

Fluid flow vs. scale of shear zones in the lower continental crust and the granulite paradox

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ABSTRACT

The external or internal origin of CO₂ and H₂O in marbles and metabasites from a large-scale granulite section is related to their structural setting: outside shear zones, within major shear zones (≈25 km wide by >340 km long), or within minor shear zones (<10 km wide by <150 km long). Outside shear zones, marbles have isotopic compositions similar to their protolith values and metabasites record mantle δ¹³C signatures. Marbles from major shear zones show little variation of δ¹⁸O but are depleted in ¹³C down to -3‰ due to exchange with mantle-derived carbon. In addition to an earlier input of mantle carbon related to mafic magmatism, major shear zones act as conduits for mantle carbon influx. They are probably mantle rooted. Oxygen was buffered by the crust. Marbles and their related skarns from minor shear zones were subjected to devolatilization reactions and water-rich infiltration from crustal sources with no evidence for a mantle carbon contribution. Large isotopic variations at the metre scale in the shear zones reflect the heterogeneous distribution of fluid flow associated with heterogeneous deformation and contrasting petrologic features and permeabilities. These geochemical-structural relationships reconcile the conflicting arguments on the origin of fluids during granulite genesis.

INTRODUCTION

Fluids in the lower crust have been addressed by many authors, leading to a variety of models concerning their composition (CO₂ and/or H₂O) and their flow (pervasive or channeled). Large-scale mantle-derived CO₂ fluxes are proposed for Bamble and Lofoten (Pineau et al., 1981; Baker and Fallick, 1988) and south India (Newton, 1987), in contrast to local crustally derived H₂O-CO₂ fluid infiltration for the Adirondacks (Valley et al., 1990) and the Ivrea zone (Baker, 1988). Most studies have not related the fluid characteristics to both local- and regional-scale tectonic structures. This paper examines fluid flow related to a crustal-scale shear-zone network within the lower crust based on a study of the granulites of Madagascar.

The Madagascar Archean and Proterozoic crust was reworked during the Pan-African event (530–630 Ma, Paquette et al., 1994). In southeast Madagascar, low-pressure granulite-facies lithologies related to postorogenic extension crop out over 100 000 km² (e.g., Nicollet, 1990). This basement is crosscut by granulite-facies vertical shear zones formed in a crustal-scale strike-slip system superimposed upon the overall granulite-facies flat strain pattern. Some of the U-Th ages of granulitic assemblages are younger (down to 400 Ma) in the major shear zones than outside (Nicollet et al., 1995). Structures have been mapped via remote sensing (Martelat et al., 1995), geological maps (Besairie, 1970), and field investigations. Deformation is localized into major (≈25 km wide by >340 km long) and minor (50 km long by 5 km wide up to 140 km long by 7 km wide) ductile shear zones (Fig. 1). Three major shear zones and half a dozen minor ones can be recognized. From field studies and petrology, no difference has been observed between minor and major shear zones. Rocks form ≈1–200-m-thick

layers parallel to the foliation. Retrograde alteration is only locally detectable. Carbonate-bearing rocks crop out widely both within and outside the shear zones. This study presents their C and O isotope geochemistry on a regional scale in order to (1) characterize the composition of the middle to lower crustal fluids, and (2) define the relationship between the structural setting and the nature and flow characteristics of the fluids.

SAMPLING STRATEGY AND PETROLOGY

Sixty-eight carbonate-bearing rocks, generally surrounded by metapelites, were sampled from outside and within three major and one minor shear zones and analyzed petrologically and isotopically. The rocks are divided into four groups: marbles with or without silicates, skarns (i.e., metasomatized limestones), carbonate-bearing metabasites, and clinopyroxenites (i.e., metasomatized metabasites).

Pure marbles with or without graphite have been rarely found either within major shear zones or to the east of shear zone A (Fig. 1). Silicate-bearing marbles (index minerals 2, 3, 4, and 5 in Table 1) with variable carbonate contents are common within and outside the shear zones. A wollastonite-bearing mylonite (4) has been observed in major shear zone C. Corona assemblages in minor shear zones show the late-stage reaction Fo + Cc + CO₂ + H₂O = Dol + various Mg silicates, which is responsible for the late production of dolomite (5). Where present, quartz is enclosed in diopside, hence the reaction Dol + Q = Di + CO₂ did not reach completion (5). Unreacted adjacent quartz and dolomite imply high XCO₂ outside the shear zones (2). The lack of quartz in the major shear-zone assemblages suggests high reaction progress triggered by deformation or fluid infiltration or both.

Data Repository item 9702 contains additional material related to this article.

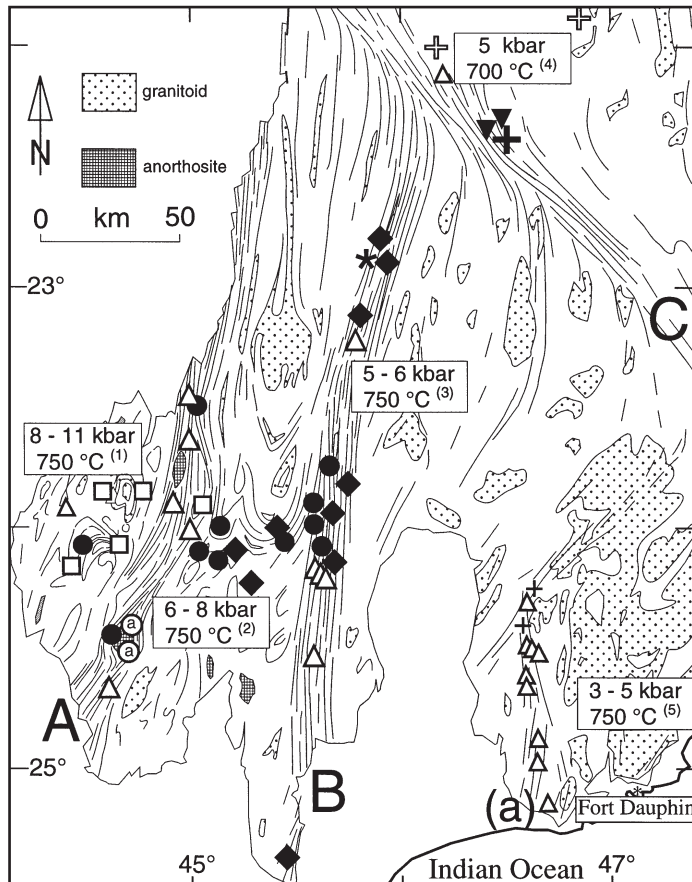


Figure 1. Structural sketch map of southeast Madagascar with foliation planes (lines), the three major shear zones (A, B, and C), the studied minor shear zone (a), and sample locations (see Fig. 2 for symbol legend). A given symbol may represent samples as much as 15 km apart. *P-T* data: (1, 2, 4) Nicollet (1990); (3) Ackermann et al. (1989), (5) Moine et al. (1985). Unmapped areas = sedimentary and volcanic rocks.

Metabasites have never been observed within a kilometre of marbles. Outside the shear zones, metabasites (index minerals 6 in Table 1) bear intergranular carbonates ($\approx 250 \mu\text{m}$) and minute replacement of pyroxene and plagioclase by carbonates (cryptic carbonatization) with no or only minor amphibole veins. Carbonatization of plagioclase mainly results from scapolite destabilization. Carbonatization of pyroxene requires metasomatic processes involving CO_2 . Carbonates from shear-zone metabasites (7) are intergranular or cryptic and are often associated with contemporaneous amphibole development in micrometre-scale veins and minute coronas around pyroxenes.

The clinopyroxenites (8 and 9, Table 1) formed during deformation of metabasites on a metre scale in structures that drained fluids (isoclinal fold hinges or interboudins), in association with amphibole + quartz pegmatites. Field investigations show no link between these carbonate-bearing rocks and any marble. In the clinopyroxenites, triple junctions between adjacent pyroxene and calcite grains show textural equilibrium, contrary to the carbonatized metabasites, which show minute secondary mineral replacement by carbonates. Phlogopite development in clinopyroxenites is related to the shearing event and lead locally to economic phlogopite deposits in the major shear zones.

STABLE ISOTOPES

Carbon and oxygen isotope analyses from carbonates are reported in per mil with reference to PDB (Peedee belemnite) and SMOW (standard mean ocean water) standards, respectively. Isotopic compositions and whole rock carbonate yields were measured¹ on CO_2 gas extracted via

H_3PO_4 (e.g., Sharma and Clayton, 1965). When present, scapolite contributes to the extracted CO_2 , but fractionations between scapolite and calcite are nearly zero (Moecher et al., 1994).

The C and O isotope composition of carbonates from marbles outside the shear zones and those within minor and major shear zones are shown in Figure 2A. Quartz is only present in the marbles with the highest isotopic values within a given structural setting. Little to no overlap occurs among the three fields. Outside shear zone values are grouped. Pure marbles and the quartz-bearing marble have higher $\delta^{13}\text{C}$ values (4 ± 1) than the silicate-bearing marbles (0.5 ± 0.5). Marbles from the minor shear zone follow a narrow band, down to $\delta^{13}\text{C} = -1$ and $\delta^{18}\text{O} = 11$. Marbles from major shear zones range from 0 to -4.4 for $\delta^{13}\text{C}$ and from 21 to 11 for $\delta^{18}\text{O}$. However, isotopic values are typically $\delta^{13}\text{C} > -3$ and $\delta^{18}\text{O} > 17$, except for one wollastonite-bearing mylonite (Fig. 2A). Within the marbles with $\delta^{18}\text{O} > 17$, no relationship is observed between either $\delta^{13}\text{C}$ or $\delta^{18}\text{O}$ values and silicate content. Most of the skarns display isotopic compositions in the same range as those of the minor shear-zone marbles (Fig. 2B).

Carbonates from mafic rocks (Fig. 2B) have $\delta^{13}\text{C} = -5.7 \pm 0.5$ for those located outside and -4.8 ± 1 for those located within shear zones. The $\delta^{18}\text{O}$ values are much more variable, related to petrology and structural setting as follows. Carbonated metabasites and clinopyroxenites are located outside shear zones and have $\delta^{18}\text{O} = 10.7$ to 13.5; carbonated and amphibole-veined metabasites and clinopyroxenites are located inside the major shear zones and have $\delta^{18}\text{O} = 14$ to 17.8. Mafic rocks located within major shear zones have been infiltrated by hydrous fluids resulting in formation of amphibole and phlogopite, and higher $\delta^{18}\text{O}$ values. A hydrothermal calcite-diopside lens associated with a giant phlogopite deposit in major shear zone B (Fig. 1) is of the same composition as carbonates from metasomatized mafic rocks (Fig. 2B).

DISCUSSION

Rocks Before Shearing Event and Fluid Reservoirs

The pure marbles outside shear zones, without silicate-driven decarbonation, are most likely to have been the least modified by infiltration and devolatilization. Their C and O isotope compositions could be inherited from their protoliths. The field shown in Figure 2A for marbles outside shear zones is in fact indistinguishable from that of Precambrian limestones (Veizer and Hoefs, 1976). The isotopic compositions of the silicate-bearing marbles outside shear zones (silicate $< 25 \text{ wt}\%$) could have been modified from their protolith values during devolatilization. Unreacted dolomite and quartz are preserved in the only high- $\delta^{13}\text{C}$ silicate-bearing marble. The initial carbonate composition is estimated by assuming that the rock was composed of carbonate plus quartz, and only forsterite formed according to the reaction $2\text{DoI} + \text{Q} + 2\text{Cc} = \text{Fo} + 4\text{Cc} + 2\text{CO}_2$. Among the possible reactions (Spear, 1993), the above one produces up to 25 wt% Fo and leads to the largest C and O isotope depletions. From their model forsterite content (i.e., all silicates taken as Fo [Table 1]), these marbles could have lost as much as 9 vol% of carbonate. Estimated δ values of the initial limestone, based on a Rayleigh distillation model, are enriched by $< 1.3\text{‰}$ in ^{13}C and $< 0.9\text{‰}$ in ^{18}O compared to their final values.

For the potential mantle reservoir, the range of C and O isotope values for carbonates in equilibrium with mantle rocks is taken as the reference ($\delta^{13}\text{C} = -3$ to -8 and $\delta^{18}\text{O} \approx 7$, Fig. 2B, e.g., Javoy et al., 1986). Carbonates are strictly minor to trace minerals in the metabasites (Table 1). The tightly clustered $\delta^{13}\text{C}$ values of the metabasites outside shear zones are within the mantle range and have most probably been controlled by their mantle source (Fig. 2B). The calculated $\delta^{18}\text{O}_{\text{calcite}}$ values in equilibrium with mantle-derived rocks ($\delta^{18}\text{O}_{\text{feldspar}} = 6$) at mantle to lower crustal temperatures are < 8 . The higher values require exchange with crustal rocks, probably via

¹GSA Data Repository item 9702, isotopic data of carbonates from Madagascar granulites, is available on request from Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301. E-mail: editing@geosociety.org

TABLE 1. SUMMARY OF RELEVANT MINERAL ASSEMBLAGES WITHIN THEIR STRUCTURAL SETTING

Lithology *	Index minerals †	Carbonate content (wt%)
<i>Outside shear zones:</i>		
Marble (4)	1: ± Graphite	94 - 100
Silicate-bearing marble (6)	2: Fo or Di ± Q	65 - 86
Skarn (2)	11: Di ± Sc	0.4 - 40
Metabasite (5)	6: Am ± Sc + Cc	0.1 - 2.0
Clinopyroxenite (5)	8: Am ± Sc + Cc	0.1 - 4.0
<i>Major shear zones:</i>		
Marble (1)	1: Graphite	92
Silicate-bearing marble (14)	3: ± Sc ± Q ± Graphite	43 - 95
	4: Wo ± Kf (mylonite)	59
Skarn (1)	11: Di + Sc	4.4
Metabasite (5)	7: ± Sc + Cc	0.2 - 1.1
Clinopyroxenite (8)	9: Ph + Cc ± Am	0.2 - 4.9
Carbonate lens (1)	10: Cc + Di	95
Anorthosite (2)	13: Pl + Di + Cc	0.1 - 1
<i>Minor shear zones:</i>		
Silicate-bearing marble § (6)	5: Fo or Di ± Sc ± Q	47 - 91
Skarn (8)	12: Fo or Di ± Sc ± Ch	1 - 18

* Number of samples given in parentheses.

† Index minerals that are referred to by number in the text.

§ Wt% silicate/Dol relationship: 9/12, 20/13, 27/5, 53/21.

Am: amphibole; Cc: calcite; Ch: clinohumite; Di: diopside; Dol: dolomite; Fo: forsterite; Kf: K-feldspar; Pl: plagioclase; Ph: phlogopite; Px: pyroxene; Q: quartz; Sc: scapolite; Wo: wollastonite.

water infiltration. Mass-balance calculations rule out closed-system exchange between carbonate and metabasite silicates (Chiba et al., 1989). The local development of clinopyroxenites with carbonates of a similar isotopic composition to that in the parent metabasites implies that at least part of the mantle-derived carbon was mobile on a metric scale outside the shear zones during granulite-facies metamorphism.

Minor Shear Zones

A well-defined isotopic trend for the minor shear zone is shown in Figure 2A. The $\delta^{18}\text{O}_{\text{calcite}}$ values vary by up to 4‰ on a metre scale. Rayleigh devolatilization can generate coupled $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ variations, as illustrated by the curved arrow. It represents an upper limit for our marbles with 22 vol% decarbonation and 53 wt% of diopside for the reaction



This model is not sensitive to temperature variations for $T > 350^\circ\text{C}$. The $\delta^{13}\text{C}$ values of our marbles can be explained by decarbonation reactions. The substantially larger observed $\delta^{18}\text{O}$ variations require additional water infiltration. Either infiltration follows decarbonation (reaction 1) or coupled isotopic variations occurred according to the reaction carbonate + silicate + $\text{H}_2\text{O} = \text{hydrous phase} + \text{silicate} + \text{CO}_2$. The correlation of the C-O isotopic shifts with the increasing silicate content, the disappearance of the high-XCO₂ paragenesis Dol + Q, and the decreasing, then increasing dolomite content (5, Table 1) support that infiltration was controlled by the degree of devolatilization. However, it is still unclear whether infiltration triggered or has been enhanced by devolatilization. Two other possible mechanisms are ruled out by the above relations: (i) a mixing process between our "un-sheared marble end-member" and a pole near $\delta^{13}\text{C} = -2$ and $\delta^{18}\text{O} = 11$, which is not known to be characteristic of any fluid reservoir or buffer, and (ii) the infiltration of fluids whose isotopic heterogeneity was externally controlled.

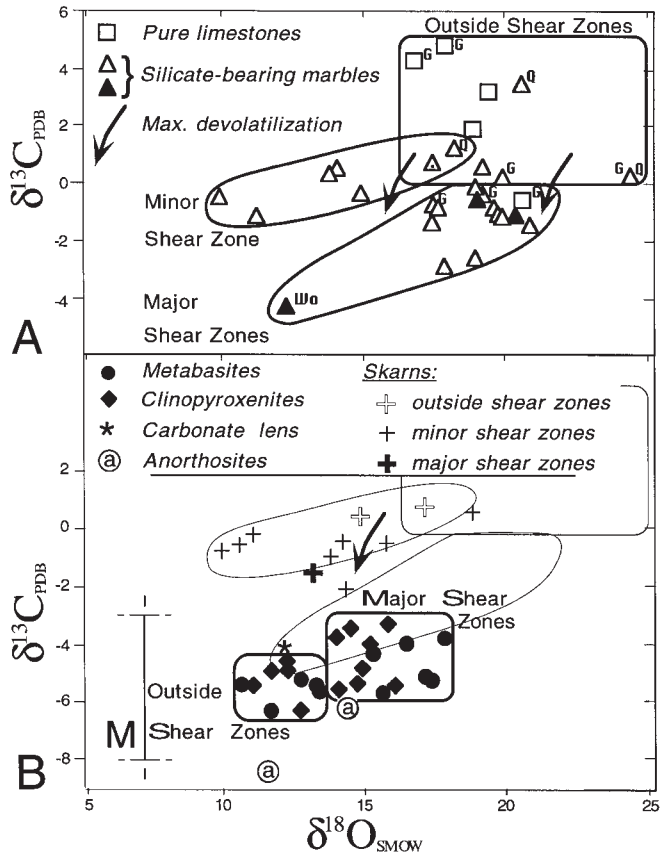


Figure 2. $\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ diagrams of carbonates. A: Carbonates from pure and silicate-bearing marbles. Fields refer to tectonic setting: outside, inside minor, and inside major shear zones. Superscript Q for dolomite + quartz, Wo for wollastonite, and G for graphite-bearing assemblages. The open triangle with dot comes from outside shear zone. Filled triangles are low strain to mylonitic samples from a 25 m traverse. Curved arrow: Rayleigh decarbonation. B: Carbonates from skarns, metabasites, and clinopyroxenites. M: Carbonates of mantle derivation.

Skarns show isotopic compositions either similar to those of the marbles from the minor shear zone or easily explained via Rayleigh devolatilization (Fig. 2B). We infer that metamorphism in marbles and metasomatism in skarns were related to comparable mechanisms based on devolatilization reactions and influx of H_2O within the minor shear zones. The initially homogeneous aqueous fluid had a calculated $\delta^{18}\text{O}_{\text{water}} \leq 11$ at 750°C (O'Neil et al., 1969); the lower the $\delta^{18}\text{O}_{\text{water}}$ value, the lower the fluid/rock ratio. Such relatively high $\delta^{18}\text{O}_{\text{water}}$ values imply that the external fluids were buffered by or originated from supracrustal rocks. The crystallization of abundant syntectonic migmatitic granites is a likely source for the fluid. Since it failed to homogenize the $\delta^{18}\text{O}$ rock values within the shear zones, even on a metre or finer scale, permeabilities must have been locally quite variable, arguing for a network of centimetre-wide channels rather than a larger single fluid path.

Major Shear Zones

Both the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ variations within the major shear-zone trend (Fig. 2A) are too large to be explained only by devolatilization. The maximum devolatilization effect on the carbon values is shown by the most depleted samples from the minor shear zones and is only attained via water-rich infiltration. Exchange with graphite is not responsible for ^{13}C depletion in major shear zones because all these marbles do not contain it (Fig. 2A). Graphite also occurs in some marbles outside shear zones, but these marbles are less depleted than within major shear zones. Infiltration of an externally derived $\text{CO}_2\text{-H}_2\text{O}$ fluid with a relatively low $\delta^{13}\text{C}$ value is required for all the

samples from major shear zones. The complete range of C and O isotope variations is observed in three variably deformed samples (Fig. 2A) collected along a 25 m section. Heterogeneous deformation is related to heterogeneous fluid flow. The most strained marble is the wollastonite-bearing mylonite (0.2 mm grain size), depleted in ^{13}C by 4‰ and in ^{18}O by 10‰ relative to the two coarse-grained (2 mm), isotopically similar samples, 15 and 25 m away. Wollastonite probably results from decarbonation reactions at lower $X\text{CO}_2$, such as $\text{Cc} + \text{Q} = \text{Wo} + \text{Cc} + \text{CO}_2$ but the observed ^{13}C and ^{18}O depletions are larger than those predicted by a Rayleigh model that produces 33 wt% Wo (i.e., all silicate taken as Wo). Thus mantle-type CO_2 infiltration is required in addition to crustal H_2O . The lowest $\delta^{18}\text{O}$ values recorded in major shear zones are similar to those from the minor ones and to the minimum ^{18}O enrichment in metabasites (Fig. 2B). This is consistent with a similar crustal control attributed to H_2O -bearing fluid circulation with $\delta^{18}\text{O}_{\text{water}} \approx 11$. Wollastonite development with $\delta^{18}\text{O}_{\text{calcite}} = 11.4$ thus corresponds to an extremely high water/rock ratio.

The only pure marble sampled in a major shear zone shows $\approx 4\%$ ^{13}C depletion (Fig. 2A) relative to its outside shear-zone counterparts: $\text{CO}_2 (\pm \text{H}_2\text{O})$ has been infiltrated. In the absence of devolatilization for enhancing permeability, fluid infiltration was nevertheless effective. The pure marble is among the marbles least depleted in ^{18}O , emphasizing that the silicate-bearing marbles have been more affected because of loss of $^{13}\text{CO}_2$ by devolatilization, which creates porosity that in turn enhances infiltration. In contrast to the H_2O -dominated process unraveled in the minor shear zone associated with high ^{18}O variations, isotopic evolution of marbles from within the major shear zones is limited for oxygen but larger for carbon, implying that CO_2 infiltration played a major role in this context. However, the skarn sampled in a major shear zone (Fig. 2B) records the same H_2O -rich metasomatic process and devolatilization as marbles from within the minor shear zones.

The amphibole veining and the giant phlogopite deposits suggest H_2O infiltration in the major shear zones. This is supported by the large $\delta^{18}\text{O}$ increase with little or no $\delta^{13}\text{C}$ variation of the metabasites compared to their counterparts outside the shear zones (Fig. 2B). Increasing $\delta^{18}\text{O}$ implies lower exchange temperatures, at least for the most-enriched samples. The marbles, metabasites, and pyroxenites are coherent in supporting the presence of mantle-derived CO_2 within the major shear zones plus mixing processes with marble-derived oxygen and carbon (Fig. 2B).

CONCLUSIONS

Metabasites with euhedral scapolite and the development of clinopyroxenites from metabasites both outside and within shear zones record early but local CO_2 circulation related to mafic magmatism that was independent from shearing events. However, there is no isotopic evidence for widespread, pervasive infiltration of CO_2 . Postdating the magma-related carbon influx, the regional shearing structures, divided into minor (<140 km long by 7 km wide) and major (>340 km long by 25 km wide) shear zones, focused crustal H_2O - CO_2 for the former and crustal H_2O - CO_2 plus mantle-derived CO_2 for the latter. The major shear zones are thus probably rooted in the mantle. The extent of infiltration was highly variable on a metre or finer scale, and the largest time-integrated fluxes were restricted to a network of metre- to centimetre-wide channels within the shear zones. The nature of the circulating fluids was controlled by the tectonic root of the shear zone, whereas variations in fluid flow must have been controlled by the local permeability, closely related to both deformation and the extent of decarbonation in carbonate rocks. The ^{18}O composition of the rocks from both the major and minor shear zones was controlled by supracrustal oxygen. $X\text{CO}_2$ values were locally highly variable. The apparent conflicting results for fluid composition, origin, and flow derived by previous authors can be reconciled by studying a large-scale section of the lower crust with an integrated geochemical and structural approach.

ACKNOWLEDGMENTS

Partially financed by the INSU-CNRS "Tectoscope" and DBT "Fluides dans la croûte" programs. We thank M. Émery for technical assistance, K. Rosen and R. Rakotondrazafy for kind collaboration and providing additional samples, and J. Ague and D. Rumble for helpful reviews.

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Manuscript received June 3, 1996

Revised manuscript received September 9, 1996

Manuscript accepted October 21, 1996