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

## Overview

This project addressed 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 was 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 aimed 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.

The project objectives were successfully achieved and the project's final results have addressed significant measurement challenges in the nuclear decommissioning sector in terms of rapid and automated radiochemical destructive analysis of solid samples, remote gamma- and alpha-screening and mapping of contamination, materials characterisation and waste sentencing as well as monitoring of repository sites. Uptake and application of the novel methods and prototype instrumentation developed within the project will allow the user community to facilitate and accelerate the process of on-site radiological characterisation, ultimately leading to safer, cost efficient and environmentally friendly decommissioning of nuclear facilities.

## Need

Previous to the start of the project, first-generation nuclear power plants and reprocessing facilities were coming to the end of their working lives. As per November 2020, a hundred nuclear power reactors have been shut down for decommissioned in the EU and most of the remaining 116 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 was that nuclear sites have been operating for many decades, so the disposition and quantities of hazardous materials were not well known. There was significant progress towards studies on the scientific bases of novel methods for material and waste characterisation within the frames of the previous EMRP projects ENV09 MetroRWM and ENV54 MetroDECOM. The present project built on these bases and focussed on bringing the techniques into use on nuclear sites and developing of robust and efficient procedures and instrumentation for radiological characterisation of decommissioning wastes and materials.

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

These needs were reflected in EU Council Directive 2011/70/EURATOM which focuses on encouraging 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 was 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 were:

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 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**

As a step towards improved mapping of contamination inside nuclear facilities, the spectrometric performance of a gamma camera equipped with new generation pixelated chips Timepix3 Si and Timepix3 CdTe has been successfully tested and evaluated. An additional strand to this work resulted in the development of a measurement technique that uses a network of sensors for dose mapping which was validated with a range of radiation sources. Detecting of contamination by alpha-emitting radionuclides has been also developed beyond the current state of the art by building and testing a system for remote alpha mapping that works well in ambient lighting, it is robust enough for use on nuclear sites and shows a sensitivity up to 10 times better than the target value of 10 kBq.

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. To address this, calibration methods for pre-selection and free release measurement were developed for different measuring geometries including containers, drums and ‘big bags’ and validated by measuring of real decommissioning waste.

Novel instruments have been developed for rapid and automated monitoring of radioactive discharges from waste repositories and nuclear facilities, including a liquid scintillation counting system capable of automatic sampling and measurement of $\alpha$- and $\beta$-emitters in aqueous streams and quantum-cascade cavity ringdown IR laser absorption spectrometer for measuring $^{14}CO_2$ and $^{14}CH_4$ in air. A compact prototype of the laser absorption spectrometer has been built and tested on-site at a nuclear power plant proving to be very suitable for the monitoring of radiocarbon emissions, with an unprecedented temporal resolution.

**Results**

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

For remote monitoring of gamma emitters, testing of a GAMPIX gamma camera with nuclides covering the expected energy range for measurement has been carried. The energy calibration of the GAMPIX camera using the $^{241}$Am, $^{57}$Co and $^{60}$Co sources has been performed. A new set of radioactive point sources including $^{54}$Mn and $^{134}$Cs and $^{133}$Ba extended sources has been produced by drop deposition, followed by evaporation and sealing. Mono-energetic and multigamma radioactive sources were used to evaluate and validate the performance at energies from 60 keV to 2600 keV, and an uncertainty budget determined. It has proved that, contrary to previous generation of detectors, Timepix3 equipped with CdTe sensors are able to reconstruct the total absorption peaks of Co-60 which was a major breakthrough, ensuring the capability of the Timepix3-based gamma camera to have better performances in terms of sensitivity and energy resolution for high-energy radionuclides.

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. A new user-friendly software for the alpha scanner has been developed and the system was enhanced for operation under daylight conditions. UV-based stand-off alpha scanner has been successfully trialled, with the detector performance exceeding the target sensitivity. Sensitivity was further improved with the addition of nitric oxide, with measurements not sensitive to normal lighting conditions. Two validation campaigns were performed with realistic and calibrated test samples, achieving a sensitivity up to 10 times better than the target value of 10 kBq.

Wireless compact battery-operated dosimeters were used to produce the first maps generated by application of approximation and statistical methods. A test setup to test mapping has been built and validated using Cs-137 sources before conducting measurements over fixed distance to apply statistical methods for mapping. Wireless networks for dose mapping have been developed and tested in a field demonstration with a waste management company. The impact of the number of sampling points and different statistical methods for mapping has been investigated.

The applications of borate fusion have allowed the development of a method for automated digestion of up to 5 g 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, acceptable results for U with a chemical recovery about 50 %, and Sr showing lower recoveries, which still allow confident determination. Target chemical recoveries of >60 % was achieved by NPL for Ca-41 and Zr-93, with a measurement uncertainty of <10 %, focusing on measurement by mass spectrometry. 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 has been upgraded to process higher samples masses up to 5 g. The procedure has also been validated on various concrete matrices by ICP-MS and LSC measurements performed for $^{41}$Ca analysis. The NiV automated radiochemistry system has been built for Sr-90 separation, with >95 % recoveries from test solutions. A semi-automated microfluidic device has been designed for more complex separations and successfully tested using the separation procedure developed for U, Pu and Sr-90. With these results, the project successfully achieved the objective regarding the development and implementation of rapid methods for destructive and non-destructive analysis of the radioactivity content of materials on a nuclear site.

*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. It was demonstrated that the system can be used successfully on a decommissioning site and reach standards required for software and operation in the nuclear industry.

Calibration methods for homogeneously contaminated waste and hot-spots were improved by using new set of sources with Co-60 for Petanque balls reference material and a new set of EG2 sources with Eu-152 as well as Cf-252 and Am-Be neutron sources.

Calibration methods were developed for a new ‘Big-Bag’ measuring geometry of real decommissioning waste streams including concrete, soil and sand. Selected decommissioning waste materials were measured in laboratories and at the pre-selection and free release facility and the results compared to validate the system and confirm the target uncertainties. With these results, the project successfully achieved the objective regarding the 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.

*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*

The *waste characterisation* system deployed at JRC (Ispra) has been shown to be applicable for 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 for direct determination of the fissile material component. Combining measurement techniques enables identification and quantification of radionuclides present in the item with an unprecedented accuracy.

The gamma measurement station including Segmented Gamma-ray Scanner (SGS) and Tomographic Gamma Scanning (TGS) has been tested and characterised for free release measurements with a particular focus on the SGS part of the facility. Measurements of multiple waste drums of various densities have been performed with certified radioactive sources. The neutron measurement capability has been tested and to demonstrate traceability to national standards, leading to a simplified measurement strategy. A measurement campaign on 220 litre simulated waste drum using well-characterised radioactive and nuclear standards, and standard containers of different matrix materials including metal, concrete, and low-density process waste has been carried out. With these results, the project successfully achieved the objective regarding the 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.

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

Two novel instruments have been developed for rapid and automated monitoring of radioactive discharges from waste repositories and other nuclear facilities. The first instrument, WILMA, developed by LL is a liquid scintillation counting system capable of automatic sampling and measurement of radioactivity (e.g. $^{3}H$, $^{14}C$, $^{99}Tc$,$^{137}Cs$, $^{90}Sr$-$^{90}Y$, $^{226}Ra$, $^{241}Am$) in an aqueous stream. An option exists to pair WILMA with a bubbler system to enable in-direct measurement of radioactivity-in-air (e.g. HTO, HT, $^{14}CO_2$, $^{14}CH_4$). The system was tested at a fracking site to investigate its use for monitoring naturally occurring radioactive materials (NORM) (in particular, $^{226}Ra$) in produced and flowback waters. The trial was successful, with WILMA able to measure gross alpha and beta decays above background rates despite the high salinity of the samples. The system was also tested on-site at the Culham Centre for Fusion Energy (CCFE) to investigate its use for monitoring tritium outgassing from stored tritiated waste. A new measurement configuration was successfully developed and demonstrated through the trial. The second system, developed by VTT, is a quantum-cascade cavity ringdown IR laser absorption spectrometer for measuring $^{14}CO_2$. The system includes an air sampling unit and catalytic furnace to oxidise methane enabling indirect measurement of $^{14}CH_4$. A compact prototype has been built and tested on-site at the Loviisa Nuclear Power Plant (Finland) to investigate routine monitoring of $^{14}C$ emissions in comparison to established monitoring technology installed at the plant. The cavity ringdown spectroscopy method proved to be very suitable for the monitoring of radiocarbon emissions, with an unprecedented temporal resolution. Fluctuations of the radiocarbon activity concentrations, which could not be captured with currently available methods were successfully measured. Further, the novel sample-processing unit allowed speciation of the radiocarbon molecular forms.

Thermo-mechanical characterisation procedures suitable for optical fibers and DSTS systems have been developed and tested. A stretching facility has been designed and adapted on a newly contructed tubular furnace of 6 m length. A newly designed in-situ temperature calibration system has been integrated in the concrete structure of Andra’s Underground Laboratory (Bure, France) and successfully tested and validated. Uncertainty of such in-situ temperature calibration system does not exceed 1.3 °C. With these results, the project successfully achieved the objective regarding the development and implementation of onsite measurement systems and methods for monitoring the condition of radioactive waste repositories, including airborne radioactivity and temperature/strain measurements.

**Impact**

As part of disseminating the project results, a dedicated blog was set up and regularly updated to provide up to date results and information. A Stakeholder Committee was established to participate and gave feedback and steered the work of the consortium. Within the course of the project five open access peer-reviewed articles have been published in relevant scientific journals and four more have been submitted. The main results and outcomes of the project were presented at 27 international and national conferences and five training courses have been provided to the project consortium, stakeholders and university students.

*Impact on industrial and other user communities*

The main impact of this project on the nuclear decommissioning community was through the delivery of methods, instrumentation and procedures developed to ensure that radioactive waste is accurately characterised and safely managed. More specifically, the portable instrumentation prototypes and procedures developed, tested and validated for in-situ measurement of gamma- and alpha-emitting radionuclides could be adopted for use by decommissioning personnel on nuclear sites to screen and measure radioactive contamination and plan decommissioning activities. The semi-automated micro-fluidic system for radiochemical separation has the potential for significant cost and time saving for the determination of difficult to measure radionuclides in radioactive waste and materials originating from nuclear licence sites. The set-ups for laser-spectroscopy measurement of radiocarbon and automated liquid-scintillation counting of airborne-radioactivity had routes to the market through the instrumentation companies that were partners during the project, but also with early adopters likely to be nuclear power plants, decommissioning sites and low-level waste repositories in the UK, Finland, France and other EU countries.

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

*Impact on the metrology and scientific communities*

To address the measurement challenges in decommissioning (different radionuclides, different materials & activity levels), the project activities resulted in the development of novel advanced methods for radioactivity measurements, contributing to the establishment of harmonised, international measurement infrastructure to support nuclear decommissioning. Highly efficient procedures have been developed for the analysis of radionuclides of significant relevance for nuclear decommissioning and radioactive waste characterisation such as actinides, $^{90}Sr$, $^{41}Ca$, $^{3}H$, $^{14}C$ with potential application in other scientific areas, i.e. environmental monitoring.

*Impact on relevant standards*

The project will have impact on standards being developed by the International Organisation for Standardisation (ISO). ISO standards are of essential importance for laboratories and measurement scientists throughout the nuclear industry and have been adopted in many countries as national standards. The technical findings have been fed back for incorporation in standards under development and new work items covering topics such as the use of sensor networks at ISO/TC85/SC2/WG17 and improved radioanalytical methods for radioactivity measurements ISO/TC147/SC3. The work contributes to national good practice guides, such as the proposed UK Nuclear Industry Code of Practice for site characterisation.

The project partners have also contributed to the preparation of international standards in three IEC/ISO committees: i) IEC/TC86/SC86C/WG2 Fibre optic sensors, ii) ISO/TC85/SC2/WG14 Air control and monitoring, iii) ISO/TC147/SC3/WG14,15&16 Radioactivity measurements.

*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 waste. 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.

Furthermore, the project results will have impact on distributed strain and temperature sensors based on optical fibres, which 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, fusion research centres or defence facilities.

**List of publications**

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E. Braysher, B. Russell, S. Woods, M. García-Miranda, P. Ivanov, B. Bouchard, D. Read. Complete dissolution of solid matrices using automated borate fusion in support of nuclear decommissioning and production of reference materials. J. Radioanal. Nucl. Chem. 321(1), 183-196 (2019). https://doi.org/10.1007/s10967-019-06572-z

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This list is also available here: https://www.euramet.org/repository/research-publications-repository-link/

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| Project start date and duration: | 01 September 2017, 42 months | |
| Coordinator: Peter Ivanov, National Physical Laboratory, Tel +44 (0)2089438598 E-mail: peter.ivanov@npl.co.uk<br>Project website address: http://empir.npl.co.uk/metrodecom/ | | |
| Internal Funded Partners:<br>1 NPL, United Kingdom<br>2 CEA, France<br>3 CIEMAT, Spain<br>4 CMI, Czech Republic<br>5 ENEA, Italy<br>6 LNE, France<br>7 PTB, Germany<br>8 VTT, Finland | External Funded Partners:<br>9 ANDRA, France<br>10 EDF, France<br>11 JRC, Europe<br>12 LL, United Kingdom<br>13 MAGICS, Belgium<br>14 NNL, United Kingdom<br>15 NUVIA, Czech Republic<br>16 TAU, Finland | Unfunded Partners: |
| RMG1: CIEMAT, Spain (Employing organisation); FTMC, Lithuania (Guestworking organisation)<br>RMG2: CIEMAT, Spain (Employing organisation); ENEA, Italy (Guestworking organisation) | | |