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Direct Analysis of Americium in Non-Separated Spent Nuclear Fuel Using High Resolution ICP-OES and Sector Field ICP-MS

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Complementary analysis employing instrumental techniques based on different physical detection principles is a key issue for quality assurance. Lacking matrix-matched certified reference materials, accurate, straightforward analytical procedures for the determination of americium (Am) in non-separated spent fuel employing high resolution ICP-OES and sector field ICP-MS were developed and cross-validated.

Using a commercial high resolution ICP-OES spectrometer, the eleven most sensitive Am emission wavelengths were investigated in detail for potential isotopic and elemental analysis. Calibration of the instrumental response was carried out through dilution of a ^{241}Am stock solution (1 550 mg kg⁻¹) that was prepared in-house via chemical separation of the element from a concentrated Pu solution (115 g L⁻¹).

Sector field ICP-MS, λ - and λ -spectrometry served to characterise extensively the employed Am and Pu solutions for their actual concentration, potential impurities and isotopic purity. Hyperfine splitting of relatively broad peaks (~10 pm) or multiplets, i.e. multiple peaks, were observed for most tested Am ICP-OES emission signals with ^{241}Am and ^{243}Am revealing identical signal patterns. The isotopic shift between ^{241}Am and ^{243}Am was generally small with a maximum difference of only ~3.7 pm found at $\lambda=450.945$ nm and $\lambda=469.970$ nm, respectively. As a consequence, the extraction of Am isotopic information from the emission spectra was limited. The use of a high efficiency sample introduction system helped to lower detection limits (LOD) for Am up to two orders of magnitude compared to published values. While the emission wavelength $\lambda=283.226$ nm provided the lowest LOD of 0.07 mg kg⁻¹, the ICP-OES signal at $\lambda=392.625$ nm (LOD 0.71 mg kg⁻¹) was best suited for the analysis of Am in spent fuel predominantly containing U and Pu. Both external calibration and standard addition yielded comparable Am concentration results for various spent fuels employing ICP-OES.

Calibration of the sector field ICP-MS response for spent fuel Am analysis was accomplished using both ^{232}Th and ^{238}U signals as well as applying the corresponding mass bias. This calibration methodology - frequently used in absence of an Am stock standard solution - was tested against the home-made ^{241}Am stock standard solution underpinning the validity of the calibration approach.

One spent (Th,Pu)₂O₇ fuel and two irradiated metallic fuels, having different chemical and isotopic compositions, were analysed for their Am content. Sector field ICP-MS analysis essentially confirmed the Am concentrations obtained using ICP-OES in the three spent fuel solutions with mean values overlapping within their standard deviations. Considering isotopes of other actinides such as U, Pu and curium (Cm), the Am isotopic abundance (^{241}Am , ^{242}Am , ^{243}Am) was established in the non-separated fuel solutions comparing reasonably well with model calculations based on the ORIGEN-2 code.

The excellent comparison between ICP-OES and ICP-MS results underpins the accuracy and reliability of the developed analytical procedures and adds substantially to the quality assurance of Am analysis in spent fuel.

Primary author: Dr KRACHLER, Michael (European Commission, Joint Research Centre, Institute for Transuranium Elements)

Co-authors: Dr CARBOL, Paul (European Commission, Joint Research Centre, Institute for Transuranium Elements); Dr ALVAREZ-SARANDES, Rafael (European Commission, Joint Research Centre, Institute for Transuranium Elements); Dr MALMBECK, Rikard (European Commission, Joint Research Centre, Institute for Transuranium Elements); Dr VAN WINCKEL, Stefaan (European Commission, Joint Research Centre, Institute for Transuranium Elements)

Presenter: Dr KRACHLER, Michael (European Commission, Joint Research Centre, Institute for Transuranium Elements)

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