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Origin of Calcites Filling Vugs in the Dolostones of the Upper Cenomanian El-Heiz and Lower Middle Eocene Naqb Formations, Bahariya Oasis, Western Desert, Egypt: Fluid Inclusions, Stable Isotope and Elemental Chemistry Evidences.

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ABSTRACT

Calcites filling vugs have been studied in the dolostones of the Upper Cenomanian El-Heiz and Lower Middle Eocene Naqb formations at the northern part of the Baharyia Oasis (Gabal El-Tobog and El-Harra area, in particular). These vugs were observed abundantly in the dolostones of the uppermost parts of the studied formations. They are varied in size from 1 to 50 cm. These vugs are ellipsoidal, oval and irregular in shape. Petrographically, the calcites filling vugs of both of the studied formations exhibit two forms (early stage sparry calcite and late stage blocky calcite).

A combination of elemental chemistry, stable isotopes ($\delta^{18}O$ and $\delta^{13}C$) and fluid inclusions analyses showed that the early stage sparry calcite and late stage blocky calcite have a uniform elemental and isotopic compositions independent of their host dolomites. The early stage sparry calcites were found lining the vug walls, and formed as a result of dedolomitization of the marine El-Heiz and mixed-water Naqb host dolomites by aqueous fluids of temperature 35-40°C and salinity of 0.8-3.43 NaCl eq. Wt.%. On the other hand, the late stage blocky calcites were grown toward the vug centers, and precipitated from meteoric water supersaturated with $CaCO_3$ at a temperature 24-30 °C, without interaction with the host rocks.

The present study also helped in realising that the El-Heiz and Naqb formations in the study area were subjected to subaerial exposure and affected by meteoric water. This took place after the precipitation of the El-Heiz and Naqb formations at the Late Cenomanian and Early Middle Eocene ages, respectively.

INTRODUCTION

Calcites filling pores, vugs and cavities in the carbonate rocks exhibit variable fabrics (Flugel, 1982), and can occur under variable geochemical conditions (Meyers and Lohman, 1985; Holail, 1988 and Hays and Grossman, 1991). They are also recorded in a wide variety of depositional and diagenetic environments range from subtidal marine to vadose fresh water (Buchbinder and Friedman, 1980 and Tucker and Wright, 1991). Calcites filling small cavities in carbonate rocks were used as indicators of continental palaeoclimate (Esteban and Klappa, 1983; James and Choquette, 1984; Andrews *et al.*, 1994 and Muchez *et al.*, 1998) and karstification of carbonate rocks (El-Aref ,1994).

Trace elements and stable isotope analyses in conjunction with fluid inclusions studies of calcites filling pores, vugs and cavities in carbonate rocks have been increasingly used to know their origin in many geological settings world-wide (e. g., Walls *et al.*, 1979; Frank *et al.*, 1982; Harris *et al.*, 1985; Barnaby and Rimstidt, 1989; Goldstein, 1990; Kenneth *et al.*, 1996 and Holail, 1998). At the Bahariya Oasis, although the calcites filling vugs have been recorded in the Upper Cretaceous dolostones (Holail *et al.*, 1988 and Khalifa and Abu El-Hassan, 1993), and in the Middle Eocene carbonate rocks (El-Aref *et al.*, 1987), no studies have been done (as far as the authors are aware) to discuss the origin of those calcites on the basis of their stable isotopes and fluid inclusions. Therefore, the present work is pointed to explain the possible origin and mechanism of formation of that calcite filling vugs on the basis of a combination of petrographical, elemental chemistry, stable isotope and fluid inclusions studies.

GEOLOGICAL SETTING

The Bahariya Oasis is located in the central part of the Western Desert of Egypt, about 380 Km. southwest of Cairo. It lies between latitudes 27^0 48$'$ & 28^0 30$'$ N and between longitudes 28^0 35$'$ & 29^0 10$'$ E (Fig. 1). It is oval in shape and forms a great depression surrounded by escarpments. It exhibits a major anticlinal structure with axis trending NE-SW (El-Akkad and Issawi,1963 and El-Bassyouni, 1978). The regional geology of the Bahariya Oasis is well known since Ball and Beadnell (1903), El-Akkad and Issawi (1963) and El-Bassyouni (1972). Based on the stratigraphic classifications of El-Akkad and Issawi (1963), Said and Issawi (1964), Khalifa (1977) and El-Aref *et al.* (1991), the exposed rock units at the Bahariya Oasis are represented by the Lower Cenomanian Bahariya, Upper Cenomanian El-Heiz, Turonian- Santonian El-Hefhuf, Campanian-Maastrichtian Ain Giffara, Maastrichtian-Danian Khoman, Early Middle Eocene Naqb, Late Middle Eocene Qazzun, Late Middle Eocene to Late Eocene El-Hamra and Oligocene Qatrani formations and Oligo-Miocene volcanic rocks (Fig. 1).

Concerning the geological settings and aspects of the Upper Cenomanian El-Heiz Formation, a number of studies has been carried out (e. g., Hermina, 1957; Soliman *et al.*,1970; Khalifa, 1977; Soliman and El-Badry, 1980; Franks, 1982; Allam, 1986; Holail *et al.*, 1988 and Abu El-Hassan, 1992 and 1994). As well, the Naqb Formation has been studied by many authors (e.

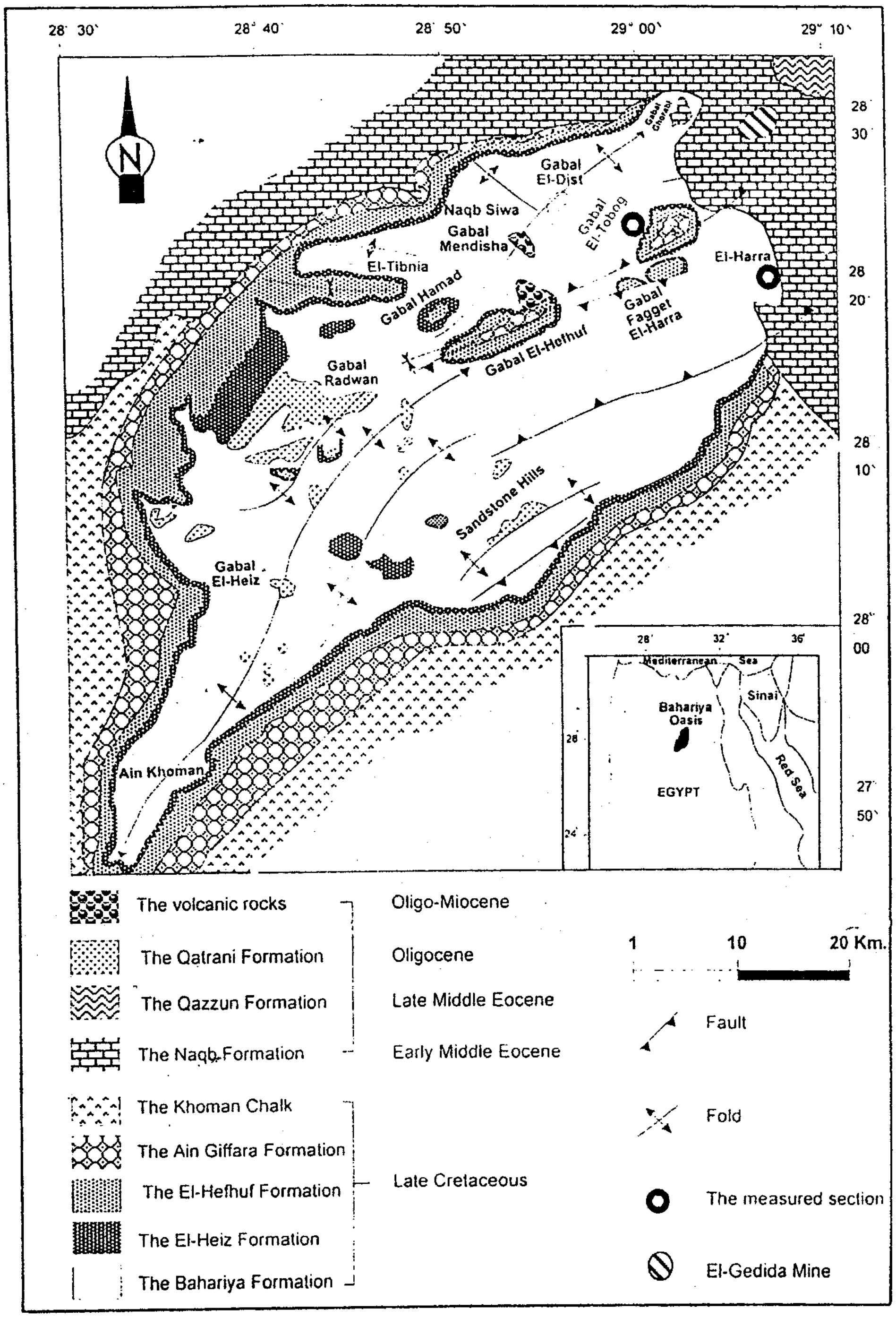


Fig. 1 Geological map of the Bahariya Oasis (after El-Akkad and Issawi, 1963 and after modification of Khalifa, 1977, El-Aref *et al.*, 1991 and Abu El-Hassan, 1994).

g., Said and Issawi, 1964; El-Aref and Lotfy, 1985; El-Aref *et al.*, 1987 and Lotfy, 1988).

ANALYTICAL METHODS

Samples of calcite filling vugs and the host dolomites were collected from the Upper Cenomanian El-Heiz and the Lower Middle Eocene Naqb formations at Gabal El-Tobog and El-Harra area, respectively. Representative samples were thin sectioned and examined under an optical microscope. Polished thin sections were stained by Alizarin redS to distinguish calcite from dolomite (Dickson, 1965). Fresh surface samples were gold-coated and investigated using A Jeol JSM-6400 Scanning Electron Microscope (SEM) equipped with an energy-dispersive X-ray analyzer (EDX). To study the fluid inclusion populations trapped within calcites, double polished thin sections were prepared using cold techniques to prevent laboratory reequilibration of fluid inclusions by overheating (Roadder, 1984). Samples with extensive open cleavage, twining and ferroan calcite were not used for analysis of fluid inclusions. Characteristics of fluid within the inclusions were determined by microthermometry using a Linkam Thm 600 heating/freezing stage provided with a thermal control unit (TMS-90, TP-90). The lower and upper limit of the stage are -190°C and 600°C.

Powders of representative calcite filling vugs and associated host dolomite samples were analyzed by Perkin Elmer model 603 Atomic Adsorption to determine the amount of trace elements (Sr, Na and Mn). Calcium, magnesium and iron contents were analyzed by using a Cameca Camebax Microbeam CX n-827 electron microprobe (EPM) equipped with a link EDS system. Operating conditions for EPM were 15 kv accelerating voltage and 30 nA beam current with 2 µm spot size. The $\delta^{18}O$ and $\delta^{13}C$ analyses were carried out according to the classical method of McCrea (1950). To perform carbon-and oxygen isotopic analysis, about 10 mg of calcites filling vug as well as the associated host dolomites were extracted from polished slabs using a 6 mm microdrill. These extracted powders were vacuum roasted for one hour at 370^0 C , and then reacted with 100% anhydrous $H_3\ PO_4$ at 60^0 C. The liberated CO_2 from calcite were collected after 24 hours, whereas for dolomite the liberated CO_2 was collected after 27 hours. The collected CO_2 was analyzed by multi-collector FINNIGAN 252 mass spectrometer. The isotopic compositions were expressed as δ values in per mil (‰) relative to the PDB standard (Craig,1957). All samples were analysed twice and the precision of the measurements is better than + 0.5‰ for both $\delta^{18}O$ and $\delta^{13}C$.

FIELD OBSERVATIONS

The Upper Cenomanian El-Heiz Formation crops out in the southern and northwestern escarpments of the Bahariya Oasis whereas in the extreme northeastern sector of the Oasis escarpments it is not present (Fig. 1). At Gabal El-Tobog, the El-Heiz Formation shows unconformable relationship with the dolostones of the overlying El-Hefhuf Formation and the clastic beds of the underlying Bahariya Formation (Fig. 2). The El-Heiz Formation consists of about 15 m of alternating beds of brown marly dolostone, and grey to olive shale. The dolostones of the lower part of the El-Heiz Formation are sandy and massive, whereas in the upper part of such formations the dolostones have numerous vugs filled with calcites. These dolostones are covered by undulatory layer of calcrete (Plate 1A). Such calcrete layer occurs at the contact between the El-Heiz dolostones and its overlying El-Hefhuf Formation (Plate 1A).

The Lower Middle Eocene Naqb Formation covers only the northeastern and northwestern parts of the Bahariya Oasis (Fig. 1). At El-Harra area, the Naqb Formation is unconformably underlain by the clastic strata of the Lower Cenomanian Bahariya Formation. The upper contact of the Naqb Formation still exposed to the surface until now and is covered by ferricrete duricrusts. The Naqb Formation is made up of yellowish brown dolomitic limestone of 20m thick, that are vuggy in its upper part (Fig. 2). Calcites filling vugs studied here were collected from the dolomitic rocks of the uppermost parts of the both of the studied formations, where the calcites filling vugs are abundant (Fig. 2).

As mentioned above, the dolostones of the studied formations have numerous vugs in their uppermost parts (Fig. 2). Most of these vugs are filled with calcite and they are varied in size from 1 to 50 cm. These vugs are ellipsoidal, oval and irregular in shape (Plate 1B and Plate 2A,B). Iron oxide coating is often encountered in the contact between vug wall and host dolostone (Plate 1B, C and D). In hand specimen, the calcite filling vug appears variable in crystal size which increases generally from the vug wall toward its centre (Plate 1B and Plate2A,B). These calcite crystals commonly stand sub-perpendicular to substrate (dolostone), and form radiating pattern that coalesce toward the vug center (Plate 1B and Plate2A,B).

PETROGRAPHY

The petrographical investigations have been carried out to emphasize and demonstrate the different fabrics of calcite filling vugs. To achieve this petrographical investigation, the calcite fillings and their host dolomites were thin sectioned and examined under a polarizing microscope. The calcite fabrics have been classified according to Flugel's terminology (1982). However, terminology proposed by Friedman (1965) is used to describe the dolostones.

The petrographical investigations showed that the host dolostone of the El-Heiz Formation differs from that of the Naqb Formation. The El-Heiz dolostone is essentially made up of fine dolomite rhombs (40-80 um) exhibiting idiotopic to hypidiotopic fabric and equigranular texture. Most of these dolomite rhombs are zoned containing iron-rich rhombohedral cores surrounded by clear outer rims (Plate 3A). The other rhombs are clear without iron cores. The Naqb

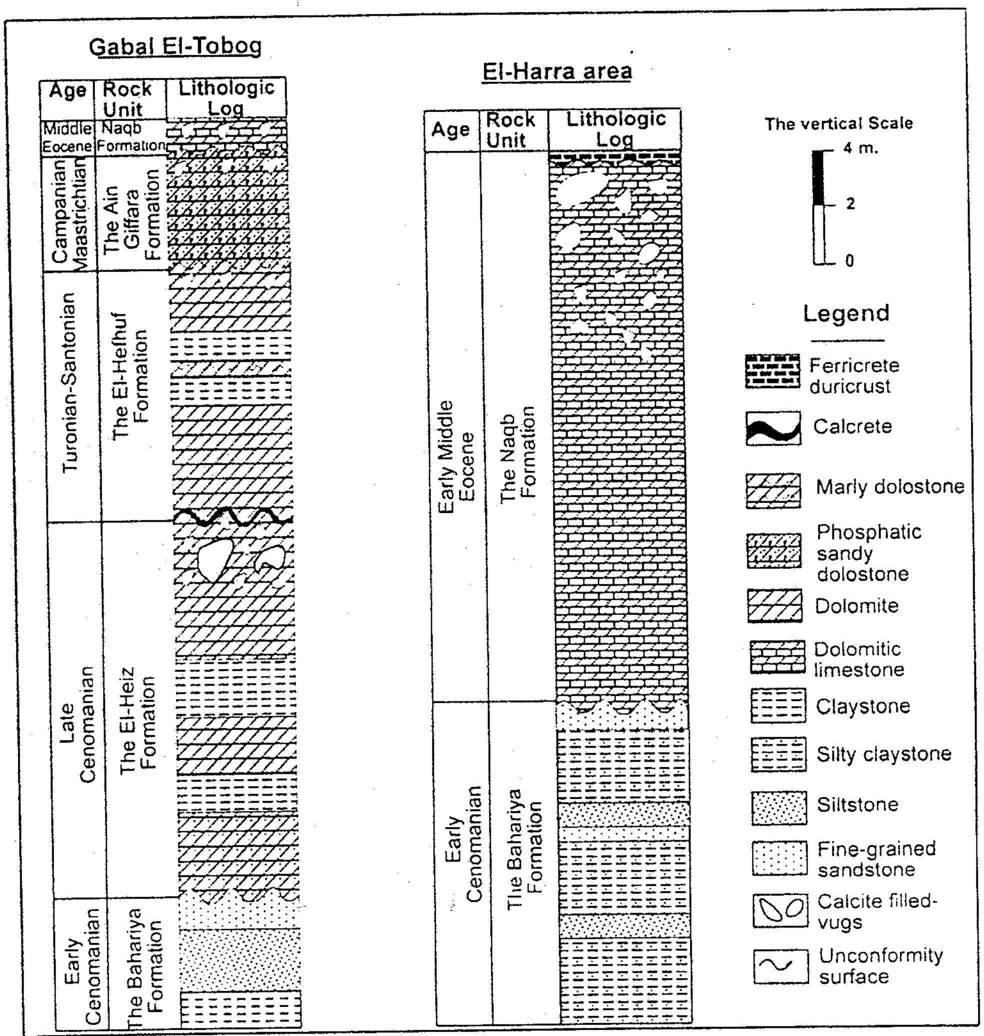
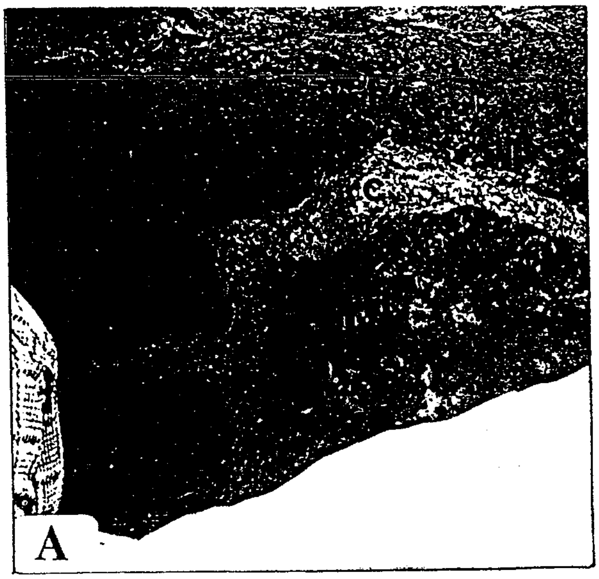
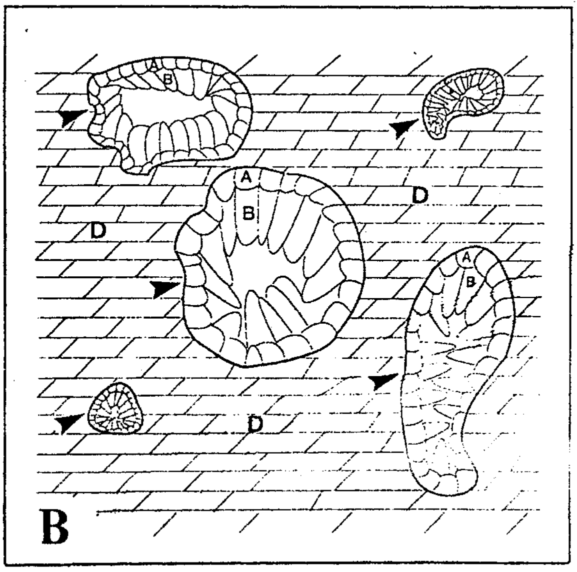


Fig. 2 Lithostratigraphic sections of the El-Heiz Formation at Gabal El-Tobog and Naqb Formation at El-Harra area, showing the occurrences of the calcite filled-vugs.

Plate 1

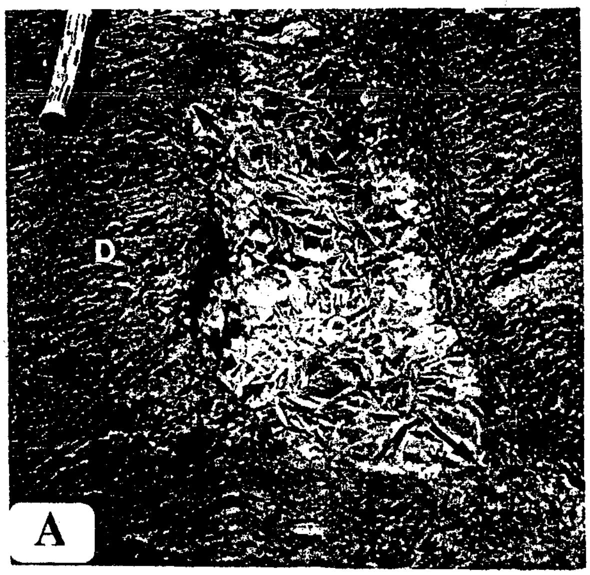


(A) Field photograph showing an undulatory layer of calcrete between the El-Hefhuf Formation (F) and the El-Heiz Formation (H) at Gabal Tobog.

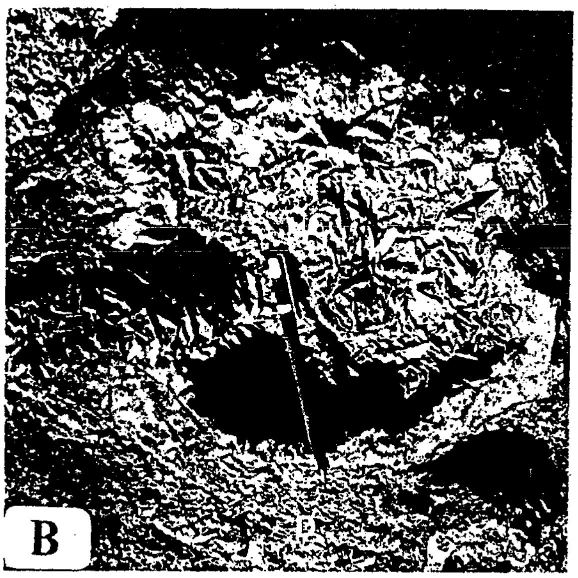


(B) Sketch model showing the different sizes and shapes of calcite-filled vugs in the host dolomite (D). Notice, the calcite crystals increase in size from the vug walls toward the vug centers, forming a sparry calcite (A) lining the vug wall and a blocky calcite (B) filling the vug center. Notice, the occurrence of black Fe and Mn oxides rim at the boundary between vug wall and host dolomite (arrows).

Plate 2



(A) Field photograph showing a calcite-filled vug (c) in the host dolomite (D) of the El-Heiz Formation at Gabal Tobog. Notice, the occurrence of black Fe and Mn oxides rim at the boundary between vug wall and host dolomite (arrows).



(B) Field photograph showing a calcite-filled vug (c) in the host dolomite (D) of the Naqb Formation at El-Harra area. Notice, the occurrence of black Fe and Mn oxides rim at the boundary between vug wall and host dolomite (arrows).

dolostone consists of xenotopic dolomite crystals containing cores of iron oxide patches and clear outer rims (Plate 3B).

Thin section study revealed that there are two main generation of calcite filling the vugs. These are sparry and blocky calcites. The sparry calcite coats the vug wall without a sharp contact with the host rocks, and is herein called "early stage calcite" The sparry calcite consists of subhedral to anhedral crystals (2-4 mm in size) exhibiting hypidotopic to xenotopic fabric and equigranular texture enclosing relics of dolomite rhombs and iron oxide patches (Plate 3A and B) The SEM image showed that the sparry calcite has relics of corroded dolomite rhombs (Plate 4A). Such occurrence of dolomite relics in the sparry calcite which is adjacent to the host dolostone indicates that the sparry calcites filling vugs were formed from dedolomitization of host dolostone (Buchbinder and Friedman, 1980; Holail *et al.*, 1988 and Khalifa and Abu El-Hassan,1993). This is also confirmed by the occurrence of the black coloured- iron and manganese oxide patches (as confirmed by microprobe analysis) at the boundary between the sparry calcite and the associated host dolostone (Plate 2A and B). This iron and manganese oxides boundary were most probably resulted from leaching of iron and manganese from host dolomites during their dedolomitiztation to form sparry calcites (Khalifa, 1977). The blocky calcite was grown toward the vug center following the first generated sparry calcite. The blocky calcite shows large euhedral prismatic crystals (4-6 mm in size) without any mineral inclusions (Plate 4B). This blocky calcite is herein defined as "late stage calcite"

FLUID INCLUSIONS

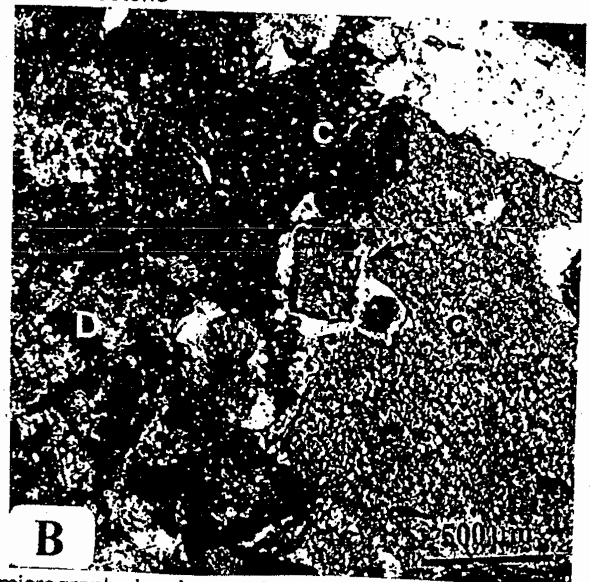
Calcite is one of the most useful minerals for the observation of fluid inclusions in sedimentary rocks and can be used to estimate temperatures and compositions of paleofliuds during diagenesis. However, the methodology of fluid inclusion microthermometry in calcite has been poorly investigated, unlike detailed studies of inclusions in quartz, sphalerite and halite (see review in Hollister and Crawford, 1981 and Roadder, 1984). Gratier and Jenatton (1984) and Prezbindowski and Larese (1987) demonstrated experimentally that fluid inclusions trapped in calcite may reequilibrate by stretching during heating beyond the homogenization temperature. Here, the fluid inclusions in calcites are used by means of defining how the populations of homogenization temperature (T_h) and melting temperature (T_m) are for the diagenetic fluids.

Fluid Inclusions Petrography

All of the fluid inclusions of the studied samples are H_2O mono- or two-phase and their size are variable, but most are between 4 and 20μm (Plate 5A and B). Many healed fractures contain secondary fluid inclusions that are mostly two phase H_2O with consistent liquid to vapour ratios (the vapour phase is about 10-15 vol.%). Primary fluid inclusions are identified by their distribution along

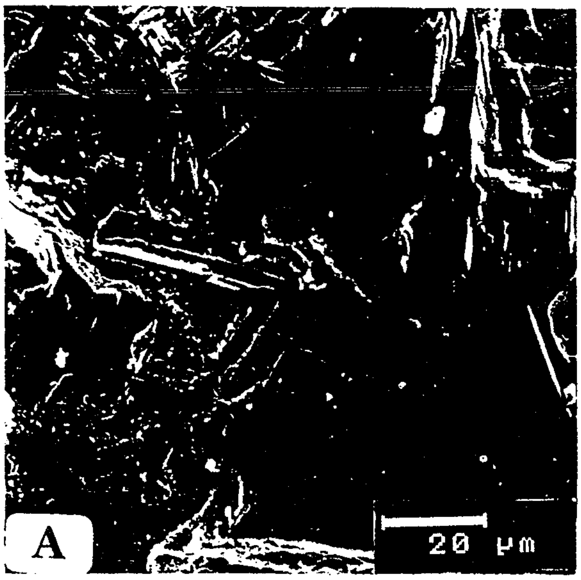
Plate 3

(A) Photomicrograph showing a calcite calcite (c) filling vug of the dolostone of the El-Heiz Formation at Gabal Tobog. This calcite contains relics of dolomite rhombs (arrows), and exhibits irregular contact with the host dolomite (D). Notice, the xenotopic to hypidotopic dolomite rhombs of the host dolostone

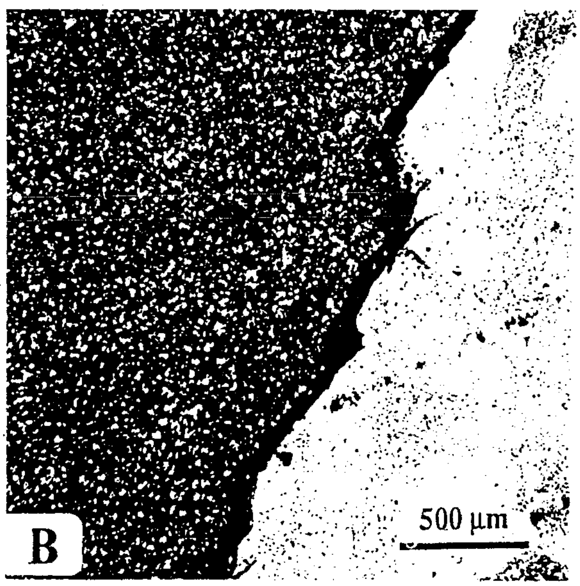


(B) Photomicrograph showing a calcite calcite (c) filling vug of the dolostone of the Naqb Formation at El-Harra area. This calcite contains relics of dolomite rhombs (arrows), and exhibits irregular contact with the host dolomite (D). Notice, the xenotopic to hypidotopic dolomite rhombs of the host dolostone

Plate 4

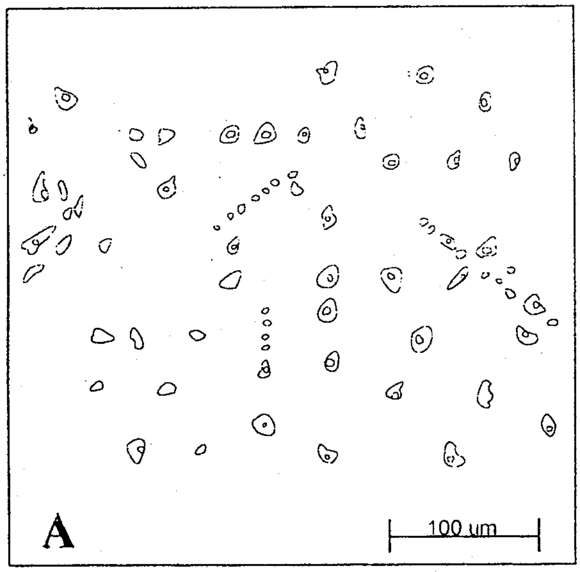


(A) Scaning Electron Microscope (SEM) image of the sparry calcite (c) of the El-Heiz Formation enclosing relics of dolomite rhomb (arrow).

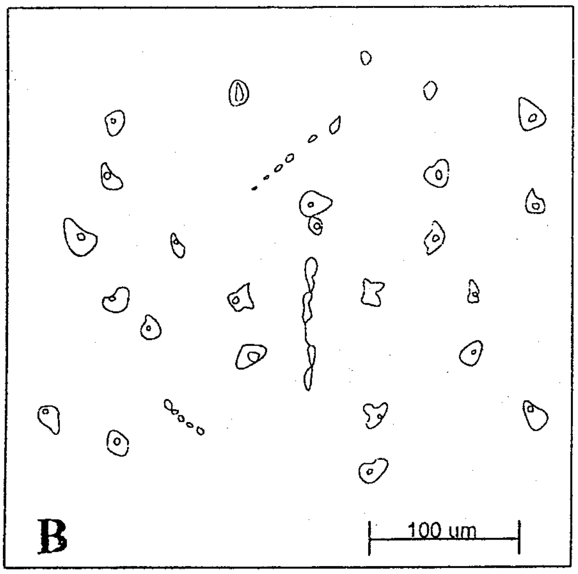


(B) Photomicrograph showing parts of two blocky calcite crystals that filling the dolostone vugs of the Naqb Formation at El-Harra area.

Plate 5



(A) Sketch of the mono-and bi-phase H_2O fluid inclusions in the sparry and blocky calcites filling vugs in the dolostones of the El-Heiz Formation at Gebel Tobog.



(B) Sketch of the mono-and bi-phase H_2O fluid inclusions in the sparry and blocky calcites filling vugs in the dolostones of the Naqb Formation at El-Harra area.

growth zones and between or parallel cleavage planes. Primary fluid inclusions are mostly one phase H_2O and in some calcite crystals they dominate, suggesting that they are not the result of necking down of two phase inclusions. The mono-phase H_2O inclusions do not appear to contain significantly metastable 'stretched" H_2O because they are in the same size range as many of the two phase inclusions, and bubbles could not be nucleated during cooling. Studied fluid inclusions have various shapes including tubular, negative crystal and irregular outlines.

Microthermometry

The microthermometry data of the investigated fluid inclusions were obtained from a prepared doubly polished sections to a thickness of 100 μm (Cleavage sections were broken to a thickness of <1 mm and used without polishing) and are represented in table (1).

Table (1) Microthermometry data of the investigated mono-phase H_2O fluid inclusions in calcites of the studied formations.

Rock unit	Sparry calcite °C		Blocky calcite °C	
	T_h	T_m	T_h	T_m
The Naqb Formation	39	-1.5	27	0
	38.7	-1.0	27.5	0
	39.3	-0.5	28	0
	36	-1.3	30	0
	40	-0.9	25	0
The El-Heiz Formation	37	-1.3	24	0
	35	-1.1	27	0
	38.9	-2.0	26	0
	36.5	-2.1	28	0

It is important here to refer that all the measured fluid inclusions were selected as mono-phase to avoid the fluid inclusions which affected by stretching (bi-phase inclusions) and metastability (notably the small mono-phase fluid inclusions).

Homogenization temperature (T_h) Heating fluid inclusions to temperatures greater than the T_h can cause subsequent Th determinations to yield higher values due to reequilibration (Burruss, 1987; Prezbindowski and Larese, 1987). It is not a general rule and seems to depend on the value of T_h and the shape, size and composition of the inclusion as well as the duration and value of overheating. Overall, these observations show that overheating must be avoided. Lawler and Crawford (1983) demonstrated that low-temperature and low-salinity fluids trapped in sphalerite are likely to stretch during freezing. High salinity fluids do not expand on freezing, and so do not cause stretching, and high-temperature of homogenization inclusions have large vapour bubbles which provide space for ice to expand into. In this study, similar results have been found for calcite: for the same inclusion, T_h values measured after determining the melting temperature (T_m) were generally greater than values measured before T_m. The difference of the two determinations is not correlated with the shape of the fluid inclusions or the first value of T_h but seems to increase with the size of the fluid inclusions. Therefore, in order to avoid overestimation of the T_h for calcite studied here, the T_h was determined before the Tm. In the calcite studied here, T_h values of aqueous fluid inclusions in sparry calcite at the contact with dolomite are 35-40°C and for blocky calcite filling vugs are 24-30 °C.

Melting temperature (Tm) In H_2O fluid inclusions, Tm is defined by the final melting of the last crystal of ice in the presence of the vapour bubble during warming. For the calcite studied here, the vapour phase is rarely present during freezing but appears between the temperature of eutectic (first melting temperature) and the melting temperature. Tm determination in this study (Table. 1) displays a range of -0.5 to -2.1 °C for aqueous inclusions of the sparry calcite (showing bulk molar volume 18.12-18.03, density 0.99-1.02 and 0.8-3.43 NaCl eq.wt.%), while Tm of aqueous inclusions in the blocky calcite filling vugs have 0 °C (indicating bulk molar volume 18.16, denesity 0.99 and 0 NaCl eq.wt.%). The bulk molar volume, denesity and NaCl eq.wt.% are calculated according to Brown and Lamb (1989) equation of state.

Fluid Inclusions Discussion

For the large aggregates of sparry calcite, fine but discontinuous growth banding, coarse and fine solid inclusions ?, euhedral termination, growth asymmetry, and mode of occurrence in solution pockets strongly support a vadose to possibly phreatic calcite speleothem origin; most of these features are well known from speleothems that form in modern vadose zones (Kendall and Broughton, 1978). Therefore, the observations suggest filling of karast cavities took place in a low-temperature environment that was close to a subaerial surface. The simple petrographic observations of fluid inclusions indicate that the calcite precipitated in low-temperature (less than 50°C, notably mono-phase H_2O inclusions) vadose zone, and then probably was heated as a result of deep burial. The pressure increases in fluid inclusions, caused by heating after calcite precipitation, resulted in re-equilibration of some low-temperature fluid inclusions by leakage and stretching of the fluid inclusion cavities. The all-liquid H_2O inclusions faithfully preserve the record of fresh water conditions of precipitation, whereas the two phase H_2O inclusions were formed during thermal re-equilibration. Therefore, in a general sense, these data indicate that although some of the original fluid inclusions re-equilibrate when low-temperature calcite are subjected to burial heating, some of the original inclusions preserved. One phase, all -liquid H_2O inclusions survive burial heating to provide a record of the temperature and original salinity of low-temperature calcite precipitation.

ELEMENTAL CHEMISTRY

Major elements. Elemental analysis of Mg and Ca from electron microprobe demonstrates that the El-Heiz and Naqb dolomites are non-stoichiometeric containing $CaCO_3$ that ranges from 52.93 to 56.12 mole% and $MgCO_3$ that ranges from 41.97 to 45.12 mole% (Table 2). The microprobe data also show that the early stage sparry and late stage blocky calcites filling vugs of the studied formations are chemically similar (Table 2). The sparry calcites have relatively high Mg CO_3 (8.69-10.33 mole%) relative to the blocky calcites which are depleted in Mg CO_3 (0.48-0.49 mole%). As evidenced by petrographical and SEM studies, the relatively high content of Mg CO_3 mole% in the sparry calcites may be related to abundant distribution of dolomite rhombs from the host dolomites (see Plate 3A, B and Plate 4A). This is in turn indicates that there was an interaction between the host dolomites and the fluids responsible for sparry calcites formation. On the other hand, the very low value of Mg CO_3 (less than 2 mole%) in the blocky calcites is more typical to cementation from meteoric water (Tucker and Wright, 1990). Furthermore, the more depletion of Mg in the blocky calcites relative to the above cited sparry calcites may indicate that there is no interaction between the meteoric water and the host dolostones during formation of the blocky calcites. This is in harmony with that deduced by Meyers and Lohman (1985) who suggested that the depletion of Mg in calcite within host dolomites represents very low degree of host rock-meteoric water interaction.

Iron and Mn contents of the El-Heiz dolomites and calcites filling their vugs are relatively higher than those of the Naqb Formation (Table 2). In both formations, there is a decrease in Fe and Mn contents from host dolomite passing toward sparry calcite to blocky calcite (Table 2). Such progressive depletion in Fe and especially Mn may be related to an increase of degree of host rock-meteoric water interaction during sparry calcite formation rather than during precipitation of blocky calcites.

Trace elements Study of the trace elements, Na and Sr, in particular, are the most useful indicators of salinity during deposition of carbonates (Land and Hoops, 1973 and Land, 1980). The samples analyzed for Na showed that the El-Heiz dolomites have high Na contents relative to that of the Naqb dolomites (Table 2). The Na concentration in the El-Heiz dolomites (560-740 ppm) is very similar to marine water dolomites (Land and Hoops, 1973 and Mitchell *et al.*, 1987). On the other hand, the Na contents in the Naqb dolomites (370-450 ppm) is very close to the Na concentration of mixed-water dolomites (Land *et al.*, 1975; Randazzo and Cook, 1987 and Holail, 1989).

Table 2. Chemical analysis data and stable isotope results of the calcites filling vugs and their associated host host dolomites of the El-Heiz Formation at Gabal El-Tobog and Naqb Formation at El-Harra area.

Rock unit	Sample No.	Minerals	$CaCo_3$ (Mole%)	$MgCo_3$ (Mole%)	$FeCo_3$ (Mole%)	Sr (ppm)	Na (ppm)	Mn (ppm)	^{18}O (%PDB)	^{13}C (%PDB)
	1AN	Host dolomite	55.43	42.22	1.48	144	450	690	-1.46	-0.65
	2AN	Host dolomite	56.03	41.79	1.19	93	290	580	-2.17	-1.20
	3AN	Host dolomite	57.18	41.92	1.11	120	370	760	-1.98	0.23
	Average		**56.12**	**41.97**	**1.26**	**119**	**370**	**676**	**-1.87**	**-0.54**
	1BN	Sparry calcite	92.07	7.53	0.61	55	260	470	-8.48	-3.84
	2BN	Sparry calcite	89.92	9.39	0.46	32	180	360	-5.85	-5.16
	3BN	Sparry calcite	90.35	9.16	0.51	60	200	280	-7.61	-3.98
	Average		**90.78**	**8.69**	**0.52**	**49**	**213**	**370**	**-7.31**	**-4.32**
	1CN	Blocky calcite	98.80	0.89	0.23	55	160	200	-10.44	-7.18
	2CN	Blocky calcite	99.30	0.35	0.06	38	120	330	-10.30	-6.20
	3CN	Blocky calcite	99.33	0.22	0.29	13	80	265	-10.50	-6.14
	Average		**99.47**	**0.48**	**0.19**	**35**	**120**	**265**	**-10.41**	**-6.50**
	1AH	Host dolomite	53.49	44.58	1.91	152	560	2140	-2.57	0.67
	2AH	Host dolomite	52.28	45.37	3.07	140	670	2180	-2.43	0.90
	3AH	Host dolomite	53.03	45.42	2.59	144	740	1780	-1.89	0.63
	Average		**52.93**	**45.12**	**2.52**	**145**	**656**	**2033**	**-2.29**	**0.73**
	1BH	Sparry calcite	88.10	11.14	0.58	65	240	730	-9.62	-3.38
	2BH	Sparry calcite	89.80	9.76	0.64	70	190	450	-9.84	-4.57
	3BH	Sparry calcite	88.89	10.09	0.06	82	310	630	-8.70	-5.94
	Average		**88.93**	**10.33**	**0.42**	**72**	**246**	**603**	**-9.38**	**-4.63**
	1CH	Blocky calcite	98.44	0.45	0.51	70	130	500	-11.73	-8.98
	2CH	Blocky calcite	97.21	0.82	0.43	53	170	310	-12.02	-7.20
	3CH	Blocky calcite	98.57	0.20	0.37	40	220	423	-10.98	-8.82
	Average		**98.07**	**0.49**	**0.43**	**54**	**173**	**411**	**-11.57**	**-8.33**

Symbols

A = Host dolomite.
B= Sparry calcite lined the vug wall.
C= Blocky calcite growing toward the vug center.
H= The El-Heiz Formation.

The Sr contents of the dolomites of El-Heiz and Naqb formations are nearly similar (119-145 ppm). Such Sr contents are low relative to those of many ancient marine and hypersaline dolomites (Behrens and Land,1972; Land, 1980 and Mitchel *et al.*, 1987). This could be resulted from the effect of meteoric water after their dolomitization process (Brand and Veizer, 1980).

The Na and Sr concentrations in the calcites filling vugs of the studied formations are nearly similar independent of their host rocks (Table 2). This may indicate that these calcites were formed from similar fluids differ from those of the host dolomites. Na and Sr results exhibit a progressive depletion from the sparry to blocky calcites (Table 2). Kinsman (1969) has shown that calcite precipitating from sea water contains Sr content of about 1200 ppm, and Land and Hoops (1973) stated that calcite of marine origin has Na content greater than 1000 ppm. Consequently, the low values of Sr and Na (35 ppm and 120 ppm, respectively) concentrations in the present suggest that the fluids responsible for formation of the studied calcites filling vugs are most probably fresh water. Moreover, the progressive depletion of Sr and Na from the sparry to blocky calcite indicates a decrease of interaction of the fresh water with the host dolostones from the vug wall toward vug centre (Brand and Veizer, 1980).

OXYGEN AND CARBON ISOTOPES

Study of stable isotopes of carbon and oxygen was considered as a strong tool in discrimination between marine and fresh water carbonate sediments. This was based on the idea that the marine carbonates are enriched in $\delta^{18}O$ and $\delta^{13}C$ relative to the fresh water carbonate which are depleted in $\delta^{18}O$ and $\delta^{13}C$ (Clayton and Degens, 1959; Keith and Weber, 1964; Tan and Husdon, 1971 and Andrews *et al.*, 1993). It is also accepted that the general trend during diagenesis of carbonate rocks by meteoric water is a depletion in $\delta^{18}O$ and $\delta^{13}C$ isotopes (Emery *et al.*, 1954; Land *et al.*, 1975; Walls *et al.*, 1979; El-Hennawi and Loukina, 1993 and Loukina and El-Hefnawi, 1997).

In the present work, in order to reconstruct the original conditions under which the calcites filling vugs were formed in the host dolostone, the oxygen and carbon isotope compositions from 12 samples of sparry and blocky calcites filling vugs and 6 samples of the associated host dolostones have been determined (Table 2). These samples represent the host dolomite and calcite filling of six vugs, three of them from the El-Heiz Formation and the others from the Naqb Formation. The results showed that $\delta^{18}O$ and $\delta^{13}C$ values of calcites filling vugs and the host dolomites of the studied formations are very similar (Table 2 and Fig. 3).

In order to facilitate the interpretation of $\delta^{18}O$ and $\delta^{13}C$ isotope data to know the type of water responsible for formation of calcites filling vugs in the host dolomites, the authors assume that the calcites filling vugs and their host dolomites were precipitated together in the same equilibrium system. Therefore, the host

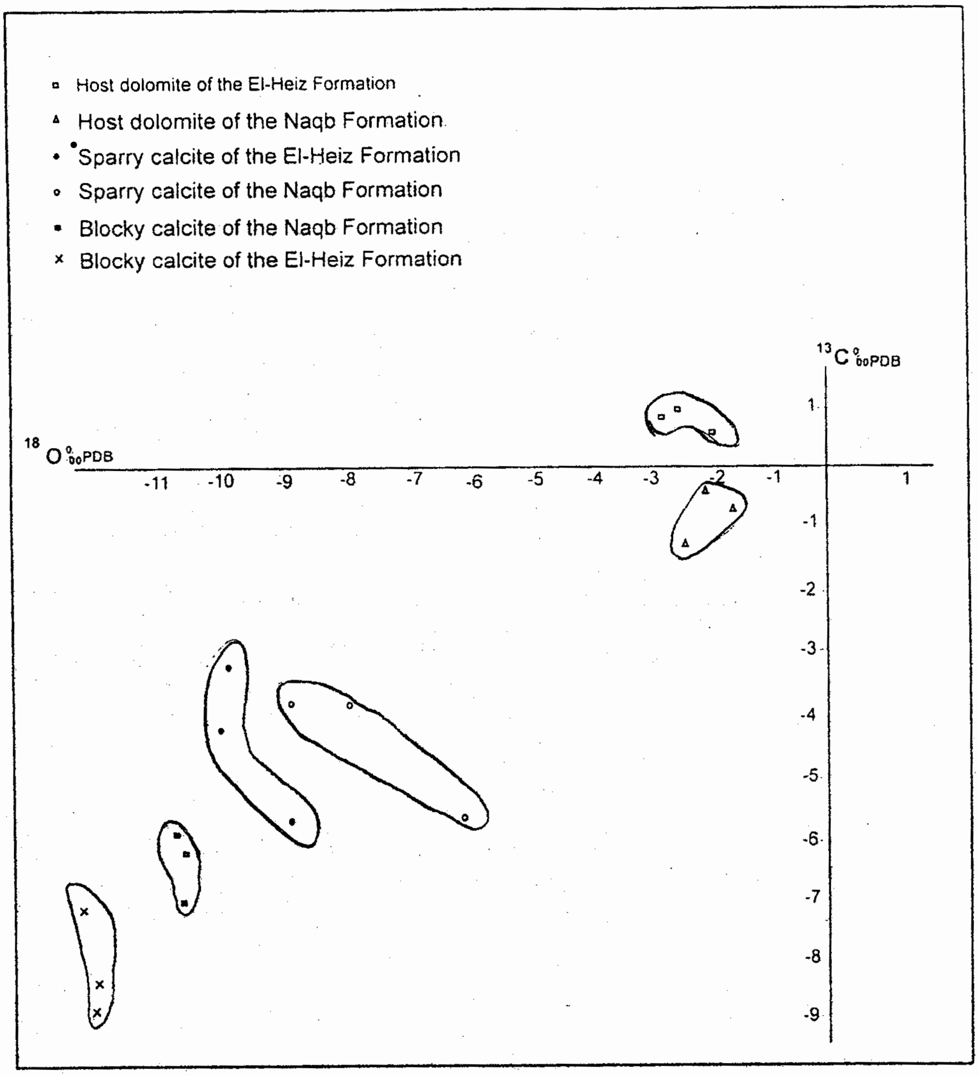


Fig. 3 Carbon and oxygen isotopic compsition of the calcites filling vugs and their host dolomites in the El-Heiz Formation at Gabal El-Tobog and Naqb Formation at El-Harra area.

dolomites should be about 3% enriched in $\delta^{18}O$ values relative to $\delta^{18}O$ values of the calcites (Land, 1985). This fact has not been observed herein and the host dolomites have heavier $\delta^{18}O$ values relative to calcite (Fig. 3). Such a significant wide spread in $\delta^{18}O$ values between host dolomite and calcite filling vugs indicates that each of which has been formed under a distinct water. The $\delta^{18}O$ values of host dolomites in the studied two formations (from -2.92 to -1.87‰ PDB) may exclude any dolomitization by hypersaline water (Kinsman and Patterson, 1973). The $\delta^{18}O$ values of the El-Heiz dolomite (0.73 ‰ PDB) are compatible with the Upper Cretaceous marine dolomites in other geological settings (Scholle and Arthur, 1980). On the other hand, The $\delta^{18}O$ values of the Naqb dolomite (-0.54‰ PDB) are closely similar to mixed-water dolomites (Land *et al.*, 1975). The $\delta^{18}O$ values of the early stage sparry calcites filling vugs in both of the studied formations range from -9.83 to -7.31‰ PDB, and from -11.57 to -10.41‰ PDB for the late stage blocky calcite (Late stage calcite). Since the best estimate of $\delta^{18}O$ values of marine calcite during the Upper Cretaceous is about -2 to -1‰ PDB (Scholle and Arthur, 1980 and Lohman and Walker, 1989), and that of the Eocene marine calcites is 1.4 to -2.9‰ PDB (Lohman and Walker, 1989 and Holail,1998), the isotopic compositions of the studied Cretaceous and Eocene calcites filling vugs are not to be marine, but are closely similar to those of calcite formed by meteoric water (Meyers and Lohman, 1985; Platt, 1989; Goldstein, 1990 and Hayes and Grossman, 1991). Also, the more depletion of $\delta^{18}O$ value (-8 to -10‰ PDB) of blocky calcites relative to their marine and mixed-water host dolomites clearly supports that they were deposited as a separate phase from circulating solution of meteoric origin without interaction with the host dolomites (Alfonsi *et al.*, 1991). Moreover, such more depletion of $\delta^{18}O$ values of the blocky calcites may indicate an increase of rain fall during their formation (Muchez *et al.*, 1998).

Since carbon isotopic composition of calcite cements in carbonates is controlled by the relative amount of CO_2 supplied by soil-zone processes and/or by calcium carbonate dissolution (Andrews *et al.*, 1993). The more depleted $\delta^{13}C$ value of the studied sparry and blocky calcites (-8.33 to -3.38‰ PDB) represents an isotopically light composition which refers to a precipitation from fluids rich in soil-gas CO_2 that form during subaerial exposure (Allan and Mattheus, 1982).

THE PROBABLE MECHANISM OF FORMATION OF THE CALCITE-FILLED VUGS

Different mechanisms of calcite filled vugs in carbonate rocks of the geological record have been discussed by different authors. The calcite filled vugs can be originated from an alteration of fossil molds by fresh water (Bathurst, 1964; Hudson, 1962 and Richter and Fuchtbauer, 1978). Longman (1980); Ford (1985); Craig (1985); El-Aref, *et al.* (1987) and El-Aref (1994) ascribed the formation of calcite filled cavities or vugs in carbonate rocks to the near surface-meteoric diagenesis (karstification process). Egemeier (1981) linked the development of calcite filled cavities or vugs in carbonate rocks to the action of hydrothermal water. Queen *et al.* (1977) suggested a model for the genesis of calcite filled caves or vugs in host carbonates. He stated that the calcite was replaced gypsum due to later meteoric water phase.

In the present study, it is clear that : (1)- the studied calcites filled vugs were not originated from a calcitization of fossil molds as evidenced from the following:- (a)- lack of any remanant of shell fossils in the vugs. (b)- occurrence of the calcites as drusy cement without preserved relics of internal structure of shells. (c)- very low values of Sr (35 ppm), Na (120 ppm), Fe CO_3 (0.19-0.43 mole%) and Mg CO_3 (less than 2 mole%) and more depletion of stable isotopes of $\delta^{18}O$ value ($^{\delta 18}O$ =-8 to -10‰ PDB, $\delta^{13}C$ = -8.33 to -6.50‰ PDB) of calcite filling vugs which in turn indicate meteoric water action and increasing rain falls, (2)- dominance of the calcite filled vugs in the uppermost parts rather than lowermost parts of the studied dolostones (Fig. 2), (3)- the studied calcites filled vugs are not of hydrothermal water origin (as evidenced from the foregoing fluid inclusions study), (4)- formation of the early stage sparry calcite from dedolomitization of the host dolostone (as shown by the petrographical investigation), (5)- formation of the late stage blocky calcite from fresh water supersaturated with $CaCO_3$ without host rock interaction (as evidenced from the trace elements and stable isotope data), (6) occurrence of calcrete bed (which is a good indicator to paleosols, Wright and Tucker 1991) at the upper contact of the El-Heiz Formation (Plate 1A), and (7) the occurrence of ferricrete duricrust (which is a good indicator to paleokarsts, El-Aref, *et al.,* 1991 and El-Aref, 1994) on upper contact of the Naqb Formation which still exposed to the surface until now.

From the foregoing discussion about the different mechanisms of formation of calcites-filled vugs in the carbonate rocks, and the characteristics of the studied calcites-filled vugs cited above, the authors suggest that the most probable mechanism for vugs formation and their filling with calcites is the interplay between the meteoric water and the host dolostones during subaerial exposures after the precipitation of the El-Heiz and Naqb formations at the Late Cenomanian and Early Middle Eocene ages, respectively. This probable mechanism may be developed in two stages: (1) meteoric water-selective dedolomitization and dissolution of the subaerial exposed the El-Heiz as well as Naqb dolomites forming empty vugs lined by sparry calcite. Later on, (2)- the early formed sparry calcite followed by precipitation of blocky calcite grown toward the vug centre by a meteoric water supersaturated with $CaCO_3$ without interaction with the host dolostone.

The deduced origin of the calcites filling vugs in the El-Heiz Formation confirms the opinions of Khalifa and Abu El-Hassan (1993) concerning an occurrence of subaerial exposure by the end of deposition of the El-Heiz Formation. On a similar way, the deduced origin of the calcite filling vugs in the Naqb formation supports the discussion of El- Aref *et al.*, 1987 and El-Aref (1994) about an occurrence of an intensive karstification of the Naqb limestone of northern part of the Bahariya Oasis at the Early Middle Eocene.

CONCLUSIONS

In the dolostones of the Upper Cenomanian El-Heiz and Lower Middle Eocene Naqb formations, the calcites filling vugs were observed dominantly in their uppermost parts. These vugs occur in different shapes and sizes. Petrographically, the calcites filling vugs exhibit two forms (early stage sparry calcite and late stage blocky calcite). The early stage sparry calcite lined the vug wall and are separated from the host dolomite by manganese and iron oxides rim. The late stage blocky calcite follows the first generated sparry calcite and grows toward the vug center.

It is quite clear from fluid inclusions data that the different forms of calcite growth (sparry and blocky) exhibit different salinities and subsequent time span of the aqueous fluids which were most probably responsible for calcite precipitation. From the T_h and T_m it is clear that the sparry calcite was formed under aqueous fluids of very low salinity (0.8-3.43 NaCl eq.wt.%) at temperature ranges from 35-40^{0}C. Later on, the blocky calcite filling vugs was precipitated from fresh water at 24-30^{0}C. The simple petrographic observations of fluid inclusions indicate that the calcite precipitated in low temperature (less than 40°C) vadose zone.

The oxygen and carbon isotopic compositions of calcites filling vugs in the host dolostones showed that there is more depletion of $\delta^{18}O$ and $\delta^{13}C$ isotopes in the calcites relative to their host dolostones. Consequently, it is clear that the sparry calcite formed from dedolomitization of the host dolomite by meteoric water effect, whereas the blocky calcite was probably formed from meteoric water supersaturated with $CaCO_3$ without interaction with the host rock.

Combination of the pertrographical, fluid inclusions, elemental analysis and stable isotopic ($\delta^{18}O$ and $\delta^{13}C$) data of the calcite -filled vugs emphasizes that the El-Heiz and Naqb formations were subjected to subaerial exposures after their formation at the Late Cenomanian and Early Middle Eocene ages, respectively.

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أصل فجوات الكالسيت فى صخور الدلوميت لمتكون الحيز (السينومانى العلوى) ومتكون النقب (الوسط أيوسينى السفلى) فى الواحات البحرية باستخدام أدلة المحصورات المائية والنظائر الثابتة واالعناصر الكيميائية

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اهتمت هذه الدراسة بتفسير أصل فجوات الكالسيت التى تتركز فى الجزء العلوى من صخور الدلوميت لمتكون الحيز (السينومانى العلوى) ولمتكون النقب (الوسط أيوسينى السفلى) فى الجزء الشمالى من الواحات البحرية (جبل التوبيج ومنطقة الحارة). وجد أن هذه الفجوات ذات شكل بيضاوى واشكال أخرى غير منتظمة. ومن الدراسات الميكروسكوبية للكالسيت الموجود داخل الفجوات تم تقسيم الكالسيت الى نوعين:- ١- كالسيت ذو بللورات صغيرة يبطن جدار الفجوات ٢- كالسيت ذو بللورات كبيرة يملأ الفجوات ناحية الداخل.

وبدراسة المحصورات المائية والنظائر الثابتة (الكربون-١٣ والأكسيجين-١٨) والعناصر الكيميائية تم التوصل الى أن النوع الأول من الكالسيت تكون من عملية اعادة الدلمتة لصخور الدلوميت المحيط بالفجوات عن طريق مياه ذات درجة حرارة من ٣٠-٤٠ درجة مئوية وملوحة من 3.43 - 0.8 NaCl eq. Wt%. أما بالنسبة للنوع الثانى من الكالسيت فقد تكون من عملية الترسيب المباشر بواسطة مياه الأمطار المتشبعة ببيكربونات الكالسيوم وذات درجة حرارة من ٢٤-٣٠ درجة مئوية.

وقد فسرت هذه الدراسة أن صخور الدلوميت لمتكون الحيز (السينومانى العلوى) ولمتكون النقب (الوسط أيوسينى السفلى) فى الواحات البحرية قد تعرضت للسطح فى نهاية ترسيبها وتأثرت بمياه الأمطار.