Experimental study of the thermal stability of pyrophyllite, paragonite, and clays in a thermal gradient

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Abstract: Tube-in-tube experiments involving a strong thermal gradient have been conducted in the system Na$_2$O-Al$_2$O$_3$-SiO$_2$-H$_2$O, using a starting mixture made of quartz ± paragonite ± pyrophyllite. Whatever the starting proportions pyrophyllite:paragonite:quartz or the location of the starting assemblage in the thermal gradient, new clay minerals crystallized along the thermal profile. The newly formed phases were beidellite ± quartz at the cold extremity and, depending on the paragonite to pyrophyllite to quartz starting proportions, mica or pyrophyllite ± diaspore crystallized at the hot extremity of the tube. Rectorite was observed to crystallize at an intermediate position (intermediate temperature). SEM observations and microdiffraction data indicate that Na-beidellite broke down into rectorite at about 360°C, and rectorite broke down into pyrophyllite + paragonite at about 420°C. These results are consistent with synthesis experiments, using a gel of beidellite composition as starting material. On the other hand, attempts to reverse these reactions by isothermal, isobaric experiments failed or gave contradictory results. Although dealing with larger experimental uncertainties, tube-in-tube experiments seem to be well-adapted to investigate the phase relations and the temperature-composition stability relations of minerals which are difficult to obtain by reversing experiments. The experimental results are consistent with thermodynamic calculations and indicate that local solid-solution heterogeneous equilibria control the composition of solids that crystallized as well as their distribution along the thermal profile.

Key-words: beidellite, rectorite, thermal gradient, tube-in-tube experiments, system NASH, experimental petrology, clay minerals.

Introduction

Hydrothermal tube-in-tube experiments involving strong thermal gradients (Goffé et al., 1987; Robert & Goffé, 1989; 1993; Vidal, 1994; Vidal et al., 1995) were initially designed to determine if mass transfer between solids and solution and mass transport induced by thermal gradients within closed systems could account for the spatial distribution of naturally occurring hydrothermal sequences of crystallization. In early experiments, the transport of elements in solution appeared to be significant, as shown by the abundance of secondary minerals crystallizing in the middle part of the tube (initially empty at the beginning of the experiments). The spatial distribution of newly-formed minerals and the variation of their compositions along the thermal profile was in fair agreement with those observed in nature. An interesting issue that has not been addressed in the previous studies is the use of tube-in-tube experiments to study the isobaric phase relations among minerals as a function of temperature. This technique may be used to determine the equilibrium conditions for univariant P-T reactions that are difficult or impossible to