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Separation and isotope ratio measurements of lanthanides and uranium, using HPIC-SF-ICP-MS, for characterization of spent nuclear fuel

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Elemental and isotopic characterization of spent nuclear fuel is of crucial importance in nuclear reactor facilities for various purposes, including the management of spent nuclear fuel [1]. Characterization of spent nuclear fuel samples requires highly sensitive and precise mass spectrometric techniques, such as sector-field inductively coupled plasma-mass spectrometry (**SF-ICP-MS**) and thermal ionization mass spectrometry (TIMS) to accurately measure the isotopic compositions of uranium (**U**), plutonium (**Pu**) and neodymium (**Nd**), especially to determine the burnup [2]. A major issue in mass spectrometry, i.e. spectral overlap of isobaric nuclides, can be overcome by the separation of the analyte elements from one another. Unlike TIMS, SF-ICP-MS can be hyphenated with online separation methods [3], e.g., high performance ion chromatography (**HPIC**), thus substantially simplifying the analytical protocol and enhancing the sample throughput.

A Dionex ICS5000+ dual quaternary gradient pump HPIC instrument, equipped with Dionex CS5A mixed bed ion exchange chromatography column, and CG5A as guard column, was connected to both a UV-Vis detector and an Element 2 sector-field ICP-MS instrument, partly installed inside a glove-box. Separation of **lanthanides** (La, Ce, Pr, Nd, Sm, Eu, Gd, Lu) and uranium from the concomitant matrix components and from one another was achieved when 0.1 −0.3 M oxalic acid and 0.5-1.0 M HCl mobile phases were used in gradient and isocratic elution programs respectively. Chromatograms obtained in this way, as well as the analytical challenges involved in optimizing the **HPIC-UV-Vis** [5] and **HPIC-SF-ICP-MS** methods [6] will be presented. In addition, an overview of both data acquisition parameters (incl. the number of nuclides monitored per run, dwell time, number of replicates, integration window, etc.) and analytical method characteristics (incl. calibration and method(s) to correct for mass discrimination while handling transient signals) to arrive at isotope ratio determinations with the best obtainable accuracy and precision will be described.

In conclusion, HPIC hyphenated to a SF-ICP-MS unit in a glove-box, for the separation and isotope ratio measurements of lanthanide elements and uranium, will be presented. Isotope ratio measurement uncertainties, determined at a later stage, will allow the evaluation of the HPIC-SF-ICP-MS method against TIMS. Method validation will be done using environmental samples.

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