Mechanisms of isotopic equilibration during partial melting: an experimental test of the behaviour of Sr

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Introduction

It is generally admitted that isotopic equilibrium is attained during partial melting and that the isotopic composition of melt produced is identical to that of the source rock (e.g. Faure and Powell, 1972; Hofmann and Hart, 1978). In order to investigate mechanisms and kinetics of isotopic equilibration, we have set up an experimental program testing the behaviour of Sr during the initial stages of partial melting of a model feldspar-mica source mineral assemblage. Mineral couples having contrasted Sr concentrations and isotopic compositions were partially melted and the products (including melt quenched as glass) analysed with SIMS.

Experiments and analyses

Starting materials consisted of natural plagioclase single crystals of gem quality and of stoichiometric fluorphlogopite monocrystals, synthesized with a specific batch melting technique (Hammouda *et al.*, 1994). Fluorphlogopites were doped in Sr during synthesis, using a mixture of $^{87}SrCO_3$ and $^{86}SrCO_3$. Sr concentration was chosen to match the available values for Kd_{Sr} between mica and plagioclase. Sr isotopic composition was selected to be in sharp contrast with that of plagioclase, and also to approach compositions of old natural micas. Partial melting experiments were performed using plagioclase-fluorphlogopite couples at 1 atm in air between 1100 and 1250°C for durations up to 336 h. Mineral couples were introduced in the furnace directly at run temperature and quenched into water at the end of the experiment.

Run products were examined by SEM. They were analysed for major elements by electron microprobe and for Sr by SIMS (concentration and isotopes). A Cameca *ims 3f* ion microprobe was used with a primary beam of negatively charged oxygen ions, accelerated to 10 kV with a current of 5–7 nA. Spot diameter ranged from 10 to 25 μm. Mass resolution was set to 4500. Two counting procedures were used, respectively for

the measurement of Sr isotopic ratios and Sr concentrations. $^{87}Sr/^{86}Sr$ ratios were obtained from counting on 5 masses (84.6 for background correction, 85 to determine the correction to be applied because of the presence of trace Rb, 86, 87 and 88). Mass 88 was used as a reference. Instrumental fractionation factor was determined through the measurement of $^{86}Sr/^{88}Sr$ of the plagioclase. An offset of −60 V was applied for Sr concentration measurements which were determined from a calibration curve of $^{88}Sr/^{30}Si$ constructed using standards of known Sr concentration. Sr concentrations are precise to ~1 ppm and isotopic compositions to 0.6 % (2σ) from measurements on a plagioclase standard. Scanning across samples of synthetic mica and starting plagioclase show that they are fairly homogeneous in Sr (mica : 77 ± 7 ppm; plagioclase : 601 ± 4 ppm, Table 1). Analysis of the starting plagioclase yields $^{87}Sr/^{86}Sr = 0.701 \pm 0.003$, in agreement with results from mass spectrometry (0.703552 ± 18). Determination of $^{87}Sr/^{86}Sr$ of the mica is hampered by its low Sr concentration especially when working at high mass resolution. Average of 14 measurements yields $^{87}Sr/^{86}Sr = 4.0 \pm 0.4$ in agreement with mass spectrometry determination (4.254 ± 8).

Results

Run products include melt quenched to glass, a newly formed plagioclase, spinel, and occasionally forsterite. Depending on run temperature and duration, the mica is either totally or partially consumed, whereas plagioclase always persists in the product assemblage. The melting reaction may be described as

fluorphlogopite + plagioclase1 = liquid + spinel/forsterite + plagioclase2

Major elements, Sr concentrations and isotopic compositions have been determined along traverses through the reaction zone. Two cases need to be distinguished depending on the presence or absence of the mica in the product assemblage.

In the first case (mica still present), the glass displays a strong chemical zonation characterized from the mica to the plagioclase interface by decreasing K_2O, SiO_2, F, MgO and increasing Al_2O_3, CaO and Sr (1200°C, 10 h). The glass is also zoned isotopically with $^{87}Sr/^{86}Sr$ ranging from 1.23 to 1.18 from the mica to the plagioclase interface. In the heterogeneous region consisting of a mixture of Plag2 and glass, $^{87}Sr/^{86}Sr$ continues to decrease progressively. When the unreacted plagioclase is encountered the $^{87}Sr/^{86}Sr$ decreases abruptly to that of the starting plagioclase. $^{87}Sr/^{86}Sr$ of the mica remains unchanged. It is worth noting that measured Sr isotopic ratios of the liquid are markedly higher (1.18−1.23) than that of the bulk starting assemblage. Taking the masses and Sr concentrations and isotopic compositions of reactants, a value of $^{87}Sr/^{86}Sr$ = 0.859 is obtained for the mica-plagioclase experimental source assemblage. The isotopic ratio of the liquid does not seem to vary strongly with time ($^{87}Sr/^{86}Sr$ = 1.25 , T = 1200°C, t = 1 h).

In the second case (mica totally consumed) the chemical (major elements and Sr) zonation of the liquid is weaker. Sr isotopic compositions show no systematic zonation from the plagioclase interface (T = 1200°C, t = 24 h : average value 1.09 ± 0.01). They evolve systematically with time towards lower $^{87}Sr/^{86}Sr$ (T = 1200°C : 0.88, 96h and T = 1250°C : 0.98, 4 h; 0.89, 24 h). The $^{87}Sr/^{86}Sr$ of the melt still differs but approaches the one of the bulk starting assemblage (0.859). The newly formed plagioclase has a Sr isotopic composition (T = 1250°C, t = 24 h, $^{87}Sr/^{86}Sr$ plag2 = 0.94) comparable with the liquid ($^{87}Sr/^{86}Sr$ liquid = 0.89). The unreacted plagioclase retains its initial isotopic composition.

Discussion

Our experiments show that the kinetics of melting of the different mineral phases and the stoichiometry of the melting reaction are the main factors controlling the Sr isotopic composition of the liquid. For example at 1200°C, $^{87}Sr/^{86}Sr$ of the liquid is the highest for the shortest run duration (1.25, 1 h) then decreases progressively with time (1.09, 24 h and 0.88, 96h). This reflects the initially high proportion of mica consumed in the melting reaction. Values of $^{87}Sr/^{86}Sr$ of the liquid are always higher than expected in the case of isotopic equilibrium between source and melt. Therefore, our experiment demonstrates the possibility (for polymineralic rocks not previously homogenized for Sr isotopes) of a control of the Sr isotopic composition of the liquid by the nature and proportion of minerals consumed, rather than bulk source assemblage.

Dissolution kinetics of most minerals are controlled by chemical diffusion in the liquid, isotopic homogenization of the liquid by tracer diffusion in the liquid and isotopic equilibration of the minerals by tracer diffusion in the solids. From the available data we expect that (1) the melt tends to homogenize faster in Sr isotopes than in major and trace elements, and (2) that minerals melt faster than they equilibrate isotopically. This is consistent with our observations concerning the respective magnitude of isotopic and chemical zonations of the liquids and the fact that the isotopic compositions of the solids remain unchanged while melting occurs.

Given the slow tracer diffusivities in solids, long residence times of liquids at their source are required for isotopic homogenization of residual minerals by diffusion. If isotopic homogenization occurs through another process (like dissolution-precipitation) then timescales of isotopic equilibration may be reduced.

Conclusion

We have experimentally determined the Sr isotopic composition of liquids produced by partial melting of a model feldspar-mica assemblage. The results demonstrates the possibility of a control (at least for experimental timescales) of the Sr isotopic composition of the liquid by the nature and proportions of minerals consumed, rather than the experimental bulk source assemblage. For source rocks not previously homogenized for Sr isotopes, kinetics of melting of minerals and stoichiometry of melting reactions must be taken into account. Liquids having isotopic compositions differents from the bulk source can be formed in this way. These liquids need to equilibrate by diffusion with the residual mineral assemblage to approach bulk isotopic equilibrium.

References

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