Final Publishable JRP Summary for ENV54 MetroDecom Metrology for decommissioning nuclear facilities

Overview

As the number of nuclear facilities being decommissioned in Europe increases sharply there is a need for safe and reliable methods to restore these sites and dispose of radioactive waste. The treatment and subsequent storage of radioactive material is expensive, but the financial burden can be reduced with accurate measurements and guidelines for decision-making on 'safe' disposal and contamination levels. Reliable measurements and strict criteria are important for ensuring public trust in the decommissioning process and the wider nuclear industry. Measurements of large quantities of nuclear waste also need to be fast and precise to help categorise and dispose of it safely, and in a cost-effective manner. There is also a requirement for ongoing monitoring of radioactive waste. This project addressed the needs of the decommissioning process by the development and implementation of new radioactivity measurement techniques, instruments, calibration standards and reference materials. It also ensured that knowledge was transferred to regulators, the nuclear industry, instrument makers and standardisation bodies, thus supporting the 'safe' decommissioning of European nuclear facilities.

Need for the project

More than 200 power reactors are being decommissioned in the EU, or will be in some phase of the decommissioning process, by the year 2025. Nuclear decommissioning covers many activities such as: the shutdown of the facility; environmental restoration of the site; and monitoring of the disposed radioactive waste. The decommissioning process relies on the ability to characterise and segregate all waste material into various radioactive waste categories precisely and rapidly. This allows safe, or free, release of cleared waste into the environment; recycling or volume reduction of contaminated waste; further treatment of contaminated waste or long-term storage and monitoring in repositories.

The decommissioning process is not only very costly but is currently being carried out in the context of low public confidence in the safety of nuclear technology. Therefore the restoration of public trust relies on adoption of standardised and traceable methods for measuring radioactivity, and the demonstration of compliance with the legislation contained in:

- EU Directive 2011/70/EURATOM: Management of spend fuel and radioactive waste
- EU directive 2013/59/EURATOM: Protection against ionising radiation

Also, compliance with the recommendations contained in:

- Commission recommendation 2006/851/EURATOM: Management of financial resources for the decomissioning of nuclear installations, spent fuel and radioactive waste
- Commission recommendation COM(2005)666: Prevention and recycling of waste

In addition, compliance with the International Atomic Energy Agency safety standards on radiation protection and radioactive waste (NS-G-2.7, GSR part 5, SSR-5, GS-G-3.3, WS-G-2.5, TECDOC-1537).

This project extended the experimental methods and devices developed in the previous EMRP project *Metrology for Radioactive Waste Management* (ENV09) to the decommissioning of nuclear facilities. Alpha and gamma radiation were measured, sampling techniques explored and safe ways of monitoring long term waste repositories investigated. This involved implementing and testing industrial measurement devices both

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on site and under real conditions. The results then addressed the need for decommissioning measurements to be carried out using standard methods with traceability to national standards, improving uniformity and the accuracy of measurements across Europe. This will ultimately help meet EU regulations and have a positive impact on the environment and public health.

Scientific and technical objectives

The project's objectives were:

1. To develop methods for the radionuclide characterisation of different types of materials present on the site being decommissioned.
2. To develop traceable methods for the pre-selection of waste materials prior to measurement for repository acceptance or possible free release.
3. To develop and implement free release measurement technologies on a decommissioning site.
4. To develop methods for monitoring in radioactive waste repositories.
5. To develop reference materials and standard sources for calibration, validation and testing of devices, instruments and procedures developed in the previous objectives.

Results

1 Development of methods for the radionuclide characterisation of different types of materials present on the site being decommissioned.

Mapping of contamination inside nuclear facilities:

GAMPIX technology is a gamma radionuclide imaging technique used to locate radioactive hot spots. GAMPIX was shown to have the potential for quantification of gamma dose rates, and particularly for automated remote gamma contamination inside nuclear facilities. Development of alpha radionuclide detection technologies showed the potential for alpha contamination remote mapping. A new metrological method and software, based on the modelling of the transmission of beta radionuclide particles for the improvement of the traceability and the accuracy of surface beta contamination measurements were also developed. These improved detection technologies for alpha, beta and gamma radionuclides represent significant improvements over the current state of the art and will be vital for clearing potentially contaminated waste during decommissioning. A test case in a real decommissioning project of Belgium Reactor 3, a pressurised water reactor, looked at contamination distribution mapping using the project's new detection technologies. The results showed that there were clear advantages in reducing the number of measurements needed and the uncertainty, as well as the costs of the pre-decontamination characterisation process.

Statistically valid sampling strategies and methods:

Existing radioactive sampling strategies for potentially radioactive decommissioning waste and statistical analysis procedures were reviewed and summarised. Based on this the project developed general guidance for planning sampling campaigns for such material. The guidance was optimised to minimise the numbers of radioactive samples taken, without introducing additional risk in terms of false negative results (that may result in the unplanned release of radioactive material) or false positives (which unnecessarily use up space in radioactive waste repositories). The outcomes of this task are in the public area of the project website.

Semi-automated rapid radiochemical analysis procedures:

A novel method to dissolve graphite samples quickly for radiochemical analysis was developed. Currently, difficult to measure radionuclides such as zirconium (Zr^{-93}), samarium (Sm^{-151}), strontium (Sr^{-90}), and uranium and plutonium isotopes can only be measured by destructive analysis. However, these radionuclides are usually found in concrete, graphite or steel. New measurement methods for the semi-automated, rapid and simultaneous analysis of selected radionuclides using radiochemical techniques were developed by the project and validated with characterised blank matrices and radioactive and stable standards. These new methods will reduce sample turn around and analysis times and will be beneficial for rapid measurement, in particular for concrete, graphite or steel on a nuclear decommissioning site.

Scaling factors for determination of radioactivity of low-level waste packages:

Scaling factors are the parameters derived from the mathematical relationships between the radioactivity of difficult-to-measure nuclides with that of the key radionuclide determined from sampling and analysis data, as defined within ISO 21238:2007. The project reviewed existing information and databases for the use of scaling factors, along with techniques for deriving new factors. Based on the results, gaps were identified in the knowledge base that may be an issue to EU wide decommissioning issues, including the early French and British gas-cooled/natural uranium/graphite moderator power reactors.

Overall, this objective improved the current ability to map contamination inside nuclear facilities. The project has determined statistically valid sampling methods, effective sampling points, new semi-automated methods for quicker radiochemical analysis and scaling factors to aid the faster characterisation of materials on decommissioning sites. Using these new measurement techniques decision makers and operators will now be able to simplify decommissioning processes and help to minimise worker exposure.

2 Development of traceable methods for the pre-selection of waste materials prior to measurement for repository acceptance or possible free release, and

3 Development and implementation of free release measurement technologies

Reduction of decommissioning costs of nuclear facilities requires precise and quick segregation of the various waste categories. Reducing the current conservative approach which is based on disposal instead of free release, or higher repository category selection than appropriate, will save money but needs reliable data.

A new measurement system for the pre-selection and free release of radioactive waste based on unique lead-free shielding blocks was developed and implemented on a decommissioning site. The new measurement system improves the throughput, accuracy, reliability, density and radioactivity distribution scanning and portability of the measurement.

Detection of radioactivity can be optimised for specific uses by integrating individual detectors into an array, rather than using several different detectors separately. The project investigated this array detector system and found that the minimum detectable activities were 2 to 5 times lower than could be previously achieved with single detectors, depending on the radionuclide. Traceability of the new array method to primary activity standards was achieved by the development of new calibration procedures and from this the project was able to reduce the standard uncertainties for radioactivity measurements to less than 20 % for pre-selection of wastes and 10 % for free release measurement.

The validation of the array detector system at a real waste measurement campaign on a decommissioning site demonstrated that the successful use of the method and these results will allow future decommissioning measurements to be performed using standard methods with traceability to national standards of radionuclide activity. This should also contribute to a reduction in the quantity of very low level, and low level radioactive waste being incorrectly sent to repositories (including recyclable waste) and will help to guarantee the integrity and cost-effectiveness of waste clearance and disposal processes with improved safety and accuracy.

4 Development of methods for monitoring in radioactive waste repositories

Radioactive waste and the facilities that store radioactive waste must currently be monitored for 10's to 100's of years to ensure the safety of workers, the local community and the environment. This includes monitoring radioactive gas emissions that may be hazardous to health, thermal power of waste packages containing nuclear waste to ensure the structural integrity of the package, and the temperature of the waste throughout the storage facility to ensure the structural integrity of the facility as a whole.

Three distinct, prototype technologies were developed by the project for the monitoring of ^{14}carbon (^{14}C) and/or ^{3}hydrogen (^{3}H) gas; the two radioactive gases most likely to be generated and released from radioactive waste. The first prototype technology was a novel mid-infrared laser spectroscopy system, the second was an instrument for real-time cryogenic separation of carbon dioxide (CO_2) from air for subsequent $^{14}CO_2$ measurement and the third instrument was an automated ^{3}H/^{14}C trapping and measurement system. The successful automation and testing of all 3 prototype technologies for sampling and measurement was demonstrated and has the potential to provide significant labour cost savings.

Raman-Distributed Temperature Sensing (DTS) systems use optical fibres for measurement of temperature and stress inside of repositories to ensure the structural integrity of the repository. Raman-DTS is a cost effective, durable solution for the long-term monitoring of temperature throughout a large concrete structure, such as a disposal facility. The thermal power output of each waste package can be measured using Raman-

DTS to avoid hot spots inside the repository which can be dangerous for the structural integrity of the repository and of individual waste packages. The project developed facilities for the characterisation of the DTS. LNE developed a horizontal furnace of 25 meters length and rectangular thermal enclosures for the metrological characterisation of Raman-DTS systems and has enabled a new calibration service to be offered to both industrial and academic users, as well as DTS manufacturers. LNE is currently the only European national metrology institute able to provide such a calibration service in this field.

The structural integrity of waste packages must be ensured for waste to be stored safely for extended durations. New calorimeter prototypes for the measurement of the thermal power of nuclear waste packages of at least 175 litres, as well as 'power reference packages' for their calibration by electrical substitution, were designed. These calorimeter prototypes will enable a range of waste packages to be calibrated and support the development of new packages. The measurements performed with these calorimeters demonstrated the feasibility of thermal power measurements up to 500 W with a low-level of uncertainty (lower than 5 %), making the systems suitable for high level waste that can generate >2 kW/m³.

5 *Development of reference materials and standard sources for calibration, validation and testing of devices, instruments and procedures from the previous objectives*

A series of standardised radionuclide sources and reference materials were produced by the project:

- 1100 inactive steel balls and 1100 inactive plastic balls with drillings to insert 1100 Eu-152 or 1000 Co-60 point sources, for testing and calibration of the free release measurement system.
- A set of 9 point-like standard sources of Eu-152 with activities in the range from 100 Bq to 1 MBq for hot-spots simulation in phantoms filled with 'inactive' steel, building or light material (steel balls, low activity gravel, plastic balls) for testing and calibration of the free release measurement system.
- A Co-60 source with an activity of 95 MBq and 6 different non-active materials to test the scanning facility.
- Three blank materials (concrete powder, steel solution, graphite) in combination with five standardised single nuclide solutions (Sr-85, Zr-93, Sm-151, U-236, Pu-241) for testing and validation of the new developed radiochemical procedures.
- Gaseous reference sources with $^{14}CO_2$ and $^{14}CH_4$ in-air for testing the ^{14}C monitors.
- A set of 8 large area sources produced by quantitative dropping of calibrated solutions of ^{60}Co and ^{137}Cs for testing and calibration of surface contamination monitors.

These were designed to characterise the radioactive waste and meet the reliability and traceability requirements the EU Directives, Recommendations and standards listed in the Need section of this document. The radionuclide sources and reference materials were used to calibrate and test the new measurement devices and methodologies which were developed within the project in objectives 1-4.

The methods for the production of the radionuclide sources and reference materials developed in the project are published and can also be used to build future calibration sources for the whole field of decommissioning measurement devices.

Actual and potential impact

Dissemination of results

The results of the project have been presented to stakeholders at national and international conferences. The project submitted 39 conference presentations and 22 publications including 18 peer-reviewed journal papers. 7 articles were also published in trade journals to disseminate beyond the specialist nuclear decommissioning industry, and are available on the project website.

Four good practice guides were developed and are available on project website http://www.decommissioning-emrp.eu/?page_id=1031

- Measurement of surface activity and mapping the contamination
- Radionuclide scaling factors (based on ISO 21238:2007)
- Segregation measurement of solid waste

- Free release measurement

A training course for network operators was held and lectures were given at the 8th International Summer School on Nuclear Decommissioning and Waste Management. Three workshops for nuclear decommissioning operators, nuclear regulators, standardisation bodies and measurement instruments manufacturers were also held in Trebic (Czech Republic), Ispra (Italy) and Warrington (UK) which covered the new measurement techniques, best practice guides, reference materials, and on-site demonstrations of the new free release and pre-selection waste measurement systems developed by the project.

Input to standards

The results of the project were disseminated to international and European standardisation bodies and working groups including IAEA Safety Standards and supporting publications (Safety Reports, TECDOCs), European Commission Directives 2011/70/EURATOM and 2013/59/EURATOM and Recommendations 2006/851/EURATOM and COM(2005)666, nuclear agency (Expert Group on Fukushima Waste Management and Decommissioning R&D), IEC TC45, ISO TC85 (SC5) and IEC TC/SC 86C Standardisation Committees working on radioactive surface contamination monitoring, and French Standardisation Association (AFNOR).

Work from the project was incorporated in the published documentary standards: IEC 60325:2002 Radiation protection instrumentation and ISO 7503:2016, ISO 8769:2016 Measurement of radioactivity. The facility for the characterisation of Raman-DTS systems will enable LNE to contribute to the standardisation committees related to the distributed temperature measurements ie. AFNOR UF86 “photonics” and IEC TC/SC 86C/WG2 “Optical fibre sensors”.

Actual impact

The uptake of project’s outputs will build confidence in the decommissioning process, and will lead to commercialisation of the measurement techniques. Examples of impact are:

- The partner and nuclear specialist company NUVIA has invested in a new measurement system prototype built during the project for radioactive waste pre-selection and free release. The system was constructed and implemented on a decommissioning site and validated during measurement campaign with real radioactive waste.
- Electricity of France (EDF) and French National Radioactive Waste Management Agency (ANDRA) developed methods for the metrological characterisation of distributed temperature sensing systems based on optical fibres, based on the work in objective 4.
- The prototype gamma camera GAMPIX, developed in objective 1, was utilised by CANBERRA, a measurement company for the nuclear industry, for upgrade of their industrial gamma camera iPIX.
- An agreement with the LabLogic Systems Ltd. concerning development of an integrated counter for monitoring of radioactive waste repositories has been made. This technology will decrease the measurement time and provide cost savings. Several prototype systems have been sold for monitoring of ^{3}H stack emissions, a critical part of monitoring discharge of nuclear facilities (objective 4).
- A cryogenic system for CO_2 trapping was designed with the cooperation with Air Liquide, a supplier of industrial gases. ^{14}C is very toxic, and this technology will enable ^{14}C labelled CO_2 to be sampled from the air to help ensure radiation protection of workers (objective 4).
- An air flow calorimeter prototype for the measurement of thermal power generated by large radioactive waste packages was developed in cooperation with the French National Radioactive Waste Management Agency (ANDRA) (objective 4).

Potential impact

The outputs from the project are also being explored for future products and services:

- Radioanalytical companies as SPEX SamplePrep, Agilent Technologies and Triskem International SAS are interested in the radiochemical research outputs from objective 4.

- SPEX SamplePrep produces the furnaces which were used to prepare radioactive samples by dissolving concrete and graphite, Agilent Technologies make instruments for measuring radioactivity and Trisken International SAS produces extraction chromatography resins.
- Negotiations about the uptake of the mid-infrared spectroscope prototype for on-line on-site measurements of radiocarbon emissions from nuclear power plants and repositories are on-going with a European company developing instrumentation for the nuclear sector.

There are also opportunities for other producers to launch a new generation of radioactive waste monitors with special shielding, based on the work in objective 2 and 3.

The project improved the throughput, accuracy and reliability of on-site measurements which are key for the safe decommissioning of nuclear facilities. Future, more reliable decision making about the safe release of radioactive waste into the environment can now be supported by the information gained in this project. The European decommissioning industry, and both the waste management and the long term operation of waste storage facilities, will also be able to improve the management of waste generated during decommissioning. Finally, the implementation of new and more precise measurement methods and techniques will result in more effective protection of the general population and workers against undesirable additional radiation exposure. This will ultimately build public trust and acceptance of nuclear technologies.

List of publications

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