Proceedings World Geothermal Congress 2020+1
Reykjavik, Iceland, April - October 2021

Characterization of an Exhumed High Temperature Paleo-Geothermal Reservoir by Clay Minerals and Secondary Phases Identifications in Terre-de-Haut Island (Les Saintes Archipelago, Guadeloupe)

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Keywords: Hydrothermal alteration, Clays, Geothermometry, Paleo-geothermal reservoir, Guadeloupe, Fluid inclusions

ABSTRACT

Due to its geodynamic location on the Lesser Antilles arc, Basse-Terre of Guadeloupe displays an active volcanism which is favorable to the development of geothermal energy associated to power generation. Thus, twenty years ago, the Bouillante geothermal field was brought into production and now produces 15 MWe (Bouchot et al., 2010).

An exhumed analog of the Bouillante geothermal reservoir has been identified on Terre-de-Haut island (Les Saintes archipelago, 35km SE of Bouillante; Verati et al., 2016), which represents a key area to study the paleo-hydrothermal alterations, and the interactions between the paleo-hydrothermal fluids and the surrounding rocks.

To characterize the paleo-fluids in terms of composition and temperature, and to determine the mineralogical and petrographic features of the paleo-hydrothermal alterations, we propose to couple crystallographic, geochemical and geothermometric approaches, i.e. X-ray diffraction (XRD), microprobe analysis (EMPA), chlorite thermometry and in-quartz fluid inclusion study (FI) on selected samples.

The clay minerals display a specific concentric distribution at the scale of the island, with chlorite crystallization in the core, smectite in the surroundings, and illite in an intermediate halo. Considering the data obtained in boreholes in Bouillante, which show that illite and chlorite are in the deeper parts, chlorite being located in the heart of the geothermal reservoir (Bouchot et al., 2010), the clay distribution allows to identify the temperature profile. Hence, thanks to the exhumation, we are looking to a horizontal section through the paleo-system of Terre-de-Haut, where we observe a lateral temperature gradient responsible for the paleo-hydrothermal alteration.

Chlorite geothermometry, based on a model specifically developed for low temperature contexts (T < 350°C) and pressures below 4 kbar (Bourdelle et al., 2013), has been applied on chlorites from both Terre-de-Haut paleo-system and from the active geothermal system of Bouillante. The results show a strong difference between the temperature estimates for chlorite formation on Terre-de-Haut (around 120°C) and for Bouillante (around 230°C, in agreement with the temperature measured in boreholes in Bouillante; Mas et al., 2006).

Observations of newly formed euhedral quartz, sampled in a geode located in a N70 oriented fracture, reveal the presence of numerous primary fluid inclusions. Preliminary results show that quartz underwent at least two growth stages recorded in the crystal core and clear overgrowths. Data indicate very low salinity (2% NaCl), and a minimum trapping temperature of around 250-280°C in inclusions located in the core, and around 70°C or less in the outer growth zones. These two events can be interpreted as a record of the fluid cooling during system evolution. Moreover, fluid inclusions from a second quartz sample (in another N70 fracture) are composed of nearly pure CO_2 with very low density, indicating a shallow CO_2 paleo-circulation episode.

This study shows that the clay mineral zonation cropping out in Terre-de-Haut is similar to that found by drilling in the active system of Bouillante. However, the temperatures of formation of some newly formed minerals indicate that some alteration episodes occurs at lower temperature than the fluid circulation occurring in Bouillante's active geothermal system.

Hence, these results show that clay minerals study and geothermometry of newly formed minerals (chlorite and quartz) are key steps to provide new thermal constrains on the paleo-geothermal reservoir of Terre-de-Haut and its evolution.

1. INTRODUCTION

Terre de Haut island (Les Saintes archipelago, French West Indies; Figure 1A) is described in the literature as exhibiting an exhumed paleo-geothermal system (Jacques and Maury, 1988a,b; Verati et al., 2016; Beauchamps et al., 2019), giving access to a sub-horizontal 2D cross-section into the system. The present study focuses on Terre de Haut's paleo-hydrothermal system in terms of clay minerals, which are well known for being good indicators of hydrothermal alterations (Patrier et al., 2003; Mas et al., 2006; Bouchot et al., 2010) and of thermicity in order to compare it with Bouillante which is still active and exploited for geothermal production of electricity. To this aim, secondary quartz crystals were sampled for microthermometry of fluid inclusions together with chlorite-bearing altered andesite that has been described by Beauchamps et al. (2019).

2. GEOLOGICAL CONTEXT

The Guadeloupe archipelago, located in the Lesser Antilles arc (Figure 1A), is the result of the subduction of the North American plate under the Caribbean plate at a velocity of approximately 2 cm yr^{-1} (Hawkesworth and Powell, 1980; DeMets et al., 2000;

Symithe et al., 2015). One ancient volcanic arc and one active result from this subduction (Bouysse, 1983). Terre-de-Haut island, belonging to the Les Saintes archipelago, is located on the active arc.

Regarding structural data, Verati et al. (2016) recognise four families of fault systems, active from 3 to 2 Ma: (1) N050-N070, (2) N130-N140, (3) N090-N110 and (4) N000-N020 trending fault systems. These four families of fault systems affecting the Les Saintes archipelago are compatible with the global tectonic framework of the entire Guadeloupe archipelago (Verati et al., 2016).

Three main subaerial volcanic phases have been defined by Zami et al. (2014), and are constrained between 2.98 ± 0.04 Ma old and 2.00 ± 0.03 Ma. Their lithology consists of dacitic lava flows and explosive breccia, basic andesitic lava and phreato-magmatic flows, and dacitic domes (from the oldest to the youngest).

The occurrence of a hydrothermal zone has been identified early by Jacques et al. (1984) and Jacques and Maury (1988), and further described more recently by Verati et al. (2016) and Beauchamps et al. (2019). This zone displays a succession of parageneses that are characteristic of high-temperature hydrothermal alteration in epithermal settings and its retrogression during cooling (Verati et al., 2016).

According to Zami et al. (2014), the age of hydrothermalism is constrained within a time span because the oldest volcanic rocks affected by the hydrothermal activity are dated at 2.08 ± 0.03 Ma, and none of the intrusions dated at 2.00 ± 0.03 Ma are hydrothermally altered, even when located in the hydrothermally altered zone (Figure 1B, red spots). Hence, Zami et al. (2014) propose that the maximal timescale activity of the Les Saintes paleo-geothermal system lasted around 80 ky. However, for Verati et al. (2016), considering the oldest volcanic rocks affected by the hydrothermal alteration are dated at 2.40± 0.04Ma, the duration of the hydrothermal activity is around 400 ky.

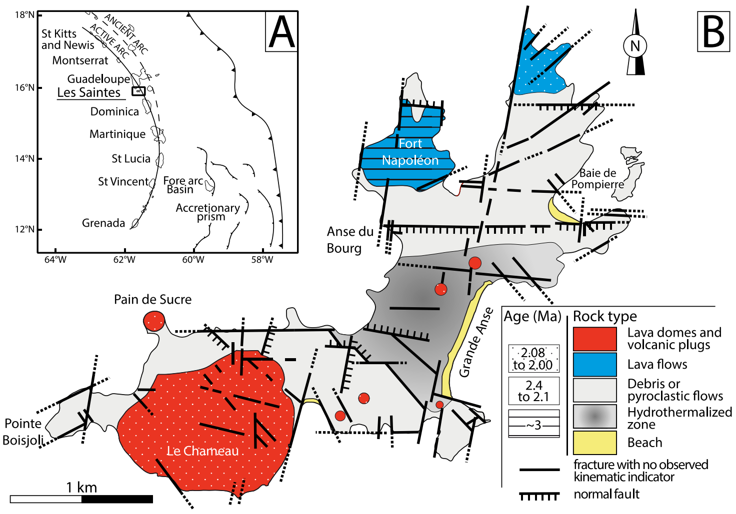

Figure 1. A) Simplified geodynamic map of the Lesser Antilles Arc. B) Simplified geological map of Terre-de-Haut (Les Saintes archipelago) indicating structural data, volcanic rock types and their ages (modified after Verati et al. 2016; Zami et al. 2014; Jacques and Maury 1988a, b; Jacques et al. 1984). Symbols refer to age of the rock formation, whereas colours refer to rock type (Figure from Beauchamps et al., 2019).

3. RESULTS

3.1 Clay minerals distribution

The clay fraction (< 2µm) of 100 samples (Fig. 2) distributed all over Terre-de-Haut island has been characterized by XRD. The 100 diffractograms allow to distinguish five main clay assemblages. The first one and most widespread over the island is made of smectite ± kaolinite ± halloysite. The second assemblage consists of smectite + illite ± kaolinite ± halloysite, and characterizes the so-called 'illite zone'. A third assemblage, similar to the second one, with presence of illite/smectite mixed-layer minerals, is also found in the 'illite zone'. A fourth assemblage, which consists of smectite + chlorite + illite ± kaolinite ± halloysite, is only observed on the east side of the island, north of Grande Anse (Fig. 2), and characterizes the 'chlorite zone'. According to this study, this assemblage has the narrowest geographical distribution. A fifth assemblage, equivalent to the previous one, but with chlorite/smectite mixed-layer minerals, is also found in the 'chlorite zone'. No difference is apparent in the mineralogy between samples from fracture infillings and the surrounding rocks. Consequently, the clay mineral distribution allows us to distinguish the three main different zones that are represented in Figure 2.

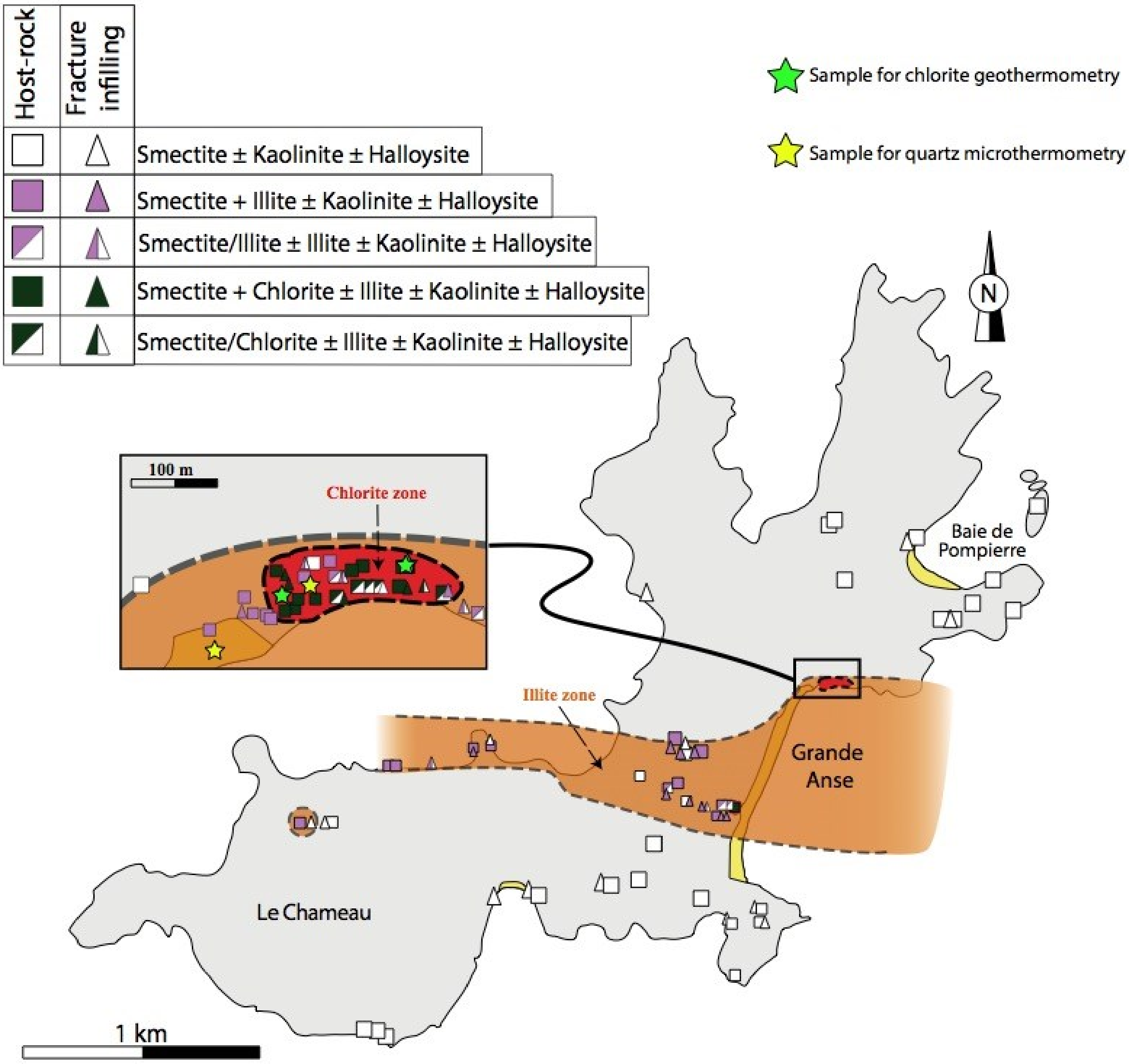

Figure 2 - Location and distribution map of the clay minerals in Terre-de-Haut island (Guadeloupe) for samples extracted from fracture infillings (triangles) and their host rock (squares). Illite zone (orange) and chlorite zone (red) are indicated. The rectangle zooms on the chlorite-bearing zone (North of Grande-Anse). The two yellow stars represent the quartz samples used for fluid inclusion analysis, the green stars represent the chlorite bearing samples analyzed for geothermometry.

3.2 Thermometry

3.2.1 Chlorite thermometry

Chlorite geothermometry was applied on the Terre-de-Haut in order to estimate the temperature of chlorite crystallisation. Because of its applicability domain (specially designed for low-T contexts, i.e. < 350 °C), the selected chlorite geothermometer is the one developed by Bourdelle et al. (2013).

Chemical analyses (EMPA) have been performed on 3 samples located in the previously described "chlorite zone" (north of Grande Anse beach; Figure 2) defined by Beauchamps et al. (2019). Samples GEC 236 and GEC 237 were collected 50cm from one another while GEC 270 comes from 150m away to the west.

Chlorite being developed at the expense of ancient pyroxenes with size up to 400µm in length, they have an euhedral shape; their basal and longitudinal sections are still visible. Some pseudomorph are entirely composed of chlorite, some are composed by chlorite and patch of calcite. Other chlorite minerals are present in the matrix with a much smaller size and in vein associated to calcite. On sample GEC270, illite and kaolinite are sometimes associated to chlorite. 75 analyses done on a total of 18 chlorites were selected for thermometric estimations.

Regarding Fe/Mg ratio, chlorites analyzed on sample GEC 270 are magnesian with a very narrow distribution (Fe/Mg = 0.1±0.02), whereas GEC 236 and GEC 237 have intermediate compositions between Mg and Fe poles with larger distribution (0.52 <Fe/Mg<1.15 for sample GEC236; 0.41<Fe/Mg<0.61 for GEC 237) (Figure 3).

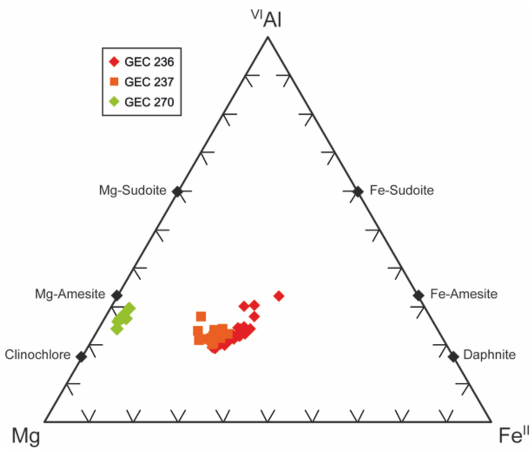

Figure 3 - Chlorite compositions plotted in a ternary diagram (Mg, ^{IV}Al, Fe^{II}). Red: GEC 236; orange: GEC 237; green: GEC 270.

The analysed chlorites were plotted in the Sudoite-Al-free chlorite-Clinochlore/Daphnite field in the Si-R^{2+} diagram developed by Bourdelle et al. (2015). Sample GEC 236 and 237 show a similar composition, with a main formation's temperature estimation around 120°C ± 20°C. These samples, as well as sample GEC 270, show a trend toward a diminution of their R^{2+} content, associated to a decrease of formation temperature until 72°C ± 20°C (Figure 4).

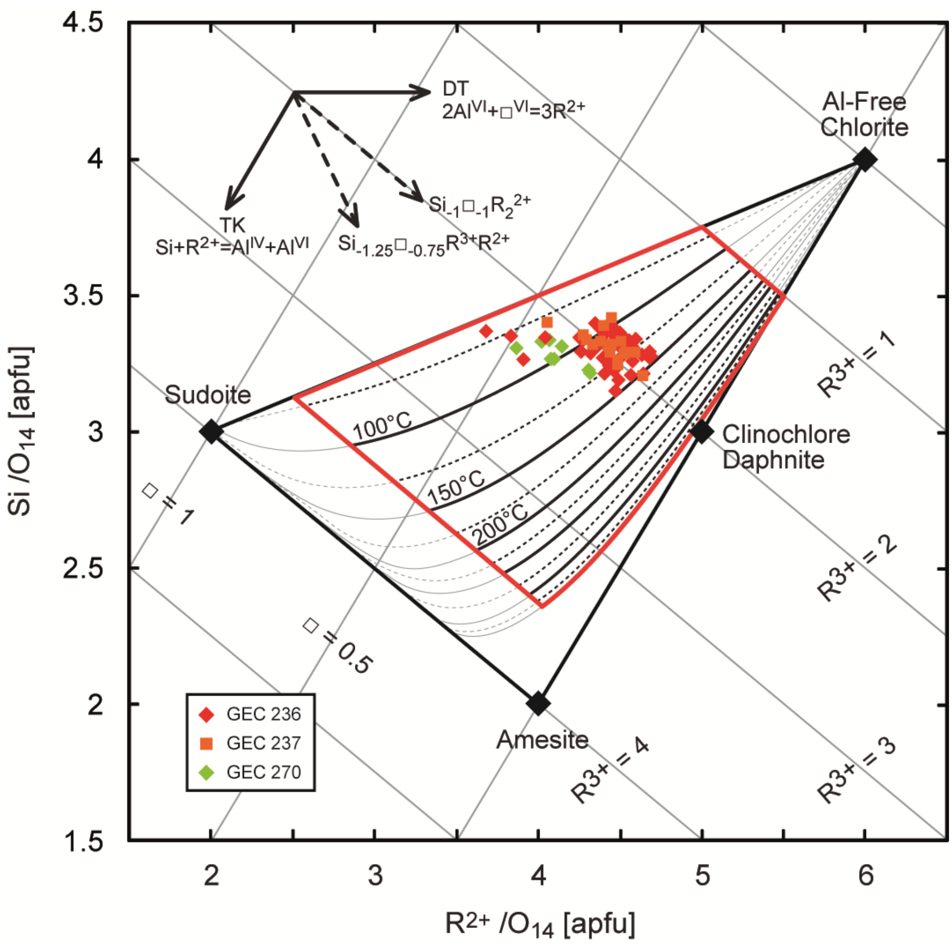

Figure 4 - Composition of the chlorite analysed from North Grand-Anse, plotted in a Si - R^{2+} diagram with their corresponding formation temperature (diagram adapted from Bourdelle et al., 2015). The 3 samples provide data in the same range of composition (in the field Sudoite-Al-free chlorite-Clinochlore/Daphnite) and thus temperature (72 to 158°±20°C).

3.2.2 Microthermometry on fluid inclusions in quartz crystals

The study of fluid inclusions (FI) allows characterizing the paleo-fluid responsible for the mineral crystallization by knowing its composition and the minimum pressure-temperature conditions during its entrapment. In this study, the "Fluid Inclusion Assemblage" (FIA) approach (Goldstein and Reynolds, 1994) was used. This method consists in gathering FIs all formed contemporaneously, this contemporaneity being based on petrographic criteria.

This study has been done on two newly-formed quartz samples extracted north of Grande-Anse beach, 170 meters away from one another (represented by yellow stars in Figure 2), in two parallel fractures. Sample GEC 261 has been sampled in a fracture oriented N70; 80SE (strike;dip) (Figure 5A). This fracture is filled with a brecciated rock containing in some places aggregates of euhedral quartz (<1cm in length) (Figure 5B). Sample GEC 285 has been taken from a banded quartz vein about 10 cm thick (Figure 5D), oriented N70;70SE (strike;dip), with mm-thick layered bands of quartz relatively parallel to one another, associated to hematite aggregates (black bands on Figure 5E).

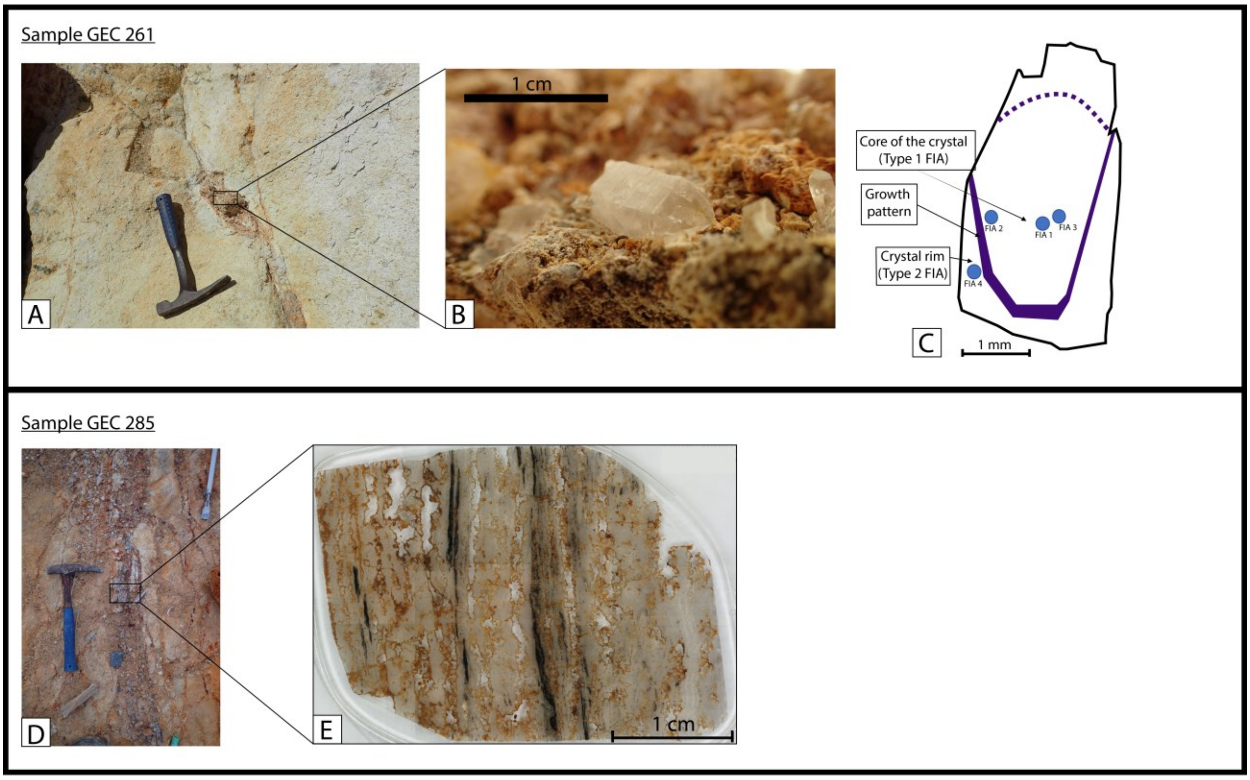

Figure 5 – Photograph of quartz samples used for fluid inclusion microthermometry. A: Fracture containing sample GEC 261; B: Macroscopical view of one of the euhedral quartz; C: Sketch of the distribution of the different type of FIAs within the crystal; D: Fracture containing sample GEC 285; E: Section of the sample GEC 285 where the subparallel quartz bands are visible.

In sample GEC 261, two types of FIAs can be distinguished:

-Type 1 contains only two phases (liquid+vapour; LV) with consistent vapor filling ratio (Flv) of about 20%.

-Type 2 is composed of all-liquid (L) fluid inclusions associated with two-phase LV inclusions with variable Flv.

Microthermometric data for sample GEC 261 indicate that the fluid trapped in all inclusions is almost pure water, salinity being less than 2 wt % eq. NaCl. The distribution of the two types of FIAs allows to establish a relative chronology. FIs of the first stage of precipitation of the mineral have temperature of total homogenization (Th) between 240 and 270 °C, whereas the later stage of crystallization is recorded by FIAs located in the outer part of the crystal, mainly composed of all-liquid inclusions, indicating of a formation temperature lower than 70°C (Figure 5C).

In sample GEC 285, FIAs are mainly composed of dark single-phase fluid inclusions interpreted as all-vapor (V) inclusions. Raman spectroscopy analyses showed that these inclusions are composed of almost pure CO_2 vapor, with traces of H_2S. Unmixing of all-vapor fluid inclusion has not been observed during cooling of inclusions down to -130°C, which means that the density of the gas is lower than 0.0140 g.cm^{-3} (Angus *et al.*, 1976). Hence this episode of fluid flow occurred at low depth. Few single-phase inclusions, much clearer, interpreted as all-liquid (L) fluid inclusions containing H_2O, are also present in the same FIAs.

This study has shown evidences of fracture-controlled fluid circulation through two different fractures oriented N70. Three episodes of fluid circulation can be drawn:

- In the fracture containing euhedral quartz (sample GEC 261):
  (1) A first high-temperature (240 – 270°C) fluid circulation, with a major meteoric fluid source.

  (2a) A low-temperature (<70°C) fluid circulation episode, with a major meteoric fluid source.

- In the fracture showing a cyclic filling (sample GEC 285):

(2b) Another low-temperature (<70°C) fluid circulation episode, associated to a crack and seal formation (Ramsay, 1980) with antitaxial mechanism (Cox, 1987). These structures testify of a low seismic activity (Jebrak, 1992). The inclusions have recorded an episode with two-phase CO_2-rich flow.

No relative chronology can be established between the two low-temperature episodes as they were recorded in two different parallel fractures.

4. DISCUSSION

The study of clay minerals on Terre-de-Haut island, along a near-horizontal surface shows a zonation with smectite in the outer rim of the island, illite in the intermediate rim and chlorite in the centre (Beauchamps et al., 2019). At Bouillante, 35 km NNW of Terre-de-Haut, a similar clay mineral zonation was observed vertically along the boreholes from the surface to 900m depth (in BO6 borehole) (Mas et al., 2006). Since Bouillante is an active geothermal site, the clay mineral zonation observed at Terre-de-Haut can indeed be considered as belonging to an exhumed paleo-geothermal system. The distribution of the clay minerals at Terre-de-Haut describes a paleo-geothermal gradient from the cooler smectite zone to the hotter chlorite zone, with the chlorite zone representing the central part of the reservoir, as it is observed in Bouillante (Bouchot et al., 2010).

In the Bouillante geothermal system, the chlorite zone appears at temperature around 250-260°C, associated to a pervasive alteration which predominates at 700 - 900 m depth (Mas et al., 2006). But the compositions of the Bouillante's chlorites (Mas et al., 2006) are chemically different (with lower Si content <3 apfu) compared to the ones of Terre-de-Haut which have a much lower formation's temperature. The low-temperature event revealed by chlorites from Terre-de-Haut ($T \approx 120°C$) might represent a later stage of crystallisation comparing to the one of Bouillante. Furthermore, chlorites found in Tillet, located in the Basal Complex, north of Basse-Terre, by Verati et al. (2018), presents a formation temperature in the same range as Bouillante's chlorite. Low temperature chlorite ($T \leq 150°C$) are commonly found in the literature (Bevins et al., 1991; Koroknai et al., 2008; Inoue et al., 2009, 2010; Bourdelle et al., 2013; among others)

Present-day fluid temperature in the active geothermal reservoir of Bouillante is about 250-260°C (Sanjuan et al., 2001; Mas et al., 2006; Guisseau et al., 2007; Bouchot et al., 2010). This temperature is in the range of the temperatures found during the first fluid circulation event for the formation of the euhedral quartz (sample GEC 261) on Terre-de-Haut. However, the low temperature (<70°C) event located in the outer rim of the euhedral quartz, and in sample GEC 285, reveal the cooling of the system which has not been yet experienced in the Bouillante geothermal system.

Gas composition has been determined in Bouillante's geothermal reservoir, through wells BO-2 and BO-4. They indicates values of 95 mol% of CO_2 and 3 mol% of H_2S (Sanjuan et al., 2001) which are very similar to that estimated in the single-phase CO_2-rich fluid-inclusion in sample GEC 285. This similitude leads to think of a global trend with the same source of gases, which could be from mixed origin (magmatic sedimentary and atmospheric) as it is found in Bouillante and in the Lesser Antilles Arc (Pedroni et al., 1999; Sanjuan et al., 2001). However, unlike Bouillante's geothermal fluid which is around 18-20g/L TDS composed of a mixture of sea water (58%) and fresh water (42%), the very low saline paleo-fluid of Terre-de-Haut is considered being essentially of meteoric origin.

CONCLUSION

The island of Terre-de-Haut exhibits a strongly hydrothermally altered east–west trending zone in the central part of the island. This area is characterised by the occurrence of several clay minerals that allow us to distinguish three main different hydrothermal zones, associated to newly discovered mixed layer minerals, organised in a general concentric pattern.

At Bouillante, 35 km NNW of Terre-de-Haut, a similar clay mineral zonation was observed vertically along the boreholes. Since Bouillante is an active geothermal site, the clay mineral zonation observed at Terre-de-Haut can indeed be considered as belonging to an exhumed paleo-geothermal system.

New thermometric data allow to better constrain the thermal history of the paleo-geothermal system of Terre-de-Haut, and to distinguish 3 different episodes during the functioning and the cooling of this system, from 270-240° C to below 70° C. The high temperature episode represents the running of the geothermal reservoir. The temperature is similar and consistent to the actual geothermal fluid temperature at Bouillante. The low temperature evidences, which have not been identified yet at Bouillante, may represent the end and death of the reservoir suggesting a change of connectivity to the thermal resource.

ACKNOWLEDGMENTS

This paper is a contribution to the program GEOTREF funded by the French government in the frame of the program "Investissements d'Avenir" and tutored by ADEME. Partners of the GEOTREF project are Teranov, KIDOVA, Mines ParisTech, ENS Paris, GeoAzur, Georessources, IMFT, IPGS, LHyGes, UA, UCP-GEC.

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