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Waste management

Early Identification and Characterization of Waste

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A THOROUGH KNOWLEDGE of the waste produced is of primary importance for both an optimized predisposal management of radioactive waste and the quantification of the source term of future repositories. To this end, SCK•CEN develops destructive and non-destructive analytical techniques, and correlation models based on key isotopes.

Objectives The programme aims to measure the inventory of critical and key nuclides in different waste streams and to identify and develop correlations between those isotopes.

Determination of the disposal-critical nuclides in waste from PWR power plants The presence of long-lived nuclides poses a major safety problem for the long-term storage of radioactive waste. Unfortunately, their low content, their low specific activity, or the particular characteristics of their radiation currently make them difficult to measure because of the presence of other highly active nuclides. Advanced separation techniques are therefore necessary to allow their immediate determination without interferences.

The critical nuclides are produced in nuclear reactors either by activation (^{3}H, ^{14}C, ^{59}Ni, ^{63}Ni, ^{94}Nb) or by fission and transmutation (^{90}Sr, ^{99}Tc, ^{129}I, ^{135}Cs, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Am, ^{242}Cm, ^{244}Cm). Their concentration may be correlated to so-called key nuclides, presently measurable with a good accuracy and representative for activation (^{60}Co) or fission (^{137}Cs) reactions. We determined the scaling factors for most of the critical nuclides with respect to the key nuclides in evaporator concentrates, ion-exchange resins, and coolant particle filters from the power plants of Doel and Tihange.

In 1997, we also focused our efforts on developing a more efficient dissolution technique, based on microwave digestion. We developed suitable dissolution schemes for resins, cement, and incinerator ashes, leading to clear solutions in a minimized volume. The resulting matrix, simpler than in the fusion-dissolution method, simplifies the further use of separation techniques.

We further developed suitable separation and measurement techniques for ^{99}Tc, ^{129}I, ^{241}Am, ^{242}Cm, and ^{244}Cm and investigated several purification methods for low amounts of ^{99}Tc in complex matrices. Solvent extraction using tri-n-octylamine in xylene yielded the most promising results. The presence of an excessive amount of ^{106}Ru, however, interferes with the measurements, even with additional purification steps.

The method previously developed to separate ^{129}I is not suited to determining concentrations below 5 Bq per ml of solution. At higher concentrations, the ^{129}I can be measured by gamma spectrometry and the separation yield can be monitored with an ^{129}I tracer. For very low concentrations, however, the amount of ^{129}I is insufficient for gamma spectrometry and the salt content of the sample impedes neutron activation analysis.

Destructive measurements of alpha emitters in radioactive waste The alpha-emitters content of radioactive waste is prevalent for its classification. However, the determination of trace amounts of alpha emitters in strong beta- and gamma-emitting waste is subject to interferences. A standard procedure for their measurement needed therefore to be set up to allow one to define a reference level for a possible declassification. This procedure includes a dissolution method for different types of conditioned and unconditioned waste forms, separation techniques to isolate the alpha-emitting isotopes, and the choice of suitably sensitive measurement techniques.

Within the framework of Working Group 5 on Chemical and Radiochemical Destructive Analysis of the EC, we started an international programme, together with laboratories from Germany, Italy, Spain, and the United Kingdom, to set up a standard procedure for measuring traces of alpha emitters. We ordered a new alpha spectrometer with increased sensitivity to measure the expected low activities and should receive it in early 1998.

Modelling and development of gamma and neutron assay techniques Passive neutron coincidence counting for the nondestructive assay of fissile material or plutonium-bearing waste is commonly performed using the shift-register technique, which allows one to count the real coincident neutrons emitted by the spontaneously fissioning plutonium isotopes. To determine the plutonium content in a waste drum with the shift-register technique, the detection efficiency of the neutron counter with its sample must be known. This efficiency can however vary from waste drum to waste drum because of unknown neutron interactions in the waste, and is generally unknown. To determine the actual neutron detection efficiency for each measurement, we therefore developed the Time Interval Analysis (TIA) technique, which allows absolute neutron assays.

Neutron measurements are however subject to important biases such as the neutron die-away process, the dead-time effects, and the cosmic spallation. These effects were extensively studied and appropriate correction algorithms were developed and incorporated in the TIA technique. To validate this technique, we performed measurements with our "hexagon" neutron counter, which contains 60 ^{3}He detectors embedded in polyethylene. The measurements (Fig. 1) proved that TIA is an unbiased measurement technique for plutonium masses as low as 100 mg $^{240}Pu_{eff}$. For high-count-rate applications, Monte Carlo simulations showed that TIA has not only a more accurate dead-time correction, but also a better statistical precision than competing techniques.

Numerical simulation is an important tool for developing nondestructive techniques and systems and predicting the systems' response. To broaden our knowledge in this field, we participated in the ESARDA Reals Prediction Benchmark Exercise. This exercise aims at predicting the real neutron coincidence count rate for a fresh PWR fuel assembly placed in a neutron coincidence collar. We developed a model and working procedure to predict this count rate on the basis of parameter estimates obtained from simulations with the Monte Carlo code MCNP. This model describes the different origins of neutron correlation and the time and spatial aspects of neutron multiplication in the fuel.

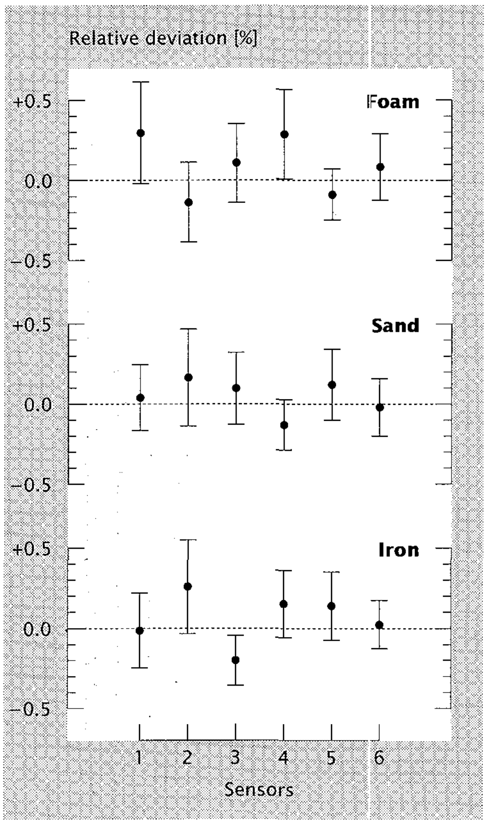

Figure 1 Relative deviation $(Pu_{measured} - Pu_{declared})/Pu_{declared}$ as measured with the TIA method for the foam, sand, and iron matrices. The $(1 - \sigma)$ error flags show the reported uncertainty.

We also contribute to an EC project on the optimization of neutron assay of waste. One of the objectives of this project is to quantify better the localized neutron activity in a 220-litre waste drum using nondestructive techniques. We therefore developed a model and the necessary numerical tools to determine the actual location of a neutron point source in a waste drum with an accuracy better than 10%. This model uses the individual count rates of the detectors of the neutron counter to derive the angular and radial position of the point source. When the neutron activity is not a point source but has a finite volume distribution, the model finds the center of gravity of the localized activity with an accuracy generally better than 15%. It allows accurate quantification of the plutonium content of a waste drum regardless of its distribution.

For another EC project, on the optimization of gamma assay of waste, we are developing a fast and user-friendly computer code, called SOLIDANG, to calculate relative system responses of gamma waste assay systems. In particular, this code calculates gamma-attenuation correction factors as a function of different sample-related parameters (waste matrix density, composition and distribution, shielding layers, activity distribution). The beta version of the code has been released among the project partners, to be tested and validated.

Measurements and validation of waste assay techniques Following the principles of sound quality assurance for nondestructive measurements concerning gamma spectroscopy by both the segmented gamma scanner and the low-level waste assay system Q^2 on 220-litre waste drums, we established working procedures and participated in two international interlaboratory tests. The first, organized within the ESARDA Nondestructive Assay working group, dealt with the determination of the uranium enrichment of UO_2 samples via gamma spectroscopy. The other is organized within an EC project on the assay of 220-litre waste drums using neutron and gamma assay systems. We already measured eight drums and expect to measure eight more and evaluate the results by the end of 1998.

Working procedures and analysis software are continuously revised and improved to attain the highest accuracy and confidence. Upgrading of the equipment is ongoing.

The decommissioning of nuclear installations produces huge amounts of waste. In view of its high disposal costs and because of environmental concerns, the waste that is apt for free release is carefully selected, possibly after radiological decontamination of the surface. The way to decide finally on commitment with free-release criteria that come essentially from international consensus is by a nondestructive assay of the items under consideration. In relation to the decommissioning of the BR3 reactor, several devices have been examined for their performance, precision, accuracy, detection limits, and measurement time. Several detector types were investigated such as a germanium detector and NaI(Tl) scintillators, that were used to perform nondestructive measurements on waste items from the dismantling operations. Data analysis is in progress, as well as an evaluation of how these measurements can support the free-release criteria.

As a routine activity, we continued to assay waste drums of SCK•CEN for waste management purposes in an overall site restoration effort, and measured more than 60 waste packages on request of external clients.

Partners, sponsors, and customers

Scientific partners Belgonucléaire (BN) — Belgoprocess (BP) — Euratom — European Commission (EC) — Franco-belge de fabrication de combustibles International (FBFC Int.) — International Atomic Energy Agency (IAEA) — Nationale Instelling voor Radioactief Afval en Verrijkte Splijtstoffen/Organisme national des déchets radioactifs et des matières fissiles enrichies (NIRAS/ONDRAF)

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