Publishable JRP Summary Report for JRP ENV09 MetroRWM Metrology for radioactive waste management

Background

The operation of nuclear facilities and their decommissioning (once their life cycle ends), must be done in a way to minimise environmental impacts. However, this can be only achieved by accurately measuring the radioactivity of radionuclides in waste and environmental samples, using standardised and traceable measurement methods based on gamma-ray spectrometry, accurate decay data and radiochemistry.

Dangerous radioactive and non-radioactive gases appear in radioactive waste repositories which could pose a risk to repository workers and the wider population in the event of gas leaks from the repository. Therefore, only the accurate measurement of the area of the repository and the tightness of individual barrels containing the waste could prevent any undesirable irradiation of people over the course of time.

Need for the project

The waste generated by nuclear facilities must be reliably separated into radioactive waste and waste that can be released into the environment. For free release measurement, the national regulators have specified very strict limits for the mass and surface activity of selected radionuclides contained in waste. At the same time, the duration of the individual measurement should not be too long because of the large number of samples to be measured. However, compliance with these mutually conflicting requirements necessitates development of a standardised traceable method based on spectrometry. To meet repository acceptance criteria, medium-active or low-active waste must be measured and the activities of radionuclides contained within the waste must be determined prior to consignment to a disposal site. This requires development of a standardised traceable measurement method.

To protect workers in the repositories and also the general public against unexpected on-site as well as off-site radiation, it is necessary to monitor gaseous radionuclides such as H-3, C-14 and Rn-222. To do this, existing radiometric techniques combined with the separation of chemical species must be developed for the repositories.

In order to monitor the vicinity of the nuclear facility, you must be able to carry out in-situ measurements, in case of an accident, or in the case of site decommissioning, in order to minimise waste management costs. These measurements should be fast and flexible and there is a need for a rapid method of preparation and measurement of samples in the place of their origin. However, this requires the development of a standardised traceable method, to be used in a mobile radiochemical laboratory.

To ensure correct and accurate measurement, all measurement systems must also be calibrated with calibration standards and reference materials traceable to national standards.

Finally, the safe and optimised management of waste relies on a precise knowledge of the variation with time of artificial radionuclide concentrations. This requires the precise and accurate knowledge of decay data, especially half-lives for long-lived radionuclides produced in nuclear facilities.

Scientific and technical objectives

This JRP addresses the following scientific and technical objectives, in order to provide validated and traceable measurements/reference methods for radioactive waste management.

- Development of standardised traceable measurement methods for solid radioactive waste free release (clearance levels verification) and for acceptance of solid radioactive wastes to repositories (acceptance criteria verification), according to international recommendations (EC and IAEA): as well as design of measurement facilities, software, calibration and testing methods.

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- Development of novel instruments and methods for in-situ measurements: this includes improved on-site radiochemical analysis, rapid in-situ screening techniques for alpha, beta and gamma emitters and measurement of activity at varying depth.
- Development of a gaseous effluent monitor/sampler for stored wastes. Rapid, sensitive methods are required to determine rates of bulk gas production, chemical composition (CH_4, CO_2 or H_2) and activity concentrations of key radionuclides (e.g. H-3, C-14, Rn-222).
- Development of standards and 'spiked' or characterised 'real' reference materials for ensuring accurate, traceable radio-assays of materials from sites (concrete, steel, aluminium, cables, wood, insulator and others).
- Improvements to decay data for selected radionuclides present in nuclear wastes, focusing on half-life measurements of long-lived fission and activation products.

Expected results and potential impact

For free release measurement, a transportable free release measurement facility design will be developed based on germanium detectors, with special shielding and appropriate evaluating methods and software. This will allow identification of individual radionuclides in waste material and lower uncertainties significantly to typically 10% and thus will lead to more effective recycling and reuse of waste materials and minimising of disposal costs.

For repository acceptance measurement, an optimised measuring device will be designed based on germanium detectors with a suitable collimator and shielding. This will allow accurate measurement over a wide range of radionuclide activities with uncertainties typically 20% and thus better radionuclide characterization of waste materials.

Since the beginning of the JRP, clearance levels and acceptance criteria legislation, requirements, standards and measurement systems used by JRP-Partners for free release and repository acceptance measurements were reviewed and reported. Free release measurement testing facility (FRMF) and Repository acceptance measurement testing facility (RAMF) were implemented. RAMF was equipped by special collimator enabling characterisation of wastes in wide dynamic measuring range. HPGe detectors for measurement efficiency calculation have been pre-characterised and validated using point standard sources and three MC codes compared. Measurement geometry optimisation has been completed for FRMF and RAMF. Spectra and activity measurement algorithms have also been developed. In addition, to this mass measurement and material density efficiency correction algorithms and homogeneity and hot spots identification and activity averaging algorithms have been produced and databases of radionuclides have been created. Metrological tools (reference materials and phantoms) for FRMF have been specified. Detection efficiency curves have been constructed for different types of waste materials in the energy range 59.5 keV to 2 MeV using MC calculations. Furthermore, FRMF and RAMF Monte Carlo models have been validated using real and spiked steel, building and light reference materials and 'point' standard sources located in phantoms.

Production of an experimental FRMF and RAMF in full working order is an important step towards the development of standardised and traceable measurement methods for solid radioactive waste free release and acceptance of radioactive wastes to repositories according to requirements of international and European recommendations.

For in-situ measurement, transportable integrated radiochemical analysis laboratory for rapid on-site measurement of pure α-, β- or X-ray emitting radionuclides will be developed and uncertainties lowered to typically 10% or better. This will allow efficient quick reaction to radiation situation on-site and reduction of analysis costs.

Novel instruments and methods for in-situ measurements are also being developed: including improved on-site measurement of radionuclides employing rapid in-situ analysis techniques for α-, β- and γ-emitters at varying depth. The radionuclides of interest have been selected and two types of concrete material identified (bioshield concrete and non-active concrete). Initial measurement work has been carried out on ^{41}Ca; interference from K-41 was identified and mitigation of this interference is in progress. In addition, validation of the PERALS system has been completed and the TriAthler system has been validated and reports on both validation exercises are due for issue. Proficiency test samples of Sr-90 and U-nat are in various stages of completion identified and dispatched, and the analysis of this material has been started. Analysis of PROCORAD 2013 is complete, using the NPL ARSE II system and the PTB 'bench' chemistry and the results are being assessed (bench procedures have been developed for analysis of material).

An integrated measurement system for monitoring of radioactive gases and vapours containing H-3, C-14, Rn-222 and non-active 'bulk' gases arising from stored wastes will be developed and uncertainties lowered to typically 10% or better. This will provide a means of alerting workers in repositories of hazardous radioactive gases and will improve our understanding of waste degradation.

A 'LSC2' liquid scintillation counter has been inspected, renovated and then tested for its response to H-3 and C-14 standard liquid scintillation sources. Concerning the gas trapping equipment, two bubbler units have been obtained and their operating procedures examined. It was concluded that it will not be practical to incorporate a bubbler unit of this type into the system envisaged due to the difficulties associated with the connection of the fluid handling system, so a new bubbler system will be built. However, a prototype gas transfer system (i.e. a system to fill and drain the bubbler bottles, mix fluid from bubblers with a scintillation cocktail and then transfer the mixture to a counter) has been built and tested. The construction of the time-integrating monitoring system for H-3 and C-14 compounds is almost complete, and the component testing and calibration work are underway. For the mid-infrared spectroscope, all the components have been purchased, experimental arrangement set up and optimised, and sensitivity improved. Finally, the development of a mid-infrared laser spectroscope for measurements of gaseous radiocarbon dioxide isotopes will enable high measurement sensitivity for these species; moreover, the instrument is compact, which will facilitate in-line, on-site measurements.

New reference materials and standard sources will also be developed for calibration of above mentioned devices. This will lead to more accurate measurement and lowering of uncertainties at activity of radionuclides measurement.

The measurement containers for reference materials measurement in FRMF and phantom containers for non-active materials and 'point' standard sources have been designed and manufactured. Reference materials and standard sources for free release measurement have been defined, produced and characterised. The reference materials are steel balls with Cs-137, steel tubes with Co-60 and Ag-110m, and gravel and clay balls with natural radionuclides. The standard sources are four sets of 'point' sources of activities from 30 Bq to 1 MBq with radionuclides Am-241, Co-57, Cs-137 and Co-60. For radiochemical analysis, inactive and active concrete material is available and characterisation is currently underway. The bioshield concrete powder from Sellafield has been selected as reference material for spiking with solutions of Sr-90, U-nat, Pu-240, Am-241, Ca-41, and Th-232 and the activity of the Sr-90, U-nat and Pu-240 solutions have been standardized. Further to this, solutions of the other radionuclides are available and the measurements are under progress. The activities of the Uranium-, Plutonium-Isotopes and Sr-90 have also been measured, and a 'concrete solution' containing Uranium, Plutonium and Sr-90 has been selected as the second reference material. For radioactive gases measurement the standards with H-3, C-14 and Rn-222 are available on for testing the instruments under construction.

Half-lives for long-lived radionuclides Ho-166m, I-129 and Sm-151 occurring in waste will be improved using mass spectrometry measurement and absolute activity measurement, with uncertainties up to 3%. This will lead to better knowledge of the variation with time of artificial radionuclides in the environment.

The activity measurements of Ho-166m are complete, a report has been prepared and declared as a EURAMET key comparison and a SIR comparison. The I-129 activity measurement exercise has also been accepted and registered as a EURAMET supplementary comparison. In addition, the mass measurements of I-129 have been finished and the Sm-151 activity measurement exercise has been accepted and registered as a EURAMET supplementary comparison.

The project has so far contributed 39 presentations to the meetings, conferences and workshops and 9 presentations or contributions to standards, regulatory bodies and working groups. 2 scientific articles have been published in the journal Applied Radiation and Isotopes and 1 in a peer-reviewed conference proceeding. 5 scientific visits and 2 workshops for about 30 stakeholders have been organised. Further to this, 4 end-users have shown an interest in the FRMF on-site installation or production and 1 stakeholder has declared interest in the monitoring system for repositories production.

JRP start date and duration:	01/10/2011, 36 months
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