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Fluid circulation during the Variscan metamorphism of diabase dikes in the Rocroi Massif (Ardennes, France): a combined study of fluid inclusions and mass transfer of chemical elements

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ABSTRACT

Greenschist facies mafic dikes in the Cambro-Ordovician basement of the Rocroi Massif (southern Ardennes, France) have been used as quantitative sensors of fluid flow and fluid-rock interactions. After their consolidation, dikes have suffered a Variscan polyphase metamorphism under greenschist conditions. The major phase is a synclavage metamorphism registered only at the dike margins. The nature and volume of metamorphic fluids were first estimated by Potdevin *et al.* (1991a) from mass balance calculations. Results indicate the infiltration of an H₂O-CO₂ fluid, which caused carbonation of the metabasite assemblage. Quartz-calcite-chlorite veins occur in the metamorphosed dikes. They are contemporary with the paragenesis of calcite, chlorite, albite, quartz and rutile of the synclavage metamorphic stage and contain abundant fluid inclusions, some of them are rich in N₂. The fluid inclusion analysis shows the circulation in tension gashes, of three main fluid types during synclavage metamorphism. These fluid types can be classified in the following systems: H₂O-NaCl, H₂O-NaCl-N₂-CH₄-CO₂-C₂H₆-H₂ and N₂-CH₄-CO₂-C₂H₆-H₂. Results on early fluid inclusions are consistent with pressure estimations from paragenesis (P = 200 to 300 MPa). Fluid pressure between 150 and 420 MPa and temperature conditions were determined by combination of paragenesis data and chlorite geothermometry. The origin of these fluids is unclear. They could be the product of the devolatilization of enclosing rocks.

KEYWORDS

Diabase, fluid inclusion, infiltration, mass transfer, metamorphism

Introduction

Fluids play a fundamental role in numerous physicochemical processes during metamorphism (Fyfe *et al.*, 1978). Indeed, most of the metamorphic reactions need a fluid phase to progress. Fluids are often a catalyst or an essential component of chemical reactions and rock deformation. Their composition may control the metamorphic rock paragenesis. They can also induce large mass transfers of chemical elements at different scales within the crust. In the monotonous quartzite-schist series of the Rocroi Massif, diabase dikes are exceptional markers of fluid rock interactions during the Variscan orogenesis under greenschist conditions. The Variscan synclavage paragenesis developed only at the dike margins (quartz-calcite-chlorite-albite; T=300-380°C, Potdevin & Goffette, 1991b), whereas the core preserved an earlier static greenschist paragenesis (actinote-epidote-oligoclase, T=400-450°C, Potdevin & Goffette, 1991b). The core preserved also the chemical composition and texture of the dike before deformation. The comparison of the chemical, mineralogical and textural differences between the core and the margins indicates a main fluid infiltration episode (Potdevin *et al.*, 1994). The nature and the volume of these fluids were estimated by mass balance calculations (Potdevin & Goffette, 1991b).

The diabase dikes show numerous and large quartz-calcite-chlorite veins. These veins are potential markers of the Variscan fluids. The mineral paragenesis in the veins and in the dike margins are similar and are posterior to the first greenschist paragenesis of the core. Moreover, the veins are dilational and seem to be contemporaneous of the Variscan deformation. Indeed they mainly developed in the rigid core of the dike due to the ductile deformation at the margins. The euhedral quartz and calcite crystals are relatively unstrained and preserve many fluid inclusions which could have been trapped during the synclavage metamorphism. These fluid inclusions in quartz and calcite were investigated in order to: (1) estimate the temperature and pressure conditions during the metamorphic stages, (2) determine the nature, composition and origin of the palaeofluids in combination with mass transfer calculation.

Geological setting

The Ardennes area belongs to the northern external part of the Variscan orogen. The Carboniferous shortening episode results in an intense folding and the development of major thrusts. From south to north, the Ardennes allochthon overthrusts the Brabant Parautochthone. In both units, a Devonian cover unconformably overlies a Cambro-Ordovician basement. The latter outcrops in the Rocroi and Stavelot Massifs of the Ardennes allochthon. This basement, previously folded, was intruded by diabase dikes of middle to late Devonian age (Goffette, 1991). From the study of the stable elements, it appears that these diabases have a tholeiitic character and are very similar to continental tholeiites (Goffette *et al.*, 1990).

A progressive and continuous mineralogical and textural differentiation occurs between the core and the margin of these dikes. The core is a dense, fine-grained, green rock. An ophitic texture is preserved but all the magmatic minerals are replaced during a first metamorphic stage in the greenschist facies. This stage corresponds to the static equilibration of the dike with the enclosing rocks ($T = 450\text{--}400^\circ\text{C}$, $P = 200$ to 300 MPa based on the petrogenesis and the presence of garnet in the enclosing rocks; Potdevin *et al.*, 1993). It is related to the Variscan stretching which occurred during the Ardennes margin development and indicates a high geothermal gradient of about $50\text{--}55^\circ\text{C}/\text{km}$.

Afterwards, a lower temperature metamorphism occurred, also under greenschist conditions. The wall of the diabase is highly strained. Towards the margins, the syncleavage paragenesis of calcite, chlorite, albite, quartz and rutile replaces progressively actinote, oligoclase, ilmenite and titanite that are only preserved in the undeformed core of the dike.

Results

Fluid inclusion study

Quartz-calcite-chlorite veins were sampled from different diabase dikes of the Rocroi Massif. Fluid inclusions were studied both in quartz and calcite crystals by microthermometry and Raman spectroscopy. When possible, particular attention was devoted to euhedral quartz crystals to constrain the fluid inclusion chronology. Raman analysis was performed on selected fluid inclusions at the "Laboratoire de Spectrochimie Infrarouge et Raman" (LASIR-CNRS, University of Lille). Inclusions were first classified according to their chemical composition. Eight types of fluid inclusions are present in quartz-calcite-chlorite veins: $\text{H}_2\text{O-NaCl}$, $\text{H}_2\text{O-NaCl-N}_2\text{-CH}_4\text{-CO}_2\text{-C}_2\text{H}_6$, $\text{H}_2\text{O-NaCl-N}_2\text{-H}_2$, $\text{H}_2\text{O-NaCl-N}_2\text{-CH}_4$, $\text{H}_2\text{O-NaCl-CO}_2\text{-N}_2$, $\text{CH}_4\text{-N}_2\text{-C}_2\text{H}_6$, $\text{CH}_4\text{-N}_2\text{-CO}_2$, and $\text{CH}_4\text{-N}_2\text{-H}_2$ inclusions. The relative chronology of these fluids and the correlation of the fluid inclusion families in the different veins are still unclear. Nevertheless, $\text{CH}_4\text{-N}_2\text{-CO}_2$, $\text{CH}_4\text{-N}_2\text{-H}_2$, $\text{CH}_4\text{-N}_2\text{-C}_2\text{H}_6$ and some $\text{H}_2\text{O-NaCl}$ inclusions are undoubtedly secondary. The other inclusions are clearly primary or pseudo-secondary. Thus, they are contemporaneous with the quartz-calcite-

chlorite assemblage and can therefore be used as geothermobarometers.

Moreover, HCO_3^- , CO_2 , CH_4 and H_2S have been detected in the liquid phase of fluid inclusions by Raman spectrometry.

Thermobarometry

The temperature range obtained from the metamorphic assemblage is $300\text{--}380^\circ\text{C}$ (Potdevin *et al.*, 1994). In addition, chlorite-quartz geothermometry (Vidal *et al.*, 2001) gives a temperature range of 350 to 440°C . Fluid pressures between 150 and 420 MPa are estimated for some early aqueous, $\text{H}_2\text{O-NaCl-CO}_2\text{-N}_2$ and $\text{H}_2\text{O-NaCl-N}_2\text{-H}_2$ fluid inclusions. They are consistent with pressure conditions estimated by parageneses. Molar fractions for $\text{H}_2\text{O-NaCl-N}_2\text{-CH}_4\text{-CO}_2\text{-C}_2\text{H}_6$ and $\text{H}_2\text{O-NaCl-CH}_4\text{-N}_2$ primary and pseudosecondary inclusions must be estimated in order to calculate isochores for these inclusions. Data on primary aqueous fluid inclusions (Homogenization temperatures) are distributed in a large range (170 to 325°C) but two modes can be distinguished: $190\text{--}200^\circ\text{C}$ and $250\text{--}270^\circ\text{C}$. These values can be used to generate temperature-pressure curves. The isochore generated by the 195°C -mode of early aqueous fluid inclusions and by the $\text{H}_2\text{O-NaCl-CO}_2$ fluid inclusions is consistent with the temperature-pressure box given by the metamorphic assemblage ($T=300\text{--}380^\circ\text{C}$; Potdevin *et al.*, 1994). The 270°C -mode of primary aqueous inclusions is more difficult to explain. It could only be explained either by a temperature increase or a drastic pressure drop. The latter hypothesis seems to be more realistic. Indeed, the large homogenisation temperature range can be explained by the stretching of the inclusions (and consequently a density drop). Moreover, the late $\text{CH}_4\text{-N}_2\text{-CO}_2$ fluid indicates low pressure conditions (less than 50 MPa) at the end of the fluid evolution. This drastic pressure drop could be due to a rapid erosion of the Variscan orogenic belt or a post-orogenic extension.

Fluid-rock interactions

The comparison of fluid inclusions data and mass transfer calculations provides information about the Variscan fluid-rock interactions within the dikes. Mass balance computations using the Gresens method provide the amount of CO_2 and H_2O consumed by the calcite and the chlorite in the dike margins (respectively 7 and 1 weight percent of the initial rock mass). They suggest that this massive calcite and chlorite precipitation requires an $\text{H}_2\text{O-CO}_2$ fluid. A small amount of CO_2 could result from CH_4 oxidation related to iron reduction during the replacement of epidote by chlorite. The high amount of CO_2 and H_2O consumed inside the dike implies a volumetric fluid/rock ratio higher than 0.5 (Potdevin *et al.*, 1991b). This fluid/rock ratio is 50 times greater than the rock porosity and demonstrates a large fluid infiltration in the dike. According to the results of the mass balance computations, the concluding remarks of the fluid inclusion study are:

- CO_2 -bearing inclusions are present in the quartz-calcite-chlorite veins but the CO_2 is generally in low proportion. This could be due to either a consumption of CO_2 to precipitate calcite or to a lack of trapping of the CO_2 -bearing

fluid. As predicted by mass transfer computation, CH₄ is a potential source of carbon in our system.

- Another surprising points are the abundance of N₂ all along the fluids history and the presence of H₂ in N₂-rich inclusions. N₂ is present in early (primary or pseudo-secondary) water-salt inclusions (H₂O-NaCl-N₂-H₂) as well as in secondary inclusions (low density CH₄-N₂-C₂H₆, CH₄-N₂-CO₂, and CH₄-N₂-H₂ inclusions). Two episodes of N₂ fluid input in the system are therefore recognised. This indicates that N₂ was a major component of the fluid system. Its origin could be searched for in the surrounding Cambro-Ordovician metapelites. The presence of hydrogen together with nitrogen in fluid inclusions could be explained by their same origin. Indeed, the release of NH₄⁺ from micas within schists could provides N₂ and H₂ to the fluid. The occurrence of hydrogen suggests that the fluid was not allowed to reequilibrate with the enclosing rocks at the time of trapping.

Conclusions and perspectives

The understanding of fluid-rock interactions during metamorphism requires a multidisciplinary approach. This study combine petrological data, mass transfer computations and fluid inclusion data from metamorphosed dikes and related veins of the Rocroi Massif to understand a major episode of fluids circulation during the Variscan orogenesis. The pressure and temperature conditions of this episode are relatively well constrained by metamorphic mineral assemblages, chlorite geothermometry and fluid inclusion data (T = 300-440 °C and P = 150-420 Mpa). The detailed fluid inclusion study of quartz-chlorite-calcite veins reveals a more complex set of metamorphic fluid compositions than predicted by mass transfer computations. H₂O (including dissolved salts) and surprisingly N₂ are the main component of the fluids. N₂ could result of an ammonium release from white micas of the enclosing schists during the Variscan metamorphism. CO₂ is present either in rare water-carbon dioxide inclusions or as a minor component in addition to N₂ and CH₄. CH₄ could be another possible source of carbon for the massive precipitation of calcite and chlorite in the dikes studied. Further studies on the stable isotopic compositions of calcite and quartz combined with the analysis of the possible nitrogen sources in the surrounding rocks could provide more information on the origin of these Variscan fluids.

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