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The prospect of standard-free “absolute” isotope analysis by TIMS using incipient emission and total evaporation approaches

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Some applications, e.g., determination of half-lives of radioisotopes by decay counting, require “absolute” isotopic ratios (i.e., determined without reference to other materials, and without making assumptions about the true values of any isotopic ratio or about the nature of fractionation). For some elements, accuracy of “absolute” isotopic measurements by TIMS or MC-ICPMS can be verified by calibration against synthetic gravimetric mixtures of enriched isotopes, but for most elements such reference materials do not exist. Furthermore, for the elements that contain one natural stable isotope, and the abundance ratio of that isotope and a long-lived radionuclide has to be determined, such standardization is usually impossible.

Two approaches can potentially provide accurate standard-free “absolute” isotopic ratios: measurement of the isotopic of composition of the vapor in equilibrium with unfractionated solid with correction using Langmuir's evaporation model – Incipient Emission TIMS, IE-TIMS (1,2), and collection of all ions of the isotopes of interest from the onset of evaporation to complete exhaustion of the sample – Total Evaporation TIMS, TE-TIMS (3). It was recently found that for potassium accurate IE-TIMS data can be obtained from single filament emitter-assisted runs but not from double filament surface ionisation runs, and vice versa for TE-TIMS (4). Here I report the outcomes of IE-TIMS and TE-TIMS measurements for samarium (for a ^{146}Sm half-life project) and manganese (for a ^{53}Mn half-life project). Two samarium materials, Ames metal and synthetic standard GBW 04605 prepared at National institute of Metrology, China, loaded in a double filament (Re-Re) assembly, yielded precise TE-TIMS results with systematic uncertainty <0.1%, while IE-TIMS data are irreproducible and inaccurate (Fig. 1). For manganese, single filament silicagel loads yielded stable and sufficiently intense ion beams and reproducible IE-TIMS data, while TE-TIMS data are more dispersed, in agreement with our finding for potassium (4). No usable ion beams could be obtained from double filament loads.

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