

Carbonate cement stratigraphy and timing of hydrocarbon migration: an example from Tigapapan Unit, offshore Sabah

MOHAMMAD YAMIN ALI
PETRONAS Petroleum Research Institute
Lot 1026, PKNS Industrial Area
54200 Hulu Kelang, Selangor

Abstract: The 'Tigapapan Unit' is one of several Miocene hydrocarbon-bearing reservoirs in the northwestern Sabah Basin. This unit consists of bioclast-rich, clastic-carbonate mixture, and has been interpreted as progradation storm shoal deposits.

Cathodoluminescence, geochemical and isotopic studies indicate that the sequence had undergone at least 9 stages of cementation which were responsible for complete occlusion of interstitial pores. Each stage represents one distinctive cement texture which was precipitated at specific temperature and burial condition. The diagenetic evolution was first introduced by (Ca1) a syndepositional, accicular and bladed iron-free marine calcite, followed by (Do1) early methane-derived dolomite resulting from degradation of biogenic methane during uplifting and induced faulting, (Ca2) bladed calcite, (Ca3/Ca4) blocky, vein-filling Fe-calcites, (Do2) clay-associated dolomite, (Do3/Do4) Fe-dolomite and ankerite, and (Ca5) late iron-rich calcite. Each cement stage is composed of distinctive Sr^{4+} , Mn^{2+} , Mg^{2+} , Fe^{2+} , Ca^{2+} , Si^{4+} , and Na^{+} concentrations, reflecting changes in palaeopore fluid systems with time. The cement stages are correlatable over the whole unit.

The cementation occurred from very early to late phases of diagenesis at near surface to 2.0 km depth. With progressive burial and temperature increased, oxygen isotope values become strongly negative (for example, stage Ca4: $\delta^{13}\text{C} = +2.70\text{‰ PDB}$, $\delta^{18}\text{O} = -5.70\text{‰ PDB}$). Oxygen isotope data indicate that the cements were precipitated at temperatures in the range of 20–75°C. Carbon isotope data suggest that the source of carbon varies from marine dissolved carbonates to fermentation of organic matter, biogenic methane, and possibly hydrothermal fluids.

Strontium isotope dating of Do1 (with $\delta^{13}\text{C} = -34.7\text{‰ PDB}$, $\delta^{18}\text{O} = +2.60\text{‰ PDB}$) and Do2 dolomites indicates that these two dolomitization events took place at 10.5 Ma and 8.9 Ma, respectively. The Do1 dolomite was precipitated from the methane-derived brine exactly at the boundary between the Serravallian and Fortonian stages after a major uplifting, induced faulting, and beginning of sealevel fall. The Do2 dolomite was precipitated at an intermediate to deep burial diagenesis during smectite-illite transformation.

INTRODUCTION

Porosity in sedimentary sequences has been destroyed mainly by concentric compositional zonings of carbonate cements. The compositional zoning has long been recognised from rocks in wide range of geologic ages and is now used as a basic

component of every diagenetic study. It has been used to establish cement stratigraphy at local and regional scales which allows the mapping of individual cement zone, constraints on precipitational environment and temperature, burial history, and timing diagenesis.

The 'cement stratigraphy' involves the application of stratigraphic principles at an intergranular and void level, correlating stages of cementation within a given basin of sedimentation (Miller, 1989). This concept was first introduced by Evamy (1969). However, the integrative cement stratigraphy was pioneered by Meyers (1974) in the Mississippian limestones of New Mexico, and followed by detailed regional studies (eg. Meyers and Lohmann, 1978). As the approach becomes increasingly popular in North America and Europe, more studies were carried out by other workers (eg. Goldstein, 1991). However, none of such studies has been conducted in other parts of the world possibly due to the lack of relevant facilities.

This paper contributes to the further understanding of carbonate cementation processes and porosity destruction in carbonate-cemented sequences by relatively date the cement and determine the relationship to palaeo-porewater system, stratigraphy, tectonic, and petroleum migration. The study was also to attempt to resolve the paragenetic and correlation problems at hydrocarbon field-scale.

GEOLOGIC SETTING AND SEQUENCE DEVELOPMENT

The 'Tigapapan unit' (Foo & McDonald, 1983) has been discovered in offshore Kudat (Fig. 1). The unit is composed of siliciclastic-carbonate mixture that shows time-equivalent to Balambangan Limestone (Fig. 2). The unit is underlain by a thick deeper marine shale of Kudat Formation equivalent.

Well and stratigraphic correlations (Fig. 3) suggest three main depositional subunits, being deposited in open-marine storm-dominated shelf (earlier, subunit 1) to restricted nearshore environment (later, subunit 3). During the deposition of subunit 1, a part of the structure was an erosional high as a result of uplifting and downslope movement of older sediments. Shoal facies developed over the palaeoridge whilst the localised topo-lows on the shelf were infilled by massflow and turbidite deposits. Off the elevated shelf area, deeper marine mudstones were deposited.

A short transgressive occurred towards the ends of the subunit 1 deposition. High area subsidence give rises to a shoal, separating restricted nearshore from shallow open marine shelf. A subsequent marine regression initiated the subunit 2 deposition in the upper shelf area and subunit 3 in the restricted nearshore environment. Progradation of the sequence seawards produced the observed vertical sequence. Shallow marine transgressive sands marked the ends of subunit 3 deposition.

Lithological Subunits

Subunit 1 is dominated by grey, very fine to fine grained calcareous sandstone with whiter, more calcareous sandstone infilling burrows and thin laminae. Carbonates occur as nodules and cements. The base contact is sharp with irregular pipes and sheets of dark non-calcareous clays. Intermittent sedimentation allows

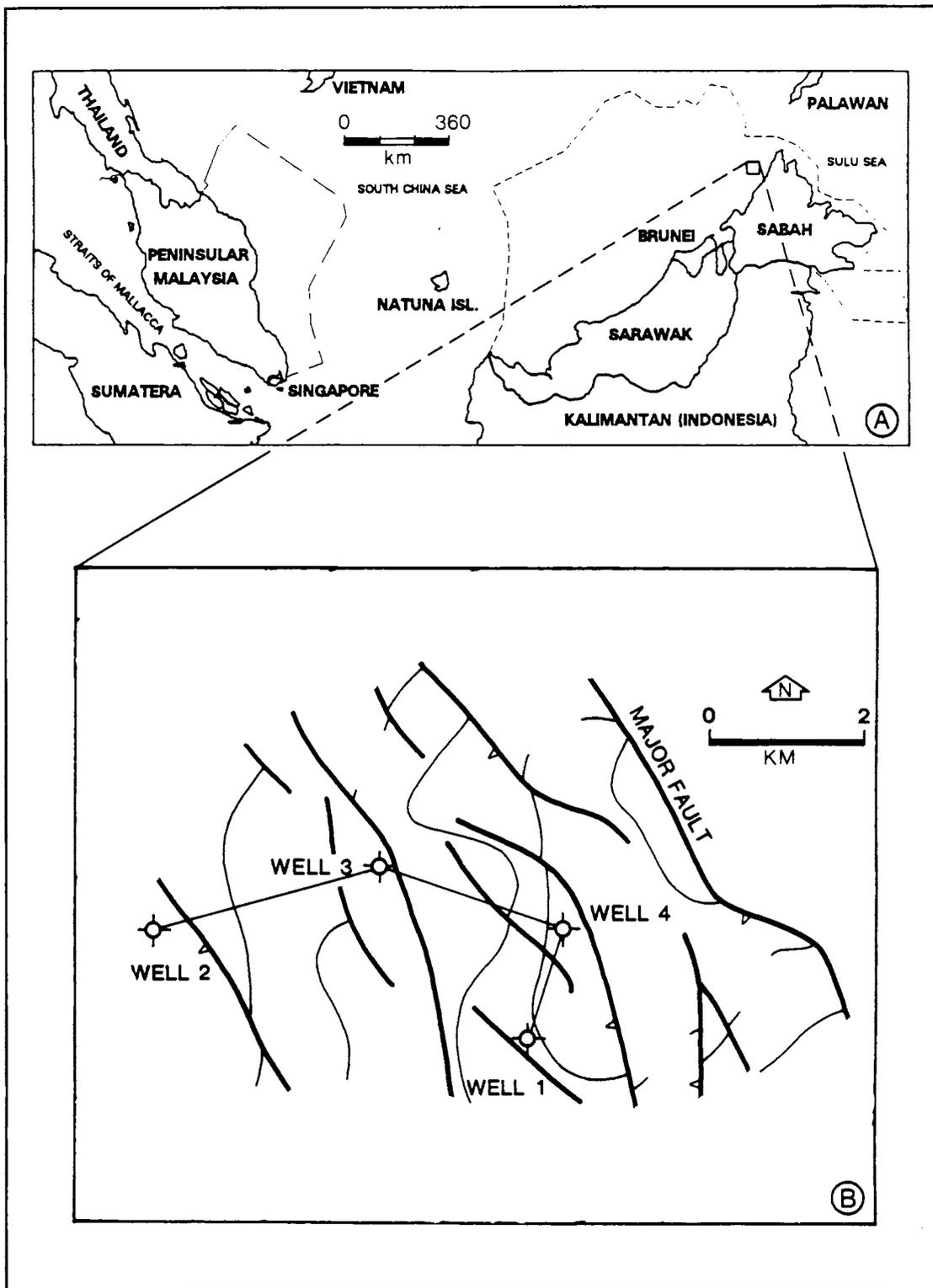


Figure 1: Location map of the study area (A) and the locality map of the wells (B).

		BANGGI ISLAND (after Wilson, 1961)	NORTH SABAH : TIME STRATIGRAPHY (after Bol & Van Hoom, 1980)	STRATIGRAPHIC SCHEME OFFSHORE (after Foo et.al, 1982)	BANGGI & KUDAT (after Tjia, 1988)				
RECENT		coastal deltaic & fluvialite alluvium Beaches & alluvium	LATE POST- GEOSYNCLINAL Stage IV		Alluvium terraces				
PLEISTOCENE						TIMOHING Fm	TIMOHING Fm	TIMOHING Fm epineritic	
PLIOCENE		BONGAYA Fm				Stage IV	BONGAYA Fm	BONGAYA Fm neritic BALAMBANGAN LST	
MIOCENE	UPPER	SOUTH BANGGI Fm				EARLY POST-GEOSYNCLINAL Stage III	KUDAT Fm	SOUTH BANGGI Fm	
	MIDDLE								BONGAYA Fm
	LOWER								
OLIGOCENE		CROCKER Fm				LATE GEOSYNCLINAL Stage II	CROCKER Fm	CROCKER Fm turbidite & mass flow	
EOCENE			EUGEOSYNCLINAL Stage I						
PALAEOCENE				CHERT-SPILITE Fm	CHERT-SPILITE Fm				
CREATACEOUS									

Figure 2: Regional stratigraphy of the study and adjacent areas.

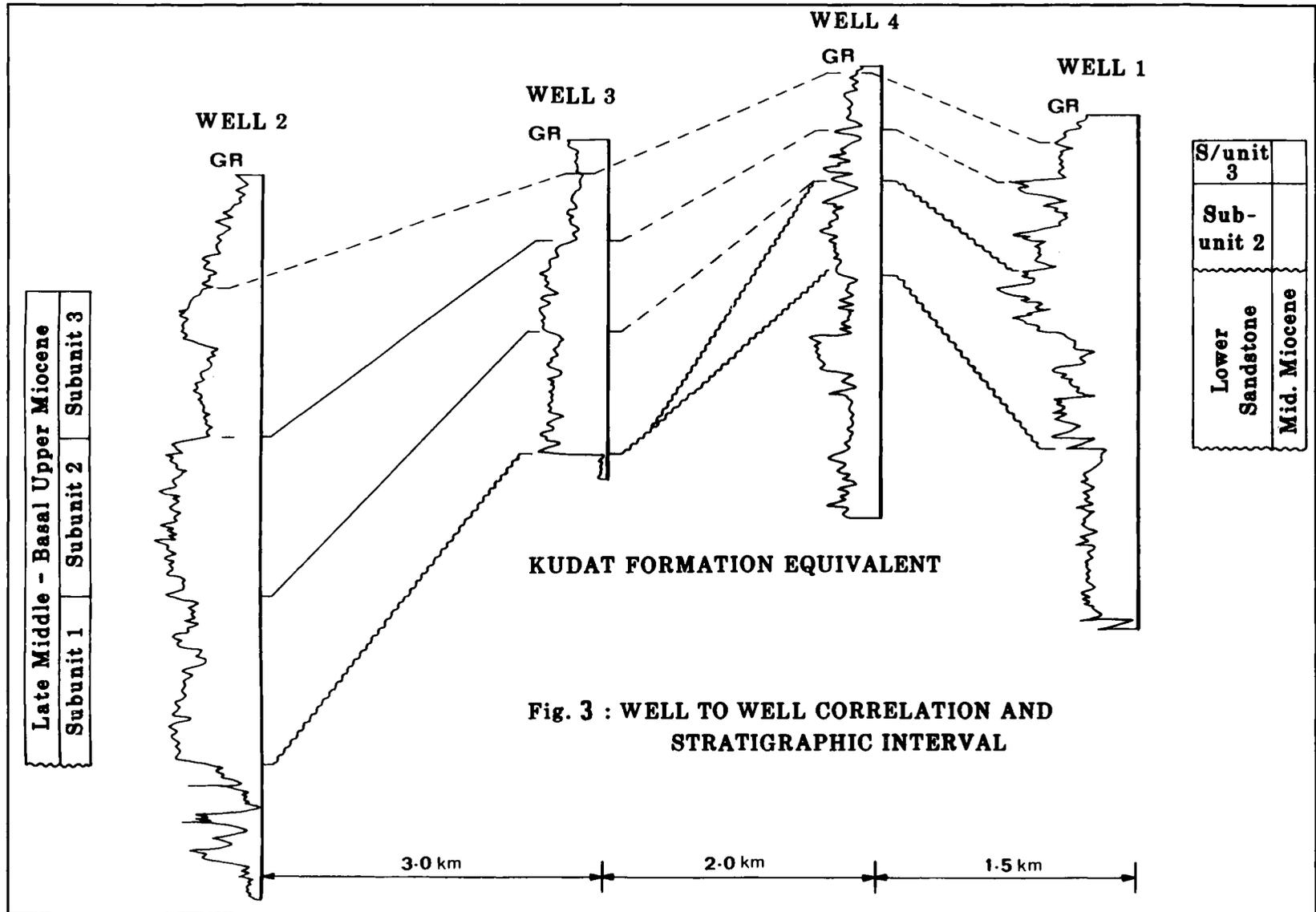


Figure 3: Well to well correlation and stratigraphic interval.

time for complete bioturbation, preserving no bedding. Sharp, scoured base and coarse-tail grading suggest turbidity currents were the main agent of deposition.

Subunit 2 displays marked alternations of tightly cemented of white, fine to medium grained sandy limestone/calcareous sandstone and porous dark grey, less calcareous sandstone. Granule-to-pebble intraclasts and nodular appearance are common. Sharp tops and bases, and coarse-tail grading are evident in places. Beds are highly bioturbated and bored by large burrows and show signs of diagenetic brecciation. Bedding planes are clear in places with some calcite-filled fractures and cracks are infilled by dark sandstone. Coarsen-upward sequence may indicate progradation, and the fine-upward sequence above it may represent traction current of a shallowing-upward sequence. Intraclasts may indicate the product of sediment dumping, while the small reverse-graded may represent true grain flow.

Subunit 3 represents brownish to grey muddy and sandy calcareous siltstone and silty calcareous sandstone. The bottom part is represented by calcareous siltstone and becoming featureless upwards. These features may indicate deposition in quiet water conditions.

METHODOLOGY AND CEMENT CORRELATION PROCEDURES

Laboratory Procedure

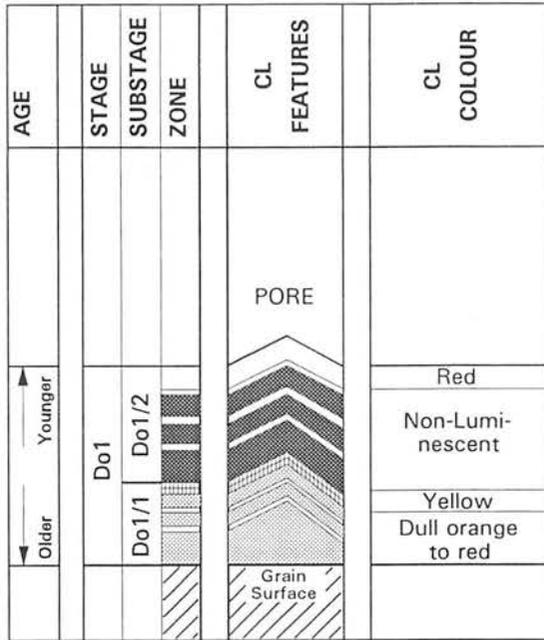
This study was based on examination of 250 thin sections with additional samples for special analyses. The samples were taken at specific depths through a 400 m section of core from two wells. Wherever core samples are not available, cutting samples were used. The study involves detailed analyses of samples using conventional petrography and cathodoluminescence microscopy (CL), scanning electron microscope (SEM), electron microprobe, and stable carbon and oxygen isotope analyses. Strontium isotope technique was used to date the dolomitization events.

Cements Correlation Procedure and Time Significant Event

The zonation pattern of cements at crystal and thin section levels were established using cathodoluminescence (CL) microscopy after textural morphology of cements is established. It was later confirmed by elemental composition and isotope measurements. Each zone of cement stratigraphic unit represents a successive change in chemistry of precipitating solution and a superpositional time-stratigraphic sequence (Goldstein, 1991). Synchronous compositions throughout its lateral and vertical extents would represent a time significant event that could be correlated across the ancient aquifer system at field and basinwide scales.

Three terms have been used to describe cement stratigraphic units for correlation purposes (Fig. 4). A *stage* is the primary unit of cement stratigraphy which consists of mineralogical homogeneity and distinctive geochemical features from adjacent cements. It corresponds to major textural differences and is correlatable over a large area.

A *substage* is a part of the stage which is characterised by distinctive compositional changes. It is a conformable cement unit, locally correlatable but



CARBONATE CEMENT STRATIGRAPHY

Definition :

- STAGE** - is the primary unit of cement stratigraphy which consists of mineralogical homogeneity and distinctive geochemical features from adjacent cements. It corresponds to major textural differences and is correlatable over a large area.
- SUB-STAGE** - is a part of the stage which has been characterised by distinctive compositional characteristics. It is a conformable cement unit, locally correlatable but often discontinuous on a basin wide scale.
- ZONE** - is the smallest individual unit in cement stratigraphy, correlatable within a small area such as a cavity or crystal. It is conformable and essentially homogeneous.

Diagenetic Environment	Description	Characteristics
1. Oxic zone	The zone within sediment/water interaction and disappearance of dissolved CO ₂ (Czerniakovski et. al., 1984)	Mn ²⁺ ≈ 0, Fe ²⁺ ≈ 0 (<0.01% wt)
2. Suboxic zone	The zone within disappearance of dissolved CO ₂ and the onset of sulphate reduction. (Froelich et. al., 1979)	Mn ²⁺ appear, Fe ²⁺ rare
3. Sulphate Reduction zone	Zone of bacterial conversion of sulphate to sulphide. (Coleman et. al., 1979)	Pyrite can be formed
4. Fermentation zone	The zone of disappearance of dissolved sulphate. (Curtis, 1978)	Fe-rich calcite would precipitate

Figure 4: Simplified features of cement stratigraphic unit and characteristics of diagenetic environments.

often discontinuous on a basinwide scale. A *zone* is the smallest individual unit of cement stratigraphy. It is conformable, essentially homogeneous and is correlatable within a small area, such as cavity.

PETROGRAPHY, CEMENT MORPHOLOGY AND DIAGENESIS

The sequence ranges from calcareous or dolomitic sandstones and siltstones, to silty and sandy limestones and dolomites. They composed of a mixture between sand grains (quartz, chert, K-feldspars, plagioclase, rock fragments), bioclast skeletal (benthonic and planktonic foraminifers, bryozoa, red algae, larger forams, echinoderms), and clay matrix. The percentages of these framework components vary from sample to sample. The sand grains represent 15–70% of the total rock composition whereas the bioclasts and clay matrix represent 3–55% and 1–5%, respectively. Secondary minerals like calcite, dolomite, and chloride are common.

The earliest phase of diagenesis involves lithification and micritization of internal sediments. Selective dissolution of mineralogical metastable bioclasts have been identified prior to the development of the first recognisable cements in the sequence. The dissolution style of skeletal grains was dependent upon the original fossil structure and mineralogy. Echinoderm (originally high-Mg calcite (HMC)) and those originally low-Mg calcite (LMC) often show evidence of partial dissolution in the form of irregular and embayed surfaces. Meanwhile, red algae (originally aragonite or a mixture of aragonite and calcite) may be represented by fabric-selective microdolomites and spar-filled moulds.

Cementation may have been influenced by an early bioturbation and boring which form hardgrounds as sites for precipitation, and microorganisms and the physico-chemical conditions of the pore fluids. Some of the carbonate dissolutions may have been the source of some of the cements. The relative timing of the early lithification and dissolution can be ascertained by the reconstruction of primary fabrics, the sequential evolution of the pore space and its contained cement stages.

The major textural hiatus which can be recognised in the cement sequence is marked by a change in crystal habits. The most common early cements are centripetal rims consisting of a mosaic of syntaxial, acicular, scalenohedral and bladed calcites. Crystals contain closely spaced inclusions which may be divergent near the substrate. Bladed crystals are length-fast with sweeping undulose extinction with crystal dimension are in order of 10–100 μ . They are followed by late blocky, pore- and vein-filling texture. The cement fabric lack consistent asymmetry or other vadose features except in well 1. Meniscus fabric and pendant fabrics are either absent or poorly developed. These occurrences indicate less vadose-zone cementation. Therefore, marine and phreatic condition of cementation were probably the most dominant.

Three types of dolomite have been identified. Type 1 dolomite is restricted within the Lower Sandstone Unit only where the entire rocks have been dolomitised. The dolomite exhibits anhedral to euhedral crystal morphology. The most likely source could be from methane-derived brines, and precipitated at shallow burial depth. The type 2 dolomite displays a sucrosic texture and is formed in association

with clay matrix, and is believed to precipitate during smectite-illite transformation during late burial diagenesis. The type 3 dolomite exhibits sucrosic texture with unclear crystal boundaries. It is iron-rich dolomite, often scatteredly distributed within organic-rich layers, possibly precipitated during late phase of diagenesis at deeper burial depth.

Majority of the pores in the rocks have been occluded by complex early and late phases of silica and carbonate cementations especially within bioclast-rich facies where bioturbations and borings are prominent. The most common remaining pore types are interparticle, intraparticle, and some fracture pores.

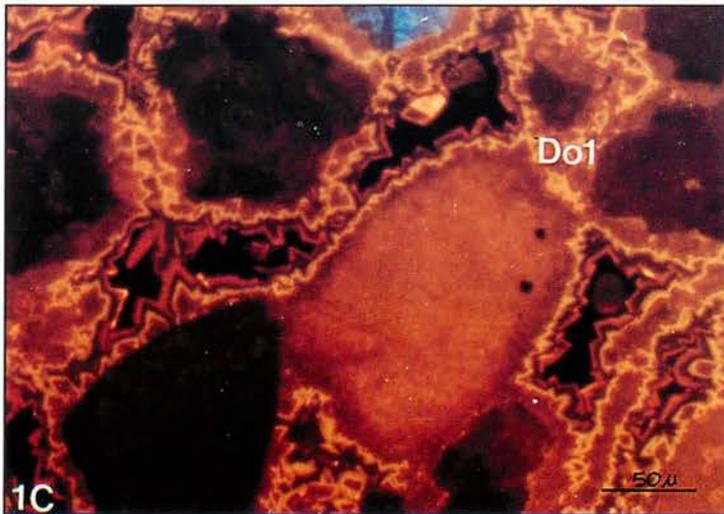
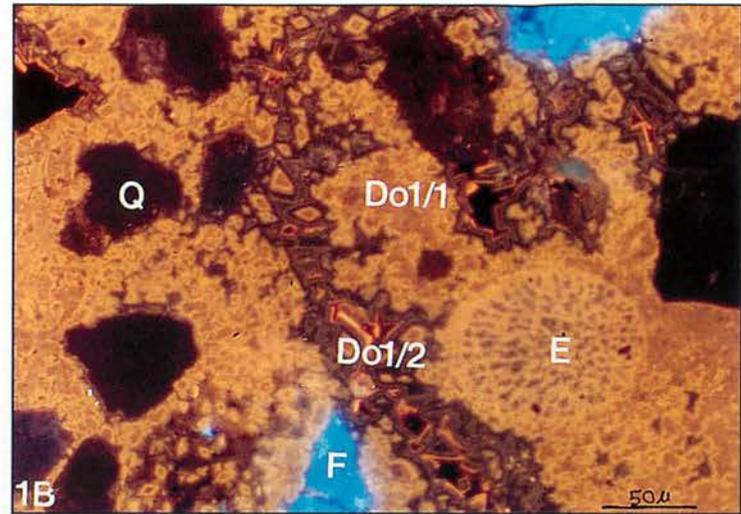
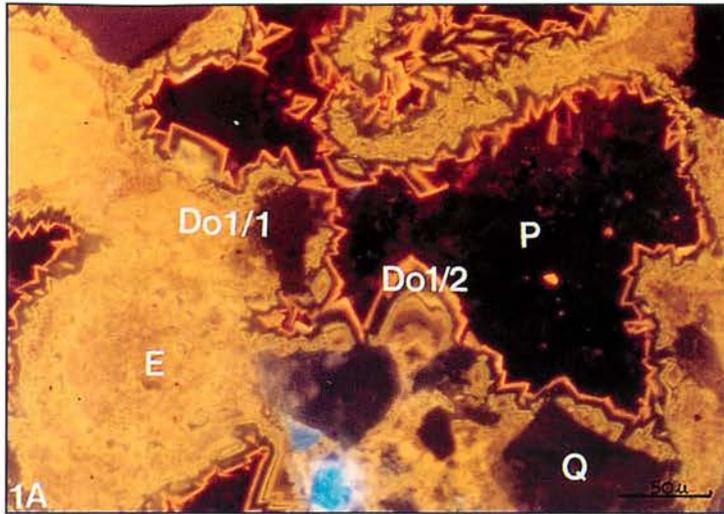
Cathodoluminescent Zonings and Elemental Compositions

Cathodoluminescent zoning of the carbonate cements is spectacular and complex, as it is determined by crystal orientation, elemental composition, and redox potential (Eh). At least nine stages of carbonate cementation have been identified. The first three stages (Do1, Ca1 and Ca2) are early, non-ferroan, and display complex zonation patterns; the remaining stages (Ca3–Ca5, and Do2–Do4) are late, iron-rich, and have broader zonation patterns. For the early stage cements, poorly luminescent zones are followed by brightly luminescent. The late stage cements, however, exhibit a single non-luminescent or bright luminescent colours. The characteristics of the cement stages are described as follows :

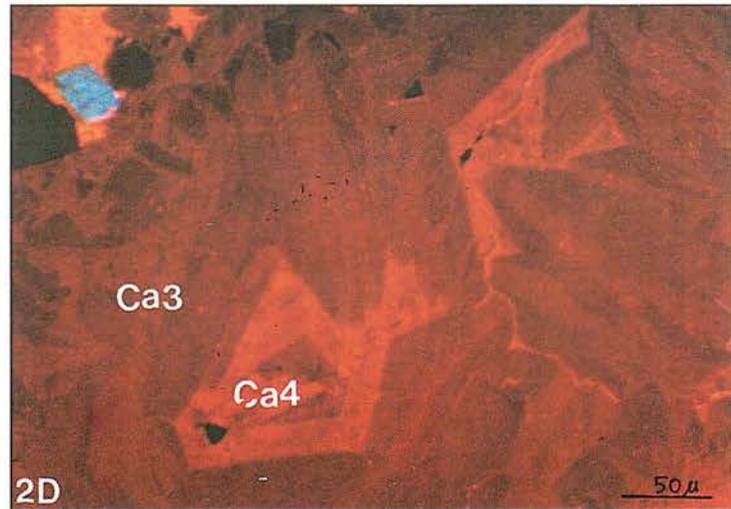
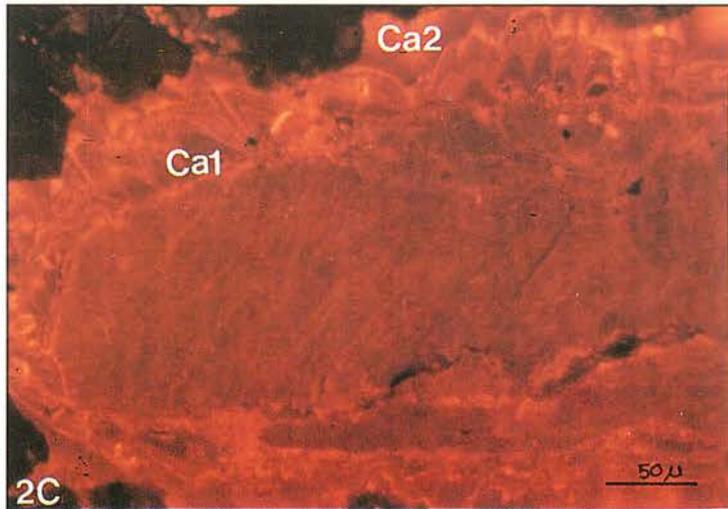
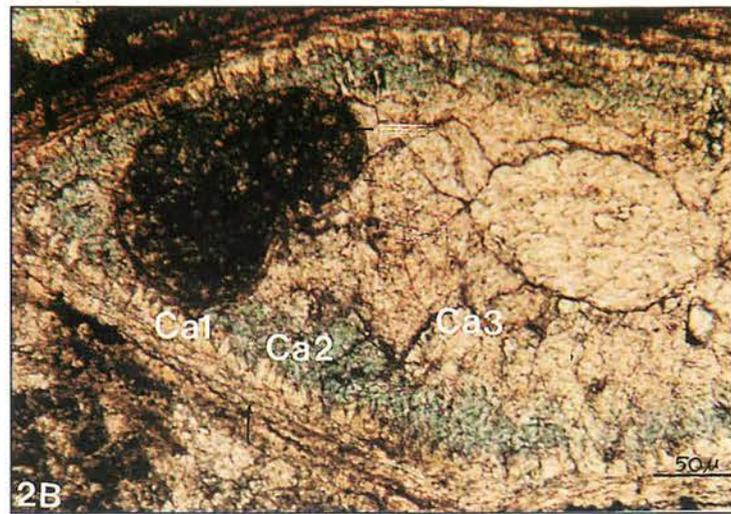
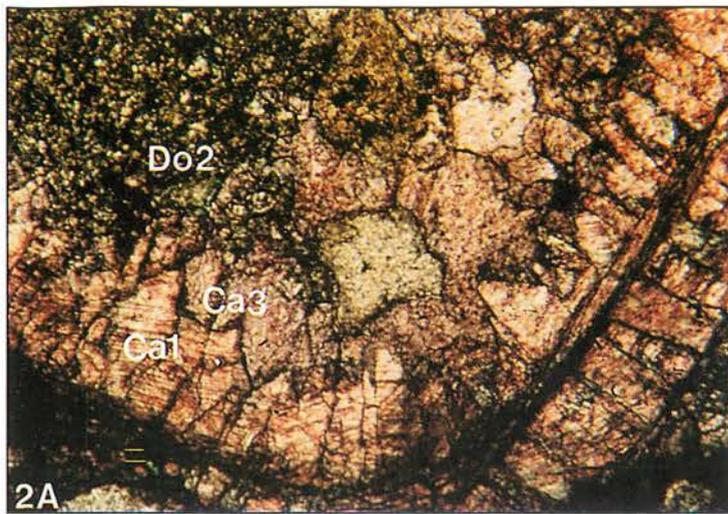
Stage Do1 (Plates 1A–1D) is an early phase of iron-free dolomite being precipitated as a result of biogenic methane degradation. The Do1 cement exhibits small rhombohedral, sucrosic texture to a continuous anhedral to subhedral crystals habits and found to be as continuous pore-filling and replacement cements. The Do1 stage cement consists of two substages (Do1/1 and Do1/2), each of them exhibit two major zones (Do1/11, Do1/12, and Do1/21, Do1/22). The major zones exhibit dull red to orange, yellow, non-luminescent (black), and pink, respectively. There is no significant different between these zones as they show low Fe²⁺ and Mn²⁺ contents (<0.01wt%).

Stage Ca1 (Plates 2A–2B,3A–3B) is an early phase, non-ferroan calcite cement, precipitated on top of fossil substrates. It represents syntaxial, isopachous, bladed, or scalenohedral cement morphology. It consists of two substages and four zones, which exhibit dull to non-luminescent, dull orange, non-luminescent, and bright orange, respectively. Each zone is iron-free (Fe²⁺ and Mn²⁺ < 0.01 wt%) but there is a significant increase in Sr⁴⁺ (0.50wt%) concentration in this stage that might be contributed by the original Sr⁴⁺ in skeletal composition.

Stage Ca2 (Plate 2A–2C,3A–3B) is an early phase of calcite with moderate amount of iron. This stage represents an accicular to isopachous and bladed texture, and comprises of two substages (Ca2/1 & Ca2/2) which are differentiated by dull orange and bright orange luminescent colours. They are differentiated from an earlier cement by slightly moderate Mn²⁺ (0.05wt%) and Fe²⁺ (0.14wt%) contents. There is an increase in Mg²⁺ (1.60wt%) and decrease in Sr⁴⁺ content compared to the Ca1 cement.



Plates 1A to 1D: Cathodoluminescent (CL) zonings of early stage Do1 methane-derived dolomite. Compositional zonings of dolomite cements are well developed within the interstitial pore space. An early substage Do1 cement is subsequently followed by a later substage Do1/2. Note : Q - quartz, F - feldspar, E - echinoderm, P - pore. CL view.



- Plate 2A:** Development of bladed stage Ca1 calcite cements (stained-pink) from the skeletal substrate and blocky stage Ca3 calcite cement (stained light purple). Some clay-associated dolomite (Do2) are also present. Normal plane view.
- Plate 2B:** Development of accicular, bladed calcite cements (Ca1, Ca2) and blocky texture of Ca3 calcite cement. Stage Ca1 and Ca2 calcites can be differentiated from their staining colours. Note that dolomite has completely replaced a planktonic foram. Normal plane view.
- Plate 2C:** CL characteristics of stage Ca1 calcite cement. CL view.
- Plate 2D:** CL characteristics of stage Ca3 and Ca4 vein-filling calcite cements. CL view.

Stage Ca3 (Plates 2A–2D, 3A–3B) can be considered as an intermediate to late phase of Fe-calcite cement. It represents blocky, equant, and pore-and vein-filling texture. It consists of two substages (Ca3/1 & Ca3/2) which show dull orange to non-luminescent features. The Ca3 cements display thick zone, and are differentiated from the earlier cements by significant increases in Fe^{2+} (0.50wt%) and Sr^{4+} (1.00wt%) contents and decrease in Mg^{2+} , with a constant Mn^{2+} content.

Stage Ca4 (Plates 2C, 3A–3D) is a late stage Fe-calcite. It represents blocky to equant, vein-filling calcites which display a thick zoning. The Ca4 calcite cement is characterised by a dull to bright orange luminescent colours. The cement has the same composition as Ca3 but with a significant increase in Mn^{2+} (0.56wt%) concentration.

Stage Do2 (Plates 3C–3D) is a burial Fe-dolomite, found in association with the clay matrix. It exhibits small rhombohedral, sucrosic texture. The early phase of this dolomite has shown non-luminescent feature, while the other substage has exhibited bright orange to yellow. They are calcian dolomites, with high Fe^{2+} and moderate Mn^{2+} contents. Stage Do2 dolomite is composed of significant Sr^{4+} and Si^{4+} that might be contributed by the clay minerals.

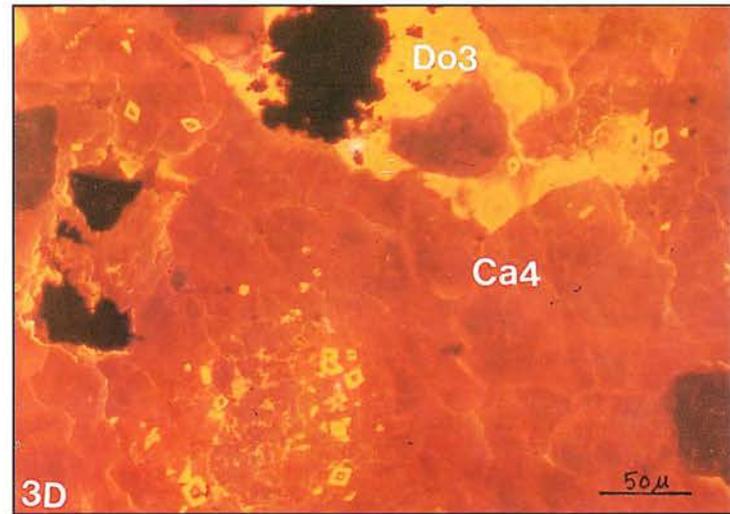
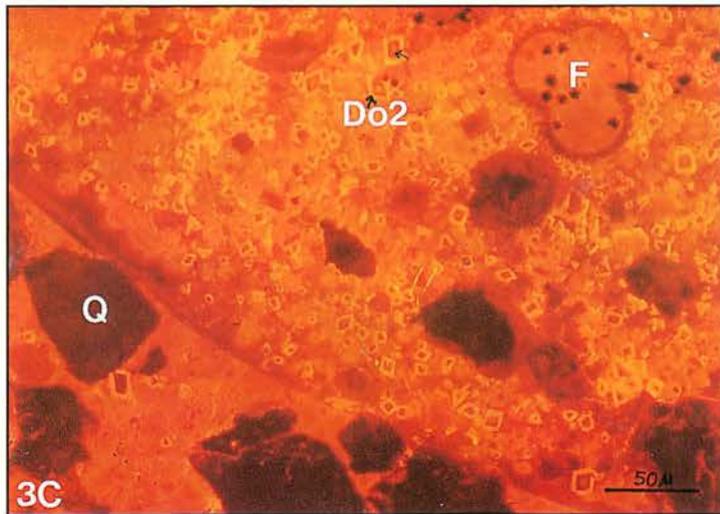
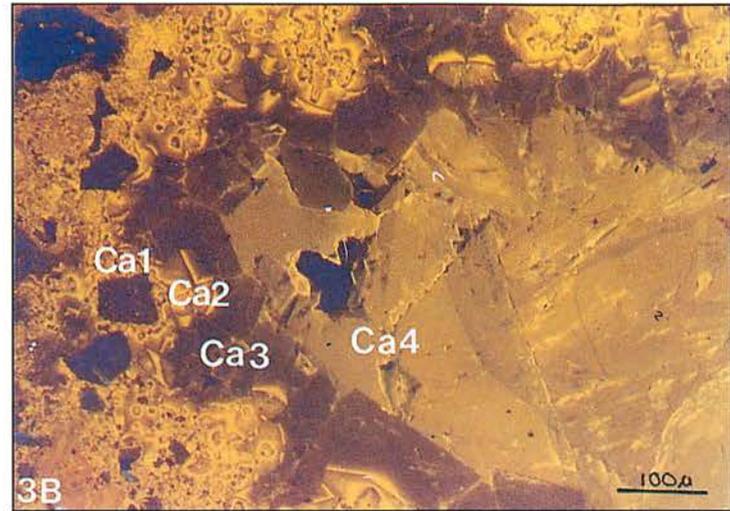
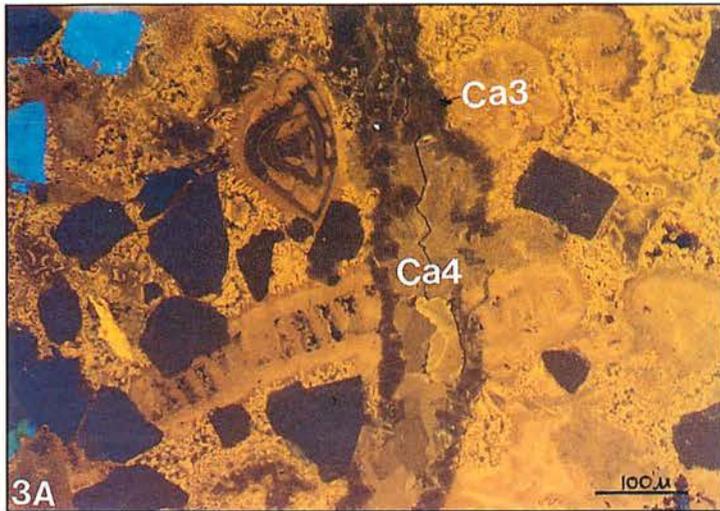
Stages Do3 (Plate 3D) represents a late phase, Fe-dolomite. It exhibits a rhombohedral sucrosic texture with unclear crystal boundaries. Stage Do3/1 exhibits finer crystals (10–30 μ) than the stage Do3/2 (30–60 μ) dolomites. They exhibit dull orange and non-luminescent, respectively. Substages Do3/1 and Do3/2 have similar composition, i.e. Mn-free ($\text{Mn}^{2+} < 0.01\text{wt}\%$). However, substage Do3/1 substage is composed of high Fe^{2+} concentration (0.57wt%).

Stage Do4 (Plates 4A–4B) represents a latest phase of iron-rich dolomite and ankerite, formed as sucrosic to subhedral with unclear crystal boundaries. They exhibit dull orange to yellow luminescent colours. Both are differentiated by different in Mg^{2+} , Fe^{2+} , Sr^{4+} , and Mn^{2+} compositions. Do4/1 dolomite is characterised by significant increase in Mn^{2+} (0.51wt%) and Fe^{2+} (3.42wt%) with consistent Na+ and Sr^{4+} contents. Do4/2 substage is characterised by extremely high Fe^{2+} concentration (10.74wt%), high Sr^{4+} (0.34wt%), and low in Mn^{2+} (0.26wt%) concentrations.

Stage Ca5 (Plate 4B) is a very late phase, interparticle pore-filling iron-rich calcite. The cement exhibits dull orange as a result of high Mn^{2+} , Sr^{4+} , and Mg^{2+} , and very high Fe^{2+} concentration. It represents a very small percentage of cements.

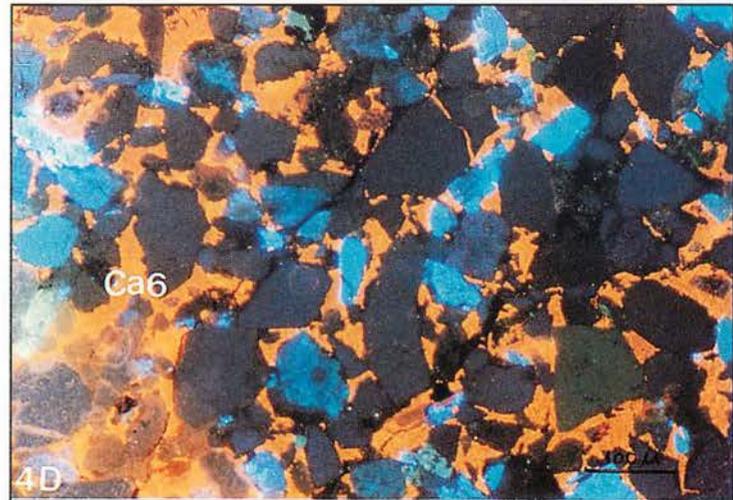
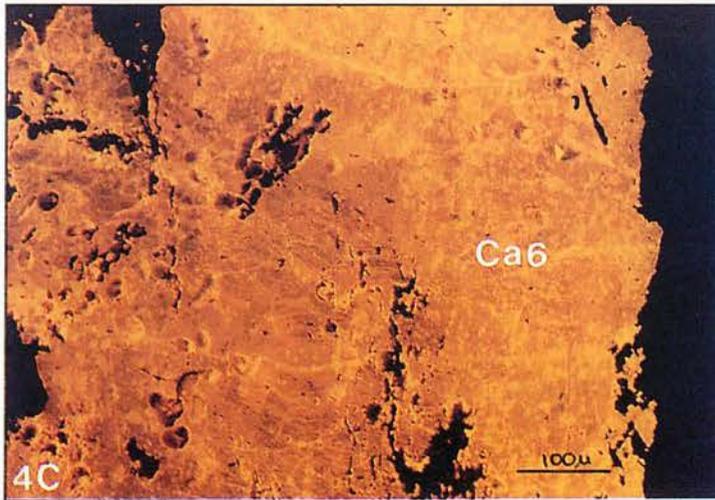
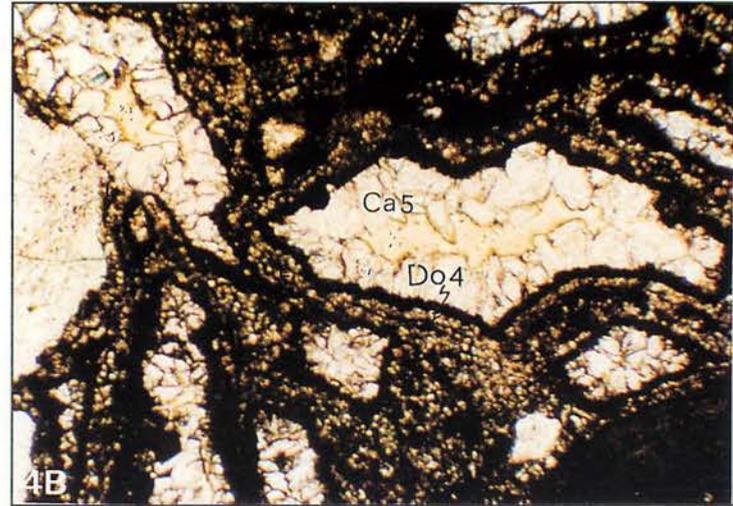
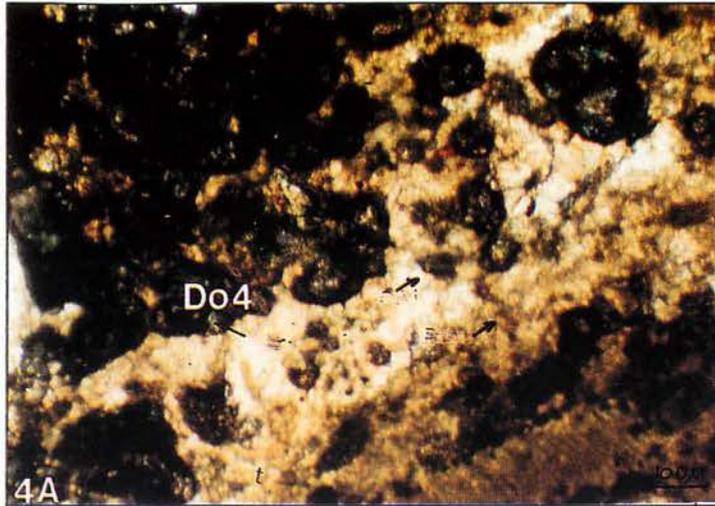
Unknown stage (Ca6) (Plates 4C–4D) is represented by two different calcites:

(1) Radial, inclusion-rich calcite with very bright orange luminescent feature. The amount of Mg^{2+} and Fe^{2+} is negligible (0.01 wt% oxides) but the Mn^{2+} concentration is extremely high (0.7 to 3.47 wt% MnO). This cement is restricted within the interval below Lower Sandstone unit which unconformably overlies the Kudat Formation.



Plates 3A&3B: Cross-cutting relationship showing the development of various stages of calcite cements (Ca1 to Ca4) that almost completely obliterate the pore spaces. Note : Q-Quartz, F- Feldspar, F-Fossil.. CL view.

Plates 3C&3D: CL characteristics of clay-associated dolomite (Do2) and later Do3 dolomite. The dolomite zones are rather simple. CL view.



Plates 4A&4B: The crystal habit of high-Mg dolomite or ankerite (Do4) and very late phase of iron-rich calcite cement (Ca5). Plane polarised view.
Plates 4C&4D: CL characteristics of unidentified stage, radiaxial (plate 4C) and blocky (plate 4D) calcite cements. CL views.

(2) Blocky, equant calcite with bright orange luminescent feature. The occurrence of this cement is restricted within subunits 2 and 3 of well no. 1.

Both calcites display similar carbon and oxygen isotope compositions.

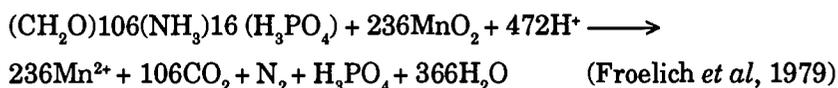
The variations in crystal morphology, cathodoluminescence characteristics, and elemental composition of each cement stage are given in Tables 1 and 2.

Palaeo-porewater System and Diagenetic Fluids

Levels of Mg in stage Ca1 cement are similar to those in echinoderms and foraminifera which were originally composed of HMC. This carbonate stage was precipitated from normal marine to hypersaline solutions. The non-luminescent feature and low levels of Fe²⁺ and Mn²⁺ (<0.01 wt%) can be attributed to precipitation from oxidised water (Curtis, 1976). Identical compositions in various substages in stage Ca1 cement suggests that the pore-fluids composition had remained essentially unchanged with respect to redox potential. Slightly higher Fe²⁺ and Mn²⁺ concentrations in stage Ca2, may suggest precipitation from oxic to suboxic pore fluids. The existence of dissolution surfaces in between these two stages further support this interpretation.

In comparison with stage Ca2, there is a significant decrease in Mg²⁺ and increase in Fe²⁺ concentrations in stage Ca3 although the Mn²⁺ content remains the same. The decrease in Mg content is probably related to depletion of Mg²⁺ in the chemically isolated pore-fluids, and substitution of Mn²⁺ into lattice sites which might otherwise have been filled by Mg²⁺. Considering that some of the inclusions in these cements have had relatively higher levels of Mg, the decrease in inclusion density which is often seen in the latter stages of calcites may be responsible for the downward trend in Mg²⁺ in the calcite lattice. Although the cement morphology is equant, it is known to be a marine cement as indicated by isotope signature.

The subsequent stage Ca4 calcite cements show consistent Mg²⁺ and Fe²⁺ concentrations with a significant increase in Mn²⁺ content. A marked increase in Mn²⁺ in Ca4 therefore indicates the onset of reducing conditions. Similar as in recent sediments, the oxidation of organic matter reactions involves :



where MnO₂ acts an electron acceptor, cause a rapid increased in dissolved Mn²⁺ at the onset of reducing conditions (Shimmield & Price, 1986). The level of dissolved Mn²⁺ in the pore fluids rapidly reaches a maximum, which is maintained unless Mn²⁺ is used in other reactions such as those which result in the precipitation of carbonate (Pedersen & Price, 1982), with Mn²⁺ constituting into Ca²⁺ and Mg²⁺ sites. These would gradually deplete the dissolved manganese in the pore fluids as reflected by the initially high Mn²⁺ levels.

The unknown stage calcite (Ca6) is composed of negligible amount of Mg²⁺ and Fe²⁺ (0.01 wt%) but extremely high Mn²⁺ (0.67 to 3.47 wt% MnO) concentrations. The high Mg²⁺ content was probably contributed by hydrothermal fluids from deep

Table 1: Textural relationship and cathodoluminescence characteristics of various cements.

CEMENT STAGE	TEXTURE OF CARBONATE CEMENTS	CL CHARACTERISTICS
Ca5	Pore-filling cement	Dull orange
Do4	Cement	Orange to yellowish
	Cement	Orange to yellowish
Do3	Fine rhombs	Dull orange / yellowish
	Coarse rhombs, overgrowth dolomite	Dull orange
Do2	Clay-associated rhombs	Bright orange to yellow
	Clay-associated rhombs	Non-luminescent
Ca4	Vein-filling	Orange
Ca3	Blocky, pore - and vein-filling	Non-luminescent
	Blocky, equant, pore-filling	Dull orange to non-luminescent
Ca2	Bladed, scalenohedral	Bright orange
	Bladed, scalenohedral	Dull orange
Ca1	Bladed, scalenohedral, isopachous	Bright orange
	Bladed, scalenohedral, isopachous	Non-luminescent
	Bladed, scalenohedral, isopachous	Dull orange
	Bladed, scalenohedral, isopachous	Dull to non-luminescent
Do1	Rhombs, cement	Red
	Rhombs, cement	Non-luminescent
	Rhombs, cement	Yellow
	Rhombs, cement	Dull orange / red
Unknown	Radial, inclusion-rich	Bright orange
Unknown	Radial, inclusion-rich	Bright orange

Table 2: Elemental composition (wt % oxide) of various cements.

Relative Timing	CEMENT			WEIGHT % CONCENTRATION (OXIDE)						
	STAGE	SUBSTAGE	ZONE	Mg ²⁺	Na ⁺	Sr ⁴⁺	Si ⁴⁺	Ca ²⁺	Fe ²⁺	Mn ²⁺
LATE ↑	Ca5			1.08	0.03	0.23	0.02	52.53	1.70	0.38
	Do4	Do4/2		12.13	0.04	0.34	0.03	32.44	10.74	0.26
		Do4/1		18.44	0.07	0.08	2.10	31.36	3.42	0.51
	Do3	Do3/2		19.62	0.05	0.06	0.02	36.14	0.08	0.00
		Do3/1		16.94	0.04	0.09	0.02	37.16	0.57	0.01
	Do2	Do2/2		18.06	0.07	0.34	0.74	35.47	1.22	0.07
		Do2/1		19.05	0.07	0.09	1.46	34.99	0.24	0.07
	Ca4			0.62	0.00	0.76	0.00	53.49	0.53	0.56
	Ca3			0.61	0.00	0.79	0.00	54.05	0.50	0.03
				0.38	0.00	1.01	0.07	54.34	0.48	0.09
	Ca2	Ca2/2		1.57	0.01	0.10	0.01	54.08	0.14	0.07
		Ca2/1		1.65	0.01	0.13	0.02	53.98	0.14	0.04
	Ca1	Ca1/2	Ca1/22	1.19	0.02	0.09	0.04	54.58	0.05	0.02
			Ca1/21	0.64	0.01	0.57	0.01	54.75	0.00	0.00
Ca1/1		Ca1/12	1.33	0.01	0.15	0.01	54.38	0.01	0.01	
		Ca1/11	1.19	0.04	0.54	0.00	54.19	0.01	0.01	
Do1	Do1/2	Do1/22	20.49	0.06	0.08	0.03	35.20	0.05	0.07	
		Do1/21	19.77	0.02	0.07	0.03	35.30	0.00	0.01	
	Do1/1	Do1/12	19.14	0.04	0.08	0.03	36.64	0.01	0.01	
		Do1/11	17.51	0.05	0.07	0.11	38.21	0.01	0.01	
EARLY ↓	Unknown			0.01	0.03	0.04	0.02	55.20	0.02	0.67
	Unknown			0.13	0.03	0.16	0.03	51.94	0.27	3.47

burial source within the sedimentary sequence or high Mn^{2+} fluid adjacent to fault (Gawthorpe, 1987). These are supported by the evidence that the occurrence of this cement is restricted within the zones below Lower Sandstone unit which unconformably overlies the older Kudat Formation rocks.

The characteristic 'black-bright-dull' CL sequence of stage Ca1, Ca2 and Ca3/Ca4 cements are interpreted as representing the passage of porewater from oxic through suboxic to sulphate reduction zones, which occurred within the marine porewaters as a result of bacterial degradation of organic matter during progressive shallow burial. The non-luminescent zone is precipitated from oxidizing waters which do not contain Mn^{2+} . The bright zone formed from reducing waters which do contain Mn^{2+} but not Fe^{2+} , or the latter is being removed as sulphide in the bacterial reduction of sulphate. The dull zone is a ferroan (manganous) calcite precipitated from more negative redox potential (Eh) waters, below the stability field of FeS_2 . This zone is precipitated during deeper burial commonly after a phase of fracturing.

Dolomitizing fluids

Petrographic and elemental/isotope analyses reveal three major types of dolomites; methane-derived dolomite (Do1), clay-associated dolomite (Do2), and iron-rich dolomites (Do3 and Do4). However, none of them has achieved its stoichiometric composition ($Ca_{0.5}Mg_{0.5}CO_3$). The departure of major elemental composition is expressed by a ratio in mole % carbonate. A strong antipathic relationship occurs between iron and magnesium, indicating substituting of iron for magnesium. All types of dolomite here are calcian. Early non-ferroan dolomite (Do1) stoichiometry varies from 59.1–64.7% $CaCO_3$, while Fe-dolomite (Do2 and Do3) stoichiometry varies from 63.5 $CaCO_3$ and 53.5 to 70.0% $CaCO_3$, respectively. The Fe-dolomite analysed shows greater stoichiometric variation than non-ferroan dolomite. Early shallow burial dolomites are less stoichiometric than deeper burial dolomites.

All substages in stage Do1 dolomite show a constant low level of Fe^{2+} and Mn^{2+} concentrations (<0.01%wt), indicating precipitation from oxidised waters. Mean carbon and oxygen isotopic compositions of -33.0 ‰ PDB and $+2.0$ ‰ PDB, respectively suggest that the carbon for dolomitizing fluids most likely being contributed by methane-derived brine and precipitation was taken place at very low temperature (about 15°C).

In the case of clay-associated dolomite (Do2), a probable source for Mg in deep subsurface is from transformation of smectite to illite resulting from compaction and shale dewatering. Solution seams would release Mg ions, as all dolomites (other than Do1) are post-dated solution seams. The most important source for Ca is the pressure dissolution of primary carbonate skeletons. Therefore, the production of dissolved calcium would have been greater in well compacted facies.

Iron oxides and hydroxides which may be adsorbed onto clay particles can be reduced to ferrous ion during the sulphate reduction process through direct or bacterial oxidation of organic matter. The ferroan iron released into pore fluids during sulphate reduction would be rapidly fixed as iron sulphide and therefore the

activity of iron remain low. The onset of bacterial fermentation is marked by the disappearance of SO_4^{2-} ions from the pore fluids, and therefore the end of pyrite formation. The dolomite texture and the occurrence of Fe-dolomite in organic-rich sediments supports the involvement of clay minerals and organic matter in iron reduction and dolomite diagenesis.

In Comparison with Do2 and Do3, there is a remarkable increase in Fe^{2+} concentration in Do4 dolomite. Extremely variable iron concentrations in stage Do4 with the maximum levels of 17.1% FeCO_3 being essentially an ankerite composition may indicate significant variation in the degree of bacterial fermentation and compaction during this time.

Carbon Isotope Signatures

The $\delta^{13}\text{C}$ values of different stages of calcite cements (Fig. 5) fall within the range of +1 or -1 ‰ PDB (PeeDee Belemnite) except for unknown stage. These values indicate that Ca1, Ca2, Ca3, and Ca4 calcite cements were precipitated in normal marine water conditions as freshwater cements are depleted in $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values compared to marine cements. However, the unknown stage cement (Ca6) shows $\delta^{13}\text{C}$ values of -8 to -12 ‰, suggesting precipitation through anoxic process within sulphate reduction environment.

Dolomite stages show differences in $\delta^{13}\text{C}$ values. Do1 dolomite has a very light carbon values -32 to -38 ‰ with positive oxygen isotope values (Figs. 5 & 6). The only source known for the very light carbon is from methane. The methane which has $\delta^{13}\text{C}$ values ranging from -60 to -90 ‰ PDB is generally biogenic origin, having been produced by specialised bacteria. The methane generated thermally during natural gas formation typically has $\delta^{13}\text{C}$ values ranging from -30 to -50 ‰ PDB (Sackett, 1978). Such enrich $\delta^{13}\text{C}$ values can also be produced via microbial oxidation of methane, as 12CH_4 is preferentially consumed. A high concentration of methane relative to other light hydrocarbon indicates a bacterial or biogenic source rather than a thermogenic source. The source of biogenic methane is probably seeping brines, and appears to be precipitated at low temperature.

Stages Do2, Do3, and Do4 dolomites have positive $\delta^{13}\text{C}$ values. The Do2 dolomite has the $\delta^{13}\text{C}$ values of +1 to +8 ‰, while the Do3 and Do4 dolomites have $\delta^{13}\text{C}$ values of +2.5 to +5.0 ‰ and +1.5 to 3.0 ‰ PDB, respectively. The $\delta^{13}\text{C}$ values of these dolomites are generally consistent (+1 to +8 ‰ PDB) but formed at different burial temperatures as shown by the significant depletion of $\delta^{13}\text{C}$ values (Table 3). Positive $\delta^{13}\text{C}$ values favours bacterial fermentation of organic matter as a source of heavy carbon (Irwin, 1980). Variable $\delta^{18}\text{O}$ signatures with more or less constant $\delta^{13}\text{C}$ values may suggest that the precipitation of different dolomites is a function of temperature rather than the source of dolomitizing fluids (Fig. 6).

Oxygen Isotopic Compositions and Palaeotemperatures

Assuming an original marine pore fluids, the depletion in $\delta^{18}\text{O}$ values from stage Ca1 to Ca4 calcites and Do1 to Do4 dolomites suggests some degrees of progress burial with increased precipitation temperatures. To relate $\delta^{18}\text{O}$ signatures to burial depth, it is necessary to know the isotopic composition of the ambient pore

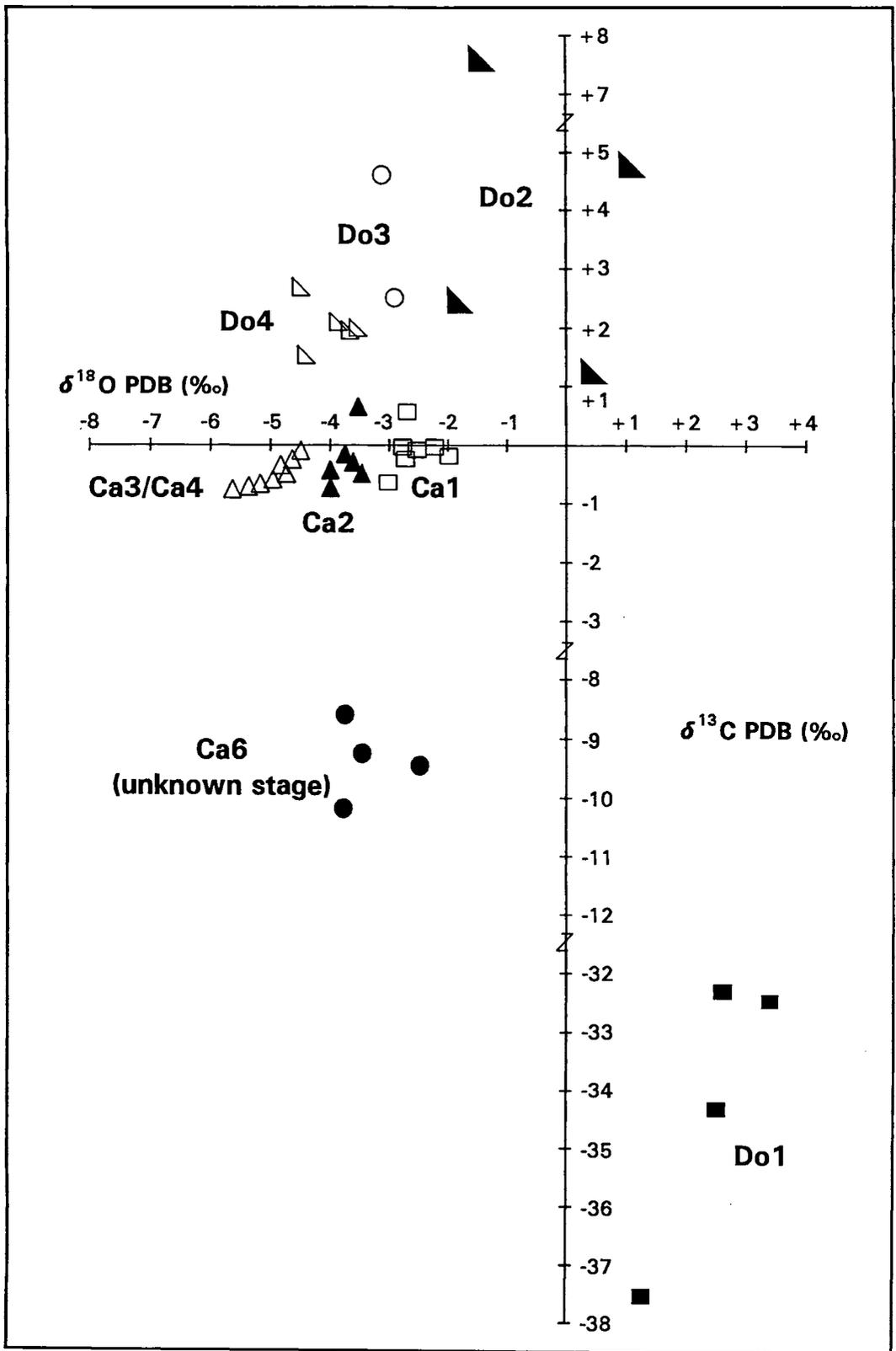


Figure 5: Stable carbon and oxygen isotopic compositions of various cements.

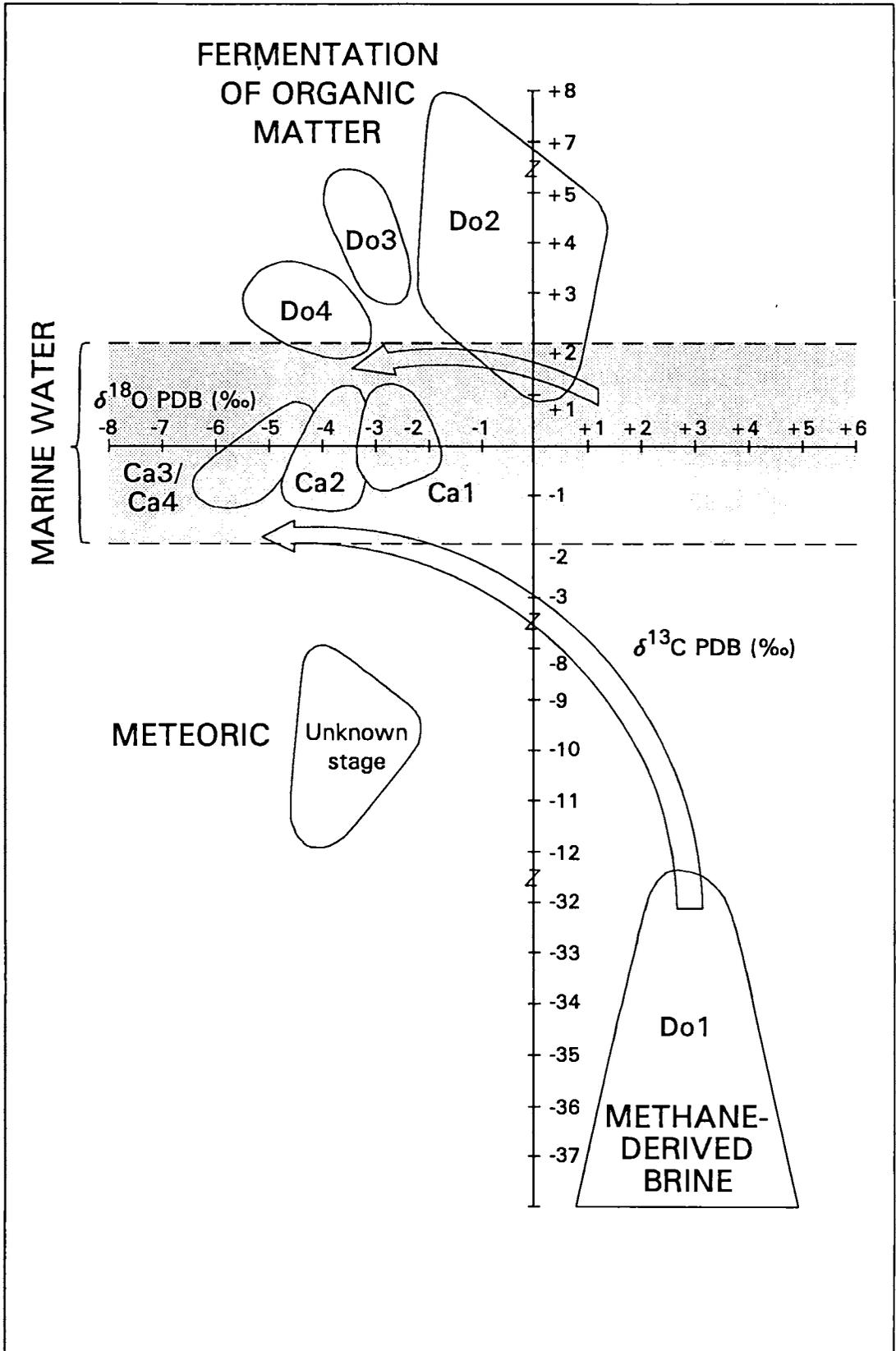


Figure 6: Possible sources of carbonate cementation.

Table 3: Isotopic composition, inferred temperature and maximum burial depth of various cement stages (average geothermal gradient 1.40°F/100ft). Tc calculated for water = -1.2‰ PDB. Td calculated for water = -4.9‰ PDB.

STAGE	SAMPLE NOs	ISOTOPE COMPOSITION		INFERRED TEMPERATURE (°C)	MAXIMUM BURIAL DEPTH (km)
		$\delta^{13}\text{C}_{\text{PDB}}$	$\delta^{18}\text{O}_{\text{PDB}}$		
Ca5	NO SAMPLE	-	-	-	-
Do4	W4.1485	+2.73	-3.89	48.0	1.54
	W4.1485b	+2.64	-3.87	48.0	1.54
	W4.1485c	+3.08	-3.78	47.0	1.50
	W4.1565	+1.70	-4.24	50.0	1.62
	W4.1530	+3.23	-4.72	54.0	1.78
Do3	W4.1470	+4.56	-3.37	45.0	1.42
Do2	W3.1668	+7.72	-1.63	51.9	1.70
	W3.1493	+2.99	-2.52	45.9	1.40
Ca3 / Ca4	W3.1500	-0.38	-4.83	34.0	0.98
	W3.1500b	-0.43	-4.81	34.0	0.98
	W1.1548	-0.65	-5.03	35.0	1.03
	W2.1714	-0.06	-4.37	32.0	0.91
	W2.1714b	-0.07	-4.39	32.0	0.91
	W2.1721	-0.51	-4.88	34.0	0.98
	W2.1721b	-0.67	-4.96	35.0	1.03
Ca2	W3.1577	-0.21	-4.08	30.2	0.83
	W3.1577b	-0.41	-3.67	28.2	0.75
	W2.1717	-1.12	-3.76	28.6	0.79
	W2.1714	-0.21	-3.75	28.4	0.75
	W2.96	-0.55	-3.36	26.6	0.71
Ca1	W1.1500	-0.72	-2.61	23.1	0.55
	W1.1528	-0.38	-2.75	23.7	0.59
	W1.1528b	-0.65	-3.04	25.2	0.63
	W4.1585	-0.01	-2.11	20.8	0.47
	W4.1585b	-0.12	-2.50	22.6	0.55
	W2.1728	-0.07	-2.84	24.2	0.59
Do1	W1.1650	-37.64	+1.19	19.6	0.43
	W1.1628	-32.45	+3.56	9.3	0.00
	W1.1640	-32.22	+2.45	13.9	0.19
	W1.1578	-32.50	+3.22	10.7	0.08
Un-known (Ca6)	W1.1690	-9.27	-3.35	26.6	0.71
	W1.1690b	-8.65	-3.68	28.2	0.73

fluids, the geothermal gradient for the time of each cement stage, and the degree of openness of the system. As the isotopic composition of the pore fluids is unknown, it is impossible to relate these $\delta^{18}\text{O}$ signatures reliable to a precise burial depth. However, the calculation below will provide some information.

$$T^{\circ}\text{C} = 16.9 - 4.21(\delta\text{c} - \delta\text{w}) + 0.14(\delta\text{c} - \delta\text{w})^2 \text{ for calcite}$$

$$T^{\circ}\text{C} = 31.9 - 5.55(\delta\text{d} - \delta\text{w}) + 0.17(\delta\text{d} - \delta\text{w})^2 \text{ for dolomite}$$

where δc or δd = isotopic composition of CO_2 produced from the calcite or dolomite at 25°C ; and δw = isotopic composition of CO_2 in equilibrium with formation water (after Epstein *et al.*, 1953; Irwin & Curtis, 1977).

Substituting a value of $\delta\text{w} = -1.2\text{‰}$ for the seawater composition prior to establishment of polar ice caps (Shakleton & Kennett, 1975) would give a palaeotemperature of each cement stage. Giving a present day geothermal gradient of $1.40^{\circ}\text{C}/100\text{ ft}$, maximum burial depth can be calculated. The results are shown in Table 3.

Strontium Isotope Dating of Dolomitization Events

A constant ratio of $^{87}\text{Sr}/^{86}\text{Sr}$ of seawater is used to evaluate changes in the strontium isotopic composition of the ocean through geologic time by measuring the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of authigenic marine cements. The strontium isotope technique was used to date the dolomitization events.

The Sr isotope ratios of Do1 and Do2 dolomites have been normalised to $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$ and to Standard Reference Material (SRM) 987 = 0.710235. A value of 9×10^{-6} has to be subtracted from the values obtained in order to get the value in terms of Ma. The results indicate that the Do1 dolomitization took place at 10.5 Ma, exactly at the boundary between the Tortonian and Serravalian stages during a sealevel lowstand. The timing has nicely fitted to the earlier events of uplifting as a result of induced faulting as interpreted from seismic section and a palaeofacies development model. Therefore, the Do1 dolomitization event was partly controlled by a sealevel change and regional unconformity that occurred at the boundary between the Middle and Upper Miocene.

Another dolomitization event, Do2 has been dated as 8.9 Ma. The timing of this dolomitization corresponds well with a burial history curve, suggesting the Do2 dolomitization took place at deep burial depth, most likely during smectite-illite transformation as supported by the petrographic evidence i.e. the occurrence of dolomite-clay mixture.

Vertical and Lateral Correlations of Cements

Some individual cement zones (Ca1, Ca2, Ca3 and Ca4) appear to be synchronous across the whole hydrocarbon field at the same stratigraphic levels (Fig. 4). However, Do1 cement is restricted in between the two unconformities in well no. 1. Similar cement does not appear in the same inferred stratigraphic level. This may indicate that the inferred stratigraphic level in well 4 does not represent the same stratigraphic level as in well 1. Another possibility is that the well no. 4

locality had been uplifted further up with lesser erosion compared to well 1 locality. If it is so, different stratigraphic levels are now affected by the same porewater system.

Well 1 has blocky, equant non-marine calcite cements with very light carbon isotope composition (-8 to -12‰). This cement was categorised as Ca6 and was only found in this well above the unconformity. This feature may indicate that well 1 was exposed above the sealevel and subjected to fresh water diagenesis.

Radiaxial calcite of Ca6 was found in well 1 and restricted below an unconformity with the Kudat Formation.

Although it displays similar carbon and oxygen isotopic compositions, it is composed of high Mn^{2+} concentration that might reflect hydrothermal brine expelled from compacting of very thick basinal shale of the Kudat Formation. The lateral and vertical extent of this cement is unknown as no sample is available to correlate them.

Stage Do3/Do4 is another localised cement that particularly occur within well 4 only. Looking at the composition of this cement, it is composed of extremely high Fe^{2+} concentration and graded to ankerite. As the sediments in the well 4 is dominated by very fine silty clay with high organic content, it is most likely that the degradation of organic matter and burial compaction are greater than any other wells.

The overall distributions of various cement stages at field scale are given in Figure 7.

Hydrocarbon Stains and Timing Diagenesis

Hydrocarbon stains were observed in some samples. They display brown stains and trapped in between the Ca4 and Do2, suggesting that the hydrocarbon migration took place at the later part of burial diagenesis about 8.9 Ma before present.

The migration of hydrocarbon has stopped the diagenesis except in the intervals where hydrocarbons are not emplaced.

CONCLUSIONS

Cement stratigraphic approach has been successful in recognising the complete history of cementation in petroleum reservoirs and mapped the distribution of palaeo-porewater system. The following conclusions appear warranted :

- 1) At least nine stages carbonate cementation have been responsible in occluding the pores. Each cement stage represents single crystal morphology, cathodoluminescence features, elemental composition, and isotope signatures.
- 2) The cements were precipitated in oxic to sulphate reduction/fermentation zone, at shallow to deep burial depths, during early to late phases of cementations.

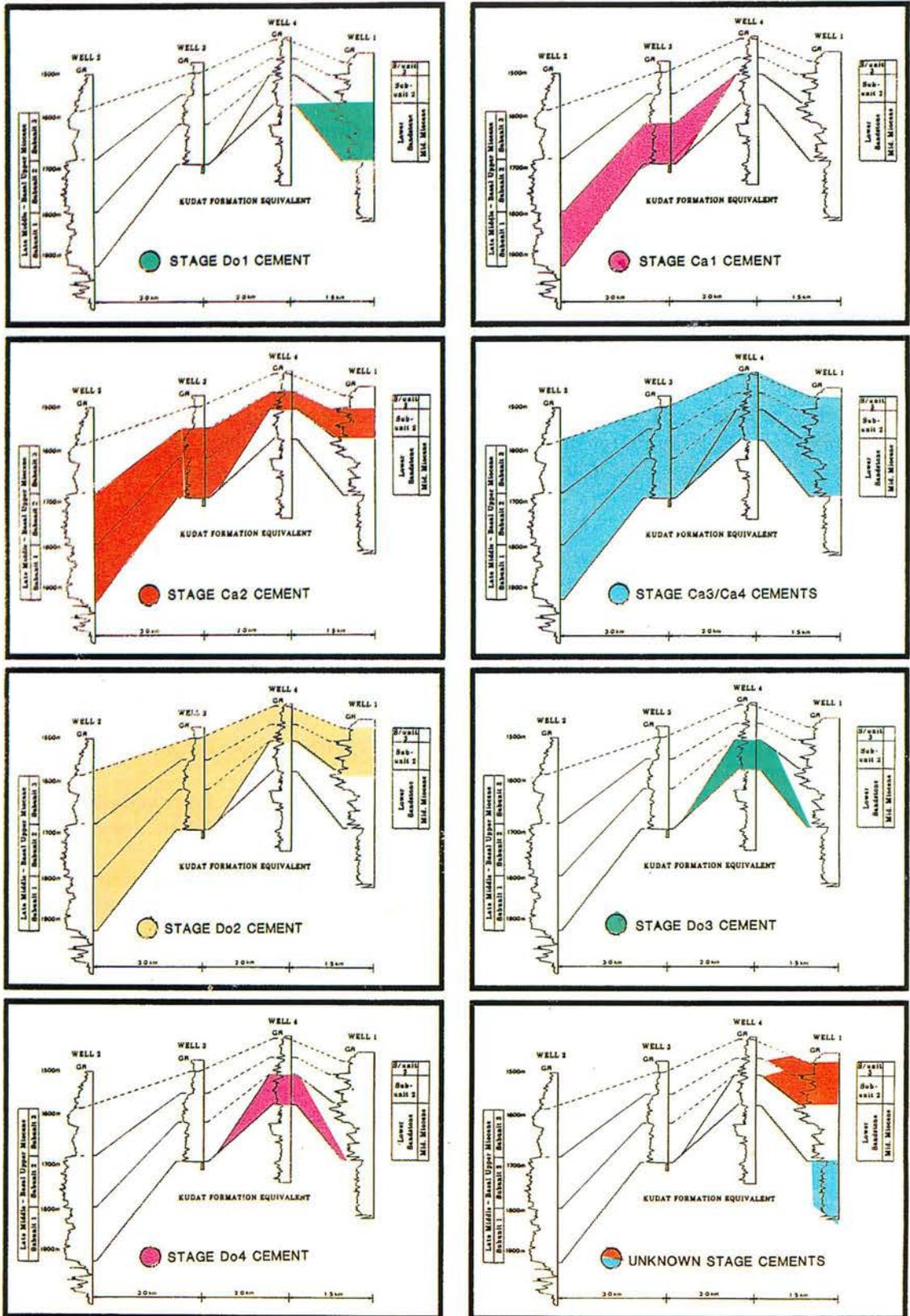


Figure 7: Lateral and vertical correlation of carbonate cements resulted from changing of palaeo-porewater system since Middle Miocene time.

The source of carbon varies from methane-derived brine, marine dissolved carbonates, fermentation of organic matter, and possibly thermogenic brine.

- 3) Three types of different origins calcian dolomites were recognised: (a) methane-derived dolomite (Do1), (b) clay-associated dolomite (Do2), and (c) iron-rich dolomite. Do1 was precipitated at 10.5 Ma, during uplifting an induced faulting, at sealevel lowstand, exactly at the boundary between the Tortonian and Serravalian stages. Do2 was precipitated at 8.9 Ma at deep burial depth during smectite-illite transformation.
- 4) Hydrocarbon emplacement took place at about 8.9 Ma before present.

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