Evaporation and isotope fractionation in a TIMS source

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Understanding the stable isotope record of meteorites requires a quantitative knowledge of isotope fractionation related to thermal events leading to vaporization of elements as refractory as Mg ($T_{50\%condensation}$=1336 K). However, so far few experiments, except for major elements are available to determine the isotope fractionation linked with the kinetic process of evaporation. In this study, we have used the evaporation of various elements (e.g. Nd, Cr) analyzed by thermal ionization mass spectrometry to characterize the isotope fractionation and the thermodynamics of evaporation from a TIMS filament under various conditions.

In the first part of this study, we investigated the thermodynamic conditions of Cr evaporation by simulating the chemical equilibrium of the mixture of the element of interest with the activator to predict the vapor pressure and speciation in equilibrium at the temperature of ion emission. This enables to predict free evaporation rate of species to be compared with the observed ion beam and to optimize the running conditions. Based on the ion beam signal, one should be able to obtain a crude estimate of the evaporation coefficients. In a second part of this study, the observed isotope fractionation was first characterized using the classical exponential law. It was found as has been reported previously that after mass fractionation correction, there is a small residual trend in the Cr isotope ratios corrected for mass fractionation plotted one against each other. A new mass fractionation law based on first principles was derived and found to remove any temporal trend mass fractionation. This mass fractionation law yields more reproducible results and allows to estimate the kinetic mass fractionation coefficients associated with evaporation.