

New developments in Ion-Laser InterAction Mass Spectrometry

M. Martschini¹, S. Merchel¹, K. Hain¹, L. Frost³, O. Marchhart¹, P. Steier¹,
A. Wieser^{1,2}, A. Wiederin¹, S.R. Winkler², R. Golser¹

¹University of Vienna – Faculty of Physics, Isotope Physics, Vienna, Austria

²Helmholtz-Zentrum Dresden-Rossendorf, Accelerator Mass Spectrometry and Isotope Research, Dresden, Germany

³JEN Jülicher Entsorgungsgesellschaft für Nuklearanlagen mbH, Jülich, Germany

Interferences from isobars typically restrict the applicability of AMS to selected long-lived radionuclides. The novel Ion-Laser InterAction Mass Spectrometry (ILIAMS) technique at the Vienna Environmental Research Accelerator (VERA) can overcome this limitation in many cases by highly-efficient isobar removal at eV-energies in a gas-filled radiofrequency quadrupole. The virtually complete suppression of isobars serves two objectives: A great number of nuclides can be measured for the first time with AMS while others become accessible at environmental levels – even on low terminal voltage AMS-systems – with the benefit of unprecedented detection efficiencies and blank values. This opens exciting possibilities in environmental radioactivity (⁹⁰Sr, ⁹⁹Tc, ^{135,137}Cs), astrophysics (⁴⁴Ti, ⁵³Mn, ¹⁸²Hf), and Earth science (²⁶Al, ³⁶Cl, ⁴¹Ca) research. [1] At AMS-15, we have reported on the excellent performance of ILIAMS for ²⁶Al and ³⁶Cl. Here we highlight the recent developments for the technique across the entire nuclear chart.

ILIAMS exploits differences in detachment energies (DE) within elemental or molecular isobaric systems by neutralizing anions with DEs smaller than the photon energy via laser photodetachment. In addition, molecular interactions with a buffer gas can further enhance isobar suppression, e.g., via breakup of ⁴¹KF₃⁻ into ⁴¹KF₂⁻ and F, or via O-pickup of ¹⁸²WF₅⁻ forming ¹⁸²WF₅O_x⁻.

With at least eleven orders of magnitude suppression of both Mg and K, ILIAMS-assisted AMS enables the detection of ²⁶Al/²⁷Al (~10⁻¹⁰, extraction of AlO⁻) and ⁴¹Ca/⁴⁰Ca (10⁻¹¹-10⁻¹³, extraction of CaF₃⁻) directly from crushed stony meteorites containing intrinsic ~1% Al and Ca, respectively [2,3]. The presence of isobars originating from the natively abundant elements (13-20% Mg, ~1‰ K) does not cause any analysis problems making radiochemical separation redundant. Measurements of ⁴¹Ca in chemically untreated concrete from nuclear decommissioning and coral sand samples clearly demonstrate the huge potential of this newly-established instrumental AMS (IAMS) technique. It is opening routes to high-sample throughput analysis, reasonable and fast provenance checks for (extra-)terrestrial origin and nuclear clearance.

At the upper end of the nuclear chart, the laser-induced suppression of U during measurements of Np constitutes the first non-chemical isobar discrimination in AMS in the actinide region.

Finally, for ⁹⁰Sr, highly efficient ILIAMS-suppression of the isobaric interference ⁹⁰Zr enables a blank value of ⁹⁰Sr/Sr < 5×10⁻¹⁶ at an overall Sr-detection efficiency of 4×10⁻⁴. This corresponds to a detection limit of <0.016 mBq, i.e., 2×10⁴ atoms or 3 ag of ⁹⁰Sr in a sample of mg of stable Sr – at least a factor 100 better than any other known technique of measurement. Recently, we have successfully demonstrated the tremendous potential of this technique for ⁹⁰Sr in the measurement of contemporary coral aragonite and seawater samples of less than 500 ml, and the analysis of ⁹⁰Sr concentration in small samples of soils and other environmental archives after adding Sr carrier.

Ackn.: This project received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 101008324 (ChETEC-INFRA) and No 824096 (RADIATE), and grants from the Austrian Science Fund (FWF): I 4803-N and P 31614-N28. (Extra-)terrestrial samples were kindly provided by A. Bischoff (U Muenster), A. Gaertner & K. Schniebs (Senckenberg Dresden), Forst- und Landwirtschaftsbetrieb der Stadt Wien, K. Pachnerová Brabcová (Czech Academy of Science, Prague).

References: [1] Martschini et al., Radiocarbon 64 (3) (2022) 555. [2] Bischoff et al., accepted for Meteorit. Planet. Sci.. [3] Bischoff et al., submitted to Meteorit. Planet. Sci..