Palygorskite-bearing fracture fills in the Kinshasa area, DR Congo – an exceptional mode of palygorskite vein development

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Abstract

Tectonic fractures in Palaeozoic strata of the Kinshasa area, DR Congo, locally host palygorskite-bearing veins and associated calcite occurrences. The palygorskite deposits are typically massive, with a varying degree of alignment of clay particles, a higher quartz content than the arkose substrate, and a variable amount of smectite (montmorillonite). The associated calcite occurrences are macrocrystalline coatings and infillings, and more fine-grained calcite veins with cataclastic texture. The calcite coatings and infillings formed from solution in earth surface conditions, as recorded by their stable isotope signature. The palygorskite-dominated deposits in the fractures formed at a later stage, in a setting without indications of authigenic mineral formation related to hydrothermal activity or to low-temperature interaction of solutions with the local substrate. The veins most likely formed by vertical infiltration of suspended matter in fractures that extended to a post-Palaeozoic palaeosurface, during or after deposition of palygorskite-bearing Upper Jurassic to Early Cretaceous sediments. This represents an exceptional mode of palygorskite vein development, unrelated to any form of mineral authigenesis that is typically invoked to explain vein-type occurrences of palygorskite and related minerals.

Introduction

Palygorskite ($(Mg,Al)_2Si_4O_{10}(OH).4H_2O$) is a clay mineral that forms in various geological contexts, including hydrothermal, sedimentary and soil environments (e.g. Singer and Galàn, 1984; Galàn and Singer, 2011). Palygorskite veins represent one typical mode of occurrence, which has been reported for various localities worldwide. Most vein-like occurrences are interpreted as hydrothermal deposits (Caillère, 1951; Stephen, 1954; Antun, 1956; Minato et al., 1969; Nathan et al., 1970; van der Wel, 1972; Haji-Vassiliou and Puffer, 1975; Furbish and Sando, 1976; Gibbs et al., 1993; García-Romero et al., 2006; Wang et al., 2006; Giustetto and Compagnoni, 2011), restricted to a low temperature range (see e.g., Golden and Dixon, 1990). Relatively few occurrences are considered to be non-hydrothermal (Evans and King, 1962; Martini, 1964; Peters and von Salis, 1965; Vernet, 1967; Müller-Vonmoos and Schindler, 1973; Tien, 1973; Imai and Otsuka, 1984; Soong, 1992; Kamineni et al., 1993; Sakamoto et al., 2006; Cantarero et al., 2014), whereby the most commonly invoked process involves Mg- and Si-enrichment of meteoric water or groundwater by interaction with the rock substrate along the fracture in which the vein deposits occur. Many other known occurrences of palygorskite veins are poorly documented and of unknown origin (see e.g. Cavallo, 1989).

The occurrence of palygorskite veins in the Kinshasa area, Democratic Republic of the Congo (DR Congo), was first mentioned by Bartholomé (1966), referring to observations by A. Egoroff of the local Geological Survey. Around the same time, their nature and origin was the subject of an unpublished short note by P. Antun (1966). According to Antun (1966), palygorskite in the Kinshasa area occurs as part of breccious infillings of northeast-oriented subvertical joints. In these joints, angular fragments of the arkose in which the fractures developed are embedded in a sandstone matrix whose composition is similar to that of a cover deposit that locally overlies the arkosic substrate. Palygorskite, with associated calcite and montmorillonite, is described by Antun (1966) as occurring in the form of veins within these infillings. Based on these observations, the infillings as a whole are considered to have formed by infiltration of sediments in open joints, and palygorskite is implied to have formed from descending solutions within these structures, whereby solution composition is influenced by smectite alteration (Antun, 1966). Since 1966, no further information has become available about the palygorskite-bearing veins of Kinshasa.

The present study is the outcome of a re-investigation of the nature and significance of these palygorskite occurrences, conducted as part of a wider study of the post-Proterozoic geological evolution of the Kinshasa area. It reports an unusual mode of development of palygorskite-dominated veins, that might be applicable to other clay mineral occurrences as part of vein-like structures.

Geological context

Palygorskite-bearing veins of the Kinshasa area occur in arkosic rocks of the Inkisi Subgroup (Frimmel et al., 2006) (Figure 1). The depositional age of this formation is currently interpreted as Palaeozoic but older than Permian (Frimmel et al., 2006; Tack et al., 2008). This ~1 000 m thick redbed sequence was deposited in semiarid lacustrine and fluvio-deltaic palaeoenvironments. The beds dip gently to the east, and they are crossed by prominent sets of subvertical northeast- and northwest-trending tectonic fractures that formed in a strike-slip context (Miyouna et al., 2018). Inkisi Subgroup deposits overlie siliciclastic rocks of the late Poterozoic Mpioka Subgroup, which overlie calcareous deposits of the Schisto-Calcaire Subgroup (Frimmel et al., 2006).

In the eastern part of the Kinshasa region, at the western edge of the Congo Basin, the Palaeozoic basement that hosts the palygorskite-bearing veins is covered by poorly consolidated to unconsolidated siliciclastic deposits, mainly of undifferentiated Cretaceous age (Figure 1), overlain by Cenozoic strata (Ladmirant, 1964). Locally, the post-Palaeozoic cover includes a ~20 m thick basal sequence of sandstone, mudstone and marl that is correlated with the lower part of the Stanleyville Group (Upper Jurassic to Early Cretaceous), as it appears in its distant type locality near Kisangani, in the northeastern part of the DR Congo (Caillaud et al., 2017). This sequence is encountered in boreholes in the Ndolo district of central Kinshasa (Egoroff and Lombard, 1962), for which the initial identification of Stanleyville Group beds was later confirmed by palaeontological studies (Defrétin-Lefranc, 1967; Cahen, 1983). The same deposits have also been reached by coring in the harbour area of Brazzaville (Nicolini and Roger, 1951; Ladmirant, 1964), across the Congo River from Kinshasa. At Ndolo, it rests unconformably on weathered Inkisi arkose, which shows an irregular surface.

Materials and methods

A field survey was conducted in parts of the Kinshasa area where the Inkisi Subgroup appears in outcrop, both along the Congo River and along the Lukaya and Ndjili valleys (Figures 1 and 2). This survey included the inspection of subhorizontal surfaces exposed along the Congo River towards the end of the dry season, between the Safricas quarry and Ile des Mimosas (Figure 1). Active quarries, presenting vertical exposures, were visited in the same area and at some more southern sites (Satraco quarry in Kinsuka, SGI quarry in Kasangulu; outside the area covered by Figure 1). Structural data were gathered for the brittle structures hosting the palygorskite veins. Samples were collected of all potential palygorskite occurrences and associated features, as well as of the local arkose substrate. Some specimens belonging to the historical collections of the Royal Museum for Central Africa (RMCA) were also analyzed, including two palygorskite samples donated by P. Antun (RGM 11281, 11708) and core samples of Stanleyville Group deposits of the Ndolo district.

Thin sections were prepared for most samples, following impregnation with a polyester resin. X-ray diffraction (XRD) analyses (Philips PW105/37, CuK_α radiation) were performed

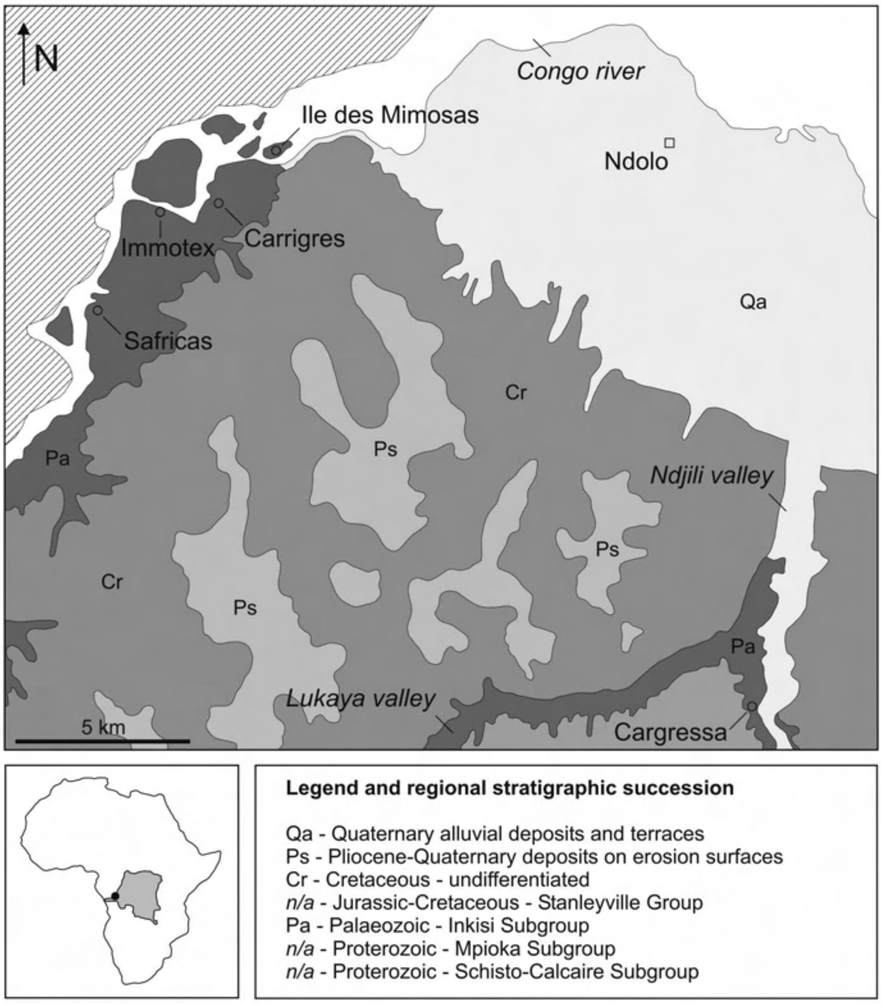

Figure 1. *Location of palygorskite vein occurrences in the Kinshasa area, with city centre in the northern part of the zone covered by this map. Geological base map after Service Géologique de la République du Congo (1963). Hatched area – Brazzaville, Republic of the Congo. Units marked as n/a do not appear in outcrop within in the area covered by the map.*

for selected samples of the veins, the enclosing arkose, and the local Stanleyville Group deposits. Non-oriented powders of bulk material were analyzed after the addition of 10% ZnO as internal standard and ten minutes of wet milling using a McCrone micronizing mill (after Srodon et al., 2001). For selected samples, oriented slides of the clay fraction (<2 µm) were analyzed, following Ca saturation, Ca saturation with glycol treatment, and Li saturation followed by heating and glycol treatment. Quantification was performed using the Rietveld-based BGMN program (Bergmann et al., 1998). For bulk samples, dioctahedral 2:1 clays such as illite, muscovite, smectite and illite-smectite were quantified as a single '2:1 Al-clays' group, because their quantification as individual species is difficult for bulk powder diffraction data, but a semi-quantitative estimation of the relative amounts of smectite and illite/mica was made based on results for the oriented slides. Scanning electron microscopy (SEM) analysis was performed with a JEOL JSM-6480LV system, using gold-coated samples. Stable isotope data ($\delta^{18}O_{V\text{-}SMOW}$, $\delta^{13}C_{V\text{-}PDB}$) were obtained for selected calcite occurrences, involving reaction with 100 % H_3PO_4 at 25°C and measurement with a Finnegan MAT 251 dual inlet mass spectrometer; the experimental error is 0.1 $^{o}/_{oo}$ for both oxygen and carbon.

Results

Macroscopic features

Five main types of palygorskite and associated calcite occurrences are recognized in the field: macroscopically fibrous palygorskite, massive palygorskite, palygorskite coatings, fine-granular calcite, and macrocrystalline calcite. The fibrous palygorskite deposits are white, non-friable, with low density. The massive palygorskite is equally a low-density, non-friable deposit, but with a white to purple colour and a recognizable sand admixture. The palygorskite coatings are thin (1 mm) white layers covering various surfaces. Among calcite types, the fine-granular type is white, friable, and massive, whereas macrocrystalline calcite refers to coarse-crystalline coatings and infillings.

Among palygorskite types, massive palygorskite (see Figure 2E) is much more common than its fibrous 'mountain leather' equivalent in the study area. Palygorskite coatings occur along planes of weakness associated with the main palygorskite and calcite occurrences, both within and along the sides of those features and along cracks extending from the main fractures, including subhorizontal planes. Massive palygorskite corresponds to what Antun (1966) described as the sandstone matrix of the veins, and pure palygorskite deposits ranging from thin coatings to thick fibrous palygorskite occurrences are what he considered to be palygorskite veins within the fracture infillings. In the present report, the entire fracture infilling is referred to as vein material.

Macrocrystalline calcite coatings along the sides of open fractures are typically composed of scalenohedral crystals with euhedral terminations. As infillings (see Figure 2D), macrocrystalline calcite has a xenotopic texture and typically clearly consist of coatings that formed along opposite sides of a fracture that is now closed. A thin layer of whitish palygorskite commonly lines the contact between both coatings. The calcite coatings are typically ~1 cm thick, resulting in infillings of up to 3 cm thickness, but one much thicker occurrence has been observed (6 cm).

All types of vein material commonly contain incorporated angular fragments of the arkose substrate. At some sites, massive palygorskite contains plate-shaped fragments of macrocrystalline calcite coatings or infillings.

The occurrence of palygorskite and calcite is largely confined to fractures with a specific orientation (see further). Thin palygorskite veins (<1 mm) occurring in fractures with other orientations, locally continuing over distances of several metres, always seem to be associated with thicker veins that are part of the set representing the dominant orientation. These thin veins are considered to belong to the category of palygorskite coatings.

Vertical variations in type of fill were observed for two veins that could be accessed at levels ~25 m apart in elevation, in the Carrigres quarry. One of these veins is composed of fine-granular calcite at the base and massive palygorskite at the top, roughly corresponding to what Antun (1966) described as a downward decrease in sand content. The other vein, 15 m from the first, consists of massive palygorskite at the base and macrocrystalline calcite at the top.

Extent of palygorskite-calcite vein occurrences and nature of the fractures

Antun (1966) observed and sampled palygorskite veins in two arkose quarries in the Kinsuka district of western Kinshasa – the Ile des Mimosas quarry, and the former Carri-Léo quarry (now Carrigres) (see Figure 1). The survey conducted for the present study demonstrates the occurrence of palygorskite over a wider area along the Congo River, but with the most prominent, thickest veins in the area identified by Antun (Carrigres quarry, Immotex site). Rare thin veins observed in the Safricas quarry (Figure 1) and the SGI quarry (25 km S of Kinshasa; 4°34'48" N, 15°11'46" E) represent the most southern confirmed occurrences. The survey also revealed the presence of palygorskite and macrocrystalline calcite veins near the Lukaya-Ndjili confluence (Cargressa quarry), ~20 km from Kinsuka (Figure 1).

Based on measurements for a total of 137 subvertical fractures containing palygorskite or calcite, their dominant orientation is found to be northeast-southwest, with a maximum frequency at N55-235°E; a subsidiary set is oriented north-northwest-south-southeast, with a maximum frequency at N160-340°E (Figure 3). Among northeast-southwest fractures, three types are distinguished:

- plume joints, i.e. extensional fractures without visible displacement, containing no calcite or palygorskite;
- extension fractures, showing no lateral displacement, which can contain macrocrystalline calcite coatings/infillings (see Figure 2D); and
- large fracture systems showing evidence of lateral movement, containing, in varying relative amounts, fragmented macrocrystalline calcite coatings/infillings, microcrystalline calcite, palygorskite, and host rocks fragments (see Figure 2C).

The fill of the fractures of this third type, which can extend laterally over tens or hundreds of meters, has a fault breccia aspect is some parts and it can include palygorskite coatings along shear planes. Left-lateral movement along this large fracture system is indicated by slightly displaced pebble fragments. The same subhorizontal displacement is also recognized for striated and stepped surfaces along the sides of palygorskite and calcite veins. In high vertical exposures, such as those of the Carrigres quarry (see Figure 2A), no vertical shifts are recognized for the subhorizontal bedding planes of the arkosic host rock along the fractures.

Mineralogical composition

All analyzed samples of massive and macroscopically fibrous vein material consist of palygorskite, with varying amounts of smectite and traces of mica and kaolinite (Tables 1 and 2). The smectite was identified as dioctahedral based on d_{060} values obtained for non-oriented samples, and as montmorillonite based on d_{010} values after Li saturation followed by heating and glycolation. White vein material typically has a high relative palygorskite content, whereas occurrences with a purplish colour and an important sand admixture generally include larger amounts of smectite. Palygorskite in all samples shows swelling behaviour following glycol treatment, affecting mainly d_{h00} values rather than the diagnostic d_{110} value (Figure 4). Based on Rietveld structure refinement, palygorskite in all samples shows a combination of orthorhombic and monoclinic forms (*cf.* Artioli and Galli, 1994; Giustetto and Chiari, 2004), in roughly equal

Figure 2. *Field appearance of palygorskite-bearing veins, in arkosic Inkisi Subgroup deposits. (**A**) Quarry wall showing two parallel long vertical veins, recognized mainly as white material on rock surface (Carrigres quarry). (**B**) Closer view of vein deposit in vertical exposure (Carrigres quarry). (**C**) Vein in horizontal exposure along the Congo River, as part of a northeast-trending left-lateral strike-slip fracture system, including a lens with abundant fragments of macrocrystalline calcite infillings (Kinsuka area). (**D**) Macrocrystalline calcite in extension fractures without lateral displacement, in horizontal exposure (Kinsuka area). (**E**) Hand specimen of palygorskite vein, composed mainly of pinkish impure massive palygorskite, with enclosed arkosic rock fragments, and with white palygorskite coating along the sides (RGM 11708, Carrigres quarry).*

Table 1. Mineralogical composition of bulk samples of selected specimens.

Type	Site	Sample n° *	Pal	Smec**	Mica**	Chl	Kaol	Qz	Ca	Kf	Pl	Hm
Massive palygorskite, white	Carrigres	RGM 16856	27	*18*	*7*	1	-	39	-	8	-	-
Massive palygorskite, white	Immotex	RGM 16862	49	*2*	*4*	-	-	31	-	14	-	-
Massive palygorskite, reddish	Carrigres	RGM 16854	15	*16*	*9*	3	1	34	3	17	-	2
Massive palygorskite, reddish	Immotex	RGM 16860	34	*4*	*5*	1	-	41	-	14	-	1
Fibrous palygorskite	Ile des Mimosas	RGM 11281	29	*20*	*5*	-	1	36	1	7	1	-
Fibrous palygorskite	Carrigres	RGM 11708	50	*9*	*3*	-	2	10	24	2	-	-
Palygorskite coating	Carrigres	RGM 16854	65	*2*	*1*	1	1	17	3	9	-	1
Palygorskite coating	Cargressa	RGM 17056	64	*13*	*4*	-	-	13	6	-	-	-
Palygorskite coating	Safricas	RGM 17057	70	*21*	*4*	-	2	3	-	-	-	-
Palygorskite coating	SGI	RGM 17058	98	-	-	-	-	2	-	-	-	-
Fine-granular calcite	Carrigres	RGM 16836	-	*4*	*3*	-	-	1	91	1	-	-
Fine-granular calcite	Carrigres	RGM 16842	2	*7*	*1*	-	-	1	89	-	-	-
Fine-granular calcite	Carrigres	RGM 16843	4	*1*	*2*	-	-	1	92	-	-	-
Fine-granular calcite	Carrigres	RGM 16844	3	*5*	*1*	-	-	2	85	4	-	-
Arkose	Carrigres	RGM 16839	-	*7*	*3*	1	1	65	-	21	1	1
Arkose	Carrigres	RGM 16856	-	*1*	*4*	2	1	61	3	27	-	1
Arkose	Immotex	RGM 16861	-	*6*	*3*	-	-	74	-	16	-	1
Stanleyville Formation	Ndolo	RGP 12219	-	*45*	*25*	-	2	24	-	3	-	1
Stanleyville Formation	Ndolo	RGP 12221	-	*31*	*21*	1	2	14	27	3	-	1
Stanleyville Formation	Ndolo	RGP 12222	-	*12*	*3*	-	1	27	49	7	-	1

Pal palygorskite, Smec smectite, Mica mica and/or illite, Chl chlorite, Kaol kaolinite, Qz quartz, Ca calcite, Kf alkali feldspar, Pl plagioclase, Hm hematite.
* RMCA registration code; for some samples, two different materials were analyzed (RGM 16854, RGM 16856).
** Quantified as 2:1 Al-clays (for non-oriented powder samples), followed by semi-quantitative estimation of the relative proportions of smectite and mica or illite (based on results for oriented slides).

Table 2. Mineralogical composition of the clay fraction of selected specimens.

Type	Site	Sample n°	Pal	Mont	Mica-Mont*	Mica	Chl	Kaol
Massive palygorskite, white	Carrigres	RGM 16856	54	36	-	8	2	-
Palygorskite coating	Carrigres	RGM 16840	95	4	-	-	-	1
Fine-granular calcite, reddish	Carrigres	RGM 16844	60	35	-	3	1	1
Fresh arkose	Carrigres	RGM 16837	-	88	5	5	1	1
Stanleyville Formation	Ndolo	RGP 12221	-	68	20	10	1	1

Pal palygorskite, Mont montmorillonite, Mica-Mont mica-montmorillonite irregular mixed layer (75:25), Mica mica and/or illite, Chl chlorite, Kaol kaolinite

proportions. No differences between the various macroscopic types of palygorskite occurrences were detected with regard to swelling behaviour or crystal structure.

The arkosic Inkisi Subgroup substrate at the Carrigres and Immotex sites has a clay mineral association that is dominated by montmorillonite, with minor amounts of mica and kaolinite. Palygorskite is absent in the arkose matrix, also in the immediate vicinity of palygorskite-bearing veins. Some arkose samples from other sites in the Kinshasa area are dominated by mica, which is more compatible with an earlier study (Bartholomé et al., 1963), and they are rich in kaolinite when weathered. In Stanleyville Group deposits of the Ndolo district, palygorskite is absent as well, and montmorillonite, mica and minor kaolinite are the only clay minerals present.

Petrographical characteristics

In thin sections, palygorskite is easily recognized as a fine-grained component with low birefringence (first order grey interference colours at standard section thickness), which is much lower than

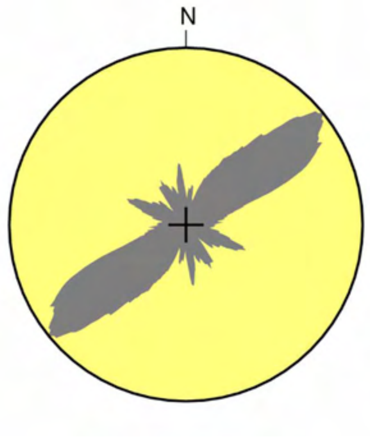

Figure 3. *Rose diagram for the orientation of 137 subvertical fractures with palygorkite or calcite (equal area, moving average with 11° aperture; maximum frequency 1.66 %).*

that of smectite and mica, the other main phyllosilicate minerals detected by XRD analysis of the vein deposits.

Fibrous palygorskite – Macroscopically fibrous palygorskite is characterized in thin section by strong parallel alignment of the clay-size particles, resulting in optical single-crystal behaviour (Figure 5A). Sand grains and rounded sparitic calcite crystals are common within the palygorskite mass (see Figure 5A).

Massive palygorskite – Massive palygorskite shows a varying degree of parallel alignment of clay aggregates, ranging from an entirely random orientation of small oriented domains (Figure 5B) to clear preferential vein-parallel alignment (Figure 5C). Alignment is commonly more pronounced in thin bands along the sides of veins or along cracks within the veins, with a gradual lower boundary of those bands and with the same nature and abundance of sand grains as in the main part of the vein (Figure 5D). Massive palygorskite contains an important admixture of sand, with components that are common in the arkose substrate (feldspar grains, rock fragments) but typically with a higher quartz content than this material. Other components are arkose fragments, in part with sparitic calcite cement (Figure 5E), and fragments of macrocrystalline calcite coatings or infillings.

Palygorskite coatings – Palygorskite coatings are typically pure clay accumulations, with parallel alignment of clay particles and a sharp lower boundary (Figure 5F). Random patterns of clay orientation and the occurrence of sand-sized aggregates of oriented clay are only locally observed. The nature of the clay in the coatings is similar to that occurring as part of the fine-granular calcite deposits, with pale and darker types (see below).

Fine-granular calcite – Fine-granular calcite veins are composed of anhedral equant sparitic crystals. Other features are a narrow to wide range in crystal size, common pressure twinning, and rare scattered quartz grains (Figure 6A). Most samples contain minor amounts of clay, occurring as thin layers along grain

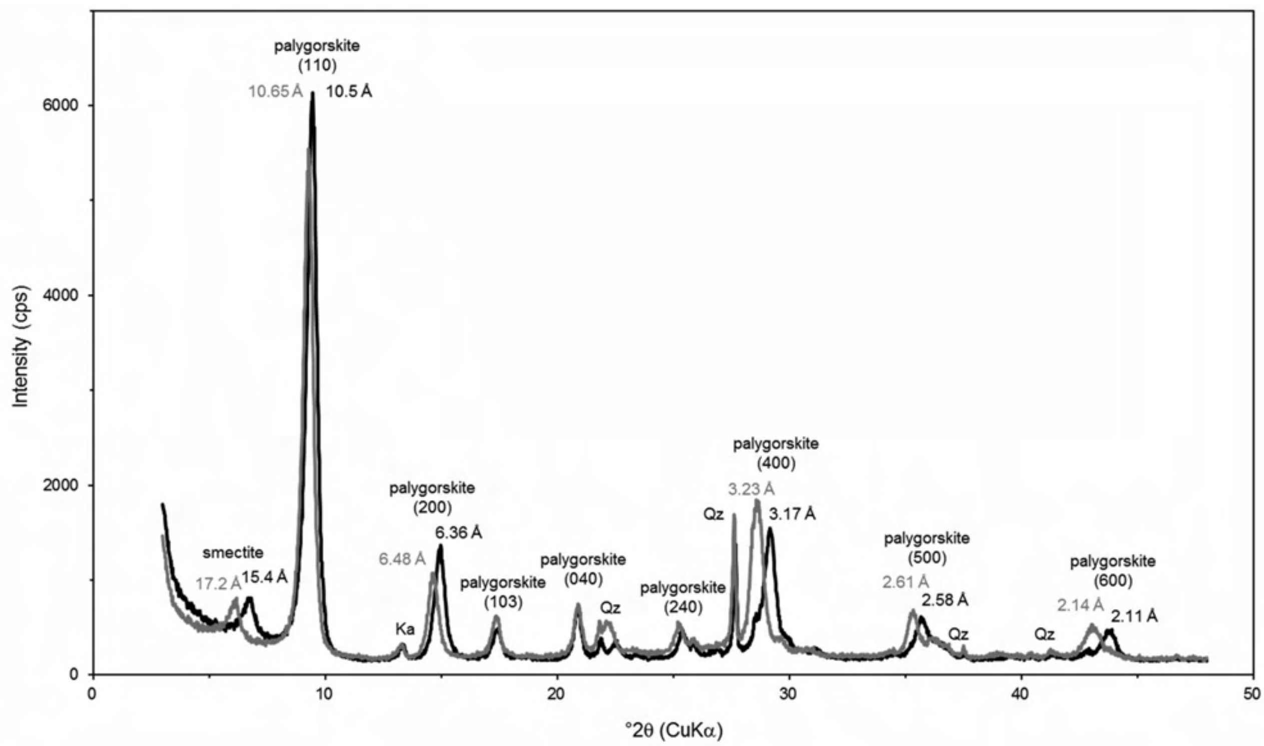

Figure 4. *X-ray diffraction patterns for the clay fraction of sample RGM 16840, for oriented slides before (black pattern) and after (grey pattern) glycol treatment, illustrating the swelling behaviour of palygorskite.*

boundaries, partly with random patterns but more commonly showing an alignment parallel to the vertical sides of the veins (Figure 6B). The clay typically has optical properties corresponding to those of palygorskite, with a pale yellowish brown colour and first order grey interference colours. In many samples, the clay in some parts has a darker colour and somewhat higher interference colours (Figure 6C). All clay in the fine-grained calcite veins typically shows perfect alignment of clay particles. One exception is palygorskite in a zone with high clay content, showing a mosaic-like fabric except along

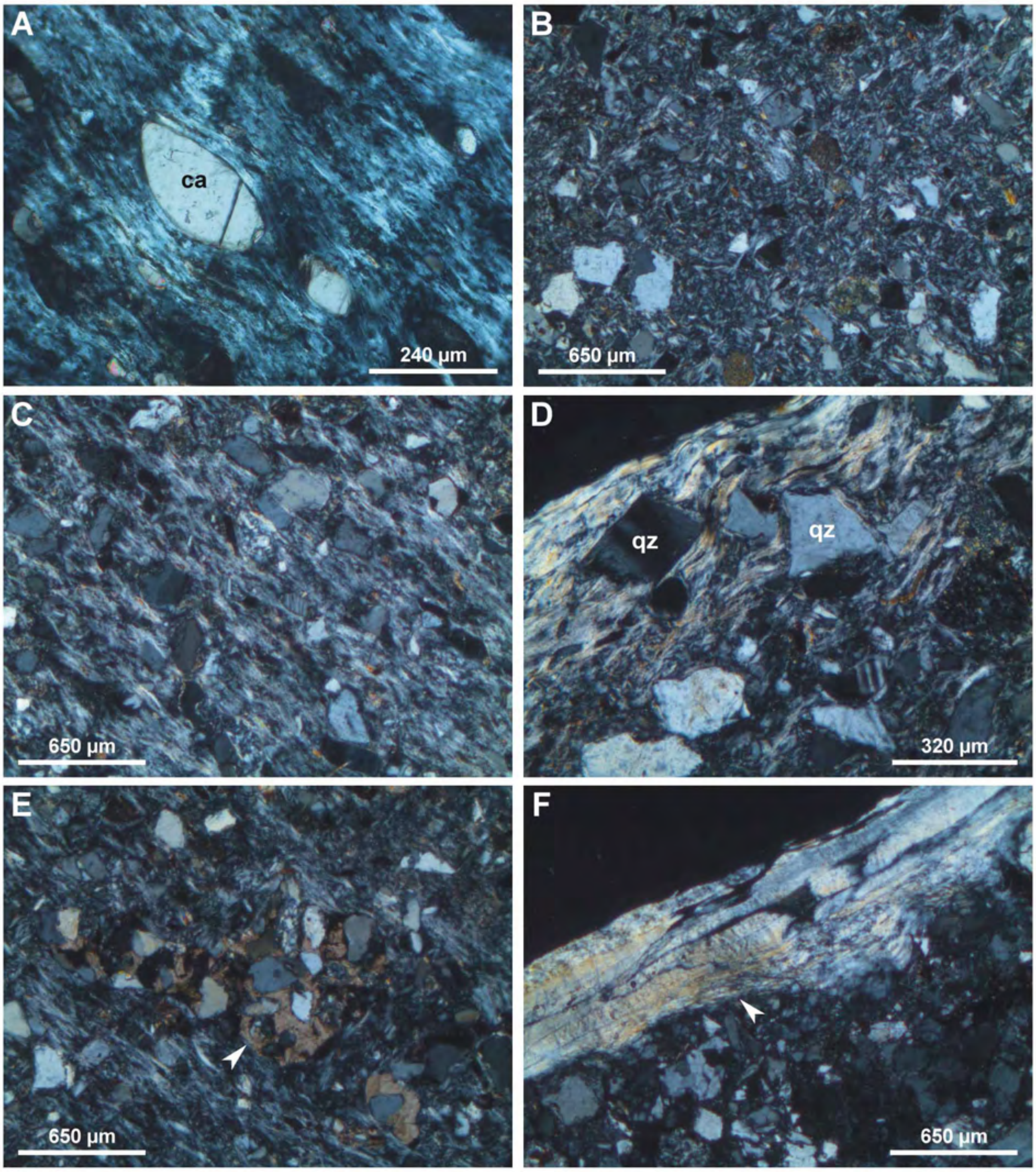

Figure 5. *Palygorskite veins and coatings. (**A**) Macroscopically fibrous palygorskite, with strong parallel alignment of clay particles and scattered rounded calcite grains (ca) (RGM 11708, cross-polarized light [XPL]). (**B**) Massive palygorskite, with random orientation of clay domains (RGM 16860, XPL). (**C**) Massive palygorskite, with moderate degree of alignment of clay particles (RGM 16856, XPL). (**D**) Massive palygorskite, with strong parallel alignment along the side of a fracture, and with quartz grains (qz) throughout the deposit (RGM 16860, XPL). (**E**) Massive palygorskite, with an arkose fragment cemented by sparitic calcite (arrow) (RGM 16856, XPL). (**F**) Palygorskite coating, showing a sharp lower boundary (arrow) (RGM 16840, XPL).*

part of the sides of this occurrence. Some clay layers enclose calcite crystals with lenticular shape (Figure 6D).

Macrocrystalline calcite – The macrocrystalline coatings and infillings consist of coarse sparitic calcite. Pressure twinning is quite common, both in coatings and infillings, and zoning is locally observed. Along the base of the calcite coatings and infillings, the arkose substrate is commonly cemented by sparitic calcite, which is absent away from the sides of the veins. The contact between calcite coatings that developed along

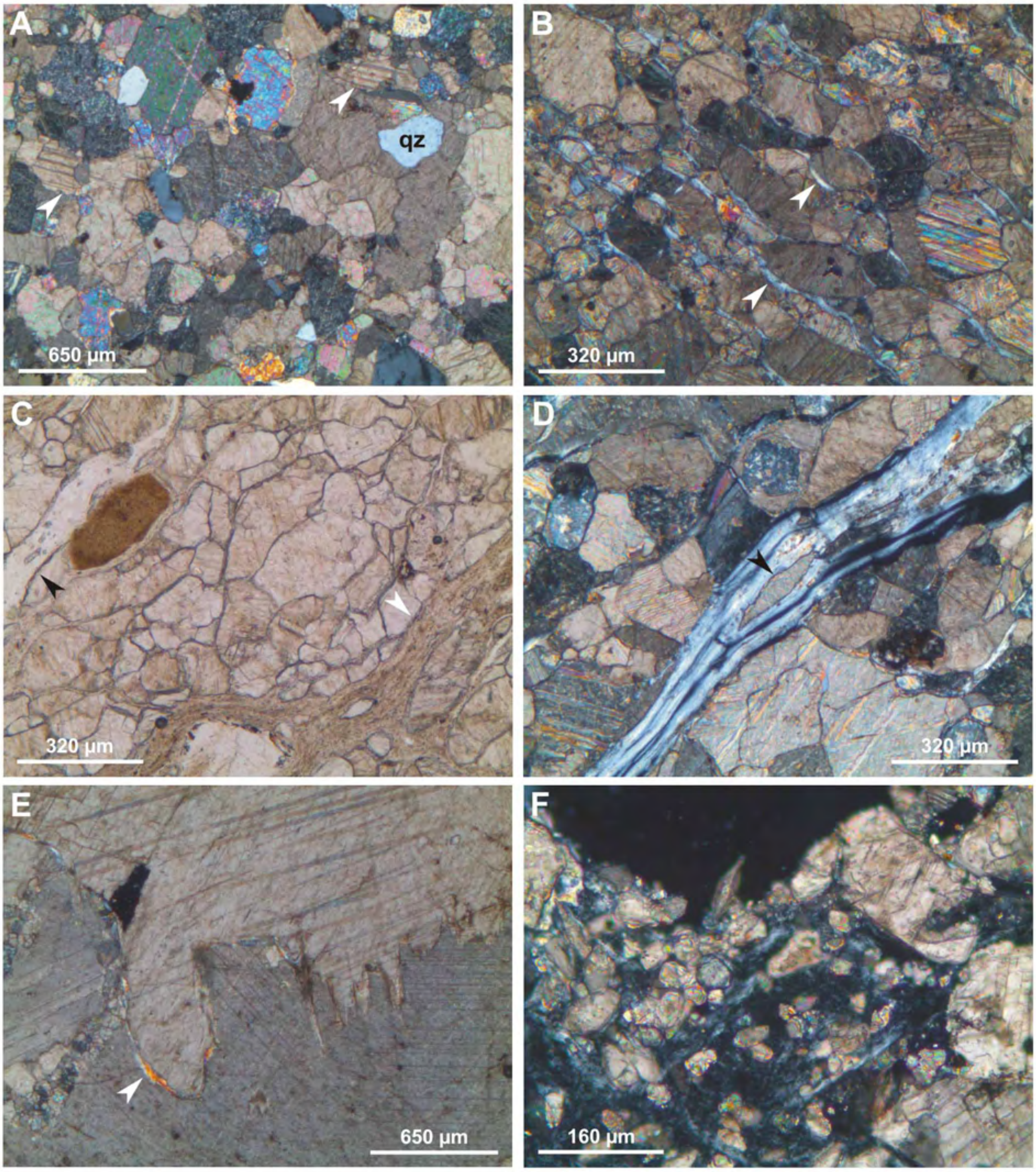

Figure 6. *Calcite veins. (**A**) Fine-granular calcite with common pressure twinning (arrows), enclosing scattered quartz grains (qz) (RGM 16841, XPL). (**B**) Parallel alignment of palygorskite occurrences (arrows) in fine-granular calcite (RGM 16843, XPL). (**C**) Pure palygorskite (black arrow), and palygorskite with smectite admixture (white arrow), in fine-granular calcite (RGM 16836, plane-polarized light). (**D**) Palygorskite band with enclosed lenticular calcite crystals (arrow), in fine-granular calcite (RGM 16836, XPL). (**E**) Palygorskite (arrow) along highly irregular contact between macrocrystalline calcite coatings in a closed fracture (RGM 16847, XPL). (**F**) Palygorskite without alignment of clay aggregates and with scattered microsparitic calcite, along the surface of a macrocrystalline calcite coating in an open fracture (RGM 16850, XPL).*

opposite sides of fractures, forming complete infillings, is commonly marked by the occurrence of a thin palygorskite layer, with strong parallel alignment of the clay particles. This contact plane is partly highly irregular (Figure 6E), and in some samples it has associated calcite that is more fine-grained than in the main part of the infilling. Clay accumulations that are locally observed along the surface of macrocrystalline coatings in open fractures are characterized by random orientation of the clay and by variable microsparitic calcite content (Figure 6F).

In SEM images, massive palygorskite shows random to parallel fibre orientations (Figure 7A). In pinkish, smectite-bearing vein material, no indications for smectite-palygorskite transformation could be recognized. Palygorskite coatings are characterized by parallel alignment, with variations between bundles (Figure 7B).

Stable isotope composition

Stable isotope analysis of three types of calcite occurrences (macocrystalline coatings, macrocrystalline infillings, fine-grained calcite) shows a strong similarity in $\delta^{18}O$ and $\delta^{13}C$ values between macroscopic types, averaging about 24.0 $\delta^{18}O_{V\text{-}SMOW}$ and -2.7 $\delta^{13}C_{V\text{-}PDB}$ (Table 3).

Discussion

Palygorskite-smectite relationship

The palygorskite veins of the Kinshasa area range in composition from relatively pure palygorskite accumulations to deposits with a considerable admixture of other clay minerals and sand grains. Because the relative abundance of smectite, with associated minor kaolinite and mica, is greatest in deposits with the highest sand content and with a pinkish colour, montmorillonite is considered to be an impurity, rather than a compound having any kind of genetic relationship with palygorskite. Areas with relatively high interference colours could represent occurrences of montmorillonite admixtures within the palygorskite deposits, but this would not be a clear indication of replacement or co-precipitation of authigenic minerals. Also, no indications for smectite-palygorskite transformation could be recognized by SEM observations. As impurity, montmorillonite could be derived from the local arkose substrate, in which it is the dominant clay mineral and which is also the source of the feldspar component of the sand fraction of the vein material. However, montmorillonite is more likely derived from the local Stanleyville Group, which typically has a much higher clay content.

Swelling behaviour of palygorskite, similar to the one observed here, has been reported in a few other studies (Watts, 1976; Jeffers and Reynolds, 1987; Lopez-Galindo et al., 1996), and it may well be more common than suggested by these few mentions, as its detection is made difficult by the limited 2θ range that is typically used for XRD analysis of glycol-treated samples. Palygorskite structure expansion has been related to palygorskite-smectite transformation (Jeffers and Reynolds, 1987), but the extent and nature of this feature are too poorly documented to understand its significance.

Palygorskite vein development

Hydrothermal activity, the most common explanation for palygorskite vein development, is most unlikely to be at the origin of palygorskite formation in the Kinshasa area. Throughout the region, there are no indications for its occurrence, as invoked in other palygorskite vein studies, such as host rock alteration (Stephen, 1954; Antun, 1956), proximity of magmatic rock bodies (Nathan et al., 1970; Haji-Vassiliou and Puffer, 1975; Furbish and Sando, 1976; Gibbs et al., 1993), and associated base metal and other mineralizations (Caillère, 1951; van der Wel, 1972; Lindqvist and Laitakari, 1981). Also the limited extent of palygorskite occurrences in the local fracture system is not compatible with a hydrothermal origin (Salter and Appleyard, 1974). The stable isotope composition of calcite, which formed in the same setting as the associated palygorskite, demonstrates that all calcite formed in earth-surface conditions. This is based on temperatures of 24 to 27°C that are obtained when the standard equation for the calcite-water system is applied (O'Neil et al., 1969), using an estimate of -4.50 $^{o}/_{oo}$ $\delta^{18}O_{SMOW}$ for regional groundwater (Melchiorre et al., 1999). The fracture-associated palygorskite occurrence nearest to Kinshasa, at Boko Songo in the Republic of the Congo (Bigotte, 1959), is located in an area with extensive fault-related Cu-Pb-Zn mineralization, similar to the one occurring at Bamba Kilenda, ~70 km south of Kinshasa, attributed to basin brine circulation rather than to hydrothermal processes (Verhaegen, 2005). However, the age of these mineralizations is only known to be younger than that of Inkisi Subgroup formation and provides no indications for the timing and nature of events that could have led to palygorskite formation in the Kinshasa area.

For palygorskite formation related to interaction between the rock substrate and groundwater or meteoric water along fractures, a major source of Mg and Si is lacking. The Inkisi Subgroup is largely non-calcareous, with feldspar and chlorite as the most abundant reactive phases, and the underlying Mpioka Subgroup deposits are equally siliciclastic (Frimmel

Table 3. Stable isotope data for calcite veins.

Type	Sample n°	$\delta^{18}O_{V\text{-}SMOW}$	$\delta^{18}O_{PDB}$	$\delta^{13}C_{V\text{-}PDB}$
Macrocrystalline calcite coating	RGM 16849b	23.71	-6.94	-3.34
Macrocrystalline calcite infilling	RGM 16847	24.35	-6.31	-2.14
Fine-granular calcite	RGM 16859	23.85	-6.80	-2.75

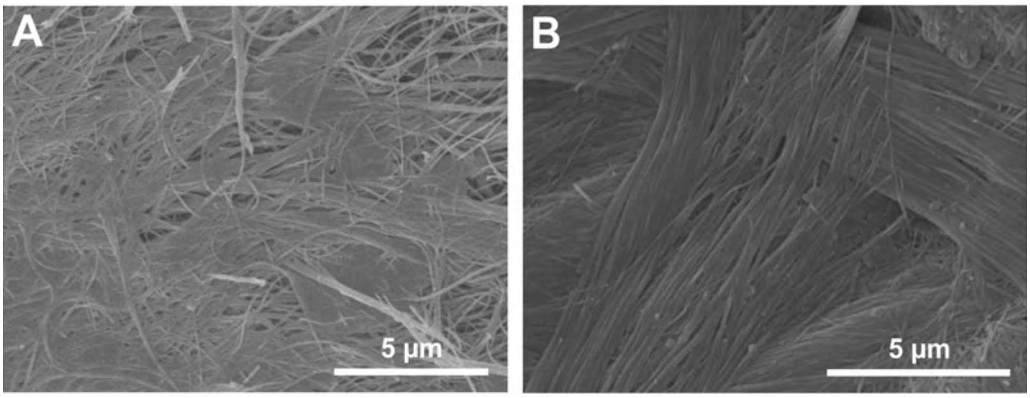

Figure 7. *SEM images. (**A**). Massive palygorskite, with random orientation of fibres (RGM 16856). (**B**). Palygorskite coating, composed of wide bundles with varying orientations (RGM 16865).*

et al., 2006). The underlying deposits of the Schisto-Calcaire Subgroup are dolomite- and talc-bearing in the upper part of the sequence, but the depth of the top of these deposits is probably in excess of several hundreds of metres. The overlying Phanerozoic deposits are in principle a possible Mg-Si source, but various features suggest another role for these formations, as discussed below. A related possible mode of palygorskite formation is transformation of a precursor whose occurrence was confined to the fractures, but no indications are recognized for such type of origin.

A third possible mechanism to be considered is palygorskite vein development by infiltration of essentially clastic palygorskite-bearing material in open tectonic fractures. A first indication is the variable degree of alignment of clay particles and aggregates in massive palygorskite veins, which implies that random orientation patterns are a feature of the original fracture infilling and that parallel alignment was only obtained by deformation. Development of clay alignment by deformation is supported by the recognition of stronger alignment along cracks and along the sides of the veins, where pressure can be expected to be commonly exerted. Deformation, with lateral movement, is also recorded by the occurrence of striated and stepped surfaces, which has also been observed in various other studies of palygorskite veins (Martini, 1964; Müller-Vonmoos and Schindler, 1973; Salter and Appleyard, 1974; Haji-Vassiliou and Puffer, 1975; García-Romero et al., 2006). Although random orientation patterns have been reported for assumed hydrothermal palygorskite (Caillère, 1951), including one occurrence with both massive and fibrous facies (Minato et al., 1969), they are uncommon for clay that forms as an authigenic precipitate in voids, suggesting a non-authigenic origin for the studied veins.

The occurrence of components derived from local cover deposits is a further indication for the development of palygorskite-bearing veins by infiltration of allogenic material. The sand fraction has a lower feldspar content than the arkose substrate, suggesting a contribution from another source, whereby an attribution of greater relative quartz content to weathering is precluded by the lack of visible feldspar corrosion. This difference in quartz/feldspar ratios was already noted by Antun (1966), who also reported a heavy mineral assemblage, with high garnet and epidote contents, that corresponds to that of the Stanleyville Group of the Ndolo district. The occurrence of palygorskite in these or other post-Palaeozoic deposits could not be confirmed by the present study, in agreement with earlier results for the Ndolo district deposits (Bartholomé et al., 1963). However, this local minor occurrence of cover deposits, representing a unit that reaches a thickness of ~450 m in other parts of the Congo basin, does not provide a full record of Mesozoic and Cenozoic sedimentation in the western part of the basin. As documented by studies of the Stanleyville Group of the Kisangani region (Vernet, 1961) and of Jurassic and Cretaceous deposits in cores obtained at Samba and Dekese (Vanderstappen and Verbeek, 1959, 1964), the Upper Jurassic and Cretaceous were periods with extensive authigenic mineral formation in the Congo Basin, mainly in the form of analcime ($Na(AlSi_2O_6).H_2O$). Phyllosilicates reported in these studies are mainly montmorillonite, mica and kaolinite, but Vernet (1961) also mentions the occurrence of minor palygorskite in two samples, which are dominated by interstratified montmorillonite-illite. Myers et al. (2011) report trace amounts of palygorskite in palaeosoil levels within the Stanleyville Group in the Samba cores, but both palygorskite identification and palaeosoil recognition in this study are uncertain. For palygorskite-rich dolomite from the Republic of the Congo that is mentioned by Le Chatelier (1914, p. 446-447), no information about location or stratigraphic position is available.

Palygorskite deposition clearly postdates the formation of macrocrystalline calcite coatings and infillings, as recorded by the presence of reworked macrocrystalline calcite fragments in the palygorskite deposits and by the occurrence of fragments of calcite-cemented arkose that is commonly associated with the calcite veins. A similar temporal relationship is suggested in several other studies by the occurrence of carbonate coatings along the sides of palygorskite veins, both hydrothermal and non-hydrothermal (Evans and King, 1962; Lowry, 1964; van der Wel, 1972;

Henderson et al., 1973; Tien, 1973; Furbish and Sando, 1976), indicating that calcite and palygorskite commonly form during a single event, in various contexts. Although this general observation is more compatible with palygorskite authigenesis, and accepting the interpretation that palygorskite veins in the Kinshasa area formed by sediment infiltration, calcite can be considered to have formed at a stage when solutions could circulate in open fractures, extending to a palaeosurface along which no sedimentation was taking place. Palygorskite-bearing sediments, derived from deposits with abundant synsedimentary or diagenetic palygorskite, entered the fractures at a later time, possibly following a transgression in a lake-marginal setting. As pointed out by Antun (1966), this occurred at a stage when Stanleyville Group sediments (Late Jurassic to Early Cretaceous) had already been deposited in the region, based on the presence, within the veins, of components derived from that formation.

Vertical sequences observed for two veins in high exposures provide no additional information about vein development. Solutions and suspensions circulated within a three-dimensional system of fractures with variations in degree of closure along each fracture plane, resulting in random vertical trends in local cross-sections.

Calcite formation and evolution

Macrocrystalline calcite coatings in the study area are clearly authigenic precipitates, which formed from solution in open fractures. Infillings of the same calcite type formed by closure of fractures that were lined by coatings, which involved deformation of initially euhedral crystals, as recorded by the partially highly irregular contact between the former coatings and by the locally associated occurrence of fine-grained calcite. Alignment of clay particles in the clay layers along the central contact plane of these infillings is an associated deformation feature, for which the non-deformed precursor is locally observed along the surface of calcite coatings in open fractures.

The fine-granular calcite veins seem to be derived from macrocrystalline calcite deformation during lateral movement, whereby their texture is essentially cataclastic. This interpretation is supported by the strong similarity in stable isotope composition between macrocrystalline calcite and fine-granular calcite, demonstrating that both are closely related. Clay alignment in the granular calcite veins is again interpreted as a deformation feature, which is supported by the local absence of alignment in parts with high clay content, and by the elongated shape of calcite crystals within the clay matrix (see also Haji-Vassiliou and Puffer, 1975).

The common occurrence of pressure twinning records that deformation or recrystallization under pressure took place at some stage. The occasional presence of twinning in coatings with euhedral terminations of the calcite crystals implies a complex evolution, whereby the fracture re-opened after having closed, followed by further calcite growth.

Palygorskite coating development

Palygorskite coatings within, along and outside the main veins, partly occurring up to a great distance from those veins, are the result of later redistribution of vein-derived palygorskite by circulation of groundwater within planar voids. This could have occurred at any stage after initial development of the palygorskite infillings, unrelated to regional geological events. The common good alignment of clay in these coatings is considered to be the result of deposition from suspension rather than mineral authigenesis, representing the only example of clay alignment in this study that is not a deformation feature.

Conclusions

The following sequence of events is recorded by the palygorskite and calcite veins of the Kinshasa area:

- development of fractures with a N55°E orientation, followed by opening without lateral movement
- formation of macrocrystalline calcite coatings from solutions that circulated through the fractures,
- infiltration of palygorskite-bearing sediments, derived from (Upper Jurassic to Early Cretaceous) continental formations containing authigenic clays, and
- deformation of the fracture fills during a stage with left-lateral strike-slip faulting, resulting in a transition from random to parallel alignment of clay particles, from macrocrystalline calcite coatings to infillings, and from macrocrystalline calcite to fine-granular calcite with a cataclastic texture.

The final period was followed by relatively minor dispersion of palygorskite along various types of planar voids.

The palygorskite veins of the Kinshasa area are essentially fracture infillings that formed by infiltration of allogenic material, although a different origin is suggested by their composition, with a high proportion of a mineral that typically forms as an authigenic phase in hydrothermal or earth-surface conditions, as well as by the occurrence of associated authigenic calcite. A similar mode of formation could also be considered for other occurrences of palygorskite and related minerals such as sepiolite and Mg-smectite in vein-like structures.

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