RARE EARTH ELEMENT ABUNDANCES IN PRESOLAR SiC.

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Introduction: Lanthanides of the rare earth element (REE) group play an important role in understanding the nature of geochemical and cosmochemical processes. The elements La to Lu show a progressive increase in mass and ionic radius, but maintain similar trivalent chemistry because the electron configuration changes through the filling of the inner 4f shell. This characteristic generally results in smooth changes in abundance patterns in terrestrial rocks and minerals. In cosmochemistry, the different volatilities of the REEs play an additional and significant role in elemental-abundance behaviour particularly in refractory inclusions, which have a high temperature origin in the earliest stages of the Solar System [1;2]. A similar high-temperature origin is proposed for the formation of mainstream SiC grains, whose highly anomalous isotopic compositions indicate an origin as condensates in outflows of low-mass carbon-rich asymptotic giant branch (AGB) stars with close-to-solar metallicity [3]. Elemental abundances (e.g., [4]) and isotopic compositions of several heavy trace elements (e.g., Zr, Mo, Sr, Ba) measured in mainstream SiC grains are, for the most part, consistent with theoretical models of slow neutron capture nucleosynthesis (the s-process) (e.g., [5]). Somewhat less attention has been paid to the nature of REE abundances in SiC and their implications for formation mechanisms. In this work, individual isotope abundances of Ba, lanthanides of the REE group, and Hf have been determined in bulk samples of fine-grained SiC from the Murchison CM2 chondrite.

Experimental: The s-process pathway through the REEs has been determined by analysing trace element abundances in bulk samples of presolar SiC. Abundances were measured by secondary ion mass spectrometry (SHRIMP RG) using a combination of high mass resolution ($M/ΔM_{99%} = 12,000$) coupled with energy filtering to exclude higher order polyatomic Si-C-O clusters that are evident in the mass region of the REEs. REE and Hf abundances were normalised relative to $^{138}$Ba$^+$ in NIST SRM 610. Relative sensitivity factors for REE/Ba and Hf/Ba appear to be appropriate for SiC as demonstrated by analysis of REE in a synthetic trace element bearing SiC ceramic. In performing this work, we fully evaluate potential isobaric interferences in the REE mass region by measuring a continuous high-mass-resolution spectrum from mass 30 to mass 210 in a synthetic silicon carbide. This spectrum shows the production of molecular interferences that have important implications for isotopic measurements of silicon carbide.

Results and Discussion: The measured abundances of Ba, Hf, and the refractory REE (La-Sm, Gd-Er, and Lu) are in excellent agreement with model predictions computed with the FUNS code (FUll Network Evolutionary Code; [6;7]) for the envelope compositions of AGB stars of 2-3 M$_\odot$ and metallicity close to solar. However, the model predictions for $^{146}$Nd appear to be high suggesting that one or more cross sections for nuclides involved in its production may need re-examination. Tm appears marginally high in normalised abundance, but this does not appear to be related to the Group II REE (ultra-refractory depleted) pattern found in refractory inclusions [8] because Lu and Hf are not depleted. The Tm abundance is prone to nugget effects because of the very low number of counts detected. Eu and Yb are depleted relative to other REE. This is similar to that in Group III Allende refractory inclusions [8] and in platy hibonite crystal fragments [9;10] and suggests that the silicon carbide grains condensed while Eu and Yb remained in the gas phase. The REE pattern depleted in Eu and Yb is consistent with depletions in Sr noted by [4], however, other less-refractory trace elements (e.g. Ba, V) do not show this depletion. The depletion of Eu and Yb, and the presence of volatile elements (e.g. Xe) in meteoritic SiC indicates that these elements must be incorporated when the SiC grains have cooled and most likely by ion implantation.