



Textural and paleo-fluid flow control on diagenesis in the Paleoproterozoic Franceville Basin, South Eastern, Gabon

Olabode M. Bankole^{a,*}, Abderrazak El Albani^a, Alain Meunier^a, François Gauthier-Lafaye^b

^a UMR CNRS 7285, IC2MP, Bâtiment B35, 5, Avenue Albert Turpain, 86073 Poitiers Cedex, France

^b Laboratoire d'Hydrologie et de Géochimie de Strasbourg, UMR 7517 CNRS, 67084 Strasbourg, France

ARTICLE INFO

Article history:

Received 25 October 2014

Received in revised form 15 July 2015

Accepted 18 July 2015

Available online 29 July 2015

Keywords:

Paleoproterozoic

Diagenesis

Fluid flow

Clay minerals

Franceville Basin

Gabon

ABSTRACT

The Paleoproterozoic (~2.15 Ga) Franceville Basin, South Eastern Gabon, is a continental sedimentary basin that host unmetamorphosed sediments. This study involve detailed mineralogy, sedimentology, and petrography of the basal sedimentary units of FA and lower FB (FB1) Formations, from the basin margin to centre in relation to mineral paragenesis and fluid flow. The FA Formation conglomerate, sandstone, and mudstone consists lithofacies of mixed fluvial and fluvio-deltaic transitional origin, while the overlying FB1 Formation includes alternating organic rich black shale and sandstone of marine deposit.

The medium- to coarse grained fluvio-deltaic quartz arenite in upper part of FA Formation is characterized by pervasive authigenic quartz cementation that reduced the porosity and permeability in the early stage of burial history. This provides a resistant framework for subsequent diagenetic modification and also inhibits fluid flow during burial diagenesis. In contrast, the clay and unstable detrital grains rich fluvial arkosic to sub-arkosic sandstones that escaped early quartz cementation show considerable pressure solution at grain contacts. These arkosics were less porous and permeable when deposited but transformed to diagenetic aquifers as a result of dissolution of detrital grains during diagenesis and subsequent precipitation of authigenic mineral cements in the resulting secondary porosities.

From the proximal to distal basin and within sample suites, there is no considerable chemical variation in the petrographic distinct generations of the precipitated illite and chlorite suggesting their precipitation from a near equilibrium homogenous pore-fluid. The predominance of 1M₁ illite polytype in most lithologies reflects precipitation of the clay minerals and probably other cements in an environment with high fluid/rock ratio. Dolomite, anhydrite, barite, and Fe-oxides are the main crystallized authigenic minerals aside illite and Fe-rich chlorite clay minerals. The mineralogical assemblages and textural occurrences of the rocks suggest that diagenesis and fluid flow in the FA Formation in the Franceville Basin are mainly controlled by depositional facies.

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1. Introduction

The Paleoproterozoic (2.5–1.6 Ga) sedimentary basins constitute a huge part of the Earth's history and serve as a major supply of almost 75% of earth's important resources such as Au, Pt, Fe, Mn, Zn, U, and Cr (Altermann and Corcoran, 2002). Many Proterozoic rocks have undergone variable degree of metamorphism and igneous activities; however, vast exposures of unmetamorphosed Proterozoic rocks are preserved in many areas and are separated from Archaean rocks by an unconformity (e.g. Thelon Basin, Athabasca

Basin, Transvaal Basin, McArthur Basin, and Franceville Basin). The unmetamorphosed Paleoproterozoic (~2.15 Ga) Franceville Basin (Fig. 1), located in south-eastern Gabon, represents one of the world oldest petroleum systems and ranked as a major Proterozoic accumulation of organic matter (Mossman David et al., 2005). The basin is globally known for its unique fossils (El Albani et al., 2010, 2014) and natural nuclear fission reactors at Oklo, and widely explored for uranium and manganese ore deposits (Gauthier-Lafaye and Weber, 2003).

Despite the vast studies on the reservoir rocks, FA Formation, of Franceville Basin (Weber, 1968; Haubensack, 1981; Gauthier-Lafaye, 1986; Mathieu, 1999; Mathieu et al., 2000; Ossa, 2010; Ossa et al., 2014, etc.), important information regarding mineral paragenesis, diagenetic fluid history, paleo-hydrologic properties,

* Corresponding author. Tel.: +33 549 454 903.

E-mail address: olabode.bankole@univ-poitiers.fr (O.M. Bankole).

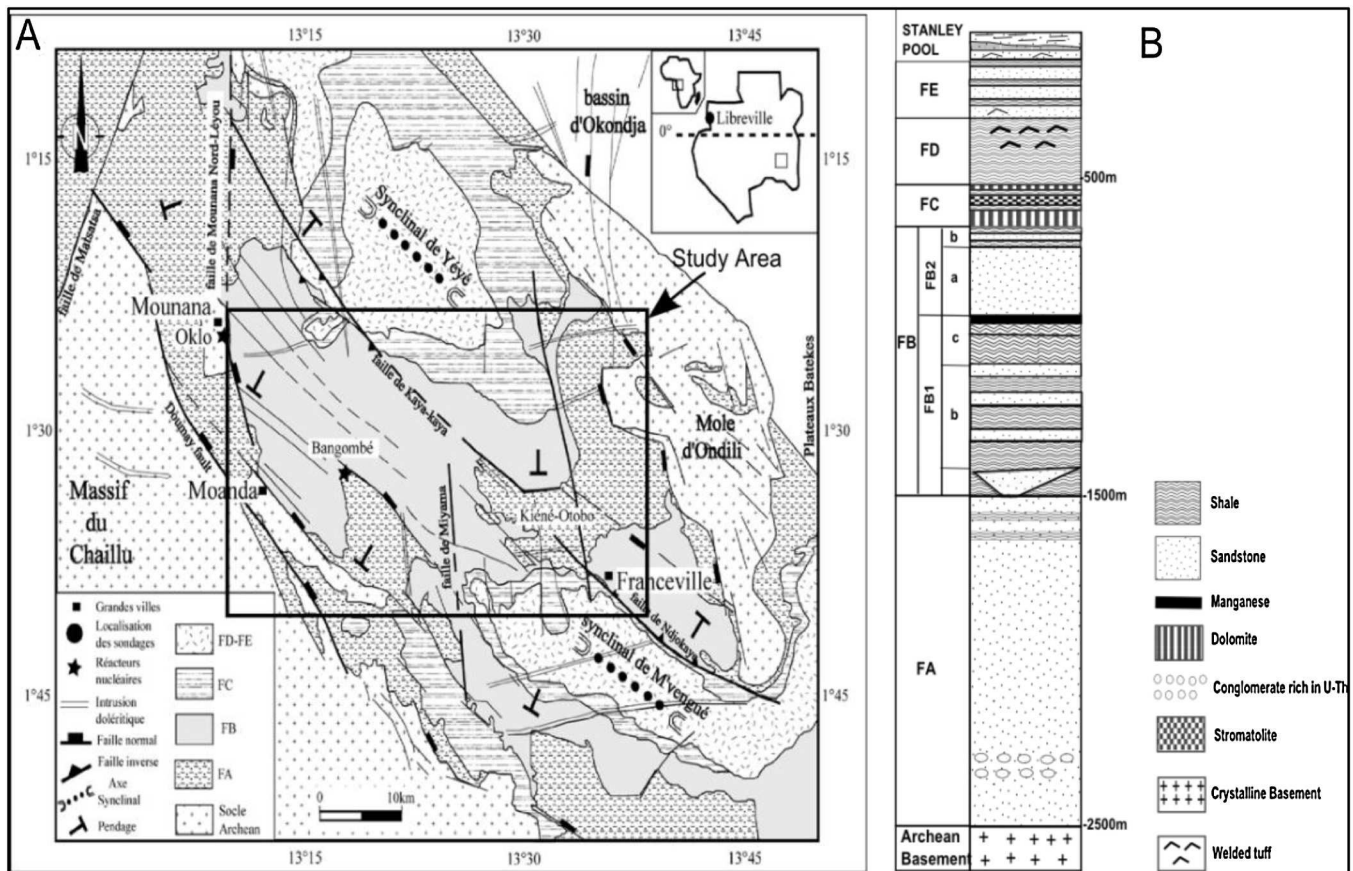


Fig. 1. (a) Regional geological map of Franceville Basin in Gabon (Ossa et al., 2013) that shows location of the study area (inset) and general geology of the surrounding area. (b) Lithostratigraphic subdivision column of Franceville Basin (modified from Gauthier-Lafaye and Weber, 2003).

and fluid migration are poorly constrained. Mathieu (1999) and Mathieu et al. (2000) detailed the paleo-fluids origin and evolution from fluid inclusions studies on mineral veins around the natural nuclear reactors zone at Oklo, Franceville Basin. Five different paleo-fluids responsible for mineralization identified by the authors are: (1) a low saline meteoric water firstly migrating through the basement and injected into the basin, (2) high chlorine brines equilibrated with carbonate and evaporitic layers in sandstones, (3) hydrocarbon-rich fluid derived from organic matter maturation in the shale, (4) a mineralizing fluid originating from the mixing of (1) and (2), (5) a hot fluid related to the nuclear reactors functioning. Their study is corroborated by Mouélé et al. (2014) who investigated the nature of the percolated fluids and alteration processes that affected the upper part of the underlying granitic Archean basement rocks in Franceville Basin. These authors concluded that same fluids of meteoric origin migrated through the basement and FA Formation rocks.

Understanding the diagenetic history and fluid flow is important as the uranium mineralization has been linked to interaction of fluids (3) and (4) at the redox boundary between FA/FB transition in Franceville Basin (Gauthier-Lafaye, 1986; Mathieu, 1999; Mathieu et al., 2000; Gauthier-Lafaye and Weber, 1989; Gauthier-Lafaye and Weber, 2003). The rocks that make up the basin-filling of Franceville Basin have only been affected by low temperature processes (Gauthier-Lafaye and Weber, 1989; Gauthier-Lafaye and Weber, 2003). Thus, these rocks provide an excellent opportunity to study the long term effects of post-depositional alteration processes and mineral paragenetic evolution on fluid flow in a Paleoproterozoic sedimentary basin.

This paper presents a contribution to the study of diagenesis and paragenesis with integrated lithological, petrographical, and mineralogical data in an unmetamorphosed Paleoproterozoic sedimentary basin. The studied basin is a good example of interaction between “petroleum” and uranium mineralization in a diagenetic system. This could allow comparison of diagenetic processes in ancient and modern sedimentary basins. This study takes a systematic approach to studying the regional lithofacies assemblages, petrography and diagenesis of the FA and lower FB1 Formations rocks, by describing and establishing mineral paragenetic sequences and their impact on paleo-fluid migration in FA Formation of Franceville Basin.

2. Geological background

The Franceville Basin (Fig. 1a) is part of the extensive Franceville Series that is exposed in four different intracratonic basins: the Plateau des Abeilles, the Franceville, Lastoursvilles, and Okonja basins in the South Eastern Gabon (Gauthier-Lafaye and Weber, 1989, 2003; Gauthier-Lafaye, 2006). The Franceville sedimentary series covers an area of approximately 35,000 km² consisting of 1–4 km (Bonhomme et al., 1982; Gauthier-Lafaye and Weber, 1989; Bros et al., 1992; Gauthier-Lafaye, 2006; Pr  at et al., 2011) or 1–2.5 km (Thi  blemont et al. 2009; El Albani et al., 2014) thick unmetamorphosed sediments. The sedimentary basin fill is composed of siliciclastic fluvial and marine deposits of Paleoproterozoic age (~2.15 Ga) that unconformably overly Archean metagranitic basement rocks within the Congo craton (Gauthier-Lafaye and Weber, 1989; Feybesse et al., 1998; Gauthier-Lafaye, 2006). The

Series is bounded to the west by the Ogooué domain (Weber, 1968; Ossa, 2010) which is a folded belt considered as a part of Paleoproterozoic (~2.5–2.0 Ga) orogen known as the West Central African Belt (Feybesse et al., 1998) and on the eastern part by the horizontal formations of the Mesozoic Congo Basin (Mathieu, 1999).

On a regional scale, the formation of the intracratonic Franceville series is attributed to the crustal shortening, thickening, thinning, and break-up of Archaean continental blocks of the West Central African Belt (WCAB) (Feybesse et al., 1998). Their formation is characterized by three main syn-sedimentary tectonic events and sedimentological evolutions separated by major unconformities (Feybesse et al., 1998; Pr  at et al., 2011). According to the proposed model of Feybesse et al. (1998), the first phase corresponds to the break-up of the Archaean continental rocks accompanied by opening of the pre-oceanic Franceville extensional basin leading to deposition of FA Formation (before 2.15 Ga). The second phase was characterized by brittle tectonism of the foreland basin with accelerated subsidence resulting in transition from fluvio-deltaic sedimentation at the top of (FA) to a marine sedimentation (FB-FC) from 2.1 Ga onward. This phase is also accompanied by the propagation of northwest-southeast faults that formed a number of grabens into which fine terrigenous sediments of FD and FE Formations were deposited.

2.1. Stratigraphic framework

Five major formations (Fig. 1b) constitute the lithostratigraphic framework of the sedimentary rocks in the Franceville Basin (Weber, 1968; Gauthier-Lafaye and Weber, 1989; Gauthier-Lafaye, 2006; Ossa, 2010; El Albani et al., 2010, 2014; Pr  at et al., 2011). Starting from the oldest, which unconformably rests on the Archaean basement, is the basal syn-tectonic FA Formation consisting predominantly of 500–1000 m thick fluvial to deltaic, arkosic conglomerate and sandstone successions (Gauthier-Lafaye and Weber, 1989). The rocks of this formation at the distal basin shift in colour from red/pink at the base, to green around the middle, and grey-black in the upper part. This colour variation commonly cuts stratigraphic boundaries and is interpreted as alteration related to diagenetic fluid flow (Haubensack, 1981; Gauthier-Lafaye, 1986; Gauthier-Lafaye and Weber, 1989). All the uranium ore deposits in Franceville Basin are hosted in the upper part of FA rocks in association with solidified bitumen (Gauthier-Lafaye, 1986; Gauthier-Lafaye and Weber, 1989; Gauthier-Lafaye and Weber, 2003).

The FB Formation discordantly overlies the FA Formation, and consists 300–1000 m thick alternating sandstone and mudstone (shale and siltstone) (Gauthier-Lafaye and Weber, 1989). The FA–FB transition boundary is interpreted to mark the onset of shallow marine deposit in the basin (Gauthier-Lafaye, 1986; Pr  at et al., 2011). FB Formation is further subdivided into FB1 (a, b, and c) and FB2 (a and b) subunits based on lithological variations (Azzibrouck-Azziley, 1986; Pambo, 2004). FB1a and FB1b are composed of interlayered shale, sandstone, and conglomerates in an upward fining succession. FB1c consists essentially of black shale with thin iron formation and capped by a thick manganese ore deposit (Gauthier-Lafaye and Weber, 2003; Mossman David et al., 2005; El Albani et al., 2014). FB2a consists of sandstone beds that are overlain by FB2b consisting of finely laminated black shale interbedded with thin siltstone.

The overlying FC Formation is interpreted as a basin edge deposit consisting of 10–40 m thick dolomite and thick-banded stromatolitic cherts (Gauthier-Lafaye and Weber, 2003; Dutkiewicz et al., 2007; Pr  at et al., 2011). FD Formation consists of 200–400 m rocks dominated by black shale capped by welded tuff (Gauthier-Lafaye and Weber, 2003; Pr  at et al., 2011), while the uppermost and youngest FE Formation consists of medium-grained sandstone with

interlayered shale (Gauthier-Lafaye and Weber, 2003; Pr  at et al., 2011). Recently, Thi  blemont et al. (2009) reported that the FE Formation could not be maintained due to the difficulty in distinguishing it from FD Formation in the eastern basins. The authors assumed that sediments probably resulted from erosion of the Ogoou   orogenic belt filling local depressions.

2.2. Diagenesis and mineralization

The mineralogy of Franceville Basin sediments has been described to reflect formation of diagenetic mineral phases during burial (Gauthier-Lafaye, 1986; Ossa, 2010). The maximum burial depth is estimated to reach about 4 km and temperature of 180–200 °C (Openshaw et al., 1978; Gauthier-Lafaye and Weber, 1989; Gauthier-Lafaye and Weber, 2003). Variable proportions of illite and Fe-rich chlorite formed from alteration of detrital aluminosilicate minerals characterize peak diagenesis in the basin (Gauthier-Lafaye, 1986; Bros et al., 1992; Gauthier-Lafaye and Weber, 2003; Ossa, 2010). Ossa (2010) and Ossa et al. (2014) suggested the occurrence of these authigenic clays through a widespread activity of migrating hydrothermal fluids in the FA Formation and gave an estimated formation temperature of 240 ± 30 °C from thermodynamic models. The overlying FB Formation is characterized by an assemblage of diagenetic illite, chlorite, and illite–smectite mixed layers (Bros et al., 1992; Ossa et al., 2013; Ngombi-Pemba et al., 2014).

The mineralization of the uranium ores in Franceville Basin has been linked to fluid migration, paleo-redox condition, and organic matter maturation in the black shale at the FB1/FA transition boundary (Gauthier-Lafaye and Weber, 2003; Mossman David et al., 2005). The inferred coincidence of uranium mineralization with the interval of hydrocarbon migration was believed to have led to the localization of uranium ores in petroleum-type structural traps in the upper part FA sandstones at Oklo (Gauthier-Lafaye, 1986; Mathieu, 1999; Gauthier-Lafaye and Weber, 1989; Mossman David et al., 2005).

3. Lithostratigraphy of the study area

The thickness of the FA Formation varies across the basin and could reach a maximum of about 1000 m towards the centre and may be absent or reduced to few metres at the edge. A simplified lithostratigraphic column of representative drill-holes from the basin edge to the centre across the studied area (Fig. 2) is presented in transect of Fig. 3.

3.1. Proximal (Mabinga – NW)

The studied well in the proximal part of the basin reaches an approximate depth of 447.45 m and correspond to 106 m thick lower FB1 overlying 341.75 m of upper FA Formation. The FB1 sediments consist of alternating greyish light green to greyish black mudstone (shale and siltstone) and fine-grained sandstone that are characterized by preserved plane laminations, convolute bedding, and load cast sedimentary structures.

The underlying upper FA is composed of basal coarse-grained sandstone with few interlayered conglomeratic sandstone (447.45–300 m), medium-grained sandstone with thick interbedded coarse-grained sandstone (300–175 m) in the middle part, and fine- to coarse-grained sandstone with minor interlayered mudstone in the upper unit (175–106 m). Upwardly, the succession represents an upward-fining to -coarsening greenish to greyish black rocks. This drill-hole exhibits a dolerite intrusion as confirmed by a dolerite sample at about 373 m depth.

3.2. Intermediate (Bangombé)

At Bangombé, the deepest one (BA 2) among the studied drill-holes reaches a maximum depth of 950 m and drilled directly to the Archaean basement (Fig. 3). The studied drill-holes at Bangombé cover about 100–150 m thick of FB1 Formation (BA 30B and BA 37) consisting of alternating mudstone, sandstone, and breccias. The rocks just above the FA Formation are very light green grading upward into greyish black facies. FA Formation reaches a thickness of about 946 m consisting of basal thin mudstones (946–941 m), thick bedded medium- to coarse-grained sandstone and conglomerate with few interlayered mudstone (941–380 m), medium-grained sandstone with thick interbedded coarse-grained sandstone (380–180 m), and capped by alternating fine- to coarse-grained sandstone with interlayered mudstone in an upward-fining succession (180–9 m).

3.3. Distal (Kaya-Kaya/Kiéné-Otobo – SE)

The drill-holes at the distal basin are essentially FA Formation rocks reaching a maximum thickness of about 1000 m at GR 15 (Fig. 3) and represent part of the basin that present the complete lithostratigraphy of FA Formation. The lower section rocks (~993 to 750 m) consist mainly of coarse to conglomeratic sandstone with an interlayered mudstone displaying fining-upward sequence. The middle section consists of sequences of fine-grained sandstone grading upward into coarse grained sandstone and each sequence is sometimes capped by conglomerate (750–600 m), and overlain by thick bedded coarse-grained sandstone with interbedded medium-grained sandstone in no particular sequence (600–500 m). The next succession consists of alternating mudstones and fine- to coarse-grained sandstone in an upward coarsening succession (500–400 m) and overlain by thick bedded fine-grained sandstone with interlayered thin beds of medium- to coarse-grained sandstone and minor mudstone coarsening-upward (400–300 m). The uppermost section (300–28 m) consists of alternating sequences of fine- to coarse-grained sandstone with interlayered mudstone fairly coarsening upwardly.

3.4. Stratigraphic correlation of FA Formation

Lateral stratigraphic correlation from the proximal to distal of the FA rocks appears difficult due to absence of continuous marker bed, lack of biostratigraphic signals (no microfossils), and very low amounts of mudstone in FA Formation. Interbedded mudstone are very few at the basin margin compared to distal basin. Also, absence of detailed subsurface data (e.g. seismic and well logs) makes horizontal stratigraphic correlation practically impossible in basin architecture like fluvial dominated FA sandstone in Franceville. However, the distal part of the basin is presumed to provide an excellent key area for understanding the stratigraphy in the FA Formation rocks because it presents complete sedimentary successions of ~1000 m thickness. In this study, we have informally divided the FA Formation rocks into three stratigraphical sections at the basin centre (Fig. 3) based on lithofacies associations and mineral composition: Lower Section (LS); Middle Section (MS); and Upper Section (US).

4. Sampling and analytical methods

4.1. Study area and sampling

Ten drill-holes covering all lithostratigraphic units in FA and lower FB1 Formations have been studied from three localities from proximal to distal part of Franceville Basin (Fig. 2). One

drill-hole at Mabinga (MAB) provides data towards the proximal location; four at Bangombe (BA 2, BA 11, BA 30B, and BA 37) represent relative intermediate position along the basin centre; and five at Kaya-Kaya/Kiéné-Otobo (GR15, GR 22, GR 23, GR 25, KA 13) from distal basin. The drill-holes thicken from ~447 to 1000 m along the proximal to distal basin in NE-SW transect (Fig. 3). The samples at Mabinga and Bangombe (except BA 2) crosscut the FB1 Formation and FA sandstones, whereas none of the samples at Kiéné-Otobo intersect the FB1 Formation in this study.

After careful examination of drill cores, about 200 representative core samples covering different characteristic features (depositional facies, textural, and colour variations) from bottom of FA through FA/FB1 transition were selected. The samples were logged and described to provide information on sedimentology of the studied area. Detailed logs were completed for representative drill-holes from each part of the basin (Fig. 3). Samples were subsequently prepared for analytical studies.

4.2. Analytical methods

Petrographical and mineralogical examinations of polished thin sections were made by optical and scanning electron microscopies for detail mineralogy, texture, and mineral paragenetic relationships. Thin sections were examined by transmitted light and reflected light microscopy using a Nikon ECLIPSE E600 POL microscope coupled with a Nikon Digital Sight DS-U1 camera and installed NIS-Element D software for scanning observations. Selected thin sections were carbon coated and examined using a JEOL JSM 6400 scanning electron microscope (SEM; University of Poitiers) equipped with energy dispersive spectra (EDS) for semi-quantitative mineral analysis in backscattered electron mode (BSE), operating at 15 kV accelerating volts, 1 nA beam current, and 16.5 mm working distance. Thin sections of selected carbonate rich and quartz cemented samples were characterized with a Technosyn cold Cathode Luminescence Mode 8200 MK II device to visualize the carbonate and quartz cements. CL images were recorded with a Nikon Digital Sight DS-U1 digital camera.

We analyzed whole rock powders and clay mineral fractions (<2 µm) of all samples with X-ray diffraction (XRD). Furthermore, clay size fractions (<1 µm, 0.2–1 µm, and <0.2 µm) of some selected samples were obtained by ultrasonic agitation (Elma S60), GR 4 22 centrifuge (<2 µm and <1 µm), and a Beckman J2 21 ultra-centrifuge (<0.2 µm) from gently hand crushed bulk samples dissolved in deionized water without any chemical pre-treatment (Moore and Reynolds, 1997). The diffraction patterns were acquired with a Bruker D8 ADVANCE diffractometer at University of Poitiers using CuKα radiation operating at 40 kV and 40 mA and 0.025 s step size for mineralogical composition. Clay sized oriented fractions were analyzed from 2 to 35° 2θ angular range after successive air drying (AD), ethylene glycol (EG) saturation, and heating treatment to 550 °C. The bulk powdered samples were examined between 2–65° 2θ angular ranges. X'pert High Score software was used for background stripping, indexing of diffraction peaks, and identification of mineral phase by comparison with International Centre for Diffraction Data (ICDD) files.

Illite and chlorite polytypes were determined for selected representative samples on randomly oriented (<2 µm, 1–2 µm, 0.2–1 µm, and <0.2 µm) clay sized powder separates. The mounts were recorded from 19 to 34° 2θ with a step size of 0.01° 2θ per 2 s counting time for illite and 30 to 65° 2θ with a step size of 0.01° 2θ per 2 s counting time for chlorite. The resulting patterns were compared with reference data (Brindley and Brown, 1980; Drits et al., 1993; Moore and Reynolds, 1997).

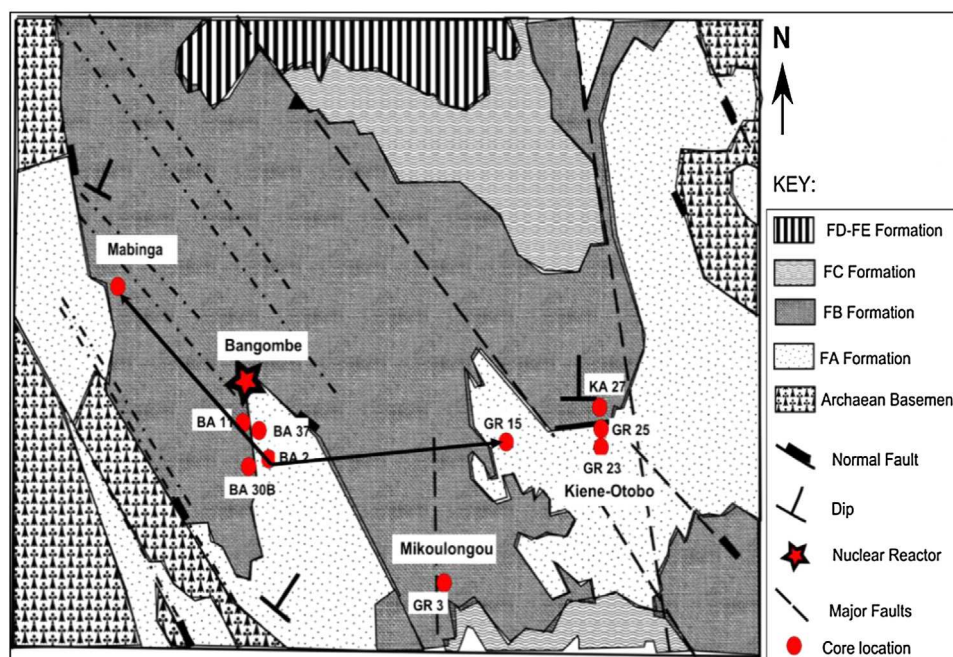


Fig. 2. Map of the study area with locations of drill-holes included in this study. The connecting thick line between sample localities represents the transect in Fig. 3.

5. Results

5.1. Lithofacies associations

5.1.1. FB1 Formation

The lower FB1 rocks are characterized by alternating sandstone and mudstone lithofacies. The rocks are mainly light green at the base and grades vertically upward into grey-black. The sandstone consists of matrix supported fine- to coarse-grained arkosic sandstone that contains poorly sorted, angular quartz and plagioclase grains. The mudstones (siltstone and shale) lithofacies are characterized by preserved plane, inclined, and convolute laminations, and load cast sedimentary structures. The black coloured rocks are mostly rich in organic matter which are often concentrated parallel to bedding.

5.1.2. FA Formation

In the study area, three broad lithofacies associations are identified: fluvial (lithofacies 1), deltaic (lithofacies 2), and tidal (lithofacies 3) in the FA Formation. All lithofacies were described from petrographic thin section and drill cores.

The **fluvial facies** (LF1) consists of coarse- to very coarse-grained conglomeratic sandstone with interlayered medium-grained sandstone and minor mudstone. This lithofacies association is common in the lower section (LF1a) and part of the upper sections (LF1b) of FA Formation. LF1a is predominantly thick coarse- to very coarse-grained sandstone with interlayered conglomerate, whereas LF1b is dominated by alternating medium- to coarse-grained sandstone with minor interlayered fine-grained sandstone and mudstone. The sandstone consists of poorly sorted, angular to sub-angular, quartz and feldspars clasts. Few of the lithofacies in the distal basin show oblique stratification. The rocks are classified as arkosic arenites. The LF1b at the proximal is coarser and poorly sorted with more interlayered conglomerate when compared to distal counterpart where it is moderately sorted, sub-angular, and medium- to coarse-grained with more interlayered fine-grained sandstone and mudstone. LF1 is well developed at the basin edge and its thickness decreases towards the basin centre.

The **deltaic facies** (LF2) is composed of sequences of alternating fine- to coarse-grained sandstone and mudstone which are sometimes interlayered with minor conglomerate, mostly in upward coarsening successions. This heterolithic lithofacies covers most parts of middle and upper sections at the distal basin. The middle section, LF2a, consists of purple/green interbedded poor- to moderately-sorted, sub-angular medium- to coarse-grained sandstone and mudstone grading in upward-coarsening successions and sometimes grades in no definite pattern. LF2a is observed only in the distal basin and the interbedded mudstone corresponds to the Otobo unit defined by (Haubensack, 1981; Gauthier-Lafaye, 1986; Gauthier-Lafaye and Weber, 1989). The interbedded sandstone are composed of plane laminated or crossed laminated medium-grained sandstone with minor coarse-grained arkosic arenites. The interbedded mudstone and fine-grained sandstone are characterized by lenticular bedding, flaser bedding, and plane laminations. Thin bedded medium grained heterolithic sandstone lithofacies predominantly of grey-black, sub-rounded, moderately- to well-sorted, quartz cemented quartz arenites is stratigraphically restricted to the topmost part of the FA Formation (<8 m). LF2b is only observed in core MAB-110.6 and BA2-21.2 just below the FB1 Formation in this study.

The **tidal facies** (LF3) consists predominantly of thick bedded cross and ripple laminated fine-grained sandstone with interlayered parallel laminated mudstone and minor interbedded medium- to coarse-grained sandstone. This predominantly red and very micaceous lithofacies is observed only in the distal basin, and the associated interbedded mudstones are characterized by slump beddings, lenticular and flaser beddings.

5.2. Petrography

5.2.1. Sandstone and mudstone mineralogy

5.2.1.1. FB1 Formation.

Petrographically, the sandstone are dominantly matrix supported arkosic arenites consisting of quartz, plagioclase (albite), and mica detrital grains (Fig. 4). The grains show moderate to strong degrees

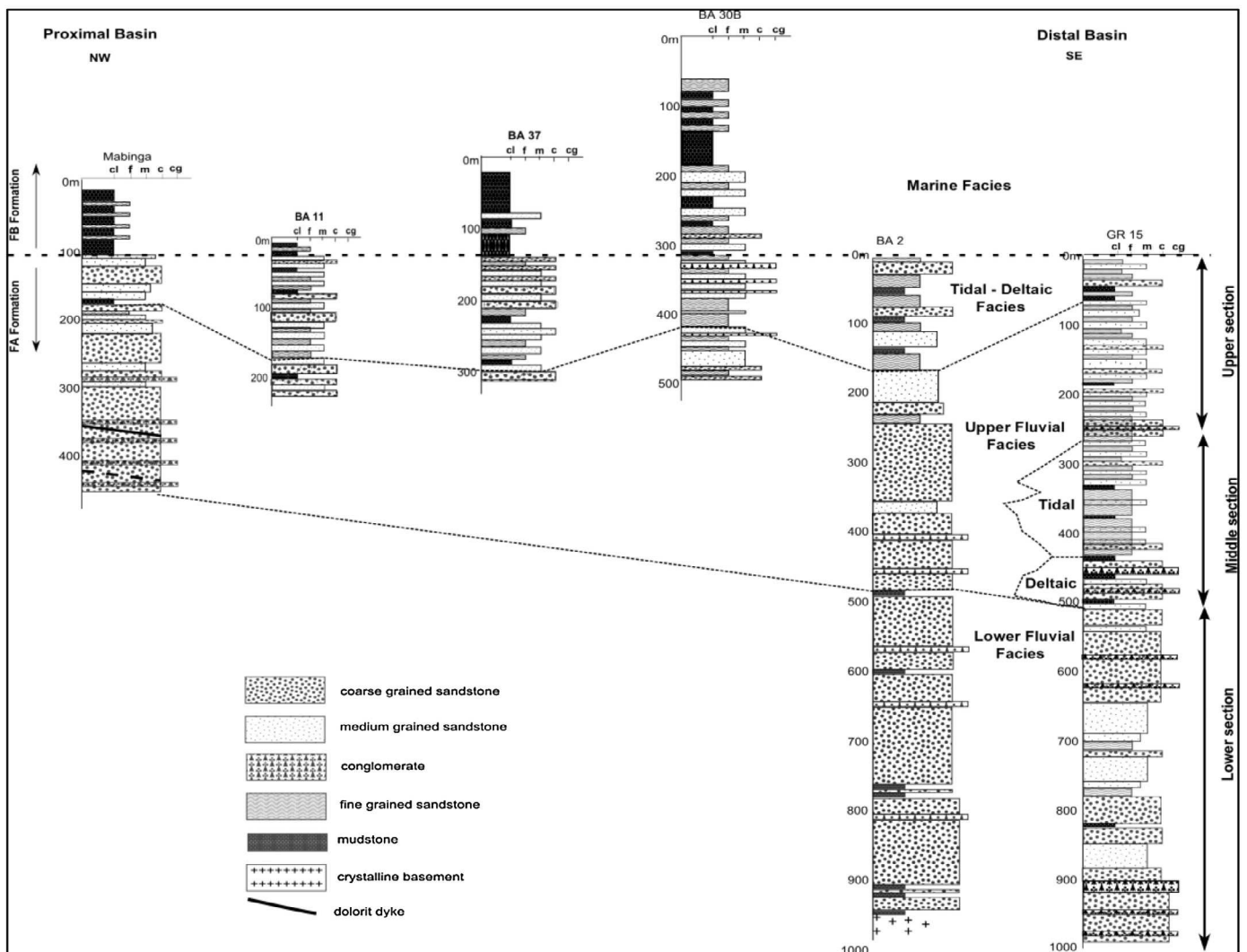


Fig. 3. Stratigraphic sections showing the vertical succession of lithofacies in six drill holes shown in Fig. 2. The drill holes are drawn and placed vertically based on inferred FA/FB boundary after core logging.

of compaction characterized mostly by long grain and stylolite contacts (Fig. 5a). The feldspar is mainly albite which is often partially altered and replaced by dolomite and illite; unaltered plagioclase is also observed. The pore space is dominated by illite, chlorite, dolomite, organic matter, in some instances pyrite (Fig. 5b), and minor quartz overgrowth. Apart from illite/smectite mixed layer (I/S), all the diagenetic clay minerals in the FB1 are also found in FA Formation.

5.2.1.2. FA Formation.

5.2.1.2.1. Mudstone composition.

Qualitatively, the detrital and clay mineralogy of the mudstone in FA Formation is similar to that of sandstone but differs in relative abundances, distribution, and microfabrics. Petrographic relationship shows that the detrital grains are suspended in matrix which is essentially dominated by illite and chlorite which commonly occurs along mica cleavages. The mudstones are richer in platy micaceous minerals which are often aligned parallel to bedding and compacted detrital quartz and feldspars. The micaceous minerals (biotite and muscovite) are moderately preserved compared to sandstone. However, in the red coloured facies, fine grains of hematite are often observed along the cleavages of the micas and stains within the matrix clay minerals. Dolomite is present in

few of the drab facies while sulphates minerals are completely absent.

5.2.1.2.2. Sandstone composition.

The FA Formation sandstones form a spectrum between texturally immature arkosic- to sub-arkosic sandstone and texturally sub-mature quartz arenites. Quartz is the principal detrital framework grain and corresponds to high monocrystalline/polycrystalline ratio (Fig. 5c). The non-quartz detrital components largely include K-feldspar (microcline) with plagioclase (albite) which is common in the middle fluvio-tidal samples (Fig. 4). The feldspars have undergone partial to complete dissolution and replacements by (Fig. 5d); however, unaltered feldspar is sometimes preserved (Fig. 5e). Dissolved feldspars and quartz are commonly replaced by carbonate, barite, illite and chlorite which fill their intragranular pores and feldspar cleavages (Figs. 5d, f and 6a, b). Micaceous minerals consist mainly of muscovite and altered biotite which are usually bent when present at grain contacts due to compaction. Chloritization of biotite is common in the middle-upper section (Fig. 5g), while its replacement by hematite and titanium oxide/titanium-iron oxides is common in the lower section red coloured rocks (Fig. 5h). Zircon and rutile appear to be the most abundant heavy mineral occurring within matrix, grain surface and contacts, whereas thorite, monazite, and apatite are in small amounts. Traces of lithic fragments are also observed in most of the samples.

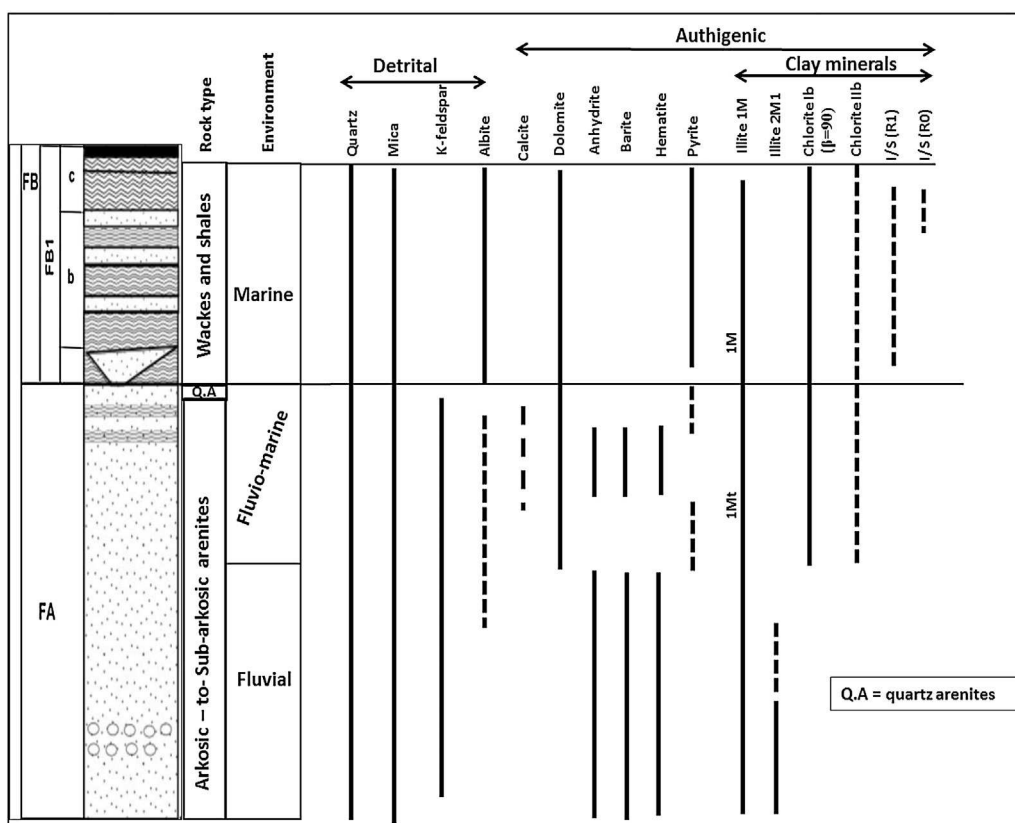


Fig. 4. Stratigraphic mineral evolution across the studied formations (Q.A.: quartz arenite).

5.2.1.3. Diagenesis and authigenic minerals.

The FA sandstones show variable degrees of diagenetic alteration depending on original detrital composition and the amount of clay matrix present. The quartz arenites show moderate degree of compaction features and have their intergranular pore spaces filled with authigenic quartz cements and minor organic matter (Figs. 5c and 6a, b). Compaction features such as long and concavo-convex grain-to-grain contacts (mechanical), and stylolite/pressure solution (chemical) are common in the arkosic sandstones (LF1, LF2a, and LF3), and they contain diverse species of authigenic minerals (Fig. 5f).

Quartz cement is the major and important authigenic phase in the quartz arenite, LF2b, where interstitial clay is less abundant. This lithofacies have experienced early burial quartz cementation as grain-to-grain relationships show few signs of compaction (Figs. 5c and 6a, b). The earliest quartz cement is observed by its pore-filling and syntaxial overgrowths on quartz grain boundaries and is easily distinguished from its detrital host by presence of dust rims which are mostly filled by organic matter in the upper black lithofacies. This quartz cement (Q1) is coined “early quartz” cement because it appears to predate significant grain-to-grain mechanical or chemical compaction features, such as pressure solution during the burial history. Quartz cements are not abundant and are less developed in the arkosic and sub-arkosic lithofacies that lacked Q1 probably due to presence of matrix clays and carbonates. These lithofacies are characterized by minor syntaxial quartz overgrowths/cements (Q2) that precipitated through compaction-driven grain-to-grain pressure solution during burial. This burial quartz cement/overgrowth (Q2) appears to pre- and postdates most authigenic cements (Fig. 6c).

Authigenic carbonate replacements and pore-fillings are seen to be widespread with no single pattern in the arkosic and sub-arkosic arenites, but absent in the quartz arenites. The carbonates

commonly occur as early macro-crystalline pore-filling in form of poikilotopic texture occluding pore space (Fig. 6d), and also as late replacements of quartz and feldspars during dissolution and fracturing (Figs. 5f and 6d). Dolomite predominates while calcite is present in lesser amount. Pore-filling calcite post-dating pore-rimming hematite is also observed in few reddish sandstone. Compositionally, the dolomite is mainly non-ferroan; however, Fe-dolomite is also observed in few cases. When present, the non Fe-dolomite appears to pre-date Fe-dolomite. Carbonate occurrence is relatively limited to the middle and upper units in the FA Formation (Fig. 4).

Sulphate cements (anhydrite and barite) are exclusively observed in the pink/red and bleached medium- to coarse-grained at the distal basin, and they are very rare to absent in drab facies. Anhydrite is the most abundant; barite is minor in most instances, and gypsum in trace amount in very few greenish facies. Most of the sulphates are pore-filling (Fig. 6c, e and f). Anhydrite and barite appear to postdate early formed calcite, illites (Fig. 6e), and quartz overgrowth (Q2) (Fig. 6c). Their preferential replacements of detrital grains, especially feldspars are also commonly observed. Several textural evidences show that anhydrite predates barite and hematite (e.g. Fig. 6f). Like carbonates, later dissolution of anhydrites results in preservation of secondary microporosities in the sandstone.

Hematite is present only in red facies of FA Formation at distal basin. It commonly occurs as matrix stains or disseminated within clay minerals and other cement phases, and in very rare instances filling and lining secondary micropores. It often occurs in association with titanium oxide both within the matrix and along cleavages of altered biotites (Fig. 5h) and feldspars. Occasionally, in the medium to coarser red facies, iron oxides occur as amorphous or euhedral micro-size crystal on grain surfaces and in fluidised form cross cutting precipitated anhydrite and other cements

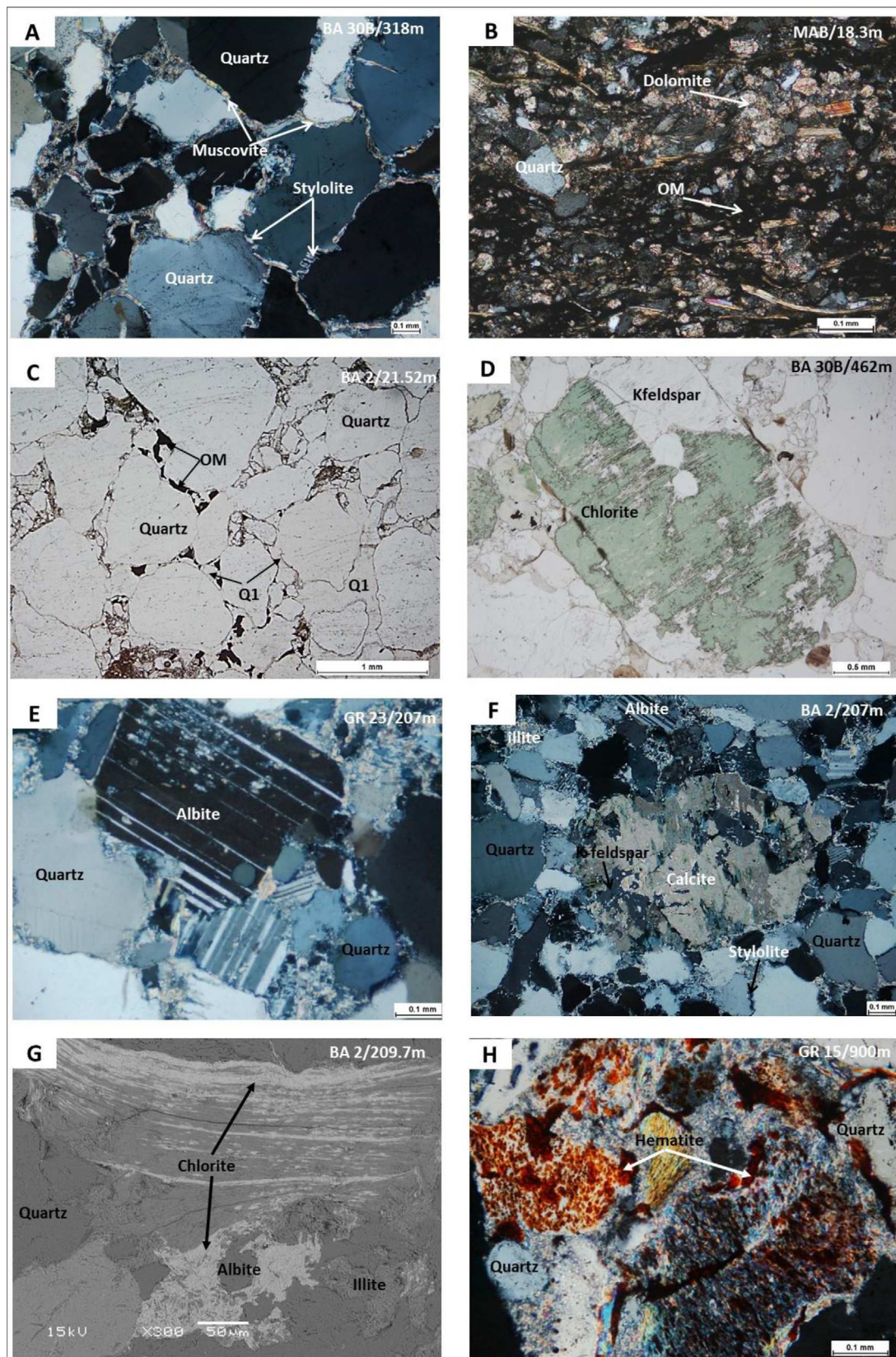


Fig. 5. Photomicrographs of representative samples and other petrologic features found within samples from FB1 (A and B) and FA (C–H) Formations: (A) sandstone with strong solution contacts between coated grains (XL); (B) detrital grains supported by dolomite, clay minerals, and organic material cements in silty mudstone (XL); (C) Quartz cemented medium-coarse grained quartz arenite with stains of organic matter on overgrowths and grain edges. The presence of floating grains and absence of pressure-solution features demonstrate that the quartz cement (Q1) predates significant compaction (PL); (D) chloritization of feldspar (PL); (E) preserved unaltered plagioclase with quartz (XP); (F) Dissolution and replacement of K-feldspar by calcite (XP); (G) Chloritization of mica and “fan-like” pore-filling coarse Fe-chlorite surrounding detrital albite (BSE); (H) Hematitization of mica in coarse grained sandstone (XP). (OM: organic matter; XP: cross polarized; PL: plane polarized; BSE: backscattered electron).

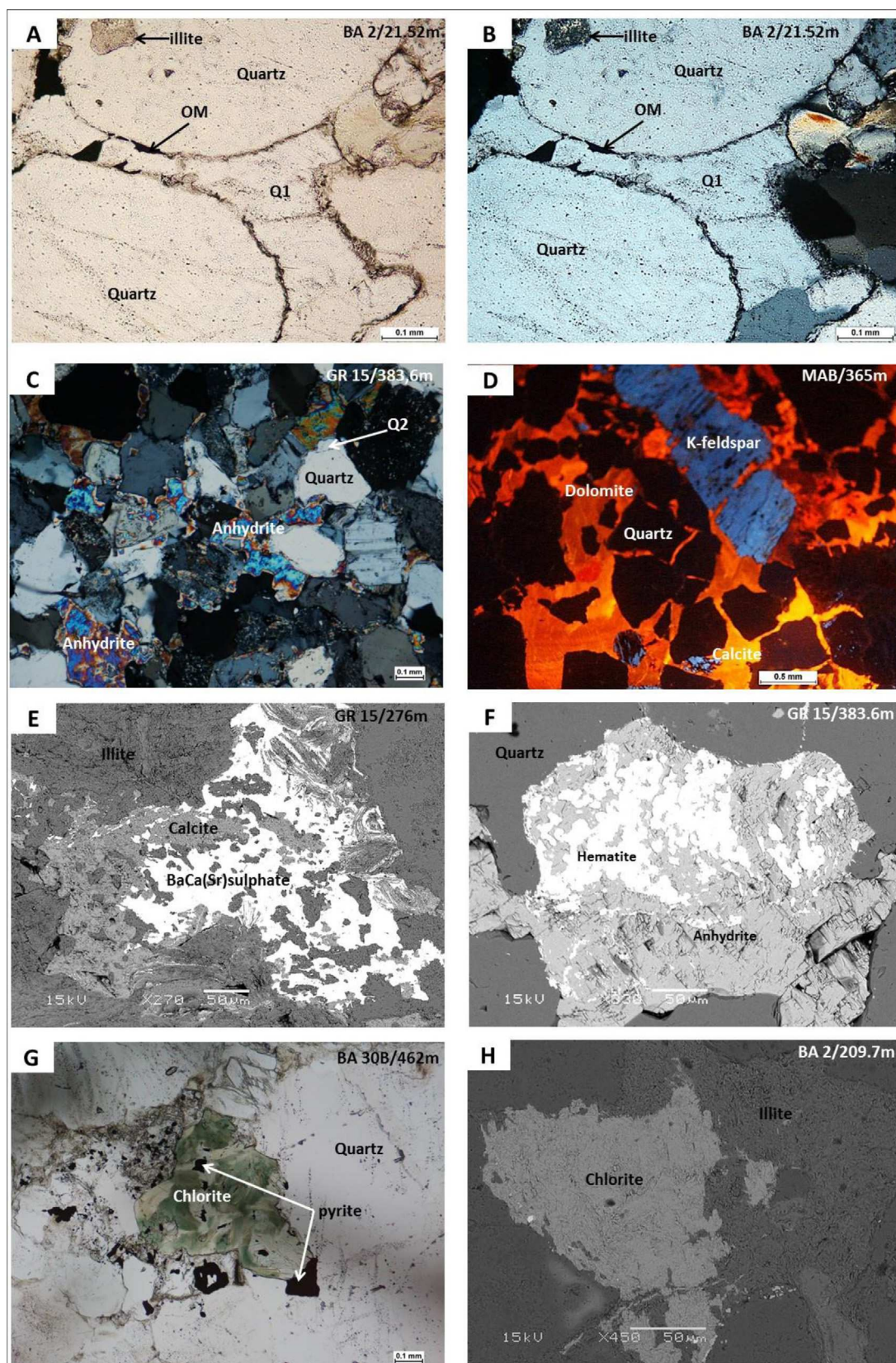


Fig. 6. Photomicrographs of representative samples in FA Formation: (A and B) pore-filling quartz cement in the moderately sorted quartz arenite in PL and XL respectively. The quartz cement (Q1) appears to predate significant grain-to-grain compaction features; (C) pore-filling anhydrite cement postdates quartz overgrowth (Q2) in red arkosic sandstone (XL); (D) Poikilotopic calcite and dolomite cements cross-cutting fractured quartz and K-feldspar. Presence of floating grains indicates that calcite precipitation began early in the burial history (CL–XL); (E) barite postdates pore-filling calcite and illite (BSE); (F) crystals of hematite post-date pore filling anhydrite (BSE); (G) Chlorite is matrix in association with postdating authigenic pyrite (PL); (H) pore-filling illite and chlorite (BSE). (OM: organic matter; XP: cross polarized; PL: plane polarized; BSE: back scattered electron; CL: cathodoluminescence).

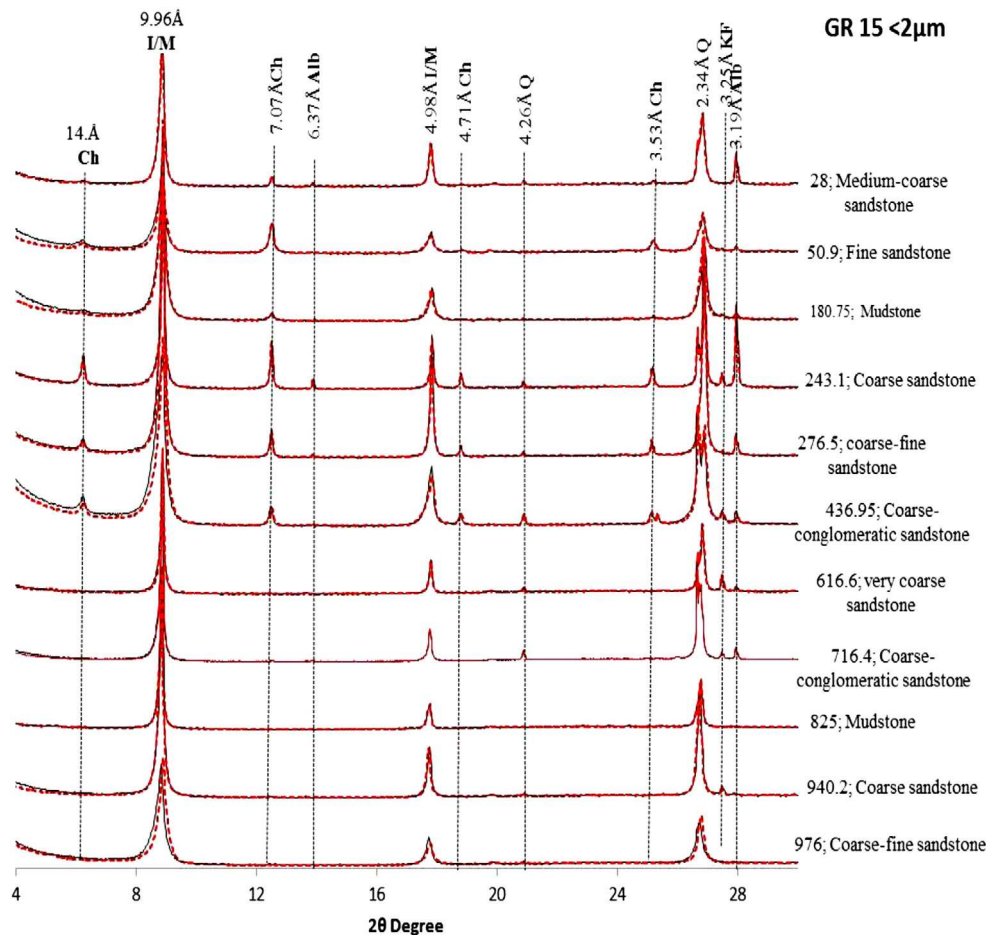


Fig. 7. X-ray diffraction patterns for oriented preparations in <2 μm clay fractions (A) FB1/FA Formation transition; (B) FA Formation. (Ch-chlorite; I/M: illite/mica; Q: quartz; KF: Kfeldspar (microcline); Alb: albite; I/S (R1): illite smectite regular mixed layer).

(Fig. 6f). Although very rare, poorly developed thin hematite grain coatings occurring before quartz overgrowth (Q2) are also observed.

Pyrite occurs as accessory mineral in the drab facies (non-red) and rare to absent in red and creamy facies. It is observed as euhedral crystals or amorphous deposit filling secondary pores (Fig. 6g) and sometimes along grain edges corroding detrital grains and their overgrowths. Occasionally, it occurs in association with matrix chlorite and postdates the chlorite (Fig. 6g).

Illite and chlorite are the only clay minerals observed in the FA Formation rocks. The clay minerals are present as either pore-filling (Figs. 5f, g and 6e, g, h) or detrital grain replacements (Fig. 5d and g). They appear to mostly postdate compaction due to absence of grain coatings clays and/or at grain-to-grain contacts. Illite occurs largely as pore-filling diagenetic cements (Figs. 5g and 6e, h), and also as feldspar and mica replacements. These illites appear to predate other diagenetic cements in the arkosic lithofacies. Chlorite commonly occurs as feldspar and mica replacements (Fig. 5d, g) and also as pore-filling cements (Figs. 5g and 6g, h). EDS analyses and XRD results revealed that the chlorites in drab facies are iron rich while red facies consist of magnesium rich chlorites (Fig. 10b). Volumetrically, illite is more abundant than chlorite in all samples. While illite occurs throughout FA Formation rocks, chlorite is restricted to the middle and upper units (Fig. 4).

Occasionally, authigenic albite, K-feldspar, titanium oxide, titanium-iron oxide, monazites, K and Na chlorides (salts), and titanite are present in trace amounts in some samples.

5.2.2. Pore space

The primary (intergranular) and secondary (intragranular) pore spaces of the arkosic arenites FA sandstones are occluded by authigenic cements (Figs. 5d–g and 6c–h), compaction and pressure dissolution, while the pore spaces of the quartz arenites are filled with quartz cement and minor clay minerals (Figs. 5c and 6a, b). However, minor intragranular (commonly associated with quartz, feldspars, carbonate, sulphate dissolutions) microporosities are sometimes preserved. Primary porosity and permeability in FA sandstone has also been totally destroyed due to cementation and compaction effects.

5.2.3. Clay mineralogy and chemistry

The clay size fractions (<2 μm) in FA Formation consists of illite and chlorite (Fig. 7), while the clay mineral assemblage includes ordered illite–smectite mixed-layer (R1) in FB1 Formation which is the significant clay mineral variation between both formations.

5.2.3.1. Illite.

Identified in XRD patterns by 10 Å, 5 Å, and 3.3 Å peaks, is the major clay mineral phase and present in all the samples. There is no significant change in either the peak position or peak profile of 001 reflection of illites after EG solvation, thus confirming the possible absence of expandable layers in the FA illites. FA Formation illites are predominantly of 1M_t polytype in the middle to upper interval with traces of 2M₁ one at Mabinga and Bangombe, whereas illite from lower intervals and Kiené-Otobo in the central part of

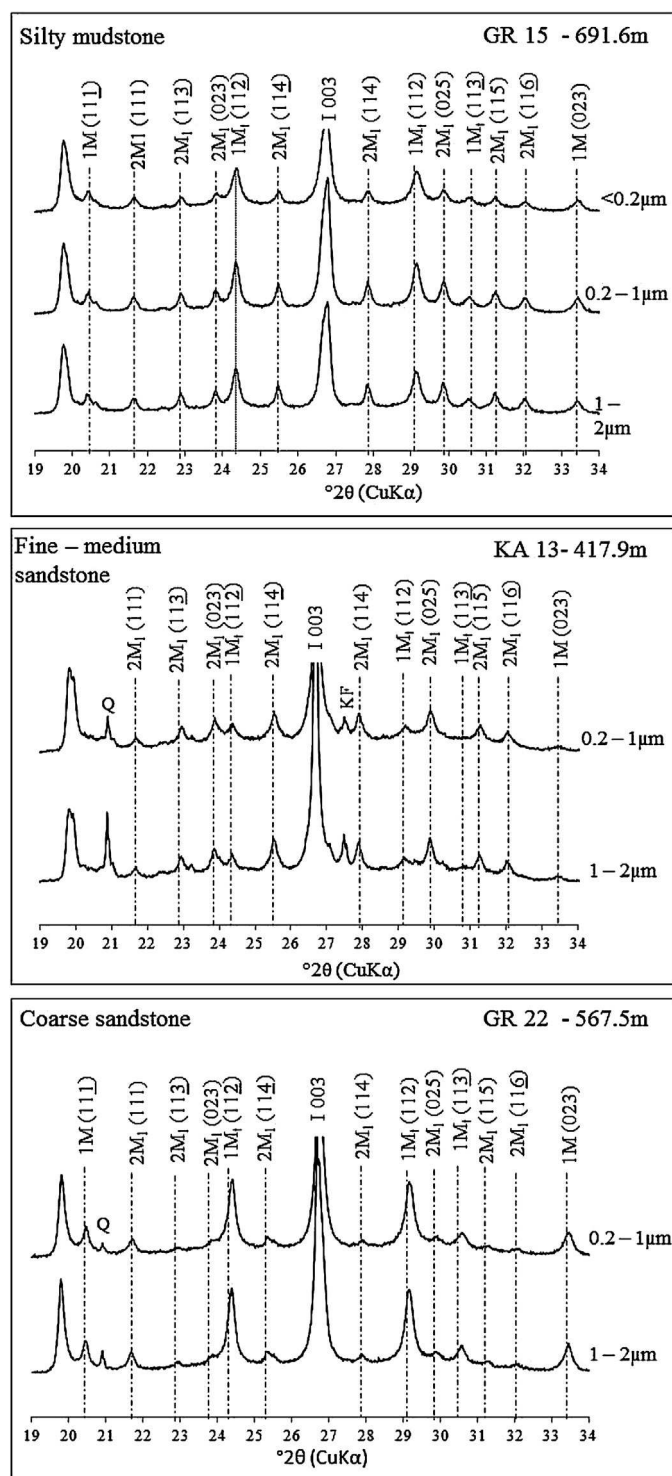


Fig. 8. XRD patterns of random illite powder mounts of various clay-size fractions: illite polytypes (Q: quartz; Ch: chlorite; Alb: albite; KF: K-feldspar).

the basin consists of mixture of both $1M_t$ and $2M_1$ polytypes in varying proportions. However, $1M_t$ polytypes dominates in most instances. The illite polytypes did not show any regular variation in proportion with size fractions and facies type. In some samples, mostly in coarser facies at middle to upper intervals, the amount of $1M_t$ polytype increases relative to $2M_1$ polytype as size fractions decreases, whereas in other samples, there is no significant change in the proportions of $1M_t$ and $2M_1$ polytypes according to change in size fractions (Fig. 8a–c). This observation is also noticed in the fine grained facies and interlayered mudstones. FB1 Formation illites

are predominantly of $1M_t$ polytype. No trace of $1M_c$ polytype was observed in all the samples.

The chemical composition of illite, measured by EDS and calculated semi-quantitatively with all Fe assumed as Fe^{3+} , shows that the illites are aluminium rich with octahedral sum of 2.0 ± 0.07 per 11 oxygen and are, therefore, dioctahedral (Table 1). Fe and Mg are minor while the interlayer charge is compensated by relatively high K^+ and very low Na^+ . The composition of the illites dispersed in the phengite domain and along phengite-celadonite mixing line on the $4Si-M^+-R^{2+}$ chemographic projection (Fig. 10a; Meunier and Velde, 2004).

5.2.3.2. Chlorite.

Chlorite is characterized by 14.2 Å, 7 Å, 4.71 Å, and 3.53 Å peaks and limited in occurrence to middle-upper FA interval within the fluvio-marine facies (~500 m depth at basin centre). The chlorite peaks are sharper and narrow which are unaffected upon ethylene glycolation (Fig. 7). The even-ordered (7 Å and 3.53 Å) peaks are more intense than the odd-ordered (14.2 Å and 4.71 Å) peaks (Fig. 7) indicating presence of Fe-rich chlorite (chamosite) (Brindley and Brown, 1980; Moore and Reynolds, 1997) in drab samples; whereas, chlorites in the red facies are Mg-chlorite (clinochlore) as pointed by stronger odd and even order peak intensities. Heating the samples to 550 °C for 4 h increases the intensity of the 001 peak with slight contraction to about 13.9 Å relative to other peaks which become weakened or disappear as expected for chlorites (Moore and Reynolds, 1997). There is no systematic change in the peak relative intensities with depth except the disappearance of the 14 Å in few samples. XRD patterns of random oriented <2 µm fractions show that the chlorites are characterized by $1b$ ($\beta = 90^\circ$) with probable trace of $1lb$ polytypes (Fig. 9).

Chlorite half unit formulae were calculated based on 14 oxygen equivalents, and all Fe was assumed to be Fe^{2+} . A significant population of the chlorite are mixed with illite as indicated by the $4Si-M^+-R^{2+}$ chemographic projection, and few of the deviations was considered to be a result of contamination from other associated mineral phases (Fig. 10a). As such, only analyses that fall on the “true chlorite” field were used in determining the octahedral cation distribution within the chlorites (Fig. 10b; Table 2). The calculated $Fe/(Fe + Mg)$ ratios increase with burial depth and range between 0.56 and 0.92 in Mabinga FA chlorite, 0.47–0.88 in FB1 Formation, and 0.76–0.89 in FA Formation at Bangombé. At the basin centre, Kiene-Otobo, $Fe/(Fe + Mg)$ range between 0.24 and 0.27 in red facies and 0.61–0.85 in drab facies (Table 2). The very low ratio of chlorite in the red facies could be related to the presence of hematite. The authigenic chlorites all plotted in the trioctahedral half vector representation of chlorite compositions (Fig. 11; Wiewiora and Weiss, 1990). The octahedral occupancy of FA chlorites is close to a fully trioctahedral chlorite in most chlorites except in few samples and some of FB1 chlorites.

There is no significant difference in the chemical composition between the petrographically distinct occurrences of illite and chlorites in all samples as indicated by random cluster distribution on Mg–Fe–Al ternary diagram (Fig. 10b).

5.2.4. Illite–smectite mixed-layer

Illite/smectite (R1) mixed layer is identified only in FB1 Formation and totally absent in FA Formation. It is characterized by asymmetry peaks at 10.63 Å in air-dried which, after ethylene glycolation, splits into two peaks at 11.0–11.77 and 9.6 Å (Fig. 7a). This property is typical of R1 type ordered illite–smectite mixed layer minerals (Reynolds and Hower, 1970). The position of the second peak at 9.6 Å indicates that the illite content is about 70%.

Table 1
Representative structural formulae of illites from the studied area.

BH	Depth (m)	Petrographic occurrence	Si	Al ^{IV}	Al ^{VI}	Al _{Tot}	Mg	Fe ³⁺	K	Na	Ca	Ti	Σ Oct	Int	
Illite per 11 O															
Mabinga (FA)	110 (2)	Replacement	3.17	0.84	1.88	2.72	0.1	0.06	0.82	0.06			2.04	0.87	
	251 (4) ^a		3.35	0.65	1.77	2.43	0.12	0.09	0.88	0.02			1.99	0.9	
	310		3.29	0.71	1.77	2.48	0.17	0.1	0.85	0			2.04	0.85	
	411.1 (2)	Matrix	3.22	0.79	1.76	2.54	0.14	0.1	0.85	0.03		0.03	2.01	0.87	
	447.47 (2)		3.27	0.73	1.74	2.47	0.17	0.13	0.9	0.04			2.03	0.94	
	110 (3)		3.29	0.71	1.86	2.56	0.11	0.05	0.75	0.07			2.02	0.82	
	135.1 (4)		3.25	0.75	1.82	2.56	0.15	0.09	0.81	0		0	2.06	0.81	
	251 (8)		3.26	0.74	1.81	2.54	0.13	0.11	0.83	0.02			2.04	0.85	
	310 (1)		3.3	0.7	1.79	2.49	0.14	0.09	0.82	0.05			2.02	0.87	
	356 (4)		3.27	0.73	1.79	2.52	0.12	0.13	0.84	0.04			2.03	0.88	
	411.1 (7)	Contact	3.22	0.78	1.77	2.55	0.14	0.12	0.87	0.05			2.03	0.93	
	251 (2)		3.24	0.76	1.75	2.52	0.13	0.13	0.92	0.07			2.01	0.98	
Bangombe															
BA 2 (FA)	946.22 (5)		3.07	0.93	1.76	2.69	0.04	0.19	0.84	0.04	0.01	0.02	2.02	0.89	
	940 (3)		3.13	0.87	1.92	2.79	0.07	0.12	0.83	0.03	0.02		2.01	0.88	
	422 (3)		3.22	0.78	1.73	2.52	0.11	0.14	0.84	0.03	0.01	0	1.98	0.88	
	206 (12)		3.25	0.76	1.7	2.45	0.13	0.15	0.84	0.03	0.01		1.98	0.88	
	32 (12)		3.18	0.82	1.55	2.37	0.14	0.23	0.86	0.03	0.01	0.07	1.99	0.9	
FB															
BA 30B	266 (3)		3.21	0.79	1.67	2.46	0.12	0.19	0.84	0.05	0.01	0.01	1.99	0.89	
Kiene-Otobo															
GR 23	28 (4)	Matrix	3.33	0.67	1.75	2.42	0.14	0.09	0.81	0.16	0	0	1.97	0.96	
	180.75 (7)		3.29	0.62	1.62	2.24	0.18	0.25	0.94	0.05			2.06	0.99	
	243.1 (2)		3.25	0.75	1.46	2.21	0.31	0.36	0.9	0.12	0.02		2.14	1.02	
	276.5 (1)		3.19	0.81	1.44	2.25	0.54	0.27	0.8	0.06			2.25	0.86	
	383.3 (5)		3.31	0.69	1.36	2.05	0.29	0.32	0.91	0.01	0	0	1.97	0.92	
	436.05 (3)		3.45	0.55	1.57	2.12	0.29	0.13	0.79	0.04			1.98	0.83	
	761 (1)		3.15	0.85	1.79	2.64	0.29	0.09	0.9	0.06			2.17	0.95	
	863 (1)		3.59	0.41	1.73	2.14	0.29	0.07	0.68	0.06			2.09	0.74	
	900 (1)		2.94	1.06	1.46	2.52	0.29	0.56	0.91				2.31	0.91	
	940 (1)		3.14	0.86	1.93	2.79	0.29	0.04	0.78	0.06			2.26	0.84	
	950 (1)	3.08	0.92	1.87	2.79	0.29	0.1	0.9	0.07			2.26	0.97		
	976 (3)	3.15	0.85	1.82	2.67	0.29	0.14	0.81	0.07	0	0	2.25	0.88		
	993.3 (3)	3.05	0.95	1.88	2.83	0.29	0	0.9	0.07	0	0	2.17	0.97		
	207 (4)	3.3	0.7	1.73	2.43	0.29	0.5	0.8	0.03	0.01	0.01	2.53	0.85		
	446.6 (2)	3.13	0.87	1.8	2.67	0.29	0.19	0.88	0.05	0	0	2.27	0.93		
	GR 15	383.3 (2)	Replacement	3.67	0.73	1.53	2.26	0.32	0.22	0.94	0	0	0	2.07	0.94
		436		3.39	0.61	1.58	2.19	0.4	0.06	0.9				2.04	0.9
		863		2.97	1.03	1.8	2.83	0.09	0.14	0.95	0.06			2.03	1.02
		940		3.07	0.93	1.83	2.76	0.07	0.1	0.91	0.07			2	0.98
950.2		3.08		0.92	1.91	2.83	0.06	0.06	0.92	0.04			2.03	0.96	
976		3.14		0.86	1.9	2.76	0.05	0.08	0.85	0.05	0		2.03	0.9	
386		3.36		0.64	1.59	2.23	0.29	0.2	0.91				2.08	0.91	
436		3.35		0.65	1.74	2.39	0.23	0.07	0.81				2.04	0.81	
950.2 (2)		3.1		0.9	1.64	2.54	0.12	0.32	0.86	0.08	0.04	0	2.12	0.93	
976		3.14		0.86	1.9	2.76	0.05	0.1	0.83	0.06	0		2.05	0.89	
276 (3)	Contact	3.31	0.69	1.49	2.18	0.29	0.25	1	0.03	0	0	2.04	1.02		
383.3 (2)		3.32	0.68	1.56	2.34	0.28	0.18	0.91	0	0	0	2.02	0.91		
940		3.06	0.94	1.84	2.78	0.06	0.11	0.9	0.06			2.01	0.97		
950.2		3.19	0.8	1.92	2.72	0.05	0.04	0.77	0.07			2.01	0.84		
976 (3)		3.13	0.87	0.87	1.74	0.41	0.87	0.82	0.04	0.12	0	2.28	0.86		
993		3.06		1.85	1.85	0.2	0.02	0.94	0	0		2.07	0.94		

^a n = number of samples average.

6. Discussion

6.1. Lithofacies associations and paleoenvironments in FB1 and FA Formation

To better understand the relationships between stratigraphic and lithofacies changes due to basin formation processes in FA Formation, we have integrated our lithofacies subdivisions and petrographic textural observations to the interpreted paleoenvironments of Haubensack (1981) and Gauthier-Lafaye (1986). Both authors identified and subdivided the FA Formation into five sedimentary units: Zone 4 (fluvial), Otobo (deltaic), Zone 3 (tidal), Zone 2 (fluvial), and Zone 1 (shallow marine) based on sedimentary structures and paleocurrent data. Recognition of relationships

between lithofacies and paleoenvironments is important in diagenesis and paleo-hydraulic properties of a basin.

The general successions of LF1 lithofacies display a fining-upward sequence that is commonly pebbly conglomerate at the base and grades stratigraphically upward into medium-coarse grained sandstones in LF1a. Minor thin-bedded mudstone separates fining upward cycles and this reflects an overall upward reduction of flow strengths or energy level in the system. Mineralogical and textural maturity of this lithofacies is considered very poor. This demonstrates that transport distance must have been relatively short due to angularity and poor degree of grain sorting, while the presence of altered to unaltered feldspars can be explained by rapid rate of erosion in relation to weathering (Einsele, 1992; Boggs, 2006; Bjørlykke, 2010) which is a common

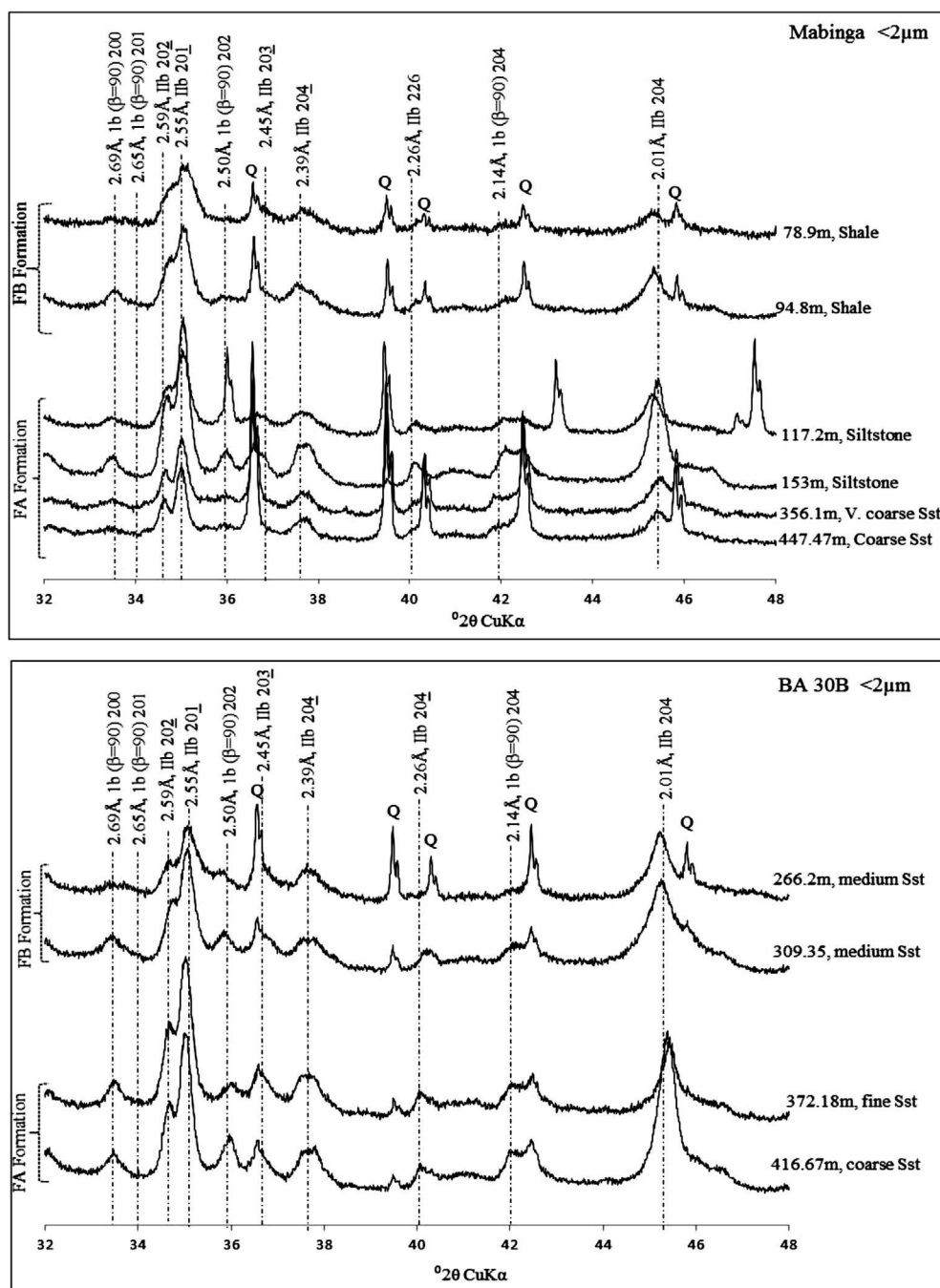


Fig. 9. XRD patterns of random powder mounts (<2 μm clay-size fraction): chlorite polytypes (Q: quartz; Ch: chlorite; Alb: albite; KF: K-feldspar).

occurrence during the Paleoproterozoic due to the absence of plants to aid chemical weathering. The attributes of the lower section coarse grained lithofacies, LF1a, probably corresponds to deposition in high energy proximal braided river deposits and upper section lithofacies, LF1b, in a low energy distal braided stream (Einsle, 1992; Selley, 2000; Miall, 2006).

The coarsening upward of the heterolithic LF2 indicates an upward increase in flow velocities. The medium-coarser and high energy sandy parts of LF2 can be interpreted as distributary mouth bars and the mudstone parts as prodelta part of a deltaic depositional system (Einsle, 1992). The uppermost quartz arenites, LF2b, is considered textural and mineralogical mature due to absence and/or very low feldspar and clay minerals contents, sub-angular to rounded clasts, and moderate to well degree of sorting. These

attributes suggest relatively long transport distance coupled with low relief and intense weathering before deposition, and/or that the sediments have undergone multiple stages of sediment reworking after deposition (Einsle, 1992; Boggs, 2006; Bjørlykke, 2010). LF2b is interpreted to be consistent with sediments deposited in beach and upper shoreface settings, such as shallow marine or lacustrine environment. LF2b₂ unit has been previously interpreted to be tidal bar deposit at Oklo (Deynoux et al., 1993).

The thick bedded cross laminated fine grained sandstones, LF3, with alternating parallel laminated mudstones suggest fluctuating flow velocities in low energy environment (Einsle, 1992; Boggs, 2006; Bjørlykke, 2010). The interbedded laminated medium-coarse grained sandstones indicate periods of increase in depositional energy. This depositional character corresponds to tidal processes

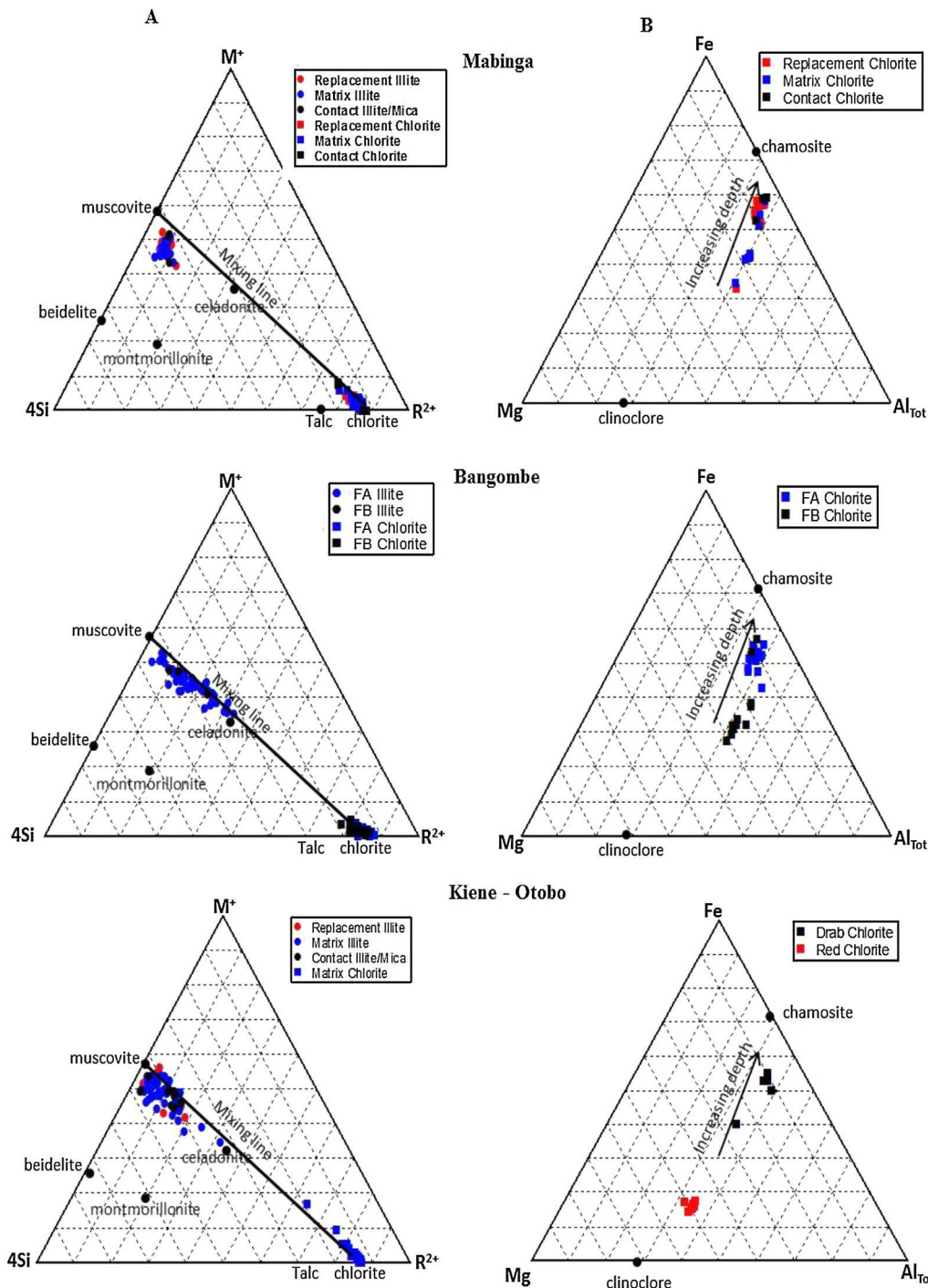


Fig. 10. (A) Chemographic projection of illite and chlorite chemical composition on 4Si-M⁺-R²⁺ ternary plot [4Si = Si⁴⁺/4; M⁺ = K⁺ + Na⁺ + 2Ca²⁺; R²⁺ = Fe²⁺ + Mg²⁺] (Meunier and Velde, 2004); (B) octahedral cationic composition of illite and chlorite projected on Mg-Fe-Al_{Tot} (Velde, 1985).

and lenticular ripple and parallel laminations in the mudstones with cross stratification suggests a subtidal depositional environment (Boggs, 2006).

The thinning of basal LF1a from proximal to distal basin suggests that its composition is likely sourced from the NW which may have been due to tectonic activity during initial stage of the basin developments. This could demonstrate that there was syn-sedimentary faulting during deposition (Azzibrouck-Azziley, 1986; Pambo et al., 2006; Pr  at et al., 2011). These faults appear to have acted as a series of normal faulting along NW to SE direction during deposition of FA Formation. It is reasonable to suggest that it is these faults that

caused the restriction of the tidal (LF3) and lower deltaic facies (LF2a) to the central part of the basin, thus making regional correlation of lithofacies difficult. The regional lithofacies relationships show that the development of Franceville Basin reflects variation in energy levels during deposition.

The observed convolute beddings in FB may reflect post deposition processes caused by liquefaction and lateral-vertical intrastratal flow (dewatering processes) while the load cast structure indicate deposition in response to unstable density contrasts when the sediments becomes liquidized (Selley, 2000; Boggs, 2006).

Table 2
Representative structural formulae of “true chlorites” from the studied area.

BH	Depth (m)	Petrographic occurrence	Si	Al(IV)	Al(VI)	Al _{Tot}	Fe ²⁺	Mg	Ca	Na	K	Fe/(Fe + Mg)	Σ Oct
True Chlorite per 14 O													
Mabinga (FA Formation)	110.6	Replacement	2.68	1.32	1.67	2.99	2.35	1.81	0	0	0	0.56	5.83
	251 (3)		2.7	1.3	1.25	2.55	4.19	0.57	0	0	0	0.88	6.01
	310		2.64	1.36	1.49	2.85	3.74	0.7	0	0	0	0.84	5.93
	356 (4)		2.68	1.32	1.37	2.69	4.01	0.6	0	0	0	0.87	5.97
	411.1 (2)	Matrix/pores	2.68	1.32	1.36	2.68	4.23	0.41	0	0	0	0.91	5.99
	110.6		2.68	1.32	1.58	2.9	2.5	1.79	0	0	0	0.58	5.87
	135.1 (4)		2.72	1.28	1.59	2.87	3	1.26	0	0	0	0.7	5.85
	411.1 (2)		2.61	1.39	1.31	2.7	3.74	0.68	0	0	0	0.85	5.72
	447		2.68	1.32	1.36	2.68	4.26	0.37	0	0	0	0.92	5.99
356 (2)	Contact	2.67	1.34	1.34	2.67	4.1	5.7	0	0	0	0.88	6.00	
Bangombe FB1 Formation													
BA 37	40 (5)	2.74	1.26	1.62	2.88	2.27	1.55	0	0.02	0.01	0.59	5.45	
BA 30B	266 (6)	2.65	1.35	1.47	2.82	2.8	1.5	0.02	0.02	0	0.64	5.76	
FA Formation													
BA 2	32 (2)	2.65	1.35	1.32	2.67	3.73	0.87	0	0.03	0.02	0.81	5.91	
	206 (3)	2.7	1.3	1.35	2.65	3.65	0.95	0	0.02	0.01	0.79	5.95	
	209 (3)	2.75	1.25	1.57	2.82	3.57	0.68	0	0	0.01	0.84	5.83	
	404 (2)	2.78	1.22	1.4	2.62	3.56	0.91	0.01	0.04	0.01	0.8	5.87	
BA 30B	422 (9)	2.68	1.32	1.36	2.68	3.85	0.73	0	0.03	0.01	0.84	5.93	
Kiene Otobo (FA Formation)													
GR 15	276.5	Matrix/pores	2.77	1.23	1.11	2.34	1.12	3.28	0	0	0	0.25	5.51
	383.6 (6)		2.77	1.23	1.09	2.32	1.1	3.33	0	0	0	0.25	5.52
GR 23	446.5		2.81	1.19	1.22	2.4	2.85	1.79	0	0	0	0.61	5.86
	207 (5)		2.74	1.26	1.33	2.58	3.76	0.77	0	0.03	0.01	0.83	5.85

6.2. Mineral paragenesis and diagenetic evolution in FA sandstone

Petrographic relationships reveal that the FA Formation rocks have been subjected to a complex history of diagenetic modifications (Figs. 5 and 6). A simplified sequence of mineral paragenesis was constrained using petrographic textural relationships and features to propose the relative timing of various diagenetic events

in the FA Formation (Fig. 12). The most striking feature is the low compaction and development of quartz overgrowths in the quartz arenites at upper FA Formation, contrasted significant compaction and very low overgrowths in the arkosic to sub-arkosic sandstones. In this study, the spatial grain-to-grain relationships have been used to define the relative timing for authigenic cementation with respect to burial depth (e.g. Bjørlykke and Egeberg, 1993; Hiatt et al., 2007).

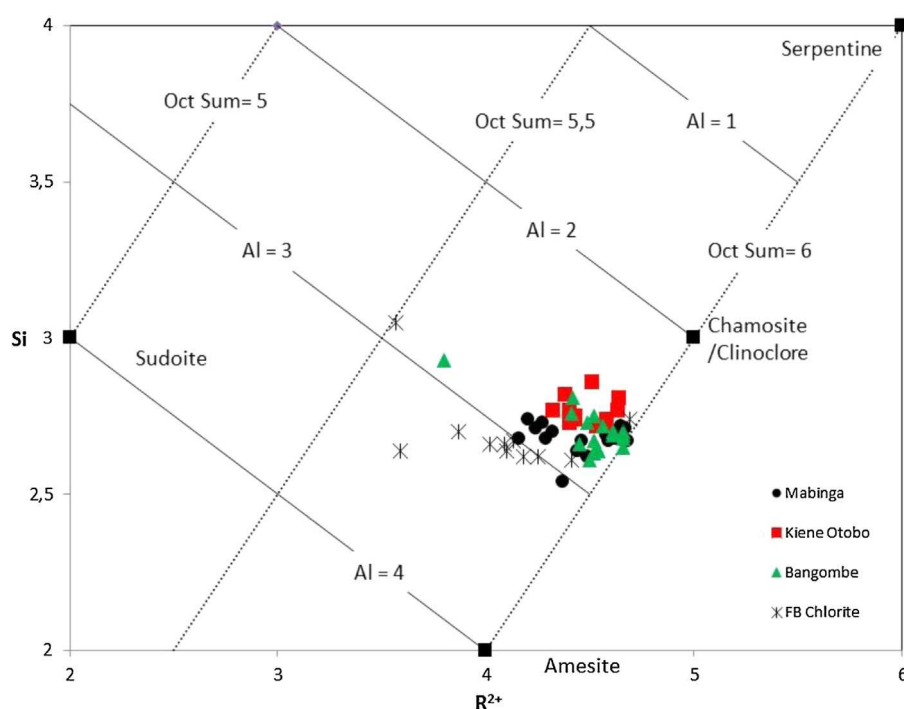


Fig. 11. Structural formula of chlorite from FA Formation and FA/FB1 boundary plotted in the trioctahedral region of the vector representation of chlorite compositions (Wiewiora and Weiss, 1990) with Si and R²⁺ as orthogonal axes and octahedral occupancy and total Al shown by contours.

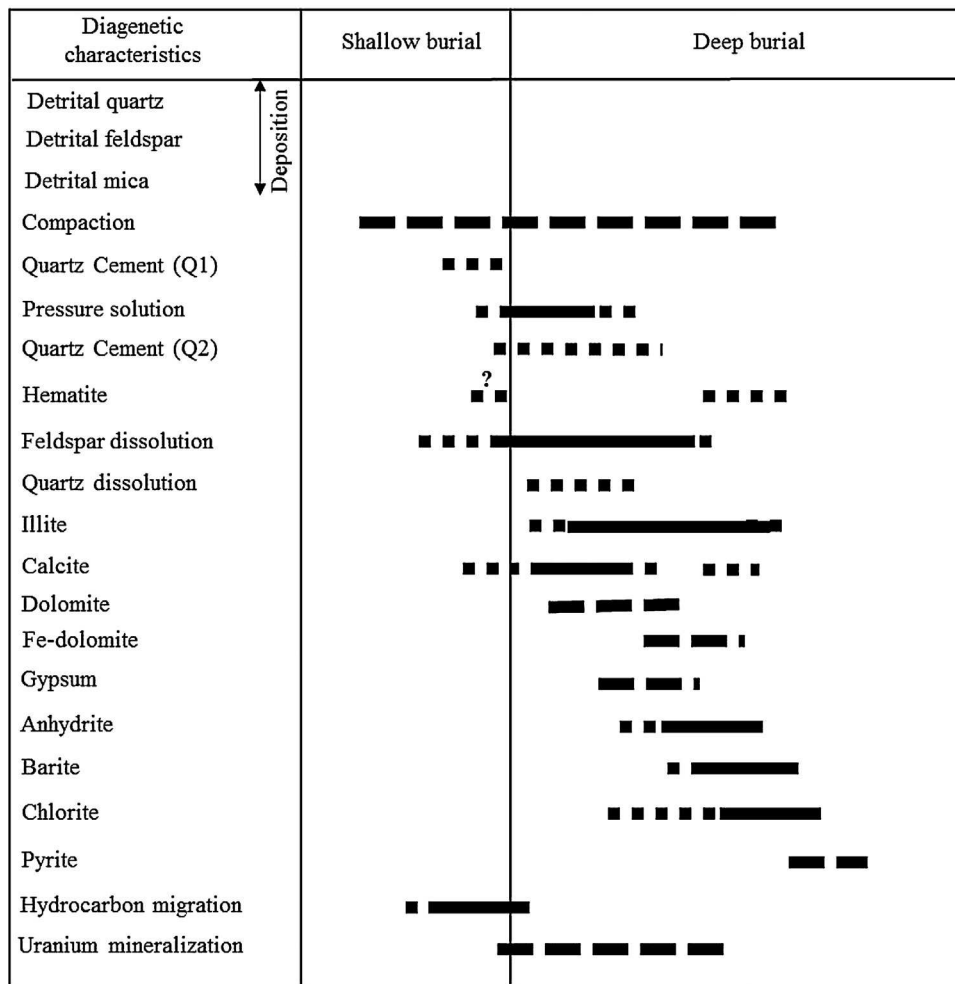


Fig. 12. Proposed paragenesis of main minerals in the FA Formation as interpreted from petrographic textural relationships.

In the quartz arenites, grain-to-grain relationships indicate little compaction features such as convex-concave to long or sutured contacts. This suggests minimal compaction before precipitation of the early quartz cements (Q1) in the quartz arenites (Fig. 5c and 6a, b). Lack of grain coatings clays (mainly illite) in this sandstone could have inhibited pressure solution development during burial. Illite grain coatings are known to facilitate pressure solution (e.g. Boggs, 2009). This presumed early quartz cement is found only in the quartz arenites (LF2) that lacked clay matrix in the upper FA Formation and FA/FB1 transition. Early quartz cementation, coupled with slight compaction, results in a more stable and resistant framework to diagenetic modifications during further burial.

As burial proceeds, compaction driven grain-to-grain pressure solution began at grain contacts and subsequent precipitation of syn-compaction syntaxial quartz overgrowths (Q2) on grains that are clay free in the arkosic lithofacies.

Increasing burial and compaction result in the dissolution and fracturing of unstable detrital silicates (e.g. feldspars) and quartz in the arkosic lithofacies, and subsequent precipitation of pore-filling and replacive illite; thus marking the onset of peak diagenesis. Dissolution and precipitation probably occur in response to change in temperature and pore-water chemistry as burial proceeds (Kyser et al., 2000). Calcite appears to post-date illite or they both precipitated concurrently as they were observed cross-cutting each other in many instances filling the matrix. However, early crystallized calcite was also observed based on floating of detrital grains and

absence of quartz overgrowths (Fig. 5d). Dolomite probably formed by replacement of calcite by migrating fluid rich in Mg. Illitization clearly predates chloritization because chlorites are commonly observed cross-cutting illite within the feldspar cleavages and also within the matrix (Fig. 6h). The authigenic Fe-chlorites could have also precipitated at the expense of illite during late diagenesis, while the Mg-chlorites in the red facies precipitated in equilibrium with hematite. Pyrite occurs mostly in secondary pore space of mainly early precipitated authigenic minerals, thus suggesting that pyrite postdates grain and cement dissolutions. Pyrite, especially those associated with chlorite (Fig. 6g) and quartz overgrowths was probably the last mineral phase to form as a result of change in paleo-redox conditions of the migrating diagenetic fluid from oxidizing to reducing.

6.3. Clay mineral authigenesis in FA Formation and FA/FB1 transition boundary

The main distinct diagenetic difference between the FA and FA/FB transition boundary is the presence of illite/smectite mixed layer clays in the FB1 Formation. Diagenesis in FB1 shales transforms the clay matrix into illite (illitization process) via a sequence of I/S MLMs. The pattern of degree of change in ordering from I/S (R0) to I/S (R1) towards the bottom indicates mineral evolution through illitization of smectite. The detailed modelling of this degree of ordering is already documented by Ossa et al. (2013). These authors inferred that the presence of unaltered K-feldspar in

this lower FB inhibits the completeness of illitization process, coupled with presence of organic matter and low permeability in these fine grain rocks. However, based on the studied samples in this study, it was observed that K-feldspar is totally absent and plagioclase is the only feldspar present in the lower FB1 Formation (Fig. 4). In this manner, the absence of K-feldspar as a possible source of K^+ for illitization, coupled with other factors inhibit the probability of illitization completeness in this organic rich fine grained paleoproterozoic rocks. The absence of K-feldspar in the lower FB1 formation could be dependent on sediment source and not likely that it is being totally destroyed in these unmetamorphosed rocks (Ngombi-Pemba et al., 2014).

The textural occurrences of authigenic illite and chlorite in the FA rocks suggest they are diagenetically formed. In sandstones, illite commonly forms as cement in primary pores (e.g. Ehrenberg and Nadeau, 1989) and its precipitation (illitization) is strongly dependent on temperature (e.g. Eslinger and Glassmann, 1993). Illite in the FA sandstone is more abundant in the pore space without any signs or traces of transformation of previously formed clay matrix. Throughout the FA Formation, XRD results of randomly oriented preparations indicate that $1M_1$ is the dominant illite, indicating that the formation of this polytype is kinetically favoured (Jahren and Aagaard, 1989), and also suggest their precipitation in an environment characterized by high fluid-rock ratio (Meunier and Velde, 2004; Laverret et al., 2006). The occurrence of $2M_1$ polytypes in both fine and coarse size fractions possibly suggests new generations of illite or could be regarded as a result of higher diagenetic stage and/or contributions from detrital micas which are present throughout the formation. This means that the last illite particle to form ($2M_1$) have crystallized at higher temperature conditions or influence by time considering the age of the rocks (Lonker and Fitz Gerald, 1990; Meunier and Velde, 2004).

The chemical homogeneity of the chemistry of illites in petrographic distinct generations and from sample to sample reflects similar regional formation conditions in a near equilibrium condition with the pore fluid (Hillier and Velde, 1991; Jahren and Aagaard, 1989; Warren and Curtis, 1989). This could also indicate that the chemical composition of the fluid did not change much during the interval of illite and chlorite formation (Kyser et al., 2000). However, the most striking minor difference between the petrographic distinct illite generations in a sample suite is the interlayer charge. The interlayer charge, predominantly K, is always higher in replacement formed illite (0.85–0.94 a.p.f.u) than pore-filling illite (0.7–0.9 a.p.f.u). This difference outlines the “microsystem” effect (Meunier, 2005). Indeed, micas being potassium sources favour the high charge illite formation. The only considerable stratigraphic variation is the increase in Mg and Fe with depth due to replacement of octahedral Al by these cations.

Chlorite is detected only in the FB1 and to middle interval of FA and absent in the lower fluvial FA. Chemical analysis data indicate there is no much difference in the chemical composition of petrographic distinct chlorites (chloritized mica/feldspar and matrix chlorites) but the Fe/Fe + Mg ratio increases with burial depth which is consistent with diagenetic formed chlorites in sedimentary basins (Curtis et al., 1985; Jahren and Aagaard, 1992). The very low Fe content in the red facies samples (Fig. 10b) in which hematite coexists with Mg-chlorite probably suggests precipitation of both mineral phases in equilibrium where there is competition for Fe ions. Fe ions prefer by far to integrate the oxide forms during oxidation and would not leave much Fe^{2+} available in the pore water for chlorite formation. Moreover, the co-existence, similar compositional trend, and observed mixing lines between illite and chlorite suggest that both minerals crystallized from near equilibrium solid-solution. The near full octahedral occupancy of the chlorites, which lie close to amesite and serpentine line (Fig. 11), coupled with the likely presence of $11b$ chlorite polytype, in a

sedimentary basin suggest their precipitation via a high temperature condition (Hillier and Velde, 1991). Ossa et al. (2014) concluded that FA chlorites were formed from migrating hydrothermal fluids.

The diagenetic illite and chlorite in the early cemented lithofacies has the lowest incorporated Fe and Mg in their crystals relative to those from other lithofacies that are stratigraphically below and more permeable during peak diagenesis. This low values of Fe and Mg in the illite and chlorite suggests precipitation from very low permeable sandstones and lack of ferromagnesian minerals in this lithofacies (Kyser et al., 2000).

The type of clay mineral diagenesis in sandstone is observed to be relatively similar to that in mudstone in FA Formation. This would further suggest that they are possibly affected by similar diagenetic processes, requiring same fluid-rock interactions to have affected both lithofacies. This could be indicative of internally sourced components for diagenetic reactions in the rock types with little influence of cross formation fluid flow on diagenesis (Huggett, 1996). Another possible explanation could be that mudstones were chemically open as the sandstones and thus underwent same diagenetic reaction simultaneously (Shaw and Conybeare, 2003).

Although, the precursor of the matrix illite in FA Formation is difficult to determine, this illite do not appear to have crystallized from mixed layer clays, and it appears that the little shift or expendability in some of the illite samples (Fig. 7) is an intrinsic property of the illite particles (Warren and Curtis, 1989). Also, the illites are aluminium rich which may reflect their formation from dissolution of kaolinite (Meunier and Velde, 2004). One possible assumption for the absence of kaolinite is that all the kaolinites could have been converted to illite due to intense illitization. However, kaolinite most often formed in dissolved K-feldspar and this was never observed in this study. This indicates that kaolinite was perhaps never formed in this old system, and this is possible if K^+ ion (supplied by K-feldspar and sea water) concentrations are too high in the diagenetic fluids. As such, illite and chlorites in FA Formation are likely to have formed as a result of dissolution and precipitation in a homogenous chemical system, facilitated by fluid migration and paleo-textural properties that is largely controlled by composition of the rocks. The absence of chlorite in the lower fluvial facies suggests that illite and chlorite do not likely precipitate together. Petrographic observations suggest that illite clearly precipitated before chlorite.

6.4. Controls on diagenesis and implications on fluid flow in FA Formation

Early diagenesis of sandstones is dependent on sediment texture, detrital mineralogy, organic content, and initial pore water chemistry which are all largely dependent on sediment source and depositional environment (Tucker, 1991; Worden and Burley, 2003). Burial depth, pore water chemistry, nature of fluid, and pore-fluid flow are the major important factors that control diagenetic evolution during burial (Tucker, 1991; Worden and Burley, 2003). It must be noted that in virtually all instances, the present day hydrologic properties (porosity and permeability) are opposite to those that would be predicted based on lithofacies characteristics as a result of diagenetic overprints on the textural properties. Like other continental Proterozoic basins (e.g. Thelon, Athabasca, Kombolgie), the FA Formation of Franceville Basin differs from those of Phanerozoic because of near absence of muddy facies during Proterozoic. As a result, cementation and compaction play important roles in the hydrologic properties of these basins relative to Phanerozoic basins.

Depositional facies appear to have the greatest influence on the diagenesis and fluid flow in FA Formation sandstones. The significant correlation between early quartz cement in the quartz

dominated arenites and subsequent diagenetic processes is related to destruction of primary porosity and permeability before significant compaction in the compositionally matured lithofacies that is poor in feldspar and/or clay matrix (LF2b). During transport and early burial, strong groundwater flow would have removed any unstable detrital minerals leaving the quartz dominated sandstone with minor feldspar. These lithofacies were thought to be initially porous when deposited, but due to pervasive quartz cementation and slight compaction became a diagenetic aquitard and significantly inhibit further fluid migrations during peak diagenesis (Hiatt et al., 2003; Beyer et al., 2011). These lithofacies have fewer deep burial diagenetic illite and chlorite in their pore spaces. This indicates that the pathways for fluid flow were not completely eliminated during early diagenesis, suggesting clay mineral precipitation during later fluid circulation. Although, most quartz cements probably formed at temperature greater than 60 °C and burial depth greater than 2 km (e.g. Bjørlykke and Egeberg, 1993); but this process of early quartz cementation at near shallow burial depths (55 °C, 1–1.5 km) has also been reported on a basinal scale in several Proterozoic basins such as Thelon Basin (Renac et al., 2002; Hiatt et al., 2003; Beyer et al., 2011), Athabasca, and Kombolgie Basins (Kyser et al., 2000).

Mathieu et al. (2000) reported that the extensive quartz cements in upper FA sandstones near the natural reactor zone at Oklo (about 30 km from the studied area) formed in low saline (2.2–8.5 wt% NaCl equivalent) aqueous meteoric fluid at 190–210 °C from fluid inclusion studies. This fluid is thought to have migrated upward along major N-S faults after being heated in the basement. According to Gauthier-Lafaye (1986) and Gauthier-Lafaye and Weber (1989) the trapped fluid of the precipitated quartz cements in the silicified sandstones have been modified by hydrofracturing during late diagenesis and they reported a temperature range of 140–180 °C. By comparison, we can envisaged that the early precipitated quartz cements (Q1) in the quartz arenites, probably of meteoric origin could have precipitated at very much lower temperature considering the burial depth of formation and the distance from the nuclear reactor. It has been estimated that the thermal perturbation around the nuclear reactors do not exceed more than 50 m from the centre of the reactor zone (Gauthier-Lafaye et al., 1996).

In contrast, the texturally immature to sub-mature lithofacies (LF1, LF2a, LF3) would have low to moderate porosity and permeability during deposition due to poor degree of sorting. These lithofacies lack extensive quartz cementation and this is probably due to presence of clays during deposition. Thus, avenues for fluid migration were present during burial and may have facilitated the dissolution of chemically unstable detrital grains within the arkosic sandstone during peak diagenesis, which in turns permits precipitation of illite and other cements within the matrix and secondary porosities. These lithofacies are the only route for subsequent fluid circulation acting as diagenetic aquifers. The precipitated authigenic pore-filling mineral cements, especially pore-filling illite due to intense illitization, resulted in significant reduction and/or closure of porosity and permeability as peak diagenesis progressed transforming them to diagenetic aquicludes (Beyer et al., 2011). The correlation between illite/sericite and very low syntaxial quartz overgrowths within the arkosic sandstone indicate that dissolution and alteration of feldspar, biotite, and lithic framework grains likely provided the necessary chemical components for the formation of authigenic minerals within the matrix indicating the occurrence of water-rock interactions during peak diagenesis.

6.5. Timing of uranium mineralization in FA Formation

In the Franceville Basin, uranium mineralization by upward migration of oxidized saline uranium rich fluid from lower fluvial conglomerate and precipitation in upper part due to change

in redox condition has been postulated (Gauthier-Lafaye, 1986; Gauthier-Lafaye and Weber, 1989; Gauthier-Lafaye and Weber, 2003; Mathieu, 1999). The timing, development, and potential preservation of porosity within the studied sandstones provide an important framework for mineralization and fluid circulation considerations.

According to Gauthier-Lafaye (1986) and Gauthier-Lafaye and Weber (1989), uranium mineralization in upper FA Formation probably coincided with migration of hydrocarbon and associated reducing fluids from already overpressured FB black shale. The mineralization was probably triggered by re-activation of extensional tectonic faults which is accompanied by hydraulic fracturing in FA sandstones. The fractured and faulted sandstones allowed the mixing of hydrocarbon with associated reducing fluids and the oxidized uranium bearing fluids in FA sediments. Therefore, uranium were thought to precipitate along hydraulic fractures and secondary pores in the FA sandstones when these migrating fluids met. The uranium is believed to be sourced by detrital heavy minerals (thorite and monazite) from the conglomerate in the lower FA Formation.

The solidified hydrocarbon (bitumen) found in the primary porosity does not contain uranium while those present in secondary porosity have high uranium content (Gauthier-Lafaye, 1986, 2006). The loss of primary porosity and permeability in the early cemented mineralized sandstones and absence of uranium in the organic matter at grain-to-grain contacts and primary pore suggests uranium mineralization occurred after the initial stages of quartz cementation in the quartz arenites. This observation demonstrates the timing of this early quartz cement and occurrence of bitumen rich uranium. It suggests that the diagenetic aquitards were developed in the FA Formation before the earliest uranium mineralization event based on paragenetic relationships.

7. Conclusions

The Paleoproterozoic FA Formation in the Franceville Basin contains three different lithofacies and few sub-lithofacies assemblages that were grouped into three facies associations representing deposition in braided fluvial, tidal, and deltaic environments. The lower parts of overlying FB1 represent deposition in marine environment. The significant difference in the mineralogical composition between FA and FA/FB1 transition boundary is the lack of K-feldspar in the FB1 and absence of diagenetic illite/smectite mixed layer in the FA Formation.

Mineral composition and petrographic textural relationships of the FA sandstone show that it has undergone multiple stages of diagenetic histories and original properties overprinted. The diagenetic transformation of minerals and fluid flow in FA sandstone occurred in a system controlled by depositional processes and paleohydrologic properties of the lithofacies. The change in hydrologic properties recognized in FA sandstones represents similar changes in Proterozoic sandstones such as Thelon, Athabasca, and McArthur basin with some mineralogical differences. The texturally and mineralogical matured quartz dominated deltaic lithofacies in uppermost part of FA Formation has pore-filling early quartz cementation, which thus serves as diagenetic aquitard for subsequent fluid migration and inhibit further diagenetic alteration processes during burial. In contrast, the other textural and composition immature lithofacies that is deficient in early quartz cementation act as diagenetic aquifers for fluid flow during peak diagenesis.

The clay mineralogy of FA Formation sandstone supports the conclusion that water-rock interaction and paleohydrologic properties have influenced the diagenesis of FA sandstones. The dominance of 1M₁ illite polytypes in the sandstones favours

precipitation in an environment characterized by high fluid-rock ratio. The chemical homogeneity in the petrographic distinct generations of illites and chlorites demonstrates that the composition of the migrating fluid was constant during diagenesis.

Acknowledgements

The authors wish to acknowledge CNRS-INSU, FEDER, the University of Poitiers, Région Poitou-Charente for financial support. Gabon Ministry of Education and Research; CENAREST; Gabon Ministry of Mines, Oil, Energy and Hydraulic Resources; General Direction of Mines and Geology; Agence Nationale des Parcs Nationaux of Gabon; COMILOG; French Embassy at Libreville for collaboration and technical support. For laboratory and other assistance, we acknowledge F. Weber; L. Ngombi-Pemba; I. Moubiya-Mouélé; N. Onanga-Mavotchy; C. Fontaine; C. Laforest; and N. Dauger.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.precamres.2015.07.008>

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