## ORIGINAL PAPER

# **Carbonate diagenesis of the mixed clastic–carbonate Galala Formation, North Eastern Desert, Egypt**

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Abstract The carbonate diagenetic history of the Late Cenomanian Galala Formation (North Eastern Desert, Egypt) successively includes marine-phreatic, mixed marine-meteoric, meteoric-phreatic, and subaerial diagenesis. The marine-phreatic diagenesis proceeded through the following path: (a) micritization of skeletal allochems; (b) shallow marine cementation (fibrous calcite and circumgranular spar cements); and (c) dolomitization of the lime mud to fine-crystalline dolostone (dolomicrite). The mixing marine-meteoric diagenesis was associated with the formation of the coarse-crystalline dolostone (dolosparite) by the aggrading recrystallization of the precursor dolomicrite. The meteoric-phreatic diagenesis comprises of the following consecutive stages: (a) the development of granular calcite, blocky calcite, and syntaxial rim cements; and (b) the recrystallization of both the carbonate matrix and bioclasts. The subaerial diagenesis is responsible for the calcitization of the precursor dolomites; whereas, the percolation of meteoric water results in the removal of the Mg ions from the dolostone and the production of calcite.

**Keywords** Carbonate diagenesis · Galala · Eastern Desert · Egypt

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### Introduction

The Cretaceous of Egypt includes eight facies belts (Sinai, Ataqa, Southern Galala, Wadi Qena, Nile Valley, Nubia-Abu Ballas, Farafra-Bahariya, and North Western Desert; Issawi and Osman 2000). The Galala Formation (Late Cenomanian) is one of the most conspicuous rock units of the Cretaceous stratigraphic sequence of the Egyptian territory. It is outcropped within both Ataqa and Southern Galala facies belts of Issawi and Osman (op.cit). The carbonates of the Galala Formation are characterized by their cyclic nature of sedimentation, besides their richness in the Cenomanian macrofauna (i.e., oysters, gastropods, ammonites, and echinoderms).

The majority of previous studies on the Galala Formation were concentrated on its stratigraphy and paleontology. Few workers attended with the digenetic history of Galala Formation (e.g., Mohammad and Omran 1992; Abu El-Hassan 1997; Khalil and Mostafa 2001; Mostafa and Hassan 2003; Essa 2005). The intention of this work is to describe and to discuss the main post-depositional changes that acted upon limestones and dolostones in order to deduce the carbonate diagenetic history of the Galala Formation.

The Galala Formation unconformably overlies the fluvial to fluvio-marine sediments of the Aptian–Albian Malha Formation. The unconformity is represented by the paleosol horizons at Gebel El-Zeit and the Southern Galala and by the intraformational conglomerates at Gebel Shabraweet. It is unconformably overlaid by the El-Khashm Formation at Gebel El-Zeit, the Northern Galala, and the Gebel Ataqa. The unconformity is delineated by the presence of undulated caliche zone. The Wata Formation is paraconformably overlying the Galala Formation at the Southern Galala. At Gebel Shabraweet, the Galala Formation unconformably underlies the Maghra El-Hadida Formation. The contact is taken at the top of the cherty dolostone of the uppermost part of the Galala Formation. The lithostratigraphic classification of the studied succession is given in Table 1. The litho-, bio-, and petrofacies of the Galala Formation reflect a shallow ramp depositional model (Hussein 2010). The main characteristics of the Galala Formation and its proposed depositional environments are highlighted in Table 2.

### Methods

To accomplish the target of this study, the following steps were executed: (a) a detailed field work was carried out, including measuring, describing, sampling, and constructing lithostratigraphic logs in order to define the boundaries between the rock units of five representative outcrops covering the study area from south to north; Gebel El-Zeit, the Southern Galala, the Northern Galala, Gebel Ataga, and Gebel Shabraweet (Fig. 1). (b) About 200 thin sections of the carbonate rocks have been prepared and examined under a polarized microscope. Selected carbonate thin sections were stained with Alizarin Red-S and Potassium Ferricyanide following the method outlined by Dickson (1966), in order to distinguish between different carbonate minerals and the ferroan and non-ferroan dolomite. (c) Scanning electron microscopy (SEM) was used to detect the microscopic textural and diagenetic features of 25 samples. It was carried out in the Central Laboratories Sector of the Egyptian Geological Survey and Mining Authority using SEM Model Philips XL 30 attached with EDX unit with accelerating voltage 30 kV and resolution for W. (3.5 nm). The samples were coated with gold. (d) Chemical analyses on 20 powdered rock samples; their compositions, expressed as % in major oxides (CaO, MgO, Fe<sub>2</sub>O<sub>3</sub>, and Al<sub>2</sub>O<sub>3</sub>) and as ppm in trace elements (Sr and Na), are important for understanding the chemistry of the studied rocks. These chemical analyses were carried out in the laboratories of the Nuclear Materials Authority and the Central Laboratories Sector of the Egyptian Geological Survey and Mining Authority. The measurements of the major oxides were done by two methods. The first method follows the method of Shapiro and Brannock (1962) with the modification of El-Reedy (1984). The samples were opened as two solutions (A and B). The solution A was used for the determination of Al<sub>2</sub>O<sub>3</sub>, using spectrophotometer technique. On the other hand, the solution B was used for the determination of CaO, MgO, and Fe<sub>2</sub>O<sub>3</sub>, using the titrimetric methods, and Na<sub>2</sub>O, using the flame photometric methods. The second method was done; whereas, the XRF analysis was carried out for powder (200  $\mu$ ) samples using a Philips X-Ray Fluorescence equipment model PW/1404, with Rh radiation tube and four analyzing crystals. Crystal (LIF-200) was used for determining Ca and Fe, while crystal (TIAP) was used to estimate Mg and Na. Crystal (PET) was used for determining Al. The concentration of the analyzed elements was determined by using the software Kernal X-40 with an accuracy of 99.5 % and a confidence limit of 95.6 %. The estimation of the major elements were done as powder pellets, which were prepared by pressing the powder of the sample in Aluminium cup using Herzog presser and a 10-ton pressure. The X-ray fluorescence technique (XRF) was used to determine the trace elements using PHILIPS X'Unique-II spectrometer. The trace elements were measured by calibrating the system under the conditions

Table 1 Litostratigraphic classification of the studied sequence of the Galala Formation

Age	Section	Gebel El-Zeit (section no.1)	Southern Galala (section no.2)	Northern Galala (section no.3)	Gebel Ataqa (section no.4)	Gebel Shabraweet (section no.5)
-	Turonian- Santonian	Wata Formation	Wata Formation	Maghra El-Hadida Formation	Maghra El-Hadida Formation	Maghra El-Hadida Formation
.ate (	C e	El-Khashm Formation		El-Khashm Formation	El-Khashm Formation	
retaceous	nomanian	Galala Formation	Galala Formation	Galala Formation	Galala Formation	Galala Formation
Ea Creta	arly iceous	Malha Formation	Małha Formation	Malha Formation		Malha Formation

Table 2 Main char	acteristics of the Galala	Formation and the sugges	sted depositional envii	ronments			
Studied section	Lithology	Sedimentary structures	Allochemes		Orthochemes	Facies associations	Depositional environment
			Skeletal	Non-skeletal			
G. El-Zeit	-Claystone (41.6 %)	-Burrows (vertical and oblique)	-Bivalvia (Oysters)	-Glauconite pellets	-Micrite	Clastics: Claystone, sandy claystone, glauconitic claystone, sandy glauconitic silistone, ferr. qz arenite, dol. qz arenite, siliceous qz arenite, evaportiic qz arenite, glauconitic dolomitic litharenite and paleosol (rooted ferruginous suh litharenite)	Shoreface to peritidal
	-Sandstone (33 %) -Dolostone (19.8 %) -Limestone (2.5 %) -Siltstone (1.5 %) -Marl. (1.1 %) -Gypsum (0.5 %)	Bioturbation -Flaser bedding -Cross bedding -Lenticular bedding -Thin laminations -Cross laminations -Fissility in claystone	-Plant remains -Phospathtic bioclasts	-Detrital quartz	-Dolomitic cement -Evap. cement -Ferr. cement -Siliceous cement		INNER RAMP
The Southern Galala	-Claystone (82.6 %)	-Cross bedding and ripple marks in sandstones	-Bivalvia (Oysters)	-Peloids	-Micrite	Clastics: Oyster claystone, glauconitic sandy oyster claystone, ferruginous qz aretnie and sandy dolomitic	
	-Dol. L.S. (6.8 %)		-Plank. forams	-Algal oncolites	-Dolomitic cement	gladocomecures Carbonates: Bioclastic foraminiferal wackestone, foraminiferal molluscan packstone, oncolitic packstone, dolomitic algal bioclastic packstone, dolomitic paloidal packstone and ferroan dolomicrite.	Shallow subtidal
	-Limestone (6.5 %) -Sandstone (2 %)		-Echinoderms -Green algae	-Detrital quartz -Detrital glauconite	-Ferr. cement		<i>INTRA-RAMP BASIN</i> MID RAMP
	-Glauco-arenite (1.3 %) -Dolostone (0.9 %)		-Gastropods -Ostracods -Cephalopods -Phospahtic bioclasts	peners			
The Northern Galala	-Claystone (53 %) -Dolostone (18 %)	-Burrows (vertical and oblique) -Thin laminations	-Bivalvia (Oysters) -Echinoderms	-Intraclasts -Oolites	-Micrite	Clastics: Claystone, oyster claystone and sandy glauconitic siltstone. Carbonates: Fossiliferous marl,	Peritidal to open marine shallow subtidal MID RAMP
	-Limestone (12 %)	-Fissility in claystone	-Plank. forams	-Aggregates		caliche, lime mudstone, bioclastic foraminiferal wackestone, dolomitic molluscan wackestone, foraminiferal molluscan packstone, ostracoda molluscan packstone, sandy dolomicrite and dolomicrite.	
	-Dol. L.S (7.4 %) -Marl (6.7 %)	-Nodular claystone	-Gastropods -Ostracods	-Detrital quartz -Detrital glauconite pellets			
	-Siltstone (3 %)		-Green algae -Phospahtic bioclasts -Bryozoa	1			
G. Ataqa	-Dolostone (39.5 %)		-Bivalvia (Oysters)	-Intraclasts	-Micrite	Clastics: Claystone.	Peritidal to shallow subtidal

Table 2 (continue)	(p;						
Studied section	Lithology	Sedimentary structures	Allochemes		Orthochemes	Facies associations	Depositional environment
			Skeletal	Non-skeletal			
	-Dol. L.S (31 %)	-Burrows (vertical and oblique) -Thin laminations	-Green algae	-Peloids		Carbonates: Fossiliferous marl, catibee, lime mudstone, dolomitic lime mudstone, dol. lithoclastic lime mudstone, algal bioclastic	CARBONATE BUILDUP
	-Limestone (26 %) -Claystone (2.2 %) Mod (1.3 %)	-Fissility in claystone Bioturbation	-Echinoderms -Gastropods Octrocode			wackestone, dolomitic molluscan wackestone, dolomici algal bioclastic packstone, dolomicitie, birds eye dolomicrite and dolosparite.	MID RAMP
	(0/ CT) 1107AT-		-Lithoclasts				
Gebel Shabraweet	-Dolostone (51 %)	-Burrows (vertical, horizontal and	-Bivalvia (Oysters)	-Intraclasts	-Micrite	Clastics: Claystone, oyster claystone, dolomitized claystone and sandy	Peritidal to shallow subtidal
	-Limestone (19 %)	-Thin laminations	-Gastropods	-Oolites	-Fibrous calcite cement	Carbonates: Line mudstone, dolomitic line mudstone, molluscan-echinoidal wackestone, dol. achinoidal wackestone, nolluscan peloidal packstone, oolitic peloidal grainstone, peloidal chinoidal grainstone, dolomicrite, sandy dolomicrite, ferroan dolomicrite siliceous ferroan dolomicrite and	MID RAMP
	-Claystone (18 %) -Dol. L.S (11.9 %)	-Thick-bedded -Mud cracks	-Echninoderms -Green algae	-Peloids -Aggregates	-Isopachous clacite cement -Granular calcite cement	dolosparite.	
	-Glauco-arenite (0.1 %)	-Fissility in claystone -Bioturbation -Nodular claystone -Slump structure	-Ostracods -Benthonic forams (Orbitoides) -Micritized forams (benthonics) -Milioidas -Bryozoa -Phospahtic bioclasts -Lithoclasts	-Detrifial quartz -Detrifial glaucomite pellets			





of Rh-target tube, LiF-420 crystal, gas flow proportional counter, coarse collimators, vacuum, 70 kV and 15 mA, for Sr. A chemical analysis (XRF and normal wet-chemical techniques) of 20 powdered rock samples for their major oxides (CaO, MgO, Fe<sub>2</sub>O<sub>3</sub>, and Al<sub>2</sub>O<sub>3</sub>) and trace elements (Sr and Na) were carried out in order to throw some light on the chemical characteristics of the studied rocks. (e) Twenty-seven carbonate rocks were micro-sampled for oxygen and carbon isotopic analyses in order to discuss the origin of dolostones and the cements. The analyses were carried out at the laboratory of stable isotope at the University of Windsor, Canada using a microscope-mounted drill assembly to extract the desired quantity (2–5 mg) of powdered samples from polished slabs. The samples for isotope analysis (n=28) were reacted in vacuum with 100 % pure phosphoric acid for at least 4 h at 25 °C for calcite and 50 °C for dolomite using the method described by Al-Aasm et al. (1990). The developed CO<sub>2</sub> gas was analyzed for isotopic ratios on a Delta plus mass spectrometer. Values of O and C isotopes are reported in per mil (‰) relative to the Pee Dee Belemnite (VPDB) standard and were corrected for

phosphoric acid fractionation. Precision was better than 0.05% for both  $\delta^{18}O$  and  $\delta^{-13}C.$ 

Marine-phreatic diagenesis

# Micritization

# **Carbonate diagenesis**

The carbonate digenesis of the Galala Formation was developed in the following four main diagenetic environments: marine–phreatic, mixed marine–meteoric, meteoric–phreatic, and subaerial environments. The carbonate diagenetic processes and their related diagenetic environments are summarized in Fig. 2. Micritization causes the transformation of the skeletal allochems into dark, structureless, and homogeneous masses of blindly massive micrite. It is well represented in the peloidal–algal–skeletal wackestones, skeletal packstones, and grainstones of the Southern and Northern Galalas and Gebel Shabraweet. Most of the skeletal constituents have been subjected with partial to complete micritization. Although the algal bioclasts are the most affected grains, also the molluscs,

Diagenetic	D	iagenetic en	vironment	
process	Marine-phreatic	Mixed marine- meteoric	Meteoric- phreatic	Subaerial
1. Micritization				
2. Cementation:				
a. Fibrous calcite				
b. Circumgranular spar				
c. Sparry calcite				
d. Blocky calcite				
c. Syntaxial calcite				
3. Recrystallization:				
a. Lime mud				
b. Allochems				
4. Dolomitization:				
a. Dolomicrite				
b. Dolosparite				
5. Dedolomitization				

Fig. 2 The main carbonate diagenetic processes and their link to diagenetic environments

echinoderms, benthic foraminifera, and oolites allochems are affected by this process with variable degrees (Fig. 3a, b). Partial micritization results in creating micritic envelopes around the mollusc shell debris and echinoid spines and stems (Fig. 3c). Also, it is noticed that many pores and cavities surround the rims of the micritized allochems (Fig. 3d).

#### Marine cementation

*Fibrous cement* It is the first generation of marine cement in the diagenetic history of the Galala Formation. It was identified from the Southern and Northern Galalas and Gebel Shabraweet. It is represented by thin, closely packed fringes

Fig. 3 a Thin-section phptomicrograph shows the micritization of algae (red arrows), Gebel Shabraweet, O.L. **b** Thin-section phptomicrograph shows the micritization of a bivalve bioclast, the Northern Galala, O.L. c Thin-section phptomicrograph shows a micrite envelope around the exterior of an echinodermal bioclast, Gebel Shabraweet, O.L. d Thin-section photomicrograph explaining the phases of the micritization process. 1 Perforation of a bivalve particle by the encrusting algae leads to forming of microborings that are filled with micrite. 2 The repeating boring and infilling of the micoborings with microcrystalline precipitates results in transformation of the particle into a mass of micrite (peloid). The Northern Galala. O.L. e. Thin-section photomicrograph showing the fibrous calcite rim cement around a bivalve bioclast (arrows). Such cement is composed of tightly packed, fibrous to bladed calcite crystals oriented with their long axes normal or nearly normal to the surface of the bivalvian particle. The Northern Galala, O.L. f Thin-section photomicrograph showing circumgranular spar cement (arrows) around a bivalve allochem. Molluscan peloidal packstone. Gebel Shabraweet, O.L. g Circumgranular spar cement (arrows) enveloping ooids (oo), Gebel Shabraweet, O.L. h SEM image showing the polygonal boundaries of the circumgranular spar cement between the ooids (oo), Gebel Shabraweet



of semi-opaque fibrous calcite grow perpendicular to the outer boundaries of some bivalve shells (Fig. 3e). Such crystals are mostly exhibiting straight or planar contacts with each other. The length of these fibrous crystals ranges from 4 to 7  $\mu$ m.

Circumgranular spar cement The circumgranular spar represents the second generation of the marine-phreatic cements. It was identified from the Northern Galala, Gebel Ataga, and Gebel Shabraweet. The best-developed circumgranular cement was observed from the grainstones (oolitic peloidal and peloidal echinoi dal grainstones) of Gebel Shabraweet; whereas, the circumgranular cement borders the outer margins of both ooids and peloids. The circumgranular calcite cement is represented herein by fine-crystalline spar (average= 50 µm) with clear to cloudy appearance arranged normal or nearly normal to the outer margin of the allochemical constituents and with a length to width ratio between 1:1 and 2:1. In spite of the boundaries between the circumgranular crystals are often planar (Fig. 3f, g), polygonal boundaries of circumgranular cement were observed in some rocks (Fig. 3h).

#### Dolomitization

The dolomitization is the most prominent diagenetic process of the Galala Formation. It is more extensive at the northern part of the study area (Gebel Ataqa and Gebel Shabraweet). Two phases of dolomitization are recognized; early and late diagenetic. Since the distinction between the calcite and dolomite in partially dolomitized rocks is very difficult, this study concentrates on the pervasive dolomitization (dolomicrites and dolosparites).

#### Early diagenetic dolostone (dolomicrite)

*Petrography* The early diagenetic dolomitization results in the formation of fine-crystalline dolostone (dolomicrite). The fine-crystalline dolostone includes six lithofacies: dolomicrite, sandy dolomicrite, sandy glauconitic dolomicrite, birds eye dolomicrite, ferroan dolomicrite, and siliceous ferroan dolomicrite. The dolomicrite is composed of fine (4 to 40  $\mu$ ), idiotopic to xenotopic, interlocking mosaics associated with gypsum (Figs. 5h and 6a–c). Biogenic sedimentary structures are represented by burrowing and birds eyes.

*Geochemistry* Thirteen representative samples of dolomicrite were geochemically analyzed in order to determine the stoichiometry, major oxides (CaO, MgO, Fe<sub>2</sub>O<sub>3</sub>, and Al<sub>2</sub>O<sub>3</sub>) and trace elements (Sr and Na) (Table 4). Three of these samples represent sequence boundaries; hence, they show the totally different values of chemical composition. The dolomicrite has an average value of 30.89 wt% for CaO and 17.12 wt% for MgO. It is non-stoichiometric Ca-rich since the average

CaCO<sub>3 (mol%)</sub> is 55.13 and the average MgCO<sub>3 (mol%)</sub> is 35.79. Fe<sub>2</sub>O<sub>3</sub> has a low wt% (1.12–3.45 %), while Al<sub>2</sub>O<sub>3</sub> wt% ranges from 1.7 % to 8.6 %. Sr content ranges from 512 to 717 ppm (average=603 ppm), while Na values are ranging from 1,005 to 2,680 ppm (average=1,694 ppm).

Stable isotope analysis Ten representative samples of dolomicrite were analyzed for their stable isotope composition ( $\delta^{18}$ O and  $\delta^{13}$ C). All values of  $\delta^{18}$ O are >-2 VPDB‰ (0.95 to -1.7 VPDB‰ with average=-0.76‰; Table 4). The  $\delta^{13}$ C values of the studied dolomicrite range from 2.8 to -1.74 VPDB‰ (average=0.933‰; Table 4). The  $\delta^{18}$ O versus  $\delta^{13}$ C values of the studied dolomicrite are plotted in Fig. 7.

## Late diagenetic dolostone (dolosparite)

*Petrography* The late diagenetic dolomitization causes the formation of coarse-crystalline dolostone (dolosparite). The dolosparite is composed of dolomite rhombs ranging in size from 40 to 220  $\mu$ m with pronounced intercrystalline porosity and sucrosic equigranular fabric (Fig. 6d). The majority of the dolomite rhombs exhibits well-developed zoning.

*Geochemistry* Seven dolosparite samples were subjected to geochemical analysis. The average wt% of CaO and MgO of the dolosparite are 29.1 % and 15.8 %, respectively. The dolosparite is non-stoichiometric; whereas, the average CaCO<sub>3</sub> (mol%) and MgCO<sub>3</sub> (mol%) are 52 and 33, respectively. The Fe<sub>2</sub>O<sub>3</sub> is not detected in four samples, while it reaches its maximum percentage (3.8 %) in the dolosparite of Gebel El-Zeit. The Al<sub>2</sub>O<sub>3</sub> ranges from 2.26 % to 7.88 % (Table 4). Sr values are ranging from 133 to 244 ppm (average=199 ppm), while Na values are ranging from 244 to 441 ppm (average=389 ppm).

Stable isotope analysis The  $\delta^{18}$ O of dolosparite are showing more negative values (-4.01 to -2 VPDB‰) than those of dolomicrite (0.95 to -1.7 VPDB‰; Table 4). The  $\delta^{13}$ C values of dolosparite range from 1.5 to 2.87 VPDB‰. This, besides the lack in fossils, suggests that the dolosparite is produced from non-biogenic carbonate-rich water.

Meteoric-phreatic diagenesis

#### Meteoric cementation

Granular sparry calcite cement The granular sparry calcite cement of the Galala Formation occurs in three different textural positions. The first is intergranular precipitates; occluding the pore spaces between the allochems of the grainstones of Gebel Shabraweet (Fig. 5a). The absence of any relics of dark micrite reflects that it is cement not a neomorphic spar. The  $\delta^{18}O_{VPDB}$ % of the interparticle sparry calcite cement in the oolitic peloidal grainstone of Gebel

Studied section	Bed no.	Facies name	Notes	$\delta^{13}$ C VPDB‰	δ <sup>18</sup> O VPDB‰
Gebel Shabraweet	33	Oolitic peloidal grainstone	Granular sparry calcite cement	1.49	-4.93
Gebel Ataqa	57	Birds eye dolomicrite	Sparry calcite cement fills birds eyes	0.92	-1.98
Southern Galala	28	Oncolitic packstone	Neomorphic spar	0.17	-5.17
Northern Galala	82	Ostracoda molluscan packstone	Neomorphic spar	1.02	-4.96
Gebel Shabraweet	100	Molluscan-echinoidal wackestone	Neomorphic spar	1.23	-4.83
Gebel El-Zeit	124	Rooted ferr. Sublitharenite	Caliche	1.61	-5.72
Northern Galala	83	Claystone	Caliche	-3.38	-11.91

Table 3 Oxygen and Carbon isotopic values of representative limestone and dolostone samples

Shabraweet is -4.93% and the  $\delta^{13}C_{VPDB}\%$  value is 1.49% (Table 3 and Fig. 4). The second type fills partially or completely the cavities and internal hollows of the dissolved bioclasts (biomouldic porosity; Fig. 5b). These intragranular sparry calcites are usually inclusion-free and are sometimes well-edged. They grow coarser towards the center of the leached bioclast, and hence, they exhibit drusy calcite mosaics. The third type is the void or cavity fillings (birds eye structures in some dolostones of the Gebel Ataqa and the Northern Galala). Such calcites are limpid with euhedral to subhedral contacts (Fig. 5c). The  $\delta^{13}C_{VPDB}\%$  value of the birds eye calcite is 0.92, while the  $\delta^{18}O_{VPDB}\%$  value is -1.98 (Table 3 and Fig. 4).

*Blocky calcite cement* It is a common cement filling of the intraparticle pores, fractures, and intergranular pore spaces in the lime mudstone of the basal part of both Gebel El-Zeit and Gebel Ataqa and in the dolomicrite of the basal part of Gebel Shabraweet. It is composed of limpid to translucent euhedral crystals with an average size of 600  $\mu$ m, displaying a sharp contact relationship with the surroundings microsparry calcite (Fig. 5d).

*Syntaxial overgrowth cement* It is a continuous to discontinuous clear cement that surrounds almost all the echinoderm fragments of the Southern and Northern Galalas and Gebel Shabraweet (Fig. 5e).

#### Recryatallization

Aggrading neomorphism of lime-mud matrix The aggrading neomorphism of lime-mud matrix is a common process in the diagenetic history of the Galala Formation. It is recognized from the majority of the studied lime mudstones, wackestones, and packstones with variable degrees. The lime-mud matrix in between allochems is partially neomorphosed into patches of microsparites in the earlier stages of neomorphism (15–35  $\mu$ m) and into pseudosparites (average=75  $\mu$ m) during the more advanced stages of neomorphism. The neomorphosed spar is inclusion-rich with turbid appearance providing anhedral form with non-planar to irregular boundaries (Fig. 5f).

The isotopic analysis of neomorphosed spar of representative wackestone and packestone from the Southern and Northern Galalas and Gebel Shabraweet displays a light



Fig. 4 Cross-plot of oxygen and carbon isotopic composition of representative phases in limestone and dolostone microsamples

Fig. 5 a Intergranular sparry calcite cement (inc) between peloids and ooids, Gebel Shabraweet, O.L. b Infilling of cavities of gastropod by granular sparry calcite cement, the Southern Galala, O.L. c Voidfilling calcite cement (vfca) filling the internal structure of birds eyes surrounded by dolomicritic matrix (dol), Gebel Ataqa, O.L. d Blocky calcite cement filling a cavity in lime mudstone, Gebel El-Zeit, C.N. e Syntaxial calcite rim cement around an echinoderm, Gebel Shabraweet, C.N. f Aggrading neomorphism of lime mud. Notice: the coexistence of micro- and pseudospars (ca) and the dark unneomorphosed micrite patches (mc), Gebel Ataqa, O.L. g Bivalve shells have undergone calcitization from their center to peripheries, Gebel Shabraweet, O.L. h SEM image showing a general picture of the planar, hypidiotopic to idiotopic dolomicrite, the Northern Galala



 $δ^{13}C_{VPDB}$ ‰ values (0.17 to 1.23‰) with the average value= 0.8‰ and depleted  $\delta^{18}O_{VPDB}$ ‰ (-4.96 to -5.17‰) with the average value=-4.98‰. The  $\delta^{13}C_{VPDB}$ ‰ value of the caliche of the uppermost Gebel El-Zeit is 1.61‰ and that of the caliche of the Northern Galala is -3.38. The  $\delta^{18}O_{VPDB}$ ‰ of the caliche of the uppermost Gebel El-Zeit is -5.72‰, while that of the caliche of the Northern Galala is -11.91‰ (Table 3 and Fig. 4). *Calcitization of skeletal particles* It is a stabilization process by which the aragonite and high Mg-calcite that build up the original structure of some allochems are converted into Mgdepleted calcite under the influence of t meteoric water (Chaftez et al. 1988). The intensity of calcitization varies from the low-degree, incomplete to the high-degree, intensive calcitization. The resulted calcite crystals are easily distinguished by their turbid appearance, wavy to irregular boundaries, and patchy fabric due to the variability in the distribution of the crystal size. It is noticed that the bivalve bioclasts are the most extensively calcitized skeletal particles (Fig. 5g).

#### Dedolomitization

The dedolomitization process is infrequently recorded in the Gebel El-Zeit, Northern Galala, Gebel Ataqa, and Gebel Shabraweet; whereas, it is portrayed by the partial calcitization of the precursor dolomites. Such calcitized dolostones were distinguished in the field by their mottling appearance (lighter color within the dark gray unreplaced dolostone). The petrographic investigation on stained thin sections shows that the dedolomitization is represented by well-developed dolomites engulfed in larger subhedral to anhedral, blocky to massive phenocrystals of calcite, exhibiting a poikilotopic texture (Fig. 6e). Sometimes, these calcites contain dark relics of the

Fig. 6 a SEM image showing the dolomite overgrowths (arrows) around dolomite rhombs, Gebel El-Zeit. b SEM image showing dissolution of dolomite core. Gebel Shabraweet. c SEM image showing gypsum (gyp) inbetween dolomite rhombs (dol) of the dolomicrite, Gebel Shabraweet. d SEM image showing a general picture of dolosparite, Gebel Ataqa. e, f The dedolomitization process; the idiotopic dolomite rhombs are engulfed by blocky to massive calcite crystals (ca) showing poikilotopic texture, Gebel Shabraweet, O.L

precursor, unreplaced dolomites. The calcitization of the dolomite rhombs is proceeding from their rims towards the centers proceeds from rims towards the centers (Fig. 6f).

It is observed that the dedolomites of the Galala Formation are always identified in the dolostones that are capping the shallowing–upward cycles. Also, the degree of dedolomitization increases near the sequence boundaries. Petrographically, it is noticed that the dedolomitization goes to be more intensive nearby the fissures and cracks.

## Discussion

The micritization is the first diagenetic process in the diagenetic history of the Galala Formation (Fig. 2). It took place during or shortly after deposition in concomitant to or shortly before marine cementation (Khalifa 2005). The petrographic analysis elucidates that there are a lot of pores and cavities around the external margins of the micritized carbonate grains



Table 4 Geochemical and is	totopic dat	a of representative dolos	tone samp	les (dolomicrite and	l dolospar	ite)						
Studied section	Bed no.	Lithofacies	CaO %	CaCO <sub>3</sub> (Mole%)	MgO%	MgCO <sub>3</sub> (Mole%)	${\rm Fe_2O_3\%}$	$\mathrm{Al}_2\mathrm{O}_3\%$	Na (ppm)	Sr (ppm)	$\delta^{13}$ C VPDB‰	$\delta^{18}$ O VPDB‰
Gebel Shabraweet	10	Dolomicrite	30.3	54.08	20.11	42.03	pu	1.7	2,680	618	Ι	Ι
Gebel Shabraweet	31	Dolomicrite	28.1	50.1	18.1	37.8	2.1	3.69	2,019	550	2.77	-1.09
Gebel Shabraweet	39	Dolomicrite	32.3	57.65	21.12	44.32	nd	3.7	1,030	654	0.4	0.95
Gebel Shabraweet	42	Dolomicrite	34.4	61.4	13.1	27.4	1.12	4.6	1,308	533	Ι	Ι
Northern Galala	7	Dolomicrite	30.1	53.72	16	33.44	1.4	5.9	1,801	696	0.24	-0.44
Northern Galala	6	Dolomicrite	29.4	52.47	17.5	36.57	1.12	8.6	1,474	548	1.14	-1.47
Northern Galala	44	Dolomicrite	29.6	52.83	17.5	36.57	nd	8.3	2,011	512	2.47	-1.24
Gebel Ataga	29	Dolomicrite	31.65	56.49	19.61	40.98	1.9	3.33	1,005	653	2.8	-0.36
Gebel Ataga	38	Dolomicrite	31.4	56.04	16.5	34.48	nd	2.51	2,480	717	0.8	-0.14
Gebel Ataga	59	Dolomicrite	29.2	52.12	16.2	33.85	2.6	3.6	1,133	549	1.13	-0.88
Northern Galala (SBNG.2)	25	Dolomicrite	33.54	59.86	16.88	35.27	2.56	4.59	244	164	Ι	Ι
Northern Galala (SBNG.4)	58	Dolomicrite	32.33	57.7	14.71	30.74	1.62	2.56	375	137	I	Ι
Gebel Ataqa (SBA.4)	61	Birds eye dolomicrite	29.31	52.31	15.23	31.83	3.45	3.18	274	183	I	Ι
Gebel El-Zeit	15	Ferroan dolomicrite	I	I	I	Ι	I	I	Ι	I	-1.74	-1.29
Gebel El-Zeit	52	Sandy dolomicrite	I	I	I	Ι	I	I	I	Ι	-0.68	-1.7
Gebel El-Zeit	78	Dolosparite	29.11	51.96	14.82	30.9	2.2	5.5	244	133	1.5	-2.99
Gebel El-Zeit	80	Dolosparite	26.2	46.76	13.72	28.67	nd	7.88	408	242	I	Ι
Gebel El-Zeit	84	Dolosparite	26.33	46.99	15.38	32.14	3.8	5.28	396	167	Ι	Ι
Gebel Ataqa	48	Dolosparite	32	57.12	15.4	32.18	pu	3.02	441	178	2.87	-4.01
Gebel Shabraweet	42	Dolosparite	31	55.33	16.4	34.27	pu	4.28	389	244	Ι	Ι
Gebel Shabraweet	102	Dolosparite	28.41	50.71	16.2	33.85	1.61	6.5	440	210	2.55	-2
Gebel Shabraweet	118	Dolosparite	30.9	55.15	19	39.71	nd	2.26	407	218	1.79	-3.26

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(nd not detected)

(Fig. 3d). Consequently, the role of encrusting algae (Bathrust 1975; Harris et al. 1979; May and Perkins 1979) is adopted herein to interpret the micritization process. The grains are cemented by fibrous and circumgranular cements, respectively, in the marine–phreatic diagenetic environment shortly after micritization (Fig. 2). Many authors described and interpreted such cements from the shallow marine–phreatic diagenetic environment (e.g., James and Choquette 1983; Tucker 1993; Melim et al. 2002).

Contemporaneously, the lime mud is subjected to pervasive dolomitization to yield fine-crystalline dolostone (dolomicrite; Fig. 2). The size and fabric of the studied dolomicrite resemble the penecontemporaneous dolostone of the ancient and recent supratidal-intertidal flats. The penecontemporaneous dolomite ( $<40 \mu m$ ) is formed in early phase of dolomitization by replacing the lime mud shortly after the sedimentation of the carbonate minerals (Lee and Friedman 1987 and Amthor and Friedman 1991). The low Fe content (1.12–3.45 %) of the dolomicrite reflects that it has been formed in a near-surface oxidizing environment characteristic of carbonate ramps (Land 1985). The high wt% of Al<sub>2</sub>O<sub>3</sub> (1.7 % to 8.6 %) in comparison with the Fe<sub>2</sub>O<sub>3</sub> is attributed to the high content of claystones forming the basal parts of cycles (Table 4). Sr content of the investigated dolomicrites is ranging from 512 to 717 ppm with average=603 ppm, while Na content is ranging from 1,005 to 2,680 ppm with the average value= 1,694 ppm (Table 4). Such values fall within the range of the hypersaline to marine dolomite (Morrow 1988). The hypersaline-marine dolomite contains Sr content ranges from 500 to 900 ppm (Mitchel et al. 1987 and Wanas 2002). The Na concentration of the marine dolomite ranges from 1,000 to 3,000 ppm (Land and Hoops 1973). The  $\delta^{18}$ O values of the dolomicrite of the Galala Formation (0.95 to -1.7 VPDB‰ with average=-0.76%; Table 4) are compatible with that of the marine ancient dolomite, which range from +2.4 to -2.5VPDB‰ (Moss and Tucker 1995; Holail et al. 1997; Railsback and Hood 2001). The deviation of the  $\delta^{18}$ O to the 2563

negative values suggests that the dolomicrite of the Galala Formation was influenced by later fluids (Lonnee and Al-Aasm 2000). The  $\delta^{13}$ C values (2.8 to -1.74 VPDB‰ with average=0.933‰; Table 4) support the marine origin of the fine-crystalline dolostone (Irwin et al. 1977). The cross-plot diagram of  $\delta^{18}$ O versus  $\delta^{13}$ C values of dolomicrite indicates that they fall within the zone of marine dolomite (Al-Aasm and Veizer 1986; Major et al. 1992; Fig. 7).

The admixture of meteoric water with marine water due to sea-level fall favors the aggrading recrystallization of the finecrystalline dolomite (dolomicrite) into coarse-crystalline dolomite (dolosparite) (Fig. 2). The neomorphic origin of dolosparite can be evidenced by the presence of relics of finer dolomite within the coarser one and the variation from euhedral to anhedral dolomite crystals. Such polymodal size distribution may be ascribed to the neomorphic alteration and/ or the heterogeneous grain size distribution of the precursor limestones and/or dolomicrite. The depleted Sr content of studied dolosparite (133 to 244 ppm with the average value=199 ppm) indicates that the dolomitization process has taken place in a mixed marine-meteoric water (Table 4). The Sr content of the mixing marine-meteoric dolomites ranges from 100 to 250 ppm (Supko 1977). It is granted that the Sr content of dolostone decreases with the increase of the extent of recrystallization by meteoric water (Nielsen et al. 1994 and Al-Hodairi and El-Hadad 1997). The Na values of dolosparite of the Galala Formation (244 to 441 ppm with average value 389 ppm; Table 4) indicate that it was originated in a mixed saline-meteoric waters (Badiozamani 1973; Loukina and Abu El-Anwar 1994). The low Na content (200-500 ppm) was attributed to the mixing marine-meteoric dolostones (Sibley 1980). The  $\delta^{18}$ O values of the examined dolosparite ranges from -4.01 to -2 VPDB‰ (Table 4). This suggests that it was originated by a fluid of salinity less than that of the seawater. Also, it can be formed by an increase in temperature due to the burial effect. The average value of  $\delta^{18}$ O of dolosparite is -3%, which reflects depletion in oxygen. This suggests that it is

Fig. 7 Cross-plot of oxygen and carbon isotopic composition of representative dolostone bulk samples (dolomicrite and dolosparite)



originated by the mixing of meteoric and marine waters that promote aggrading neomorphism of the early dolomite (dolomicrite; Khalil 1999). The cross-plot diagram of  $\delta^{18}$ O versus  $\delta^{13}$ C (Fig. 7) displays that the investigated dolosparite is located within the mixed marine–meteoric, coarsecrystalline dolostones of the Upper Cretaceous Bahariya, Egypt (Holail et al. 1988) and of the Asmari Formation, SW Iran (Ranjbaran et al. 2007).

More severe sea-level fall results in the cementation of the carbonate particles by granular sparry calcite, blocky calcite, and the precipitation of the syntaxial overgrowth around the echinoidal particles in the meteoric water diagenetic environment (Fig. 2). The light  $\delta^{18}O_{VPDB}$ % (-4.93%) of the studied interparticle sparry calcite cement indicates that it was precipitated in the meteoric-influenced pore waters (Holail et al. 1997; Fig. 4 and Table 3). Sun (1990) concluded that the intergranular cement in the biosparites is precipitated from the meteoric fluids. The  $\delta^{13}C_{VPDB}$ % (0.92) and  $\delta^{18}O_{VPDB}$ % (-1.98) values of the granular calcite cement within the birds eyes are indicative of diagenesis in the meteoric pore waters (Maliva 1995; Fig. 4 and Table 3). The common fossil-leaching feature indicates an active water circulation of the meteoricphreatic zone (Tucker and Wright 1990). The sparry calcite cement is inferred to have precipitated as a low Mg-calcite cement in the meteoric-phreatic diagenetic environment (Moore 1989; Essa 2005). The blocky calcite cement filling the intraparticle voids, fractures, and pore spaces was interpreted to have been precipitated in the meteoric-phreatic zone by James and Choquette (1990) and Vincent et al. (2007). The syntaxial overgrowth cement is interpreted to be formed in the meteoric-phreatic diagenetic environment by many authors; among them were Heckel (1983) and Harris et al. (1997).

Meteoric cementation is followed by the recrystallization process (Fig. 2). Recrystallization affects both the lime-mud matrix (aggrading neomorphism into micro- and pseudosparites) and the skeletal allochems (stabilization or calcitization). The chemical instability of the aragonite and high Mg-calcite is the driving force for the aggrading neomorphism. The present authors believe that the clay minerals and/or argillaceous material is responsible for the aggrading neomorphism of the lime-mud matrix as they adsorb the Mg<sup>2+</sup> ions that obstruct the growth of the micrite and hence favor the aggradational growth of the micrite into coarser neomorphic spar. Similar examples were elucidated by Folk (1974), Khalifa and Zaghloul (1990), and Abu El-Ghar and Hussein (2005). The light  $\delta^{13}C_{VPDB}$ % (0.17) to 1.23‰) and depleted  $\delta^{18}O_{VPDB}$ ‰ (-4.96 to -5.17‰) isotopic values of the studied neomorphosed spar indicate a meteoric diagenetic environment (Holail et al. 1997; Fig. 4 and Table 3). The aggrading neomorphism of lime mud takes place in meteoric-phreatic environment (Flügel 1982; Harris et al. 1997). Many authors believe that the calcitization process goes on when the skeletal allochems are subjected to meteoric water, resulting in the replacement of these shells by the more stable neomorphic spar (e.g., Ahmad et al. 2006 and Knoerich and Mutti 2006).

Some dolostones are exposed to dedolomitization (calcitization) during the subaerial exposure and weathering periods. This results in formation of calcite at the expense of dolomite. The meteoric water mechanism is proposed herein to interpret the dedolomitization process; whereas, the percolation of the meteoric water on the exposed surface of dolostones or through the fissures and cracks leads to the removal of Mg and the formation of calcite. This interpretation goes in harmony with the opinions of Folkman (1969) and Braun and Friedman (1969).

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