Chemical fractionations induced by irradiation in early solar materials : results from laboratory experiments

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Mineral phases from circumstellar environments are subjected to irradiation by energetic particles (ions, electrons, photons). Irradiation induces fractional volatilization of chemical elements in silicates; if large scale directional flow occurs, such as predicted by numerous models in the early solar system, then large scale chemical fractionnations occur.

In series of electronic irradiation experiments carried out in both transmission and scanning electron microscopes at 300 kV and 30 kV respectively on olivines and pyroxenes, complex differential volatilization regimes have been evidenced as a function of electron fluences and accelerating voltages (Ph. Carrez et al., Phil Mag A, 2001, 81, 2823-2840; L. Lemelle, L. Beaunier, S. Borensztajn, M. Fialin, F. Guyot; an experimental study of the destabilization of olivine single crystal by 30 keV electron irradiation : a possible mechanism of space-weathering affecting interplanetary dust and planetary surfaces, submitted manuscript).

In most regimes at 300 kV, the order of electron-induced elemental loss from the silicate matrix is O>Mg>Si>Fe. An important mechanism related to that behaviour observed in the case of olivine irradiation at 300 kV is the creation of abundant Frenkel pairs in the material. In other regimes, particularly at lower accelerating voltages, the relative irradiation-induced volatilization order is O>Si>Mg,Fe.

We show that these results, particularly those obtained at 300 kV accelerating voltage, can be understood by combined effects of dielectric properties and bonding energies in the silicate materials which allow to derive tentative majoritary chemical trends in strongly irradiated materials in the early solar system characterized by:

- chemical reduction due to oxygen loss resulting in metal formation

- preferential loss of magnesium and enrichment in silicon, resulting in increasing the pyroxene to olivine ratio

- enrichment into high field strength lithophile elements
- depletion into alkaline and earth-alkaline elements.

Reactive Transport in Fractured Saprolite: Determining Diffusive Mass Transfer and Surface Reaction Kinetics Parameters

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Diffusive Mass Transfer and Surface Reactions

Column miscible displacement experiments with flow interruptions have been used to determine diffusive mass transfer parameters for non-reactive tracers. These parameters, if corrected for the effect of molecular size, may be used to discern the contribution of diffusive mass transfer from that of surface reactions to solute breakthrough in fractured saprolite.

Methods

Simultaneous non-reactive and reactive tracer injections were performed on an undisturbed saprolite soil column obtained at Oak Ridge National Laboratory (Mayes et al., 2001). On the basis of our prior findings, this research uses a two-pore-domain reactive transport model to determine both diffusive mass transfer parameters and kinetic rate coefficients of surface reactions. The public domain HBGC123D code (http://hbgc.esd.ornl.gov) was modified to include both intra-and inter-domain mass transfer processes.

Results and Discussion



Our previous effort using a one-pore-domain model is shown above. The model clearly cannot explain the decrease of tracer concentration during flow interruptions and the tailing of tracers. The effect of inter-domain mass transfer on tracer breakthrough and its contribution relative to that of surface reactions will be discussed once we have the complete analysis of laboratory and modelling data.

Reference

Mayes, M.A., P.M. Jardine, I.L. Larsen, S.C. Brooks, and S.E. Fendorf, 2001. Multispecies transport of metal-EDTA complexes and chromate through undisturbed columns of weathered fractured saprolite. J. Contam. Hydrol. 45, 243-265.