Publishable Summary for 16ENV09 MetroDECOM II
In-situ metrology for decommissioning nuclear facilities

Overview

This project addresses one of the most significant environmental challenges facing EU member states: ensuring the safe disposal of radioactive waste from decommissioning nuclear sites. The key to dealing with such wastes is quantifying the radioactivity content, so that decommissioning can be planned and implemented to minimise the risk to members of the public and the environment.

The project aims to provide nuclear site operators with measurement techniques that can be used to measure radioactivity for planning decommissioning, for segregating and checking waste materials during demolition, and for monitoring the condition of waste packages in radioactive waste repositories.

Need

The first-generation nuclear power plants and reprocessing facilities are coming to the end of their working lives. Ninety-one power plants are being decommissioned in the EU; most of the remaining 129 reactors plus fuel cycle facilities will also be in decommissioning by 2030.

The aim of the decommissioning process is to clear the site, while minimising the risk to the public and the environment from the hazardous waste arising. The cost of decommissioning and waste management in the EU is estimated to be in excess of 150 billion Euro.

The key to safe and cost-effective disposal of the waste is accurate characterisation – determining the physical, chemical, and radiological characteristics of the material. This enables nuclear site operators to plan the demolition process, assign the waste to the most cost effective disposal route and then to monitor that the waste is being stored safely. The metrological challenge is that nuclear sites have been operating for many decades, so the disposition and quantities of hazardous materials are not well known. There has been significant progress towards developing the new techniques needed in EMRP projects ENV09 MetroRWM and ENV54 MetroDECOM; this project focusses on bringing the techniques into use on nuclear sites and developing further innovative solutions based on lessons learned.

Regulatory bodies and international organisations have therefore carried out detailed studies of technical needs in the field. The common themes that have been raised are: (1) improvements in capability, (2) harmonisation and quality assurance, and (3) sharing knowledge. The improvements in capability that are required include rapid, on site measurements improving the accuracy and traceability of measurements of waste packages.

These needs are reflected in EU Council Directive 2011/70/EURATOM which aims to encourage technical co-operation to improve safe management of radioactive waste and highlights the importance of building public trust and confidence.

Objectives

The overall objective of this project is to establish a measurement infrastructure that is accepted and implemented by the nuclear industry for the measurement of radioactivity, temperature and stress needed for nuclear decommissioning.

The objectives of the project are:

1. To develop in situ methods for the rapid radionuclide characterisation of the different types of materials present on decommissioning sites. This includes the development of novel measurement techniques that improve the mapping of contamination inside nuclear facilities and the determination of statistically valid and effective sampling methods.

2. To develop and implement traceable measurement systems and methods for waste pre-selection and free release to the environment. This includes the on-site validation of existing waste pre-selection and free release measurement systems and evidence of their traceability to primary standards.
3. To develop a validated waste repository acceptance characterisation system for use on site with very low and intermediate level radioactive waste (LLW and ILW). The system will include gamma scanning and active/passive neutron measurements, and will be sensitive to fissile material.
4. To develop improved measurement systems and methods for monitoring radioactive waste repositories. This includes miniaturised, portable and rapidly deployable gas and water monitoring systems, improved water monitoring systems and systems for monitoring the temperature and strain of inside nuclear waste repositories for the long term. All the systems will be verified with on-site testing.
5. To facilitate the take up of the technology and measurement infrastructure developed in the project by nuclear decommissioning operators, measurement device producers, radiation protection regulators and standards developing organisations. In addition, to establish in collaboration with the end-user community a European network that will co-ordinate a measurement infrastructure for decommissioning nuclear facilities.

Progress beyond the state of the art

The state-of-the-art for on-site radioactivity measurement depends on the type of radionuclide. First, contamination by gamma-emitting radionuclides can be located remotely (this technology was under development in MetroDECOM). This project develops a pre-production instrument that will also identify the specific radionuclides present. An additional strand to this work is developing a measurement technique that uses cheap distributed sensors to reduce the cost. Second, contamination by alpha-emitting radionuclides is difficult to detect remotely – some prototype instruments exist that can detect the UV radiation emitted when alpha particles ionise atmospheric nitrogen, but these instruments only work in the dark and are not practical for routine use. This project aims to develop a pre-production instrument that will work in ambient lighting and be robust enough for use on nuclear sites. Third, some radionuclides are difficult to quantify using non-destructive methods. Radiochemical separation techniques must be applied. Radiochemical separation by bench chemistry is labour intensive and time consuming; this project will validate a novel automatic system based on micro-fluidic devices.

During the decommissioning process, waste arising from demolition of buildings must be packaged and then checked to confirm whether the waste is 'out of scope of the regulations' (effectively non-radioactive) or must be sent to a radioactive waste site. A wide range of waste measurement systems is available, but traceability of measurements is not well established.

Waste packages sent to waste repositories must be measured to determine the radionuclides present, including quantifying any fissile material that may be present. Again, the traceability of existing systems is not well established and the aim of this project is to validate a new capability for such measurements.

Once stored in the waste repository, the condition of the waste packages and the structure of the buildings must be monitored continuously to ensure safe storage. The absence of airborne radioactivity above the packages is a key indicator. This project aims to implement automatic remote monitoring systems (current practice for airborne radioactivity involves staff collecting samples from in situ bubblers). A system for measuring temperature and mechanical stress in repository structures is being developed (no technology exists for this at present).

Results

Development and implementation of rapid methods for measuring the radioactivity content of materials on a nuclear site

Measurements of gamma-emitting radionuclides in situ are well established, with many commercial systems available using high-resolution gamma spectrometers. These systems are quantitative but have to assume the disposition of activity. In the preceding project, an imaging system was developed that can determine the disposition of radioactivity in a facility, but these are qualitative measurements and in general indicate where the activity is, but not the amount or the radionuclide. This project combines both goals, using spectroscopic sensors and novel reference radioactive sources to test and characterise imaging systems.

For remote monitoring of gamma emitters, preliminary testing of a GAMPIX gamma camera with nuclides covering the expected energy range for measurement has been carried out. The energy calibration of the GAMPIX camera using the ^{241}Am, ^{57}Co and ^{60}Co sources is ongoing. A new set of radioactive point sources including ^{54}Mn and ^{134}Cs and ^{133}Ba extended sources is being produced by pipet drop deposition, followed by evaporation and sealing. The sources will be used to evaluate the spectrometric performances of the GAMPIX (Timepix) at different energies.

Alpha emitting radionuclides are very challenging to measure in situ, due to the short range of the particles emitted. There have been some studies and prototype instruments to use the UV light emitted when nitrogen in the atmosphere is ionised, to detect the alphas remotely. None of the instruments has so far proven to be sufficiently sensitive or suitable for use in a decommissioning environment, therefore this project aims at improving and further developing the technique to reach the technical readiness of a pre-production instrument which can be used in areas with loose surface contamination, typical of a decommissioning site.

A comparison of optical scanning and camera imaging has been carried out with different spectral filtering for stand-off alpha detection. A demonstration unit for optical remote mapping of alpha emitters was constructed. The work was focused on the control software as the earlier device version required text commands and manual calculation of several parameters to get the scanner working. Now the control is more automated and easier to be used in field conditions. The optical part is currently being upgraded to rapidly change between two optical configurations at UV spectral range, UVB and UVC. UVB spectral range offers good alpha sensitivity but requires special background lighting conditions. UVC is excellent for visible background light removal, but the alpha sensitivity is reduced.

Measurement of the spatial distribution of dose rates is a useful tool in determining the disposition of radioactivity. This project uses (for the first time) a distributed network of cheap sensors to determine the dose rate continuously as a function of location, using wireless transmission so they can be installed easily in existing buildings and provide real-time dose maps. The high number of relatively cheap data points allows an optimised uncertainty of the initial mapping results and will automate measurements.

Wireless compact battery-operated dosimeters were used to produce the first maps generated by application of approximation and statistical methods. Measurements have been done, performed on the field and demonstrated with a waste management firm. A test setup to validate mapping has been done to conduct measurements over fixed distance and to apply statistical methods for mapping.

The most time-consuming aspect of radiochemistry measurements is sample preparation, and many groups have investigated approaches to automate the process (the potential time and cost savings are very significant). A commercial device was tested previously, but was unable to process typical decommissioning samples and tests were stopped. This project restarts the trials using a new technology for automated radiochemical separations.

The applications of borate fusion have allowed the development of a method for automated digestion of up to 5g of decommissioning samples. An additional procedure for sample dissolution has been produced based on fusion of KF with LaF_3 precipitation, followed by treatment with nitric and boric acids. The radiochemical separation method for Pu, U and Sr isotopes has been developed with good recoveries for Pu, close to the 60 % chemical recovery target, variable results with U with a chemical recovery about 50 %, and Sr showing lower recoveries, which will be investigated further. The extraction and ion exchange chromatography capabilities for Zr/Nb separation, required prior to LSC measurement, have been completed for all commercially available resins. The TEVA resin has shown the most promising results, with preferential elution of Zr in 7-9 M HCl, whilst Nb was retained for aqueous samples. Direct measurement of actinide elements in dissolved steel sample has been demonstrated using ICP-MS/-MS without prior chemical separation, with detection limits in the mBq/g range. The procedure for ^{41}Ca analysis developed for 0.5 g concrete is being tested using higher samples masses, with recoveries assessed using stable Ca tracers.

Development and implementation of a novel automatic measurement system to check whether waste packages are safe for disposal or must be treated as radioactive waste

A validated, transportable, low background, automated system that has been extensively modelled using Monte Carlo techniques has been set up. It incorporates developments in shielding materials and background subtraction algorithms, plus improves throughput by automation. This project aims to demonstrate that the system can be used successfully on a decommissioning site and reach standards required for software and operation in the nuclear industry.

After long-term testing, the automatic measurement system is now ready to operate. The mechanical parts of the waste pre-selection facility and plastic scintillating detectors were transported to a decommissioning test site at CIEMAT in Madrid and installed in the reconstructed measurement hall. Background and calibration measurements have been performed and the pre-selection facility prepared for waste measurement. Improvement of calibration methods for decommissioning materials has started using Monte Carlo codes.

Development and implementation of a sophisticated radioactive waste characterisation system, suitable for use as a waste repository acceptance system for very low, low and intermediate level radioactive waste

Both gamma measurements and passive/active neutron measurements have been applied in the past for the characterisation of radioactive waste. The system available at JRC (Ispra) is more sophisticated than current equipment: the system allows a comprehensive assay of each individual waste item by combining state of the art techniques. The approach includes tomographic gamma measurements combined with attenuation estimation using an external gamma source, and passive neutron measurements for the estimation of spontaneous fission content and (alpha, n) emission, plus active neutron interrogation to determine directly the fissile material component. Combining measurement techniques is expected to enable identification and quantification of radionuclides present in the item with an unprecedented accuracy.

The characterisation and putting the gamma measurement station including Segmented Gamma-ray Scanner (SGS) and Tomographic Gamma Scanning (TGS) in operation is ongoing with a particular focus on the SGS part of the facility. Measurements of a number of waste drums of various densities have been performed with certified radioactive sources giving promising results.

The neutron measurement capability has been initially tested and is being validated to demonstrate traceability to national standards, leading to a simplified measurement strategy. This unique new capability will act as a benchmark facility, the leading facility in the field, and it will offer the opportunity for knowledge transfer. Current activities at the station include passive neutron calibration measurements of plutonium in simulated waste matrices.

Development and implementation of (on site) measurement systems and methods for monitoring the condition of radioactive waste repositories, including airborne radioactivity and temperature/strain

An automated device for measuring airborne radioactivity, developed previously, is to be fully validated to nuclear industry standards and demonstrated on site under this project, and expanded to automate the measurement of activity in water. A novel approach to carrying out such measurements is also included, using laser spectroscopy, which may have advantages over automated bubblers in some cases in terms of sensitivity and size. The use of laser spectroscopy for ^{14}C detection is completely novel and no commercial instrument using this method is currently available. In addition, the project aims to develop a method for traceability for temperature measurement remotely, and to test both, distributed temperature sensor (DTS) and distributed stress and temperature sensor (DSTS) systems in realistic environments, increasing the level of technology readiness.

Three applications have been investigated for use of the dual purpose water/air monitor (automated bubbler-LSC WILMA, including 1) tritium head-space measurement of soft waste, 2) radium in processed water from fracking and 3) Tc-99 in effluent. Applications 1) and 2) have been selected for experimental investigation. A review of existing instrumentation that may possibly be suitable for these applications has been completed. Laboratory measurements have been completed for measurement of radium in simulated processed water. Field trials at a fracking site have been completed and plans are in place for a field trial at a tritium waste respository.

Thermo-mechanical characterisation procedures suitable for optical fibers and DSTS systems are currently under testing. A stretching facility has been designed and adapted on a newly contructed tubular furnace of 6 m length. Furthermore, in-situ temperature calibration system has been designed.

Upgrades to the ^{14}C laser spectroscopy measurement system are in progress. For temperature and stress measurements, the operating constraints have been identified, leading to modifications in the techniques employed. The instrument has been upgraded with a new catalytic conversion unit and a prototype system has been built. Control software has been developed. Review of standards and test protocols is on-going. Successful field measurements were carried out at a nuclear facility demonstrating that the technology can be used on-site.

Impact

This project will have impact mainly on the nuclear industry, but also on industries that produce radioactive waste due to naturally occurring radioactivity in materials that are processed (for example, the oil and gas industries).

At this stage of the project, there is one formal document that has been published. However, the project partners have contributed to

- Four ISO documents as follows:
  - ✓ IEC/TC86/SC86C/WG2 Fibre optic sensors
  - ✓ ISO/TC85/SC2/WG14 Air control and monitoring
  - ✓ ISO/TC147/SC3/WG9 Radioactivity measurements
  - ✓ ISO/TC147/SC3/WG12 Radioactivity measurements
- IAEA draft technical document (Waste technology section) on radiological characterisation for safe management to radioactive waste
- Seventeen conferences and workshops (Czech Republic, Finland, France, Germany, Hungary, UK, USA)

These activities have engaged mainly with the nuclear decommissioning industry sector and the scientific community – mainly higher education institutes and public research organisations.

Impact on industrial and other user communities

The wider long-term impact of this project is on the members of the public and the environment, through ensuring that radioactive waste is disposed of safely. More specifically, the portable instrumentation for in situ measurement of gamma and alpha emitting radionuclides has opportunities to be licensed to instrumentation companies, for use by decommissioning personnel on nuclear sites to identify the extent of contamination and plan decommissioning activities. An early adopter of the micro-fluidic technology for radiochemical separation would be the Sellafield nuclear site, as the technology has the potential for significant cost and time saving for the determination of difficult to measure radionuclides. The pre-production airborne-radioactivity measurement systems have routes to market through the instrumentation companies in the consortium, with early adopters likely to be the low-level waste repositories in the UK and France.

The project incorporates two major facilities: the waste-package sentencing system and the waste-repository measurement system. The immediate impact from these facilities will be accurate, traceable, regulatory-compliant assessments of radioactive waste for the nuclear sites where the facilities are located, but the use of these facilities for training is intended to disseminate best practice and traceability to all nuclear sites operating waste package measurement systems.

Impact on the metrology and scientific communities

This project aims to contribute to developing a new, harmonised, international measurement infrastructure to support nuclear decommissioning. There is an established infrastructure for measuring routine discharges from the nuclear industry and radioactivity in the food chain and the environment; a new infrastructure is needed to address the measurement challenges in decommissioning (different radionuclides, different materials & different activity levels).

Impact on relevant standards

This project is having an impact on standards being developed by the International Organisation for Standardisation (ISO); these standards are referred to by laboratories and measurement scientists throughout the nuclear industry and are also adopted in national standards. The technical findings will be fed back for incorporation in standards under development and new work items covering topics such as the use of sensor networks will be proposed to ISO/TC85/SC2/WG17 and improved radioanalytical methods for radioactivity measurements ISO/TC147/SC3/WG9&12. The work will contribute to national good practice guides, such as the proposed UK Nuclear Industry Code of Practice for site characterisation.

Longer-term economic, social and environmental impacts

The main long term impact on the nuclear industry would be a reduction in the cost of disposal of wastes. The cost of disposal depends on the radioactivity content of the material; in the UK, ‘non-radioactive’ waste costs about £200 ton^{-1} to dispose, very low level waste is about £750/ton and low level waste about £3000 m^{-3}. Estimates of the cost of disposal of higher activity waste depend on the financial model used, but vary from £12000 m^{-3} for ILW to £300000 m^{-3} for HLW (or higher). Traceability to national standards and better understanding of the measurement uncertainties gives improved confidence in the results to the regulators (and also the members of the public); this enables the industry to recycle or re-use more of the waste that is produced, and to use lower category waste disposal options (current practice is to be overly conservative, which increases costs with no benefit to the environment). It is difficult to quantify the potential savings, but even a marginal improvement in dealing with the wastes will result in significant costs savings (the UK alone estimates that decommissioning legacy nuclear sites will produce 2,840,000 m^{-3} very low level waste, 1,370,000 m^{-3} of low level waste and 286,000 m^{-3} of ILW). In addition, improved traceability will reduce the need for re-work to investigate apparent discrepancies in measurements. The links to the industry-independent international measurement system will improve public trust and confidence that radioactive waste is being disposed of safely.

The project will have impact for a wider community than that concerned by the monitoring of nuclear waste repositories and power plants. Distributed strain and temperature sensors based on optical fibres have many applications in structural health monitoring. DTS and DSTS sensors have potential applications monitoring a wide range of engineering structures: ageing of rail infrastructures, mechanical and hydraulic behaviour of dams and dykes, breakage and leakage detection systems of gas, water and oil pipe-lines (the nuclear industrial sector is also interested by these technologies for monitoring the concrete structures of power plants). Such optoelectronic devices will also play a more prominent role in a smart city framework with detection of frozen roads, soil parameters analyses in agronomy, or monitoring of telecommunication/high voltage cables.

Novel radiochemical separation techniques have potential for impact in nuclear medicine, where similar technologies are being used for purifying radionuclides for use in radiopharmaceuticals for diagnostic scans and cancer therapy.

The technologies for remote measurement of alpha emitters and for imaging the location of gamma emitters have potential applications in the defence sector, particularly for use by first responders following the detonation of a so-called ‘dirty bomb’.

The technology developed for monitoring ^{3}H and ^{14}C at repositories may easily be adapted for use at other nuclear facilities, for example, for stack monitoring for radionuclide production facilities or for radiation protection on operating nuclear power plants or defence facilities.

List of publications

G. Failleau, O. Beaumont, R. Razouk, S. Delepine-Lesoille, M. Landolt, B. Courthial, J.M. Hénault, F. Martinot, B. Hay. A metrological comparison of Raman-distributed temperature sensors.
http://dx.doi.org/10.1016/j.measurement.2017.10.041

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Coordinator: Peter Ivanov, National Physical Laboratory, Tel +44 (0)2089438598 E-mail: peter.ivanov@npl.co.uk Project website address: http://empir.npl.co.uk/metrodecom/		
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