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Chemical diffusion of fluorine in phlogopite

KAI ZHANG[1], HANYONG LIU[1], JANNICK INGRIN[2], DR. BAOHUA ZHANG, PHD[3] AND XIAOZHI YANG[1]

[1]Nanjing University
[2]Univ. Lille, CNRS, INRAE, ENSCL, UMR 8207 - UMET - Unité Matériaux et Transformations
[3]Zhejiang University

Presenting Author: zhangk@smail.nju.edu.cn

Fluorine is a minor yet important component of volatiles in silicate minerals, and affects profoundly their physicochemical properties. The diffusion of fluorine provides crucial information for quantitatively understanding many of its effects, yet very few diffusion data of fluorine have been reported. We have measured the chemical diffusion of fluorine in phlogopite, by experimental studies of chemical exchange between single crystal samples. Experiments were carried out with the diffusion couple technique at 1 GPa and 700 to 800 ºC for run durations of 25 to 264 hours. Concentration profiles of fluorine in the recovered samples and compositions of other major and minor elements were determined by electron microprobe, and water contents were analyzed by Fourier-transform infrared spectroscopy. The data demonstrate that, over the studied conditions, the fluorine diffusivity ranges between 10^{-19} and 10^{-17} m^2/s, being slightly anisotropic, and the measured activation energy is 176 to 261 kJ/mol along different directions. The diffusion is fast along the direction $\perp(110)$ and slow along the direction $\perp(001)$, with the direction $\perp(010)$ falling between them. By comparison with available reports on the diffusion of other species in phlogopite and of fluorine in other minerals, the chemical diffusion of fluorine in phlogopite suggests a strong enhancement of diffusion along the interlayer direction and thus diffusion anisotropy in the presence of molecular H_2O. The modeled electrical conductivity of phlogopite, by using the Nernst-Einstein relation and our diffusion data, is notably lower than that experimentally measured in phlogopite of the same fluorine content and dominated by fluorine conduction. This indicates that the self diffusion of fluorine in phlogopite is much faster than the chemical diffusion, and/or the mobility of fluorine differs greatly between diffusion and conductivity experiments. The diffusion data provide additional constraints on the closure temperature of fluorine in phlogopite, which is critical for the kinetic analyses of preserved fluorine zonations in natural phlogopites.