

## FLUID CIRCULATION DURING THE VARISCAN METAMORPHISM OF DIABASE DIKES IN THE ROCROI MASSIF (ARDENNES) : A COMBINED STUDY OF FLUID INCLUSIONS AND MASS TRANSFER OF CHEMICAL ELEMENTS

COQUINOT, Y.<sup>1</sup>, DUBOIS, M.<sup>1</sup>, POTDEVIN, J.-L.<sup>1</sup>, LAUREYNS, J.<sup>1</sup>

<sup>1</sup>UMR CNRS 8110 "Processus et Bilans des Domaines Sédimentaires" PBDS et FDR 1818 "Milieux Naturels et Anthropisés", Université de Lille 1, 59655 Villeneuve d'Ascq CEDEX, France.

E-mail: Yvan.Coquinot@univ-lille1.fr

### Introduction

In the monotonous Cambro-Ordovician quartzite-schist series of the Rocroi Massif (southern Ardenne, France), diabase dikes are exceptional markers of fluid-rock interactions during the Variscan orogenesis. The nature and volume of metamorphic fluids were first estimated by Potdevin et al. (1991a) from mass balance calculations. Results indicate the possible infiltration of an H<sub>2</sub>O-CO<sub>2</sub>-(CH<sub>4</sub>) fluid, which caused carbonation of the metabasite assemblage at the dike margins. Quartz-calcite-chlorite and quartz-albite-chlorite veins occur in the metamorphosed dikes and contain abundant fluid inclusions which were investigated in order to:

- 1/ determine the nature, composition and origin of the palaeofluids in combination with mass transfer calculation
- 2/ estimate the temperature and pressure conditions during the metamorphic stages.

### 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, a particular attention was devoted to euhedral quartz crystals to constrain the fluid inclusion chronology. Inclusions were first classified according to their chemical composition. Preliminary results of fluid inclusions study show that a more complex set of fluid compositions than predicted by mass balance calculation infiltrated the diabase dikes during the syntectonic metamorphism. The different types of fluids can be classified in the following systems: H<sub>2</sub>O-NaCl, H<sub>2</sub>O-NaCl-N<sub>2</sub>-CH<sub>4</sub>-CO<sub>2</sub>-C<sub>2</sub>H<sub>6</sub>-H<sub>2</sub> and N<sub>2</sub>-CH<sub>4</sub>-CO<sub>2</sub>-C<sub>2</sub>H<sub>6</sub>. Nevertheless, all these fluids can be classified in three main types: aqueous fluids, CO<sub>2</sub>-rich fluids and N<sub>2</sub>-CH<sub>4</sub> rich fluids. The relative chronology of these fluids and the correlation of the fluid inclusion families in the different veins are still unclear. Nevertheless, CH<sub>4</sub>-N<sub>2</sub>-CO<sub>2</sub>, CH<sub>4</sub>-N<sub>2</sub>-H<sub>2</sub>, CH<sub>4</sub>-N<sub>2</sub>-C<sub>2</sub>H<sub>6</sub> and some H<sub>2</sub>O-NaCl inclusions are undoubtedly secondary and most of the others are primary or pseudo-secondary.

Moreover, HCO<sub>3</sub><sup>-</sup>, CO<sub>2</sub>, CH<sub>4</sub> and H<sub>2</sub>S have been detected in the liquid phase of fluid inclusions by Raman spectrometry

### Thermobarometry

Pressure-temperature conditions of the syntectonic metamorphism have been estimated using combination of fluid inclusion results but also, chlorite geothermometry, oxygen isotope geothermometry and mineral paragenesis. The temperature range obtained from the metamorphic assemblage is 300-380°C (Potdevin et al., 1994). Chlorite geothermometers (Cathelineau, 1988, Jowett, 1991) give a temperature range of 280 to 440°C and oxygen isotope geothermometry for the quartz-calcite pair gives a temperature range of 214 to 550 °C depending on the equation used. Fluid pressures between 150 and 420 MPa are estimated for some early aqueous, H<sub>2</sub>O-NaCl-CO<sub>2</sub>-N<sub>2</sub> and H<sub>2</sub>O-NaCl-N<sub>2</sub>-H<sub>2</sub> fluid inclusions. They are consistent with pressure conditions estimated by mineral paragenesis.. The late CH<sub>4</sub>-N<sub>2</sub>-(CO<sub>2</sub>) fluid indicates low pressure conditions (less than 50 MPa) at the end of the fluid evolution. This drastic pressure drop could be due to an opening of the system.

### Fluid-rock interactions

The comparison of fluid inclusion 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<sub>2</sub> and H<sub>2</sub>O 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 H<sub>2</sub>O-CO<sub>2</sub> fluid. A small amount of CO<sub>2</sub> could result from CH<sub>4</sub> oxidation related to iron reduction during the replacement of epidote by chlorite. The high amount of CO<sub>2</sub> and H<sub>2</sub>O 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<sub>2</sub>-bearing inclusions are present in the quartz-calcite-chlorite veins. Nevertheless, it is generally at low proportion except for one dike where CO<sub>2</sub> constitutes more than 80% of the gaseous phase. As predicted by mass transfer computation, CH<sub>4</sub> could be a potential source of carbon in our system.

- Another surprising points are the abundance of N<sub>2</sub> all along the fluids history and the presence of H<sub>2</sub> in N<sub>2</sub>-rich inclusions. N<sub>2</sub> is present in early (primary or pseudo-secondary) water-salt inclusions (H<sub>2</sub>O-NaCl-N<sub>2</sub>-H<sub>2</sub>) as well as in secondary inclusions (low density CH<sub>4</sub>-N<sub>2</sub>-C<sub>2</sub>H<sub>6</sub>, CH<sub>4</sub>-N<sub>2</sub>-CO<sub>2</sub>, and CH<sub>4</sub>-N<sub>2</sub>-H<sub>2</sub> inclusions). Two episodes of N<sub>2</sub> fluid input in the system

are therefore recognised. This indicates that N<sub>2</sub> was a major component of the fluid system. Its origin could be searched for in the surrounding Cambro-Ordovician metapelites like probably CH<sub>4</sub> and CO<sub>2</sub>. The presence of hydrogen together with nitrogen in fluid inclusions could be explained by a common origin. Indeed, the release of NH<sub>4</sub><sup>+</sup> from micas within schists could provide N<sub>2</sub> and H<sub>2</sub> to the fluid. The occurrence of hydrogen suggests that the fluid was not to reequilibrated 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 combines petrological data, mass transfer computations and fluid inclusion data from metamorphosed dikes and related veins of the Rocroi Massif. 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. H<sub>2</sub>O (including dissolved salts) and surprisingly N<sub>2</sub> are the main component of the fluids. N<sub>2</sub> could result from a release of NH<sub>4</sub><sup>+</sup> from white micas of the enclosing schists during the Variscan metamorphism. CO<sub>2</sub> is present either in rare water-carbon dioxide inclusions or as a minor component in addition to N<sub>2</sub> and CH<sub>4</sub>. CH<sub>4</sub> 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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