sck cen

Intensity

Intensity

Intensity

Quantification of U, Pu, Nd & Gd for the characterization of spent nuclear fuel using isotope dilution HPIC-SF-ICP-MS



Nancy N. Wanna^{1,2}*, Andrew Dobney¹, Karen Van Hoecke¹, Mirela Vasile¹ & Frank Vanhaecke²

¹Belgian Nuclear Research Centre, SCK•CEN, Mol, Belgium

²Ghent University, Department of Chemistry, Atomic & Mass Spectrometry – A&MS Research Unit, Campus Sterre, Krijgslaan 281 – S12, 9000, Ghent, Belgium *E-mail: nwanna@sckcen.be

Introduction

Post-irradiation examination of spent nuclear fuel (SNF) includes destructive analysis for the determination of long-lived actinides and lanthanides (fission products) concentrations such as U, Pu, Nd & Gd (in Gd fuels). This is done using isotope ratio (IR) measurements with inductively coupled-plasma mass spectrometry (ICP-MS) or, alternatively, by thermal ionization mass spectrometry (TIMS) & α-spectrometry. Sector field (SF) ICP-MS is a single detector ICP-MS instrument well known for flat top peaks, which increase the precision of IR measurements. Coupling high pressure ion chromatography (HPIC) online with ICP-MS rapidly eliminates notorious isobaric interferences whilst reducing the analyst's exposure to radiation.

Objectives

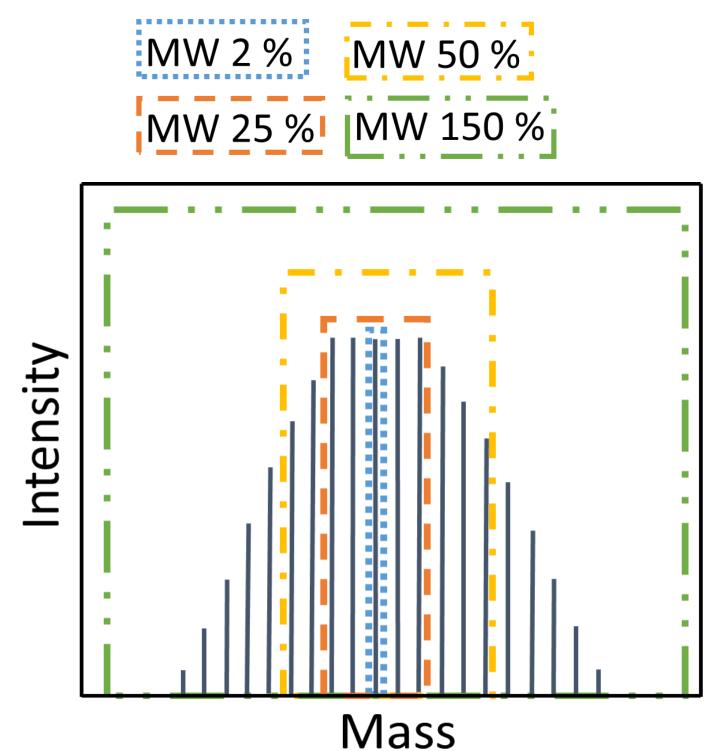
- 1. Optimize acquisition parameters of the SF-ICP-MS and select a calculation method [point-by-point (PbP), linear slope regression (LSR) or peak area integration (PAI)] to obtain the most precise IR.
- 2. Characterize two types of SNF (Uox and Gd fuel) using isotope dilution HPIC-SF-ICP-MS.
- 3. Determine the overall uncertainty budget of isotope dilution HPIC-SF-ICP-MS to compare it to that of ISO 17025 accredited isotope dilution TIMS & α -spectrometry.

Materials & methods

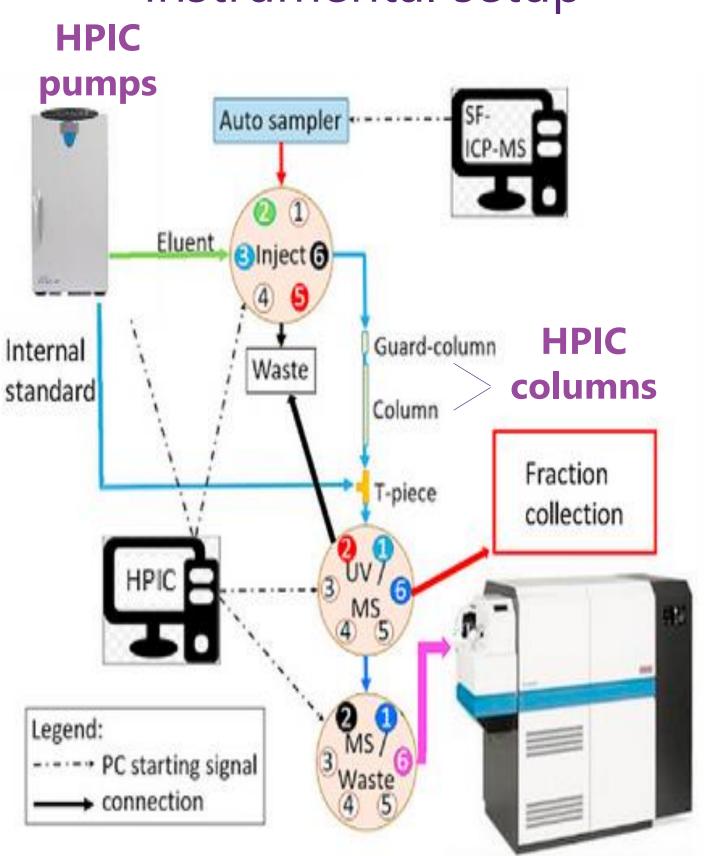
Results

IR calculation methods Flat-top peak investigated acquisition parameters Dwell times: 10 & 30 ms PbP

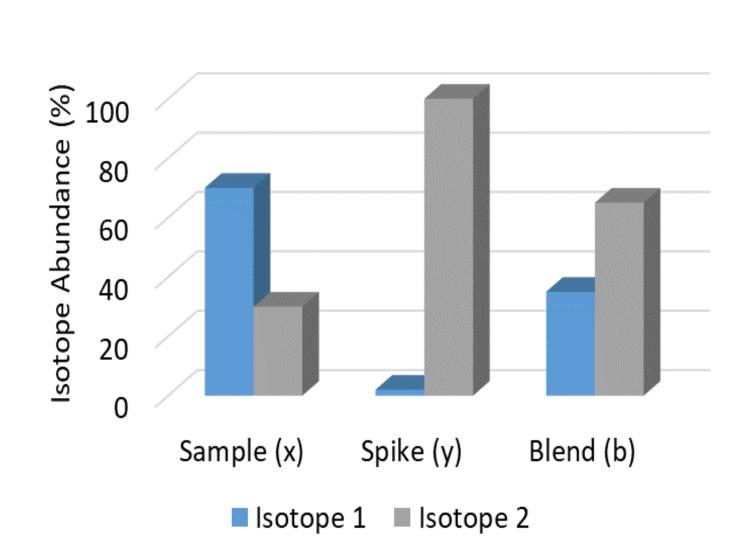
- Mass windows:

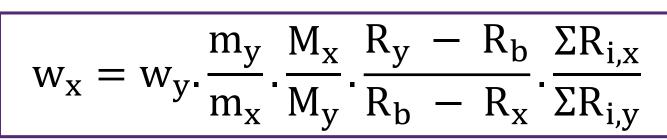


HPIC-SF-ICP-MS instrumental setup



Single isotope dilution





w: mass fraction (g.g⁻¹) m: weight (g) M: molecular weight (g.mol⁻¹) R: isotope ratio ΣR: Sum of isotope ratios

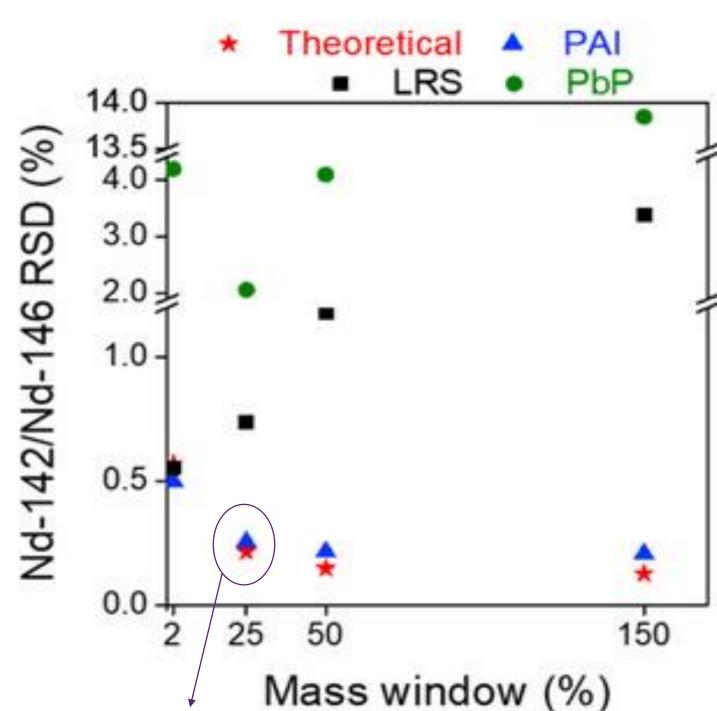
Experimental & theoretical precisions

Time

Time

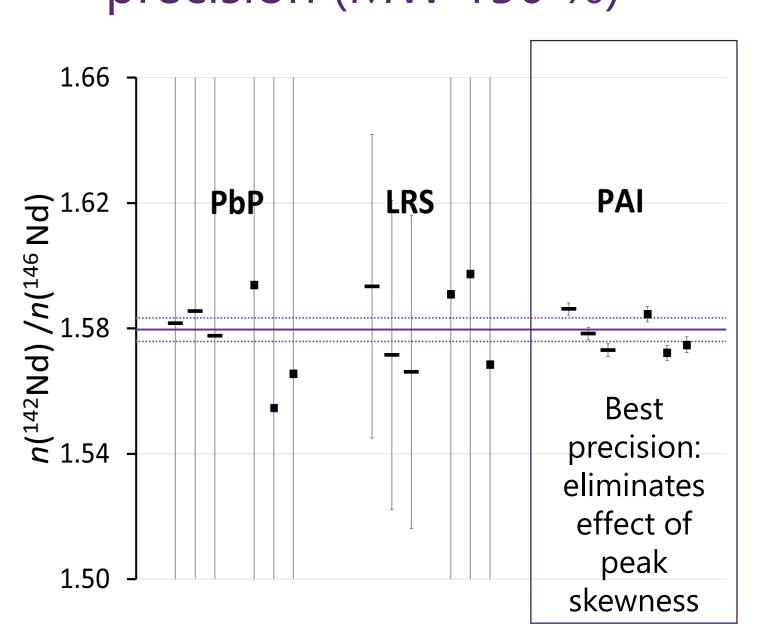
Intensity 2

PAI



Experimental precision closest to theoretical (best obtainable) precision.

Effects of dwell time & calculation method on IR precision (MW 150 %)

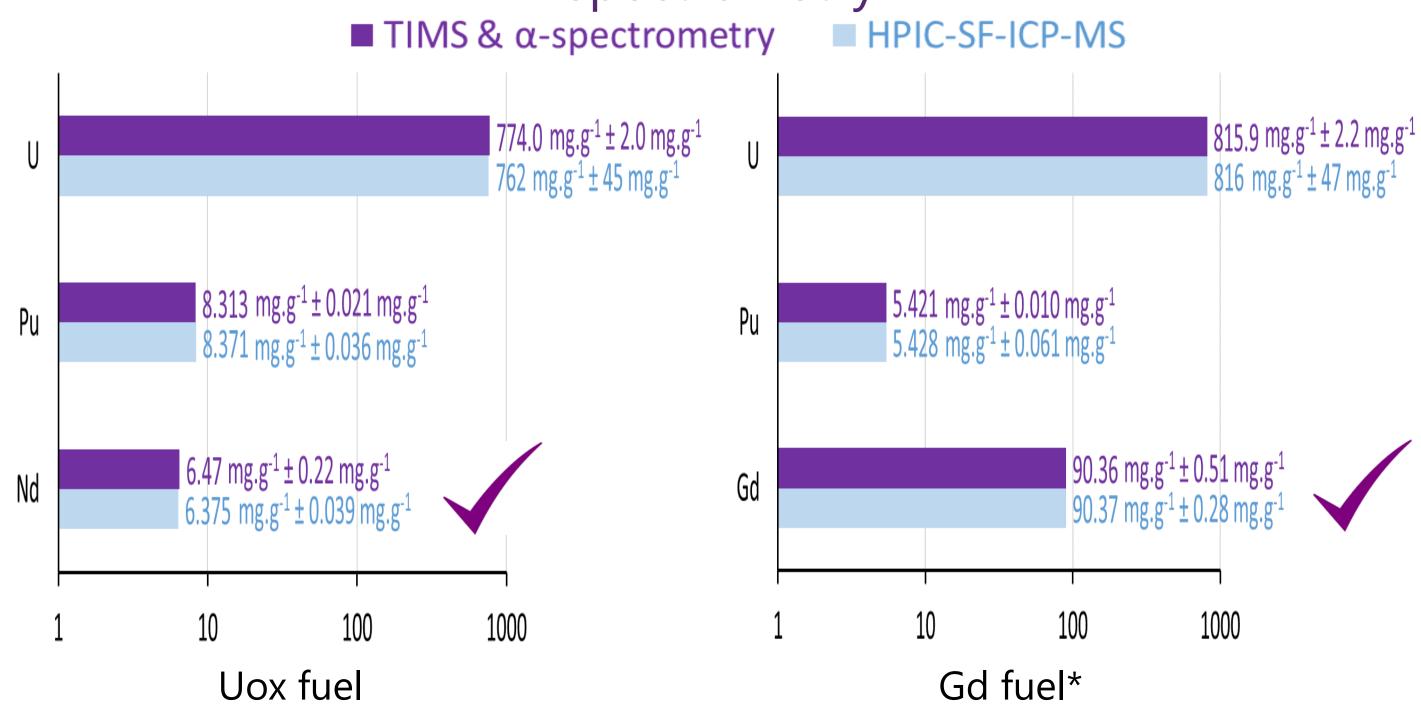


- dwell time 10 ms • dwell time 30 ms

Shorter dwell time → faster acquisition method → more precise IR with PbP & LRS

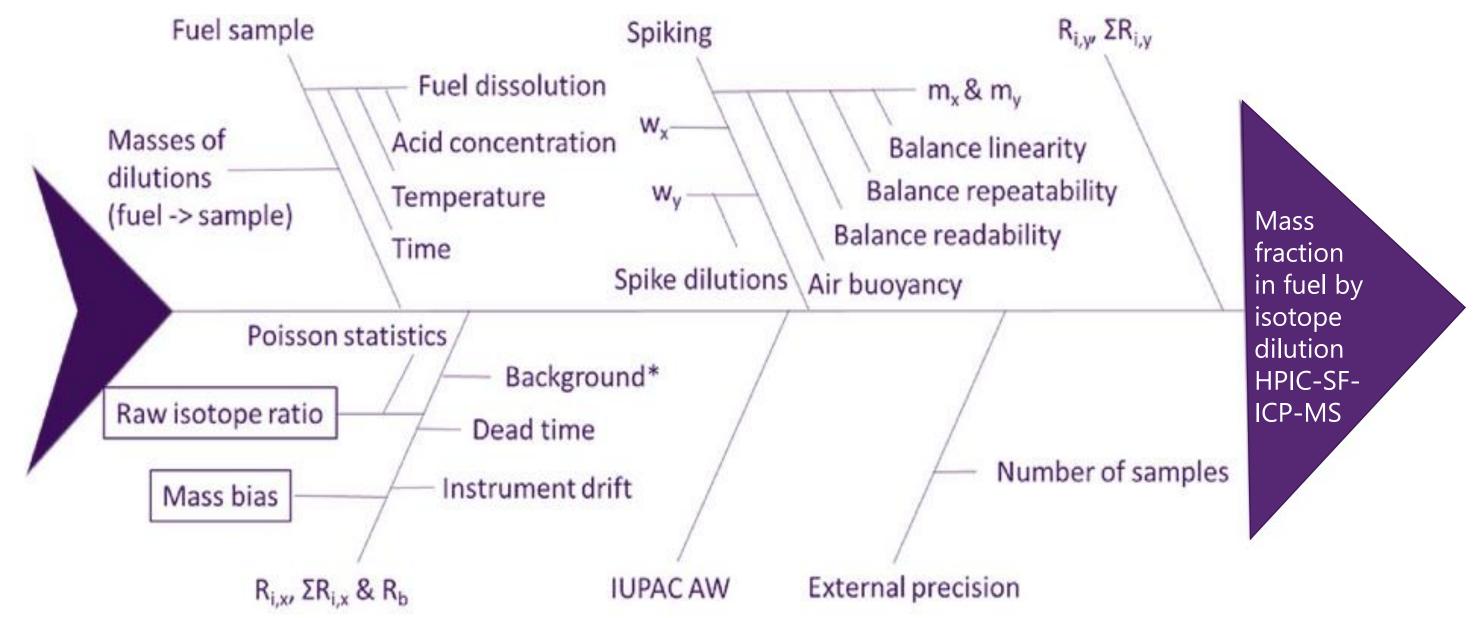
Mass fractions of elements in Uox & Gd SNF using isotope dilution HPIC-SF-ICP-MS and isotope dilution TIMS & α spectrometry

SF-ICP-MS



* The concentration of Nd in the Gd SNF was lower than the LOD of the method and hence could not be determined by HPIC-SF-ICP-MS.

Uncertainty contributors in isotope dilution HPIC-SF-ICP-MS



^{*} Background refers to instrumental background as well as signal measured in the blank and has a major effect at concentrations close to LOD.

Conclusions

- The acquisition parameters of SF-ICP-MS have been optimized to yield IR measurements with the best precision.
- Isotope dilution HPIC-SF-ICP-MS offers accurate Pu, U, Nd and Gd determination in 2 types of SNF compared to isotope dilution TIMS & α-spectrometry.

Reference

N. N. Wanna, A. Dobney, K. Van Hoecke, M. Vasile, F. Vanhaecke, Quantification of U, Pu, Nd & Gd for the characterization of spent nuclear using isotope dilution HPIC-SF-ICP-MS, Talanta, DOI: 10.1016/j.talanta.2020.121592

Exploring a better tomorrow www.sckcen.be

