

Background

Accurate and reliable analysis of the nuclear fuel composition, the identification and quantification of trace impurities, and microstructural characterisation are essential requirements for the qualification of nuclear fuel production and for solving fuel manufacturers' problems. SCK•CEN has over 20 years' experience in this field and provides analytical services for the characterization of fresh and spent experimental and commercial nuclear fuels. A significant part of the qualification of mixed oxide (MOX) fuels produced at the BELGONUCLEAIRE MOX plant situated at Dessel in Belgium was performed in our laboratories.

Objectives

The main objective is to provide analytical methodologies allowing the validation of nuclear fuels and the study of parameters that are essential for the safe handling of the fuel before, during and after burning in power reactors of various types.

Principal results

The Radio-Chemical Analysis group at SCK•CEN has considerable interest and expertise in the destructive chemical and radiochemical analysis of a wide variety of nuclear materials. Over the years our staff of 15 research scientists and analytical chemists acquired valuable knowledge and gained much experience in the analysis of fresh and spent nuclear fuels. Several analytical procedures have been developed specifically for the qualification of MOX fuels from production lines and are accredited by Belac according to the ISO 17025 standard.

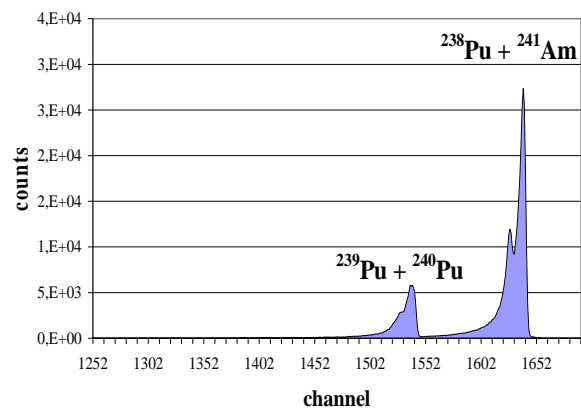
Thermal Ionisation Mass Spectrometry (TIMS) is used for analysis of nuclear fuels because it can measure isotope ratios with excellent precision (typically better than 0.02% relative for isotopes which abundance > 10%), thereby establishing the isotopic composition of a fuel very accurately. In combination with the isotope dilution technique, TIMS measurements also generate concentration values having very low measurement uncertainties (typically better than 0.3% relative for U and Pu). TIMS analysis starts with good sample preparation in which the analyte is separated as much as possible from the matrix and from any interfering isotopes (at the same nominal mass/charge ratio). Analytical procedures developed for the determination of the isotopic composition and concentration of U and Pu in fresh MOX fuels lie at the heart of our work on fuel qualification and characterization.



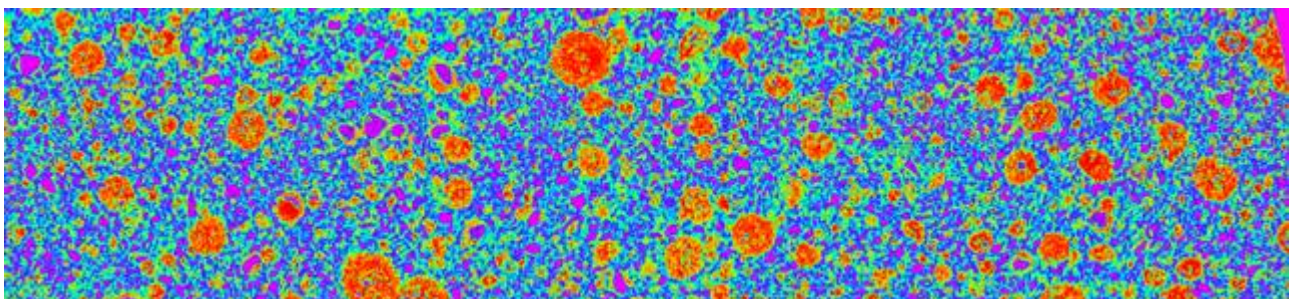
One of the parameters that requires special attention in the production of nuclear fuels is the content of trace impurities. In MOX fuels for thermal power reactors, typical impurities monitored by chemical analysis are Al, Ag, B, K, Cd, Co, Cr, Cu, Fe, Gd, Mg, Mn, Mo, Na, Ni, Pb, Si, Sn, Th, V and Zn. For the determination of the impurity content, and depending on the manufacturers' requirements, two solid-state techniques have been applied at SCK•CEN, i.e. Atomic Emission Spectrometry (AES) and Spark Source Mass Spectrometry (SSMS). In the first trimester of 2007 SSMS will be taken off-line to be partly replaced by Inductively Coupled Plasma Mass Spectrometry (ICP-MS – see figure to the right). For the latter technique the fuel has to be dissolved before the actual analysis.



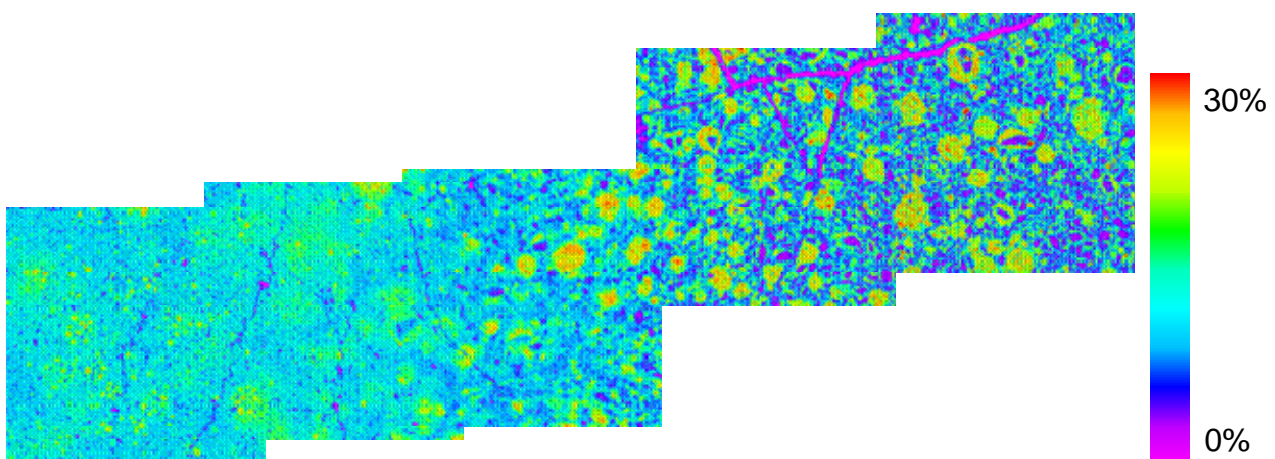
Alpha-global measurements and alpha- and gamma-spectrometry are radiometric measurement techniques that complement the chemical analysis capabilities for the qualification of MOX fuels. Specific procedures for alpha-spectrometry were developed for the determination of $^{238}\text{Pu}/(^{239}\text{Pu}+^{240}\text{Pu})$ ratios in separated Pu-fractions prepared for TIMS measurements, and for the determination of Pu-concentrations in residuals originating from dissolution tests performed on MOX pellets. A typical example of an alpha-spectrum of a Pu-loaded MOX is shown in the figure to the right. The ^{241}Am content of the Pu used in the MOX-production is analysed by gamma-spectrometry of a dissolved MOX sample.



The microstructure research group at SCK•CEN focuses on microscopy and spectroscopy of nuclear materials. Electron probe microanalysis (EPMA) of fresh MOX fuel is used to assess differences in Pu distribution caused by variations in the production process. X-ray mappings of the Pu signal are quantified and provide an immediate view of the Pu distribution across the pellet radius. A typical example is given below, in which the false colours of the image indicate the local Pu concentration from 0% (magenta) to 30% (red) in a region from pellet centre (left) to pellet periphery (right).



Post-irradiation examinations of the same MOX fuel allow a similar assessment to be made after irradiation (see below a collage of X-ray maps from pellet centre (left) to pellet periphery (right)), which immediately shows the effects of the irradiation on the local Pu distribution, with a clear homogenisation in the central region of this pellet. This depends of course on the irradiation history and the original microstructure.



In the laboratories, sample throughput is high and analysis time varies from a few hours to days after the arrival of the samples, depending on the techniques applied. The different techniques and methods discussed above are powerful and versatile measurement tools which can be combined, thereby allowing the analysis of a wide variety of nuclear fuel parameters that are important not only from the viewpoint of industrial production, but also for safety, safeguards and R&D purposes.

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