2017), it is believed that the narrow range of temperatures seen over the Middle Triassic (26–27°C; Song et al., 2019) was a subordinate control on these short-lived chemical changes. Aragonite proportions of <1% would be expected at a Mg/Ca ratio of 1 and temperatures of <15°C (Kießling, 2015), whereas a ratio of less than 0.4 at 25°C is necessary for calcite to form (Morse et al., 1997).

Accordingly, the dominant distribution of investigated bimineralic ooids during the Lower to Middle Anisian (Middle Triassic) is attributed to high Mg/Ca seawater ratios, principally controlled by low hydrothermal brine fluxes, which caused higher precipitation rates for aragonite and high-Mg calcite relative to calcite. Since the coated grains originally formed outside the Island of Hydra and evolved diagenetically after deposition in this island, it is very unlikely that their primary composition was impacted by local brines. The coeval aragonitic and high-Mg calcitic constituents of primary bimineralic ooids were precipitated during a high carbonate saturation period within the saturation realm of aragonite (i.e. the aragonite sea; Figure 14B), essentially controlled by the seawater Mg/Ca ratio with lesser effects from temperature, salinity and pCO₂. Intracortical, rapid transitions in formerly bimineralic, micritic/microsparitic ooids are inferred to result from a physicochemical threshold related to kinetic factors that favour transition from aragonite to high-Mg calcite and vice versa (Algeo and Watson, 1995). The absence of intracortical, multiphase transitions in formerly bimineralic half-moon ooids suggests that the seawater thermochemical factors linked to seawater carbonate saturation must have collectively contributed to the swift dissolution of the original cortical fabric.

The stratigraphic distribution of studied bimineralic ooids corresponds to global sea-level changes during the Middle Triassic (i.e. the low-stands; Figure 14A), implying that aragonite and high-Mg calcite nucleated instead of calcite in shallow-water nearshore settings (according to Kimmig and Holmden, 2017). From this, it can be inferred that the occurrence of bimineralic ooids at that time has the potential to be a signal of global change driven mainly by secular variations in Middle Triassic seawater chemistry, namely the Mg/Ca ratios. This conclusion also implies that the attribution of the examined grain-flow section to unusual sources of calcium carbonate precipitation in the geologic record, including aragonite seafloor fans after the largest extinction in the history of life is unlikely. This is because the seafloor carbonate fans represent anomalous (<1 Myr) environmental ocean chemistry conditions, including unusually alkaline waters and a transient restoration of Archean–Paleoproterozoic-type ocean chemistry (Pruss et al., 2008; Chen and Benton, 2012). Hence, the studied ooids are not related to the Precambrian-like seawater conditions that have been suggested for Triassic aragonite seafloor fans. Furthermore, the coated-grain facies host micritic clasts (although in small fractions), in contrast to the inhibitions placed on the formation of micrite
in the seafloor fans during intervals when the seawater was oversaturated with respect to calcium carbonate (Pruss et al., 2005).

7 | CONCLUSIONS

The Middle Triassic (Middle Anisian) outcrop at Agios Nikolaos hosts allochthonous, calcitized ooids of formerly monomineralic (only aragonite or high-Mg calcite) and bimineralic (both aragonite and high-Mg calcite) composition as well as biogenic coated grains transported basinwards from an unknown high-energy, shallow-marine setting to Hydra. The ancient half-moon ooids investigated are considered to have been originally bimineralic coated grains consisting of a small high-Mg calcite core surrounded by concentrically arranged laminae of aragonite cortices. Following early diagenetic rapid dissolution of both aragonite cortices and the more-soluble part of the high-Mg calcite core, the less-dissolved remnants crumbled to the bottom of the oomouldic cavity which was later filled by blocky calcite. When compared to analogous modern and ancient occurrences of this ooid type, and conforming to Sandberg’s (1983) criteria for recognizing ancient aragonites, the calcite cement without any relics of the original mineral filling the half-moon ooids is indicative of a likely tangential primary aragonite cortex. This aragonitic precursor may also be acknowledged by growth of a non-luminescent zone of calcite cement in the geopetal cavity which possibly inherits the signature of primary aragonite. Recrystallization of micritic calcite involving minimal or no dissolution of the collapsed remnants suggests high-Mg calcite, probably with radial fabric, as the original composition of the ooid core. The micritic to microsparitic ooids investigated were formerly bimineralic coated grains made up of concentric alternations of aragonite and high-Mg calcite laminae developed around a high-Mg calcite nucleus. Following criterion 3 of Sandberg (1983) and by comparing modern and ancient occurrences of this ooid, the cortical layers cemented by microsparite (sometimes sparry calcite) without traces of aragonite document the formerly aragonitic laminae of likely tangential fabric that were dissolved and replaced by calcite. The primary high-Mg calcite laminae of radial fabric were transformed to microcrystalline calcite through a thin-film recrystallization. The CL zoned, non-marine sequence of mould-filling calcite in the half-moon ooids and cemented oomounds is applied in order to understand the cementation history of the coated-grain facies, predicting that calcite sourced from early pre-compaction fluids, as well as pressure solution, was precipitated during shallow to deep burial. The sequence from oldest to youngest represents a progression in reducing conditions for the diagenetic pore fluids precipitating calcite. The predominance of formerly bimineralic ooids in the Middle Triassic was basically controlled by elevated Mg/Ca ratios that under low fluxes of hydrothermal brines favoured high degrees of seawater saturation with respect to aragonite and high-Mg calcite. The mineralogical transitions (from aragonite to high-Mg calcite and vice versa) in the fabric of bimineralic ooids demonstrate that short-term shifts in the chemical environment of ooid formation during the Middle Triassic are represented by fluctuations in the Mg/Ca ratios of seawater. The substantial distribution of bimineralic ooids during the Middle Triassic implies a change in the global carbonate factory, a point that underlines the significance of searching for chemical environmental factors that control the composition of these coated grains.

ACKNOWLEDGEMENTS

We thank Ashleigh Hood, the University of Melbourne and Li Tian, the China University of Geosciences for their constructive reviews. Special appreciation is given to Adrian Immenhauser and Detlev Richter of the Ruhr-University Bochum, who commented on earlier versions of this paper. We also thank Rolf Neuser, Ruhr-University Bochum for his help by carrying out the analysis with the CL microscope. We extend our deep gratitude to the preparation team at Ruhr-University Bochum for the preparation of the thin sections.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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How to cite this article: Varkouhi S, Jaques Ribeiro LM. Bimineralic Middle Triassic ooids from Hydra Island: Diagenetic pathways and implications for ancient seawater geochemistry. Depositional Rec. 2021;7:344–369. https://doi.org/10.1002/dep2.117

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