Advances in C-O-H-N-S fluid geochemistry based on micro-Raman spectrometric analysis of fluid inclusions

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Abstract: The first part of this paper focuses on the analysis of fluid inclusions by micro-Raman spectrometry. \(\text{SO}_4^{2-}\) and \(\text{HS}^-\) are the only polyatomic anions identified by this technique. \(\text{Na}^+, \text{Ca}^{2+}, \text{Mg}^{2+}, \text{Fe}^{2+}\) cations can be identified by the Raman spectrum of the corresponding salt hydrate which nucleates on cooling. Gas analysis is the most fruitful field of application of micro-Raman spectrometry. Errors in the reconstruction of the bulk V-X properties of gas-bearing fluid inclusions, arising either from Raman analysis or from the quantitative interpretation of phase equilibria, are discussed.

Geochemical constraints inferred from these analyses are considered in the second part. The V-X properties of fluids in the C-O-H-N-S system are deduced mainly from room-temperature measurements. They are shown to be representative of the fluid V-X properties in the P-T conditions of trapping, and thus, to yield representative \(\text{f}_02\) and \(\text{f}_\text{S}_2\). The paleo-redox state of fluids associated with U, Sn, W, and Au deposits is shown partly to account for the contrasted behaviour of these metals at the hydrothermal stage. Gas concentration is a key parameter for controlling metal transport and deposition properties of fluids. This is because it controls the static dielectric constant of the fluid, which in turns constrains the ion-pair stability. It is shown that some N-bearing fluids may be the end product of major redox reactions. Finally, the H\(_2\)- and O\(_2\)-bearing fluid inclusions found in three uranium deposits provide evidence for water radiolysis by alpha particles.

Key-words: fluid inclusions, micro-Raman analysis, C-O-H-N-S system, chemical equilibrium, Sn-W-U-Au-deposit.

Introduction

Major mass and heat transfer processes involved in the formation of rocks occur via a fluid phase. In many petrological studies, the composition of the fluid phase is inferred from mineral equilibria (Ferry & Burt, 1982; Rice & Ferry, 1982). However, the activity, and hence the concentration of the major fluid components, cannot often be sufficiently constrained by mineral assemblages, either because the variance of the system is too high or thermodynamic data are lacking. In addition, many studies of metamorphic terranes deal with maximum P-T-X conditions, whereas it is often difficult to decipher the retrogressive evolution from the mineral assemblages alone.

An independent characterization of paleofluids is a prerequisite for the reconstruction of the complex P-T evolution of metamorphic terranes (Hollister et al., 1979). It has been known for a long time that paleofluids are available for investigation in intracrystalline cavities designated as fluid inclusions (Sorby, 1858). However, a given crystal may have trapped several generations of fluids forming inclusions with different molar volumes and compositions. Consequently, it is necessary to determine the V-X properties of individual fluid inclusions to obtain reliable P-T-X and geochemical data. Detailed petrographical description and microthermometric study of the different fluid inclusion generations provide important complementary information. However,