Synchrotron X-ray Fluorescence: A New Approach for Determining Trace Element Concentrations in Individual Micron-Sized Presolar SiC Grains

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Introduction

The study of presolar grains has focused so far on isotopic analysis due to the wealth of information one can get from it on the nuclear processes in the stars from which the grains condensed. At the same time only one extensive quantitative chemical analysis has been done on individual grains.[1] From the chemical composition of different grains, together with nucleosynthesis and condensation calculations, one can try to infer the physical and chemical conditions in the stars that affected the condensation chemistry of the grains.

The main advantages of synchrotron x-ray fluorescence (SXRF) are that it is nondestructive and independent of secondary ion yields [in both cases unlike secondary ion mass spectrometry - SIMS[1]]. This fact motivated us to apply SXRF to the study of individual grains in order to achieve two goals: (1) combining elemental and isotopic data on the same grains, for a wider and clearer understanding of the history of the grains; and (2) using it as a "screening" method for sorting out grains with enrichments of specific elements for later isotopic analysis. We report here the first measurement of 15 individual grains by this method.

Methods and Materials

The experiment was done at SRI-CAT (sector 2). A 22.5 keV undulator-produced beam energy was chosen in order to excite K lines of elements as heavy as Mo. The beam on its path from the undulator to the sample passed through a monochromator that reduced its energy width to ~2 eV, and through a zone plate lens that focused it to a spot size of ~0.6 µm. Its intensity on the sample was ~5x10^7 photons/sec. The sample's surface was tilted at 70° to the beam, and at 20° to the single element Ge detector[2] (i.e., the detector was at 90° to the beam). Data were collected for 20-45 min for each of the 15 grains. For 8 of the grains, a nearby (2 -5 µm away) background spectrum was collected.

We developed a "universal" sample mount that could be used in all the experimental systems on which the grains are analyzed [SXRF, SEM, SIMS, RIMS (resonance ionization mass spectrometry)]: individual grains from the Murchison meteorite size fraction KJF (~1-3 µm) were glued, using diluted Duco cement, to a 7.5-µm-thick Kapton. The Kapton was held in a plastic ring (diameter of ~1.7 cm), and everything was carbon coated. Both types of plastic: (1) are composed of light elements (hence produce relatively little scattering of the x-rays, and the Ge detector is insensitive to these elements' fluorescence lines); and (2) have low vapor pressure (compatible with high vacuum systems).

The data analysis was done at CARS-CAT (sector 13) in a procedure similar to the one used for inter planetary dust particles:[3] The Compton peak of the background spectrum was scaled to that of the grain, the whole background spectrum was multiplied by the scaling factor and subtracted from the grain's spectrum (for the 7 grains that did not have their own background spectra a sum of the 8 background spectra was used). Gaussians were then fitted to the K and L lines in the reduced grain spectrum. Abundances were calculated by using the Si Kα peak as an internal reference fluorescence peak.

Results and Discussion

We detected the following trace elements in the grains (not every element was detected in every grain): Ti, V, Cr, Mn, Fe, Ni, Sr, Y, Zr, Nb, Mo, Ru and W. We also determined upper limits on the abundance of S. The results range in value from 3450 ppm for Ti (grain #22) to the detection limit of ~1 ppm measured for Sr (grains #6,12,14). Trace element distribution changed from grain to grain in both pattern and absolute abundances. All grains except for #7,12,22 (each with its own pattern) can be loosely grouped into the pattern of grain #14 - as shown in the left part of Fig. 1. The right part of Fig. 1 shows the spectrum of one of the "anomalous" grains - grain #12 (note the difference in peak intensities in the Ti-Ni and Y-Ru regions).

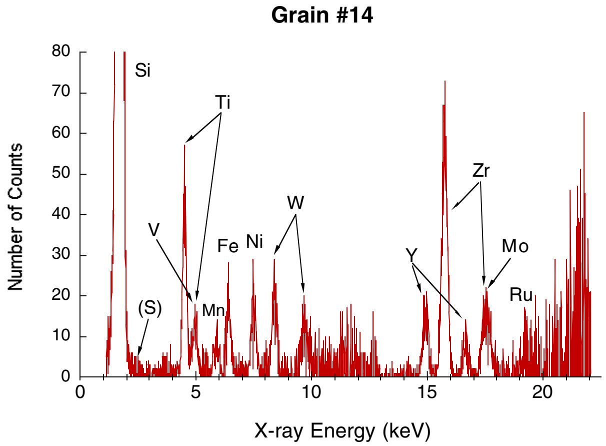

FIG. 1. Reduced XRF spectrum of grains #14 and #12. Most grains show trace element distribution somewhat similar to that of grain #14. Grain #12 is one of the 'anomalous' grains. Note the difference in peak intensities in the Ti-Ni and Y-Ru regions.

Here are some initial conclusions. (1) There is generally good agreement between our results and those of Amari et al.[1] (2) Trace element abundances of the grains can be generally understood by considering the effect of condensation temperature on the element's stellar abundance.[1,4] In other words a nuclear enrichment (or depletion) in the star's atmosphere can be further amplified or decreased in the grains due to the trace element condensation chemistry. (3) The very high W enrichment is probably due to contamination from sodium polytungstate used for the density separation of the grains.[5] (4) The detection of Ru is very interesting as it does not form a refractory carbide nor a refractory sulfide (it was detected before in presolar graphites[6]). It may have condensed into a non-iron metal subgrain; although it is a siderophile element, it does not correlate with any of the other siderophile elements detected - Fe, Ni and Mo.[7] It may also be a decay product of Tc (which forms a refractory carbide)—although the abundance of Tc in AGB stars (a factor of 30 less than Ru) seems too low to be the only source for the observed Ru8.

Acknowledgments

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