

Isotopic self-shielding of oxygen and nitrogen in the solar nebula

ROBERT N. CLAYTON

Enrico Fermi Institute, Department of Chemistry and
Department of the Geophysical Sciences, University of
Chicago, Chicago IL, USA (r-clayton@uchicago.edu)

I have proposed that the primary origin of large isotopic variations in oxygen and nitrogen in primitive solar system materials is isotopic self-shielding in the ultraviolet photolysis of CO and N₂ (Clayton, 2002). In a predissociation process, isotope-specific wavelengths are absorbed, resulting in saturation of the lines corresponding to abundant species: ¹²C¹⁶O and ¹⁴N¹⁴N, while the lines corresponding to the rarer isotopic molecules remain optically thin. The astrophysical setting for this process is still in question: Clayton (2002) proposed an X-wind site <0.1 AU from the young sun; Yurimoto and Kuramoto (2002) proposed an irradiation in the parent molecular cloud; Young and Lyons (2003) presented a model of solar irradiation at the surface of the nebula 100-200 AU from the sun. The isotopic record of the self-shielding process is found in solids condensed from the portion of the nebular cloud that has been enriched in atomic ¹⁷O, ¹⁸O, and ¹⁵N by the photodissociation process. The model predicts that the solar composition, not significantly modified by the self-shielding process, should correspond to the meteoritic and planetary components least enriched in the rare isotopes, i.e. $\delta^{15}\text{N} < -300\%$, $\delta^{17}\text{O}$, $\delta^{18}\text{O} < -50\%$. The solar wind sample, now being collected by the Genesis spacecraft, will test this prediction. The large difference in isotopic compositions between the sun and the inner solar system bodies implies that all inner solar system material underwent the heavy-isotope enrichment. In order to assess the expected magnitude of the isotope effects, it is necessary to have a quantitative treatment of the chemical trapping mechanisms needed in order to incorporate the effects into solid phases. The mechanisms depend critically on the astrophysical setting, and will provide an independent test of the origin of the isotopic effect. Photolysis of CO and N₂ may also have important consequences for chemical processes in the solar nebula by inducing non-equilibrium reactions, such as oxidation of iron and condensation of silicate liquids.

References

- Clayton R.N. (2002) *Nature* **415**, 860–861.
Yurimoto H. and Kuramoto K. (2002) *Meteoritics Planet. Sci.* **37**, A153.
Young E.D. and Lyons J.R. (2003) *LPSC XXXIV*, abstract #1923.

Matrix effect of charged environmental solutions on Pb isotope measurements by MC-ICPMS

A. COCHERIE AND M. ROBERT

BRGM, Orléans, France (a.cocherie@brgm.fr)

It has been generally accepted over the past five years or so that Pb isotope ratio determinations are as accurate using MC-ICPMS as they are using TIMS, despite the differences between the two types of instrument. Whereas the mass bias with TIMS is about 10 times lower than with MC-ICPMS, the TIMS Pb isotope measurements change slightly during the course of the thermal ionisation process. Conversely, the large mass bias of the ICP-MS instruments is wholly reproducible and can be easily corrected by adding a TI solution as an internal standard (White et al., 2000; Motelica-Heino et al., 2003). Comparisons are usually made on standard or purified Pb solutions after a passage through ion exchange columns. Here we find that using MC-ICPMS clearly requires a chemical purification process for light isotopes such as Li (Millot et al., 2003) and B (Guerrot et al., 2003) and also for ensuring precision on Pb isotope ratios close to external reproducibility (< 0.01%). However, Pb isotopes are being ever more widely used to solve environmental problems, and MC-ICPMS has the potential for rapidly determining isotope ratios in natural surface and ground waters without any prior chemical Pb purification.

The purpose of our work, therefore, was to evaluate the capability of MC-ICPMS for direct isotope measurements of natural Pb-bearing solutions, i.e. without preliminary purification. For this it was necessary to estimate the real uncertainties on the various ratios, which was done by preparing solutions synthetically charged with major cations (Na, K, Ca and Mg) and free of Pb, and adding them to known standard solutions. We were thus able to define the maximum cationic contents to ensure reasonable precision for solving environmental problems. We then compared the results obtained on real samples with TIMS after chemical separation and those obtained directly with MC-ICPMS without any prior chemical procedure.

References

- Millot R., Guerrot C. and Girard J.P., (2003), *Spectr'Atom 2003, Abs.*, P17. Pau, France.
Guerrot C., Robert M. and Millot R., (2003), *Spectr'Atom 2003, Abs.* P16. Pau, France.
Motelica-Heino M., Robert M. and Cocherie A., (2003), *Winter Conf. Plasma Spectr. Abs.*, p. 339.
White W.M., Albarède F. and Télouk Ph., (2000), *Chem. Geol.* **167**, 257-270.