Mass-independent sulfur isotopic fractionation at Lyman-alpha photodissociation of H₂S and relevance to meteorites

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Introduction

Select meteoritic classes possess mass independent (MI) sulfur isotopic compositions in sulfide and organic phases. Photochemical processes in the solar nebula have been attributed as a source of these anomalies [1-6]. Hydrogen Sulfide (H₂S) is the most abundant gas phase species in the solar nebula, and hence, photodissociation of H₂S by solar vacuum ultraviolet (VUV) photons (especially by Lyman- α radiation, one of the most prominent lines in the t-tauri phase of the sun) is a relevant process. Due to experimental difficulties associated with accessing VUV radiation, there is a paucity of data to test the hypothesis of a photochemical origin of mass independent sulfur. Here, we present multi isotopic measurements of elemental sulfur produced during the VUV photolysis of H₂S using the ALS synchrotron.

Results and Discussion

 $\rm H_2S$ photolysis was carried out at four different synchrotron bands between 90 and 157.6 nm in a differentially pumped chamber. Elemental sulfur produced in all these experiments was collected and converted to SF₆ to measure the isotope ratios (e.g., $^{33}S/^{32}S$, $^{34}S/^{32}S$, and $^{36}S/^{32}S$). Among four different synchrotron bands used, 121.6 nm photolysis products show significant MI fractionations both in $\Delta^{33}S$ and $\Delta^{36}S/\Delta^{33}S$ ratio of -3%.

Predissociative is a prominent processes in VUV photodissociation of H_2S and, introduce isotope selectivity in the resonance governed curve crossing dynamics. VUV spectrum at different locations is a variable. This scenario allows H_2S photolysis at different VUV wavelengths at different regions of the disk and result in formation of different sulfur reservoirs of unique isotopic composition within the nebula. The icy particles would incorporate these compositions while migrating within the nebular disk and finally incorporate in meteorite parent body [6].

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[5] Farquhar et al (2000) Nature 404, 50-52. [6] Chakraborty et al (2013) PNAS 110, 17650-17655. [7] MA and the ALS are supported by DOE (Contract No. DE-AC02-05CH11231), SC and MHT is supported by NASA Cosmochemistry and Origin programs.