

### Background

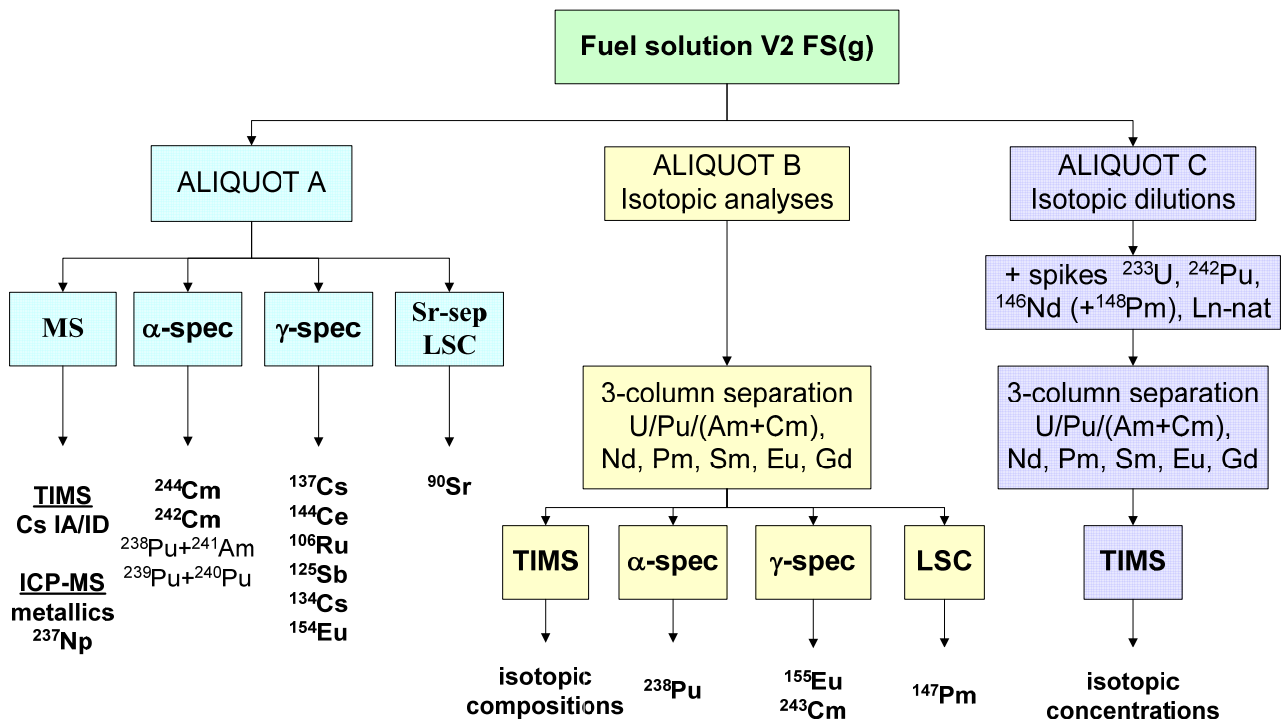
Destructive radiochemical analysis of spent nuclear fuels is an important tool to determine burn-up with high accuracy and to better understand the process of depletion and formation of actinides and fission products during irradiation as a result of fission and successive neutron capture. The resulting isotope inventories and nuclear databases that are created, are of high importance to evaluate the performance of nuclear fuels in a reactor, to evaluate computer codes applied for a safe transport, storage and disposal/reprocessing of spent fuels and to safeguard fissile material.

### Objectives

The objective is to provide chemical and radiochemical analyses procedures for an accurate determination of isotopic compositions and concentrations of actinides and fission products in different types of industrial (UO<sub>2</sub>, MOX) and experimental nuclear fuels (UAl<sub>x</sub>, U<sub>3</sub>Si<sub>2</sub>, UMo, ...). For a burn-up determination program typically 21 actinides and fission products are analyzed. For an extended characterization program this can increase to up to ~ 50 isotopes.

### Principal results

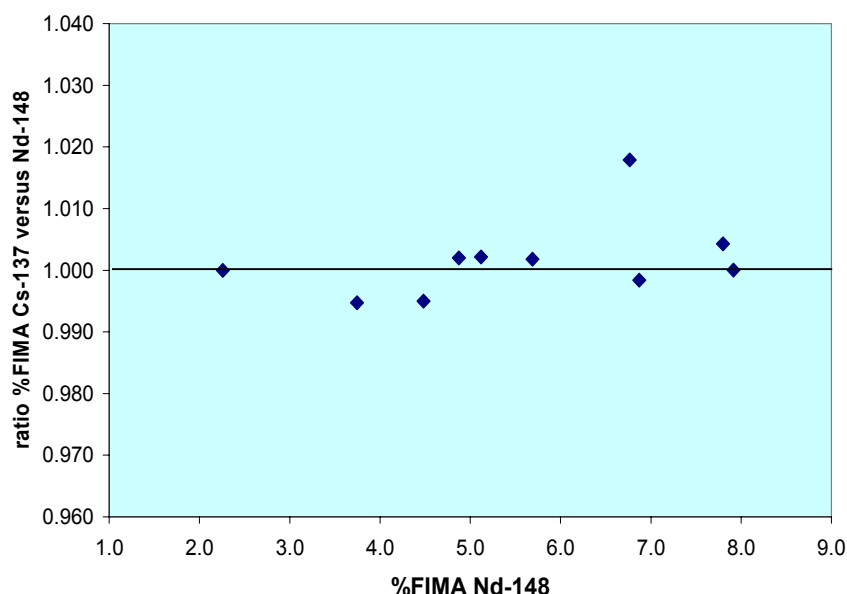
A first requirement for a successful radiochemical analysis is a quantitative dissolution of the spent fuel. The highly radioactive spent fuel samples, typically ranging from 1 to 25 g, are dissolved in a lead-shielded facility. For industrial UO<sub>2</sub> and MOX fuels with homogeneous fuel pellets that are packed in a cylindrical zircaloy cladding, a two step procedure using nitric acid as main solvent is applied resulting in a dissolution of the fuel but leaving the cladding intact. Dispersion fuels, where the fuel particles (UAl<sub>x</sub>, U<sub>3</sub>Si<sub>2</sub>, UMo) are mixed with Al-powder and confined as a thin wafer between aluminum plates, require the dissolution of the fuel together with the Al matrix and cladding. After the acid dissolution steps a small amount of residue is possibly left that contains primarily metallic fission products such as Mo, Tc, Ru, Rh, Ag and Sb-isotopes. For an extended characterization this residue is dissolved using a molten salt procedure in a shielded cell.



*The accurate detailed radiochemical analysis of irradiated fuel requires a well designed analysis scheme incorporating appropriate separations, measurements and cross-checks – as illustrated for a typical extended spent fuel characterization.*

The actinides and fission products in the fuel solutions are analyzed using mass-spectrometry (TIMS and ICP-MS) and radio analytical techniques ( $\alpha$ -,  $\beta$ - and  $\gamma$ -spectrometry). These are all techniques with analyses methodologies under the scope of QA accreditation according to the international ISO/IEC 17025 standard. Prominent  $\gamma$ - and  $\alpha$ -emitters and most of the metallic isotopes can be directly measured on a diluted spent fuel solution. However the majority of the isotopes need to be separated before they can be properly analyzed.

In radiochemistry burn-up is expressed as %FIMA, i.e. the number of Fissions that have occurred per Initial 100 heavy Metal Atoms (U and/or Pu). The number of fissions that occurred during irradiation can be derived from the concentration of selected key fission products in the spent fuel sample under investigation. At SCK•CEN, the stable Nd-isotopes  $^{143}\text{Nd}$ ,  $^{144}\text{Nd}$ ,  $^{145}\text{Nd}$ ,  $^{146}\text{Nd}$ ,  $^{148}\text{Nd}$ ,  $^{150}\text{Nd}$  and the  $\gamma$ -emitters  $^{137}\text{Cs}$  and  $^{144}\text{Ce}$  are selected as fission product monitors. Both groups of fission products are analyzed with different analysis techniques, i.e. the Nd-isotopes by isotopic dilution TIMS after a complex separation procedure and the  $\gamma$ -emitters by  $\gamma$ -spectrometry directly on a diluted spent fuel solution. The initial number of heavy metal atoms can then be calculated by summing the number of fissions and the analyzed number of actinides after irradiation, usually predominantly U and Pu isotopes.



*The excellent agreement between the burn-up derived from the Nd-isotopes measured with TIMS and the burn-up derived from  $^{137}\text{Cs}$  measured with  $\gamma$ -spectrometry illustrates the excellent performance of the analysis methodology applied at SCK•CEN over the whole burn-up range of present-day nuclear fuels.*

During the year 2005 the running programs at the analytical laboratories of SCK•CEN requiring destructive radiochemical analyses were GERONIMO (9x9 BWR fuel licensing), REBUS-PWR (criticality burn-up credit validation), RJH-UMo (reduced enrichment research reactor fuel qualification) and MALIBU (extensive radioisotope inventory assessment of spent fuel), representing a total of 12 fuel samples from which 5 samples were burn-up programs and 6 samples were extended characterization programs.

### Future work

In the near future a new shielded cell for the treatment of highly  $\gamma$ -emitting samples for radiochemical analyses will be taken into operation. For application in this new cell a closed system allowing the treatment of samples that can not be dissolved in nitric acid thus requiring the use of more aggressive acids will be developed.

Presently, the isotopes analyzed in the spent fuels are selected to address fuel performance and safety issues. In the future additional procedures for the analyses of critical radionuclides at trace-level, that are important from the viewpoint of deep geological disposal, will be developed.

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