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TABLE OF CONTENTS

1 Executive Summary

Overview
Many natural resources and raw materials contain naturally occurring radioactive elements, meaning that the industrial activities that exploit them, such as mining or water treatment, could expose people to radiation. The industries working with Naturally Occurring Radioactive Materials (NORM) can potentially produce large amounts of radioactive waste, which can be an economic and environmental burden. It is not currently possible to measure the radioactivity of NORMs accurately or in situ, and the reference materials have a high uncertainty. This project developed new methods and devices for the measurement of naturally occurring radionuclides, as well as new reference materials for their calibration. This will allow industry to assess natural radioactivity accurately and process or recycle the material appropriately and safely.

Need for the project
Due to the radioactivity content, or NORM, in raw materials exposure to them must be controlled in accordance with International Atomic Energy Agency (IAEA) Safety Standards. When NORM are being handled or processed, it is vital to be able to determine the amount of radioactivity present accurately.

The industries working with NORM, such as extraction, processing and purification, often produce large amounts of radioactive waste, which is an economic and environmental burden if not properly disposed of or re-used. Waste disposal of radioactively contaminated materials is significantly more expensive than non-radioactive materials. Therefore traceable and accurate measurements are needed to decide the appropriate recycling or re-use options. This could potentially avoid additional costs, contamination of the environment or exposing the public to risk.

Current NORM reference materials and standard sources available, and those for validating radio-analytical methods, have a measurement uncertainty level of up to 20 %. In addition the ionising radiation measurement in the recycling industry focuses on artificial radionuclides, with NORM measurements often included as part of the natural background, regardless of their concentration.

In order to measure NORMs accurately, new and validated reference materials with an uncertainty level of less than 10 % are needed, as well as improved methods for the analysis and interpretation of results. In-situ techniques i.e. at industrial sites, are needed to determine the radioactive content of raw material, waste materials and by products, particularly when the raw material are inhomogeneous. Accurate and reliable in-situ measurements are particularly important when measuring NORM raw materials or determining the type of waste, as quick decisions on the usability or disposal procedure have to be made. There is also a need to harmonise measurement procedures for NORMs across Europe.

Scientific and technical objectives
The project's scientific and technical objectives were:

1. to develop reference materials and standard sources to enable accurate and traceable calibration of measurement instruments
2. to develop measurement systems to improve the determination of the radioactive content of raw material, waste materials and by products in-situ and to build a hand-held instrument operating at ambient air pressure, integrated in to a remote expert support system.
3. to harmonise in-situ measurement procedures performed in the European NORM industry and to bring in new and innovative methods: in-situ alpha spectrometry, radon measurements and measurements of inhomogeneously distributed radiation
4. to improve the quality of available data for important alpha- or gamma- or X-ray emission intensities
5. to test the measurement procedures and measurement devices developed in this project at end-user facilities on-site/in-situ.

Results & Conclusions

Development of reference materials and standard sources to enable accurate and traceable calibration of measurement instruments

This objective has been achieved by the successful preparation of new reference materials for the traceable calibration of NORM measurement instruments. The project identified the most commonly found NORMs, in need of reference standards. Candidate reference materials included: tantalum/niobium ore processing, residue/waste from titanium dioxide production, coal ash and tuff used in building products, residue/waste from phosphogypsum processing, building aggregates, ion exchange resins from the water industry, and iron oxide/manganese dioxide sludge from the water industry and oil waste. Based on these, calibration standard sources and traceable reference materials were developed for gamma-ray spectrometry, alpha spectrometry measurements and for in-situ measurement systems.

The volumes, natural radionuclides activity concentrations, matrices and the composition of the reference materials were determined. Complete characterisation and validation was achieved through inter-laboratory comparisons.

The developed NORM reference materials and calibration standard sources were: ^{238}U (uranium), ^{235}U, ^{226}Ra (radium), ^{210}Pb (lead), ^{228}Ra, ^{228}Th (thorium), ^{208}Tl (thallium), ^{228}Ac (actinium), ^{214}Bi (bismuth), ^{214}Pb, and ^{40}K (potassium). Additionally, two standard sources for the calibration of the pixel detector developed in objective 2 were produced.

The developed NORM reference materials and standard sources now enable more accurate, cost-effective and traceable calibration of NORM measurement instruments.

Development of measurement systems to improve the determination of the radioactive content of raw material, waste materials and by products in-situ and to build a hand-held instrument operating at ambient air pressure, integrated in to a remote expert support system.

To achieve this objective, new measurement instruments for the traceable measurement of NORMs were developed. Two hand-held in-situ prototypes were successfully designed, developed and validated for use; one prototype was for gamma-ray measurement and the other prototype was an alpha spectrometer. These instruments allowed traceable and accurate on-site measurements for rapid decisions on the usability of raw materials and disposal procedures.

A measurement system based on pixel detectors (MEDIPIX/TIMEPIX) was also developed and tested successfully. This third system was used as an in-situ measurement instrument for determining important airborne radionuclides especially the activity concentration of radon and NORM alpha emitting radionuclides in air.

Automated sampling preparation is important for feasible radiochemistry and effective NORM measurements by end-users at industrial sites. To achieve this goal, an improved NORM sample preparation stage was developed, involving a chemical digestion of the sample matrix, leaving the elements of interest. This automated sample processing of NORM materials was needed for quick and reliable chemical digestion and was achieved using a Katanax K2® fusion machine which is a commercially available.

Elevated ^{220}Rn (thoron) activity concentrations have to be taken into account because of increased exposure of staff in industrial workplaces e.g. waterworks. For the NORM radionuclide ^{220}Rn, a traceable production device for the radionuclide in a vacuum chamber was designed and set-up. ^{220}Rn samples and standard sources, together with the ^{220}Rn production device, were successfully tested. In addition to this, a novel measurement system for the in-situ determination of ^{220}Rn activity was developed, and a comparison with the existing ^{220}Rn standard source showed a relative difference of 4.7 % at the maximum ^{220}Rn activity concentration, which represents an improvement on the accuracy of the currently available ^{220}Rn standard. The calibration of a commercially available ^{220}Rn monitor was successfully demonstrated. Initial tests of the instruments, equipment and tools developed was successfully performed in the laboratory, with subsequent on-site testing in objective 5.

The objective was achieved by the successful development, demonstration and validation of new automatic NORM sample preparation facilities, detectors and measurement systems for the traceable in-situ and laboratory measurement of NORM.

Harmonisation of in-situ measurement procedures performed in the European NORM industry to bring in new and innovative methods for in-situ alpha spectrometry

To achieve this objective, 37 stakeholders from NORM industries and measurement laboratories completed the project's questionnaires on specific methods used for the characterisation of residues containing natural radioactivity. The results indicated the main measurement techniques currently being used, and three of these were selected and evaluated for in-situ and laboratory measurements of NORM materials.

In addition, a novel protocol for testing and evaluating measurement methods used in European NORM industries for waste was developed. The best existing measurement practices for NORM in-situ measurement methods were also determined.

Novel methods for the measurement of airborne radionuclides using the prototype pixel detector and the hand-held alpha spectrometer (both from objective 2) have been successfully created. Best practice methods for NORM measurements for laboratory analysis were determined. Measurement procedures for the in-situ analysis of radon in waterworks and for in-situ analysis of radon emanation from building materials have been established.

Two new traceable laboratory methods for the determination of ^{226}Ra in liquid samples by LSC (Liquid Scintillation Counting) have been developed on subsamples of the same bulk sample. This method is applicable for ^{226}Ra activity measurements of industrial NORM waste water.

Additionally a new measurement procedure for the determination of the total activity of inhomogeneously distributed NORM waste in 200 litre NORM waste barrels has been developed and successfully validated. This new method supports the necessary legal identification and quantification of natural radionuclide activities in industrial NORM waste barrels. Initial tests of the new methods and procedures had been successfully performed in the laboratory, with subsequent on-site testing in objective 5.

The in-situ and laboratory NORM measurement methods developed in this project enable more accurate, cost effective and traceable NORM measurements at industrial sites. The results support the harmonisation of NORM measurement in Europe which will reduce efforts and costs for the production and transnational trade of NORM products (e.g. phosphate products, building material, rare earth materials).

Improvement of the quality of available data for important alpha- or gamma- or X-ray emission intensities in the decay of nuclides of the ^{238}U, ^{232}Th, ^{235}U decay chains i.e. ^{226}Ra and ^{210}Pb and for ^{138}La

Accurate decay data of NORM radionuclides are essential for qualified and reliable radioactivity measurements. They will enable selection of NORM raw materials and compliance with legal radiation protection requirements. This objective has been successfully reached by the revision and improvement of nuclear data for selected natural radionuclides, so that as many descendants of uranium and thorium decay chains can be accurately measured as possible.

Due to the huge diversity of the occurring radionuclides within NORM samples, gamma-ray spectrometry of specific radionuclides is challenging and requires suitable handling of spectral interferences. The project used a comprehensive study of potential spectral interferences and in-depth evaluation of selected NORM samples to provide recommendations for the choice of gamma-lines and the consideration of possible spectral interferences. Additionally a practical and sensitive method for the estimation of the ^{222}Rn leakage of sample containers was introduced.

The improved data on the decay of natural radionuclides enables accurate and cost-effective instrument calibration and activity measurement of NORM products and waste.

Test of the measurement procedures and measurement devices developed in this project at end-user facilities on-site/in-situ.

To test the applicability of the reference materials, instruments and methods developed during the project, verification criteria for measurement procedures and devices were developed. Several European NORM industry stakeholders were involved in the on-site measurements:

- Methods for determination of the total activity of inhomogeneously distributed NORM waste
- In-situ measurements of ^{210}Pb and ^{210}Po (Polonium) at a tin and lead production facility
- On-site testing of the hand-held in-situ alpha spectrometer at a metro station construction site

- Measurement procedure for in-situ analysis of radon in waterworks
- On-site testing of the prototype pixel detectors MEDIPIX/TIMEPIX
- Verification of measuring methods for an industrial site affected by NORM

On-site test measurements using the newly developed in-situ measurement methods and instruments were demonstrated and verified successfully. The on-site measurements successfully demonstrated that the new standards and techniques work in practice at industrial sites to measure low-energy gamma-ray emitters in NORM materials, surface contamination measurements, NORM in dust measurements and in-situ measurements of radon.

Actual and potential impact

The project established new reference materials and standard sources, and developed novel in-situ and laboratory measurement instruments and procedures which can be performed by the European NORM industry on building materials, drinking water and waste products. They will help eliminate discrepant NORM measurement results by establishing traceability of measurement methods and reduce end-user uncertainties to within legal limits.

The outcomes of the project, including the improvement of NORM radionuclide decay data, led to a significant reduction in uncertainties of NORM instrument calibration and measurements. This enables cost-effective, accurate and reliable end-user measurements for NORM industry. Additionally the project results have contributed to standardisation organisations (e.g. CEN) to support the harmonisation of NORM measurement procedures across Europe.

Impact on standards

The results of the project have been disseminated to European and national standardisation bodies and working groups including CEN/TC 351 Construction products: Assessment of release of dangerous substances WG3 on radiation form construction products; CEN/TC 45 Nuclear Instrumentation; the ASI (Austrian Standards Institute) and the EC Group of Experts established under Article 31 of the EURATOM Treaty. The results on NORM activity metrology and measurement have been, and will be, considered in the radiation protection and dose modelling standards for construction products, environmental measurement methods, radiation protection instrumentation and radon / thoron measurement and dose assessment methods.

Dissemination of results

The project held regular workshops for stakeholders and industrial end-users (including NORM industry facilities, nuclear regulators, standardisation bodies and measurement device and calibration source manufacturers) which covered the new measurement techniques, best practices, reference materials, decay data, on-site demonstrations, eLearning NORM and the NORM situation in EU member states. The project also established an Advisory Board of experts from the IAEA, International Committee for Radionuclide Metrology (ICRM), European Cooperation in Science and Technology (COST), European ALARA Network for NORM (EAN-NORM) and NORM industries.

The results of the project have been presented to metrologists, stakeholders, regulators and end-users at international conferences. The project has submitted / presented 47 conference presentations and 25 scientific journal papers. A joint workshop was also held with the EU COST network NORM4BUILDINGS.

Training materials for a post-graduate course is available within a NPL e-learning course. These include:

- the hazards and risks associated with work involving NORM raw materials, by-products, residues and wastes,
- national and international requirements and recommendations,
- best practices and state-of-art measurement systems regarding NORM monitoring at industry,
- measurement systems, standard sources and reference materials to perform traceable NORM analysis.

An open access NPL e-learning course on NORM metrology and measurement has been established (http://www.npl.co.uk/commercial-services/products-and-services/training/e-learning/naturally-occurring-radioactive-materials/).

Potential impact

The results of the project (traceable, accurate and standardised measurement methods and systems, in particular for in-situ applications) will help the NORM industry to decide on the use of raw material and the re- use of waste materials without increasing costs, contaminating the environment or exposing workers or the public to harm.

The improved measurement of natural radionuclides ensures that the raw materials brought into the industrial process will not cause, as far as possible, enhanced radioactivity levels in the final products and in waste. The outcomes of the project will support better characterised of NORM raw material and waste, and identification and precisely measurement using traceable standardised methods. The project will also reduce costs in the NORM industry sector and increase safety in NORM production.

2 Project context, rationale and objectives

Naturally occurring radionuclides are present in many natural resources and raw materials such as mineral feedstocks. Therefore, the industries that exploit these resources have an increased potential for exposure to NORM in their products, by-products, residues and waste. Industries working with NORM raw materials also produce large amounts of waste, and such waste materials, constitute a huge economic and ecological burden if not properly disposed of or re-used. Therefore the aim of this project was to address these issues and to develop new traceable measurement capabilities for NORM industries in order to significantly improve the industrial processing of NORM resources and waste. The new measurement capabilities included measurement systems for use in the laboratory and on-site (i.e. in situ) and reference/calibration materials for validation of such measurement systems.

The industries/branches of industry considered in this project, which process NORM and/or generate technologically enhanced NORM (TENORM) are:

- The extraction of rare earths
- Niobium/tantalum ore processing
- Titanium dioxide (TiO_2) pigment production
- The phosphate industry – thermal phosphorous production, phosphoric acid production, and production of phosphate fertilisers
- The building materials industry, including cement production
- Tin foundries, and tin/lead/copper smelting
- Waterworks, ground water filtration, and drinking water production
- The recycling industry
- The oil and natural gas industry

The overall scientific and technological objectives of the project are to accelerate the industrial innovation by development of new measurement capabilities for NORM industry applications, to improve significantly the industrial processing of NORM resources and technologically enhanced NORM residues, and to assist competitive technology throughout the industry in Europe.

The project's scientific and technical objectives were:

1. to develop reference materials and standard sources to enable accurate and traceable calibration of measurement instruments
2. to develop measurement systems to improve the determination of the radioactive content of raw material, waste materials and by products in-situ and to build a hand-held instrument operating at ambient air pressure, integrated in to a remote expert support system.
3. to harmonise in-situ measurement procedures performed in the European NORM industry and to bring in new and innovative methods: in-situ alpha spectrometry, radon measurements and measurements of inhomogeneously distributed radiation
4. to improve the quality of available data for important alpha- or gamma- or X-ray emission intensities
5. to test the measurement procedures and measurement devices developed in this project at end-user facilities on-site/in-situ.

The new traceable measurement procedures will be proposed to CEN/CENELEC for standards for industrial raw materials, products and waste. By providing improved, more reliable and accurate measurements of naturally occurring radionuclides, the project will ensure that, if these raw materials are present in industrial processes, they will not unknowingly enhance radioactivity levels in waste and finished products.

3 Research results

3.1 Development of reference materials and standard sources to enable accurate and traceable calibration of measurement instruments

Objective

The aim of this work was to develop reference materials and standard sources to enable accurate and traceable calibration of measurement instruments.

Results

New candidate reference materials have been prepared for the traceable calibration of NORM measurements using the project partners' national standards. The project partners have determined the volumes, natural radionuclides activity concentrations, matrices and the element composition of the candidate reference materials. The candidate reference materials include; Ta/Nb (tantalum/niobium) ore processing, residue/waste from titanium dioxide (TiO_2) production, coal ash and tuff used in building products, residue/waste from phosphorgypsum processing, building aggregates, ion exchange resins from the water industry, and iron oxide/manganese dioxide ($FeO(OH)/MnO_2$) sludge from the water industry and oil waste.

Beyond the state of the art, novel traceable NORM standard reference materials and sources for calibration of laboratory instruments and in-situ measurement instruments - covering the NORM radionuclides ^{238}U, ^{235}U, ^{226}Ra, ^{210}Pb, ^{228}Ra, ^{228}Th, ^{208}Tl, ^{228}Ac, ^{214}Bi, ^{214}Pb, and ^{40}K - have been prepared and applied successfully.

In total 10 standard sources and reference materials for gamma-ray spectrometry and alpha spectrometry measurement and for in-situ measurement systems for NORM have been developed by the project partners (**Figures 1 & 2**). The reference materials and standard sources were able to be fully characterised due to the methodical diversity of the project partners' metrological resources. The collaborative approach in producing and standardising the NORM reference materials and sources has produced added value that individual partners could not achieve by themselves. The activity concentrations of natural decay chain radionuclides range from some tents Bq/kg to some hundreds of kBq/kg. Further to this, two inter-laboratory comparisons using the standard sources and reference materials have been successfully carried out.

Further to this, two standard sources for the calibration of the pixel detector (objective 2, section 3.2) have been standardised and tested successfully.

Fig. 1. Developed reference NORM materials: Ionex resin (water industry), sediment (TIO_2 production), Tuff (building industry)

Fig. 2. Reference NORM sand source for use with the oil industry

Conclusions

This objective has been achieved by the successful preparation of new reference materials for the traceable calibration of NORM measurement instruments. The project identified the most commonly found NORMs, in need of reference standards. Candidate reference materials included: tantalum/niobium ore processing, residue/waste from titanium dioxide production, coal ash and tuff used in building products, residue/waste from phosphogypsum processing, building aggregates, ion exchange resins from the water industry, and iron oxide/manganese dioxide sludge from the water industry and oil waste. Based on these, calibration standard sources and traceable reference materials were developed for gamma-ray spectrometry, alpha spectrometry measurements and for in-situ measurement systems.

The volumes, natural radionuclides activity concentrations, matrices and the composition of the reference materials were determined. Complete characterisation and validation was achieved through inter-laboratory comparisons.

The developed NORM reference materials and calibration standard sources were: ^{238}U, ^{235}U, ^{226}Ra, ^{210}Pb, ^{228}Ra, ^{228}Th, ^{208}Tl, ^{228}Ac, ^{214}Bi, ^{214}Pb, and ^{40}K. Additionally, two standard sources for the calibration of the pixel detector developed in objective 2 were produced.

The developed NORM reference materials and standard sources now enable more accurate, cost-effective and traceable calibration of NORM measurement instruments.

3.2 Development of measurement systems to improve the determination of the radioactive content of raw material, waste materials and by products in-situ and to build a hand-held instrument.

Objective

The aim of this work was to develop measurement systems to improve the determination of the radioactive content of raw material, waste materials and by products in-situ and to build a hand-held instrument operating at ambient air pressure, integrated in to a remote expert support system.

Results

Beyond the state of the art laboratory and in-situ measurement systems and procedures for the measurement of NORM radionuclides and the reference materials were developed with total relative measurement uncertainties lower than 10 % (k=1) (Glacič-Cindro et al., 2014).

The project developed new metrological methodologies, measurement instruments, standards and reference materials for the measurement of natural radionuclides.

Significant improvements beyond the state of the art have been achieved by developments, designs and successful tests of a novel hand-held prototype in-situ gamma-ray measurement system, an in-situ alpha-particle spectrometry prototype by integration with a remote expert support system (Pöllänen et al., 2014, 2015a, 2015b, 2016).

Rapid detection of alpha-particle emitting radionuclides may be of utmost importance in radiation and nuclear safety, security and safeguards as well as in a nuclear emergency. As part of this project, a prototype in-situ alpha-spectrometry system was developed which can be used for all alpha emitters, including measurements of flat and smooth surfaces (**Figure 3**).

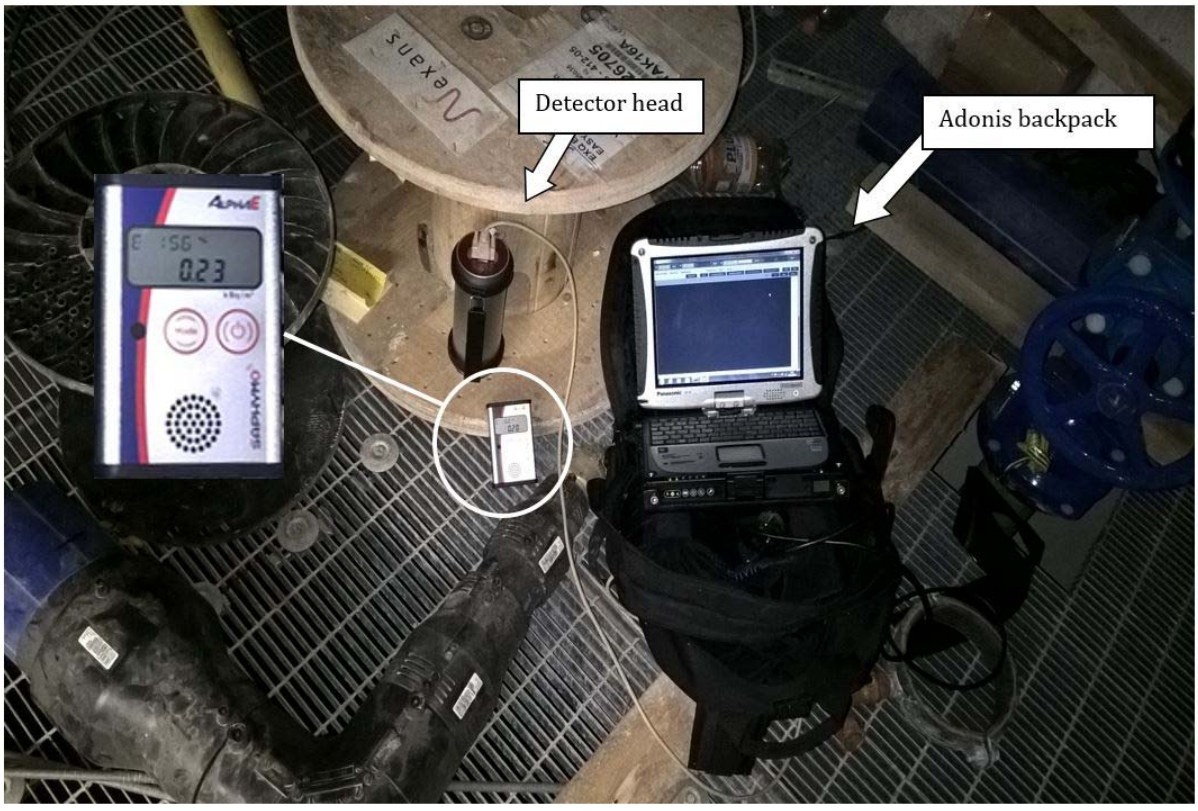

Fig. 3. The MetroNORM in-situ alpha-spectrometer system Adonis in operation

The prototype in-situ alpha-spectrometry system does not need a vacuum pump. The system also shows a reasonable energy resolution (relative energy resolution ~0.15) for radionuclide identification supported by alpha particle collimation (**Figure 4**), real-time data transfer and reachback functionality. In addition, the measurement of beta particles using the prototype in-situ alpha-spectrometry system are possible in principal. The measurement procedure for the prototype in-situ use hand-held alpha spectrometer had been established and tested successfully.

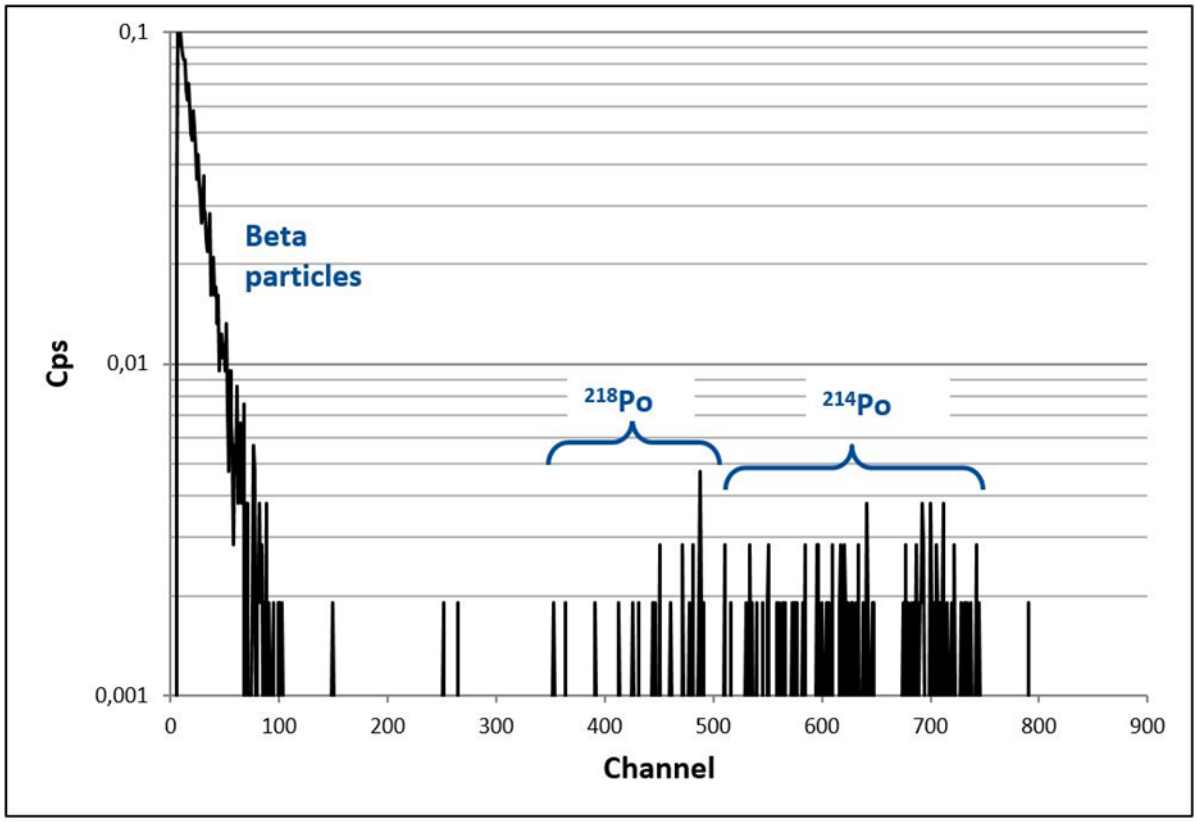

Fig. 4. Spectrum of ^{222}Rn progenies at the testing operation of the in-situ alpha spectrometer Adonis with collimator

An in-situ measurement system based on hydrid Si pixel detectors (MEDIPIX/TIMEPIX) has also been designed, developed and tested (Bulanek et al., 2015; **Figures 5 & 6**). The minimum detectable total alpha and total beta sensitivities of the developed NORM pixel detector in relation to the measuring time are shown in **Table 1**.

Table 1. In-situ pixel detector system minimum detectable activity (MDAs) at measurement of surface activity contamination by alpha-particles and beta-particles versus measuring time

Time of sample measurement (s)	MDA kBq/m^2 (total alpha)	MDA kBq/m^2 (total beta)
1	58	84
10	6.7	14.7
100	0.96	3.5
1000	0.19	0.95

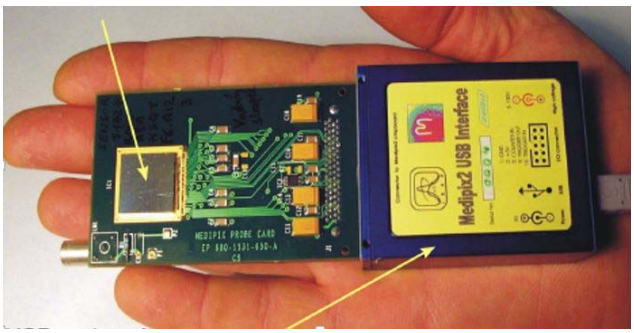

Fig. 5. Developed NORM Pixel detector with USB interface

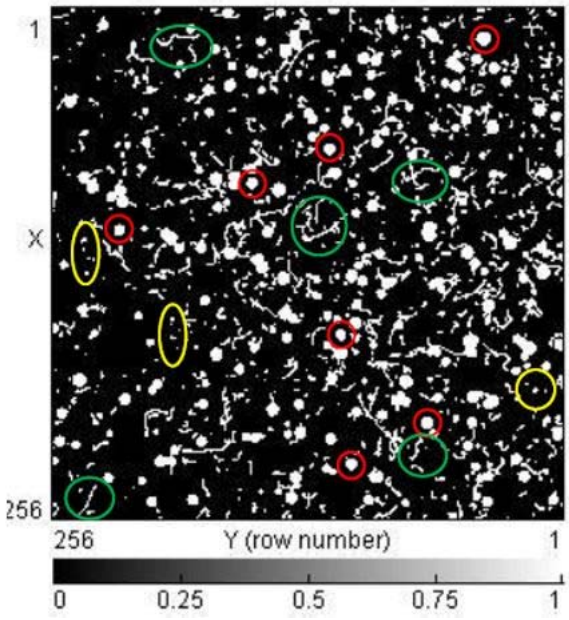

Fig. 6. NORM Alpha-particle (red), beta-particle (green) and gamma-ray (yellow) response at the pixel detector

An improved final NORM sample preparation stage was developed, as well as an automated method for the final NORM sample preparation stage using extraction chromatography with UTEVA resin (from Triskem International) and a vacuum box (**Figure 7**), which includes an appropriate initial sample preparation stage (Katanax K2® Prime) for mixed siliceous NORM matrices: scale, water, sand, wax, oil.

For ^{220}Rn, a production chain for the radionuclide in a vacuum chamber has been designed and set-up and the generators and ^{220}Rn samples/standards together with the chamber have been successfully tested (Sabot B. et al., 2015). In a collaboration between CEA and the French national public expert in nuclear and radiological risks, i.e. the Institute for Radiological Protection and Nuclear Safety (IRSN), a novel measurement system for the on-line determination of ^{220}Rn activity was successfully developed (**Figure 8**), and a comparison between the ^{220}Rn standards developed was completed. The comparison showed the relative difference was beyond the state of the art and only 4.7 % at the maximum (^{220}Rn) = 23.3 kBq·m^{-3}.

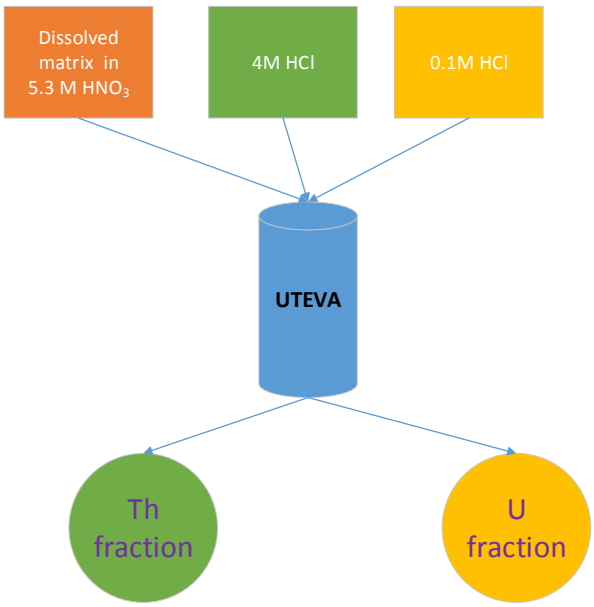

Fig. 7. NORM final sample preparation via UTEVA resin for inductively coupled plasma mass spectrometry (ICP-MS) analysis

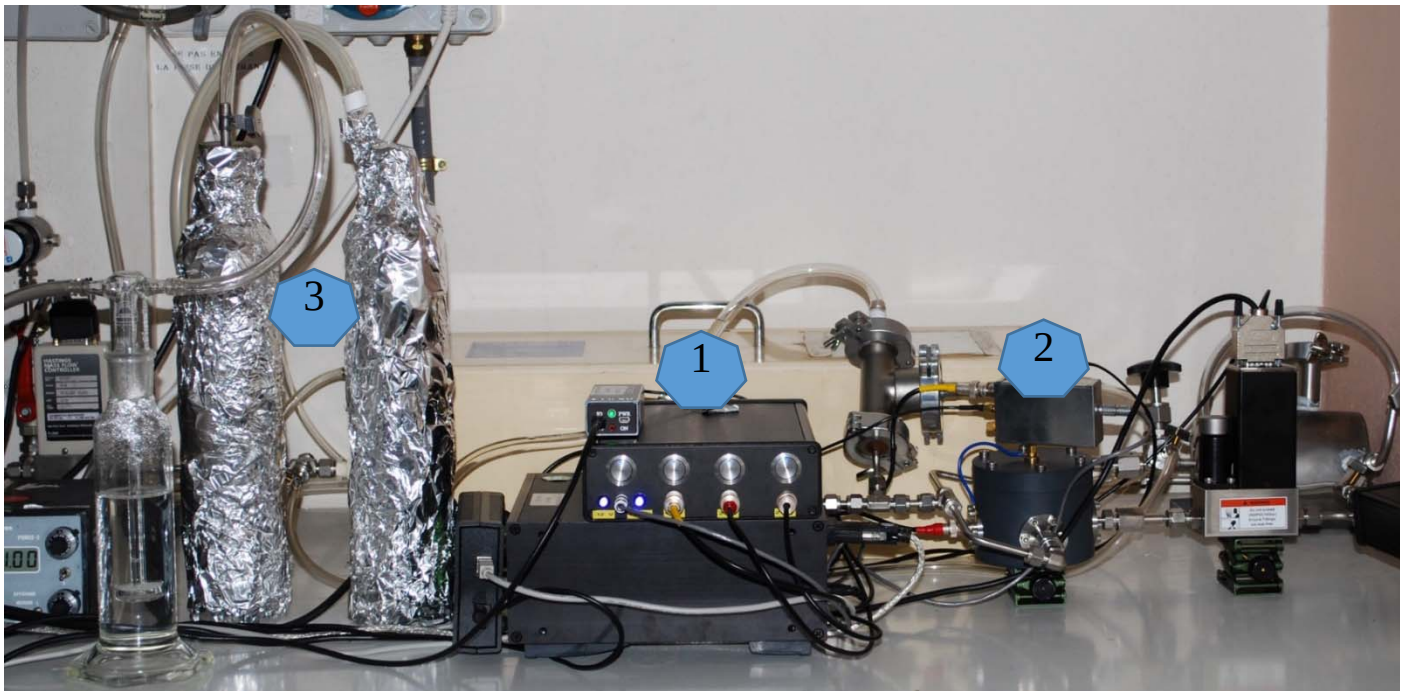

Fig. 8. ^{220}Rn measurement setup with ^{228}Th source (1), ^{220}Rn chamber (2) and gas bubblers with liquid scintillator (3)

Conclusions

To achieve this objective, new measurement instruments for the traceable measurement of NORMs were developed. Two hand-held in-situ prototypes were successfully designed, developed and validated for use; one prototype was for gamma-ray measurement and the other prototype was an alpha spectrometer. These instruments allowed traceable and accurate on-site measurements for rapid decisions on the usability of raw materials and disposal procedures.

A measurement system based on pixel detectors (MEDIPIX/TIMEPIX) was also developed and tested successfully. This third system was used as an in-situ measurement instrument for determining important airborne radionuclides especially the activity concentration of radon and NORM alpha emitting radionuclides in air.

Automated sampling preparation is important for feasible radiochemistry and effective NORM measurements by end-users at industrial sites. To achieve this goal, an improved NORM sample preparation stage was developed, involving a chemical digestion of the sample matrix, leaving the elements of interest. This automated sample processing of NORM materials was needed for quick and reliable chemical digestion and was achieved using a Katanax K2® fusion machine which is a commercially available.

Elevated ^{220}Rn activity concentrations have to be taken into account because of increased exposure of staff in industrial workplaces e.g. waterworks. For the NORM radionuclide ^{220}Rn, a traceable production device for the radionuclide in a vacuum chamber was designed and set-up. ^{220}Rn samples and standard sources, together with the ^{220}Rn production device, were successfully tested. In addition to this, a novel measurement system for the in-situ determination of ^{220}Rn activity was developed, and a comparison with the existing ^{220}Rn standard source showed a relative difference of 4.7 % at the maximum ^{220}Rn activity concentration, which represents an improvement on the accuracy of the currently available ^{220}Rn standard. The calibration of a commercially available ^{220}Rn monitor was successfully demonstrated. Initial tests of the instruments, equipment and tools developed was successfully performed in the laboratory, with subsequent on-site testing in objective 5.

The objective was achieved by the successful development, demonstration and validation of new automatic NORM sample preparation facilities, detectors and measurement systems for the traceable in-situ and laboratory measurement of NORM.

3.3 Harmonisation of in-situ measurement procedures performed in the European NORM industry to bring in new and innovative methods.

Objective

The aim of this work was to harmonise in-situ measurement procedures performed in the European NORM industry and to bring in new and innovative methods: in-situ alpha spectrometry, radon measurements and measurements of inhomogeneously distributed radiation

Results

37 of more than 50 registered MetroNORM stakeholders, from European NORM industries and NORM measurement laboratories completed the project's "Questionnaire to selected industrial companies on specific methods used for characterisation residues containing natural radioactivity" or the "Questionnaire to selected laboratories contracted by industries that request analyses for characterisation of residues containing natural radioactivity", respectively. The completed questionnaires were then evaluated to take into account the measurement methods used by European NORM industries for residues. From the results three main measurement methods were selected and evaluated for *in-situ* and laboratory measurements of NORM materials.

Based on the evaluation of the NORM industry stakeholder survey, a measurement procedure that went beyond the current state of the art for the determination of the total activity of inhomogeneously distributed NORM waste material was developed, as well as a protocol for testing and evaluating measurement methods used in European NORM industries for waste (**Figures 9 & 10**; Glavič-Cindro et al., 2014).

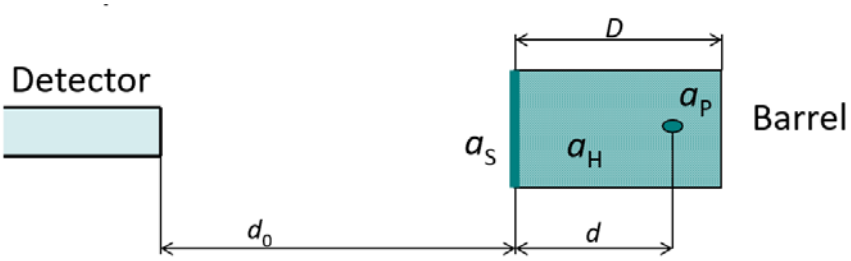

Fig. 9. Detection of inhomogeneity in NORM waste drums: Three distinct contributions to radioactivity distribution supposed: surface (a_S), homogeneous (a_H) and point source (a_P) contributions

In addition, novel methods for the measurement of airborne radionuclides using the prototype pixel detector (MEDIPIX/TIMEPIX), and for use of the prototype in-situ use hand-held alpha spectrometer (both from objective 2, section 3.2) have been successfully created. This goal was achieved through a collaborative approach between CMI, SURO and STUK, as well as a report on best practice methods for radioactivity measurements for laboratory analysis.

Further to this, good practice measurement procedures for the in-situ analysis of radon in waterworks/plants (**Figure 11**) and for the in-situ analysis of radon emanation from building materials (**Figures 12 & 13**) were established by BEV/PTP together with the researcher excellence grant (REG) from BOKU.

Fig. 10. NORM waste drum / gamma detector measurement configuration

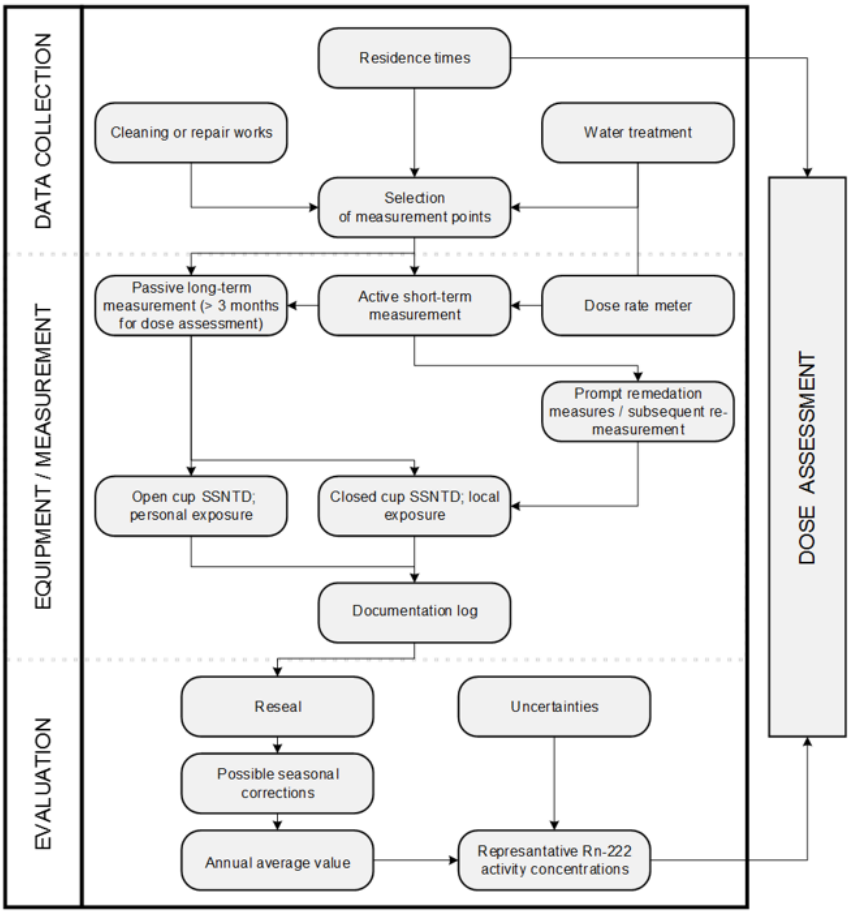

Fig. 11. Scheme of the in-situ method for exposure and dose assessment of radon in drinking water plants

Fig. 12. Radon emanation measurement of building material samples

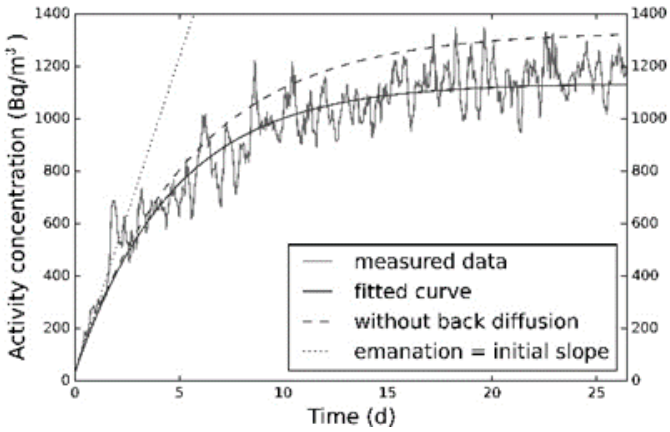

Fig. 13. Radon emanation measurement of building materials

Conclusions

To achieve this objective, 37 stakeholders from NORM industries and measurement laboratories completed the project's questionnaires on specific methods used for the characterisation of residues containing natural radioactivity. The results indicated the main measurement techniques currently being used, and three of these were selected and evaluated for in-situ and laboratory measurements of NORM materials.

In addition, a novel protocol for testing and evaluating measurement methods used in European NORM industries for waste was developed. The best existing measurement practices for NORM in-situ measurement methods were also determined.

Novel methods for the measurement of airborne radionuclides using the prototype pixel detector and the hand-held alpha spectrometer (both from objective 2) have been successfully created. Best practice methods for NORM measurements for laboratory analysis were determined. Measurement procedures for the in-situ analysis of radon in waterworks and for in-situ analysis of radon emanation from building materials have been established.

Two new traceable laboratory methods for the determination of ^{226}Ra in liquid samples by LSC have been developed on subsamples of the same bulk sample. This method is applicable for ^{226}Ra activity measurements of industrial NORM waste water.

Additionally a new measurement procedure for the determination of the total activity of inhomogeneously distributed NORM waste in 200 litre NORM waste barrels has been developed and successfully validated. This new method supports the necessary legal identification and quantification of natural radionuclide

activities in industrial NORM waste barrels. Initial tests of the new methods and procedures had been successfully performed in the laboratory, with subsequent on-site testing in objective 5.

The in-situ and laboratory NORM measurement methods developed in this project enable more accurate, cost effective and traceable NORM measurements at industrial sites. The results support the harmonisation of NORM measurement in Europe which will reduce efforts and costs for the production and transnational trade of NORM products (e.g. phosphate products, building material, rare earth materials).

3.4 *Improvement of the quality of available data for important alpha- or gamma- or X-ray emission intensities*

Objective

The aim of this work was to improve the quality of available data for important alpha- or gamma- or X-ray emission intensities in the decay of nuclides of the ^{238}U, ^{232}Th, ^{235}U decay chains i.e. ^{226}Ra and ^{210}Pb and for ^{138}La.

Results

The revision of nuclear data (Bé et al., 2011) for selected natural radionuclides was done so that as many descendants of uranium and thorium decay chains can be accurately measured as possible. To improve the required NORM related decay data, special radionuclide sources with the radionuclides ^{235}U, ^{227}Ac, ^{226}Ra and ^{210}Pb were carefully prepared and improved decay data of the selected NORM radionuclides' was determined (Rodrigues et al., 2016). An improved and beyond the current state of the art, beta spectrum for ^{138}La was calculated as well as the gamma-ray spectra of the selected key NORMs (which were measured and evaluated). A revised decay scheme for ^{138}La is shown in **Figures 14 & 15**.

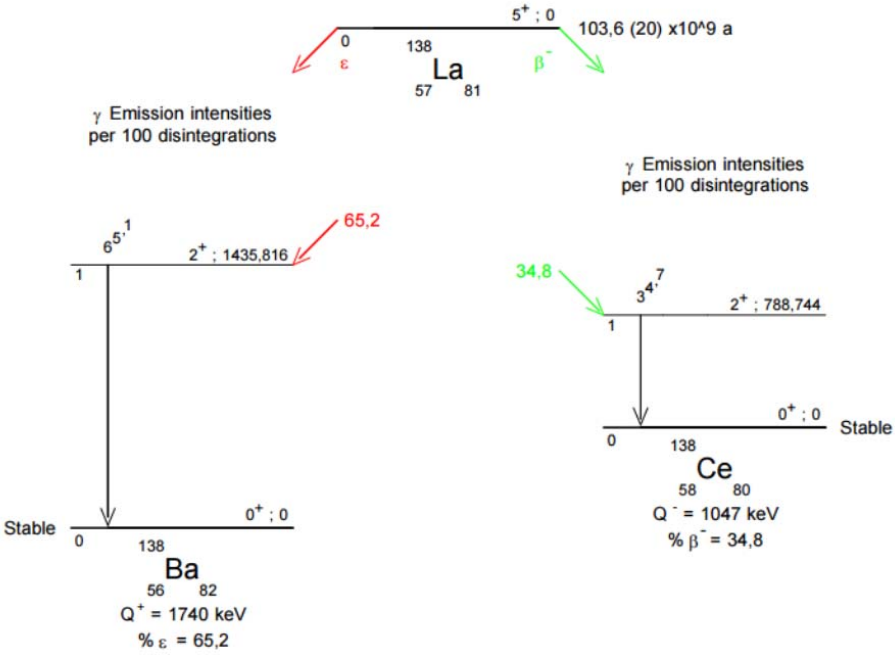

Fig. 14. Disintegration scheme of ^{138}La

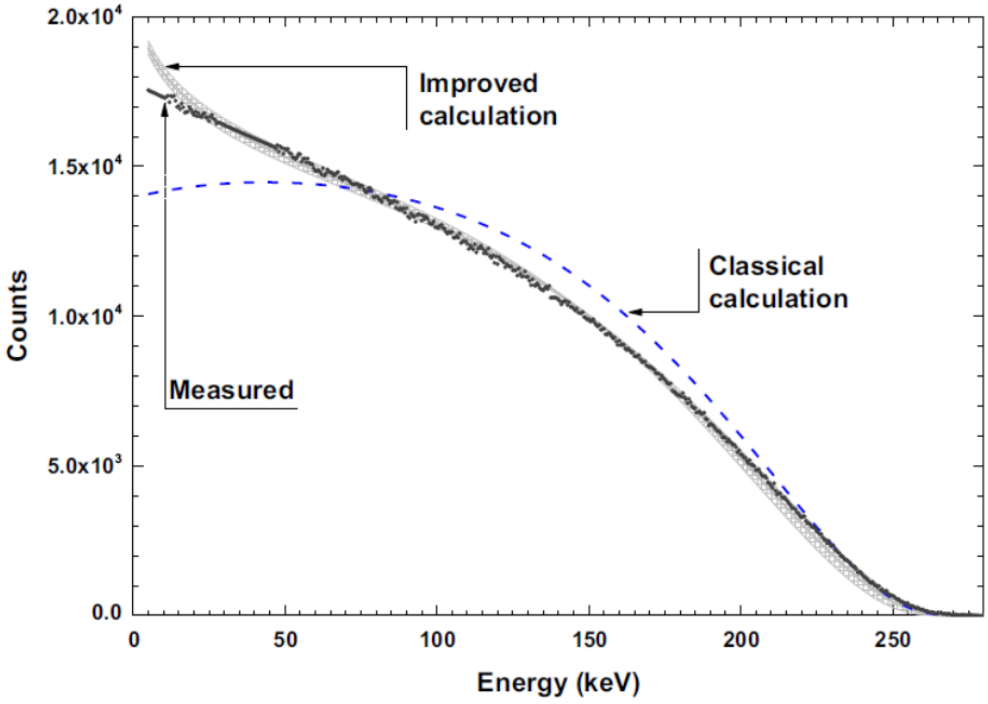

Fig. 15. Improved beta energy distribution for ^{138}La

Conclusions

Accurate decay data of NORM radionuclides are essential for qualified and reliable radioactivity measurements. They will enable selection of NORM raw materials and compliance with legal radiation protection requirements. This objective has been successfully reached by the revision and improvement of nuclear data for selected natural radionuclides, so that as many descendants of uranium and thorium decay chains can be accurately measured as possible.

Due to the huge diversity of the occurring radionuclides within NORM samples, gamma-ray spectrometry of specific radionuclides is challenging and requires suitable handling of spectral interferences. The project used a comprehensive study of potential spectral interferences and in-depth evaluation of selected NORM samples to provide recommendations for the choice of gamma-lines and the consideration of possible spectral interferences. Additionally a practical and sensitive method for the estimation of the ^{222}Rn leakage of sample containers was introduced.

The improved data on the decay of natural radionuclides enables accurate and cost-effective instrument calibration and activity measurement of NORM products and waste.

3.5 Test of the measurement procedures and measurement devices developed in this project at end-user facilities on-site/in-situ. chains

Objective

The aim of this work was to test the measurement procedures and measurement devices developed in this project at end-user facilities on-site/in-situ.

Results

In the final project phase, site specific verification criteria, requirements and procedures for selected end-user sites were developed, for testing the in situ systems, methods and reference materials from objectives 1, 2 and 3 (sections 3.1-3.4). The location, instrumentation and procedures for these on-site/in situ tests were carefully prepared. The final on-site tests were successfully carried out in a collaborative approach as follows:

- Development of in-situ alpha spectrometer (Roy Pöllänen et al., STUK - Radiation and Nuclear Safety Authority, Finnland)
- Pixel detectors and their application in NORM Industry (Jiří Hůlka et al., SURO - National Radiation Protection Institute, Czech Republic)
- ^{210}Pb activity determination using gamma-ray spectrometry (Mikael Hult et al., JRC - Joint Research Centre, Belgium)
- Direct method for ^{210}Pb activity concentration determination (Boguslaw Michalik et al., GIG - Central Mining Institute, Poland)
- On-site verification of a measurement procedure for radon in waterworks (Michael Stietka et al., BOKU - University of Natural Resources and Life Sciences, Austria)
- Method for determination of the total activity of radioactive waste in drums (Branko Vodenik et al., IJS - Jozef Stefan Institute, Slovenia)

Conclusions

To test the applicability of the reference materials, instruments and methods developed during the project, verification criteria for measurement procedures and devices were developed. Several European NORM industry stakeholders were involved in the on-site measurements:

- Methods for determination of the total activity of inhomogeneously distributed NORM waste
- In-situ measurements of ^{210}Pb and ^{210}Po (Polonium) at a tin and lead production facility
- On-site testing of the hand-held in-situ alpha spectrometer at a metro station construction site
- Measurement procedure for in-situ analysis of radon in waterworks
- On-site testing of the prototype pixel detectors MEDIPIX/TIMEPIX
- Verification of measuring methods for an industrial site affected by NORM

On-site test measurements using the newly developed in-situ measurement methods and instruments were demonstrated and verified successfully. The on-site measurements successfully demonstrated that the new standards and techniques work in practice at industrial sites to measure low-energy gamma-ray emitters in NORM materials, surface contamination measurements, NORM in dust measurements and in-situ measurements of radon.

4 Actual and potential impact

The project established new reference materials and standard sources, and developed novel in-situ and laboratory measurement instruments and procedures which can be performed by the European NORM industry on building materials, drinking water and waste products. These will help to eliminate discrepant NORM measurement results by establishing the traceability of measurement methods and reducing end-user uncertainties to within legal limits.

The outcomes of the project, including the improvement of NORM radionuclide decay data, led to a significant reduction in uncertainties for NORM instrument calibration and measurements, which in turn enables cost-effective, accurate and reliable end-user measurements for the NORM industry. Additionally the project's results have contributed to standardisation organisations (e.g. CEN) to support the harmonisation of NORM measurement procedures across Europe.

The project's results will:

- improve weakly developed end-user measuring systems and methods, in particular for in-situ use, and will provide reliable instrumentation for monitoring and raw-material selection
- support innovative processing technologies for the re-use of TENORM by-products and waste
- support the fulfilment of the requirements of the European Construction Products Directive concerning natural radioactivity in building materials
- support the cost-effective implementation of the EU Basic Safety Standards Revision concerning the NORM industry, building material industry and waterworks
- support the global competitiveness of the European NORM/TENORM industry
- help to eliminate technical trade barriers and prevent trade disputes between EU member states
- support the economically effective implementation of the revision of the European Directive on radioactivity in drinking water.

The project's results also supports NORM industries and stakeholders regarding the implementation of the European Radiation Protection Directive 2013/59/EURATOM.

The project developed optimised systems and procedures for the determination of natural radioactivity content. These also allow effective selection of input material with low radioactivity content as a prerequisite for the reduction of radioactivity content in products, residues and waste. This also makes it possible to classify reliably waste as non-radioactive waste, reducing the production costs and environmental burden of the NORM industry.

In addition to this, the project's results support the development of metrological capabilities for industrial applications with an emphasis on precise, reliable and comparable measurements. The project focused on metrological requirements in the area of NORM materials for stakeholders' needs, and this was ensured by close cooperation with stakeholders during the project's lifetime.

The project has impacted specific industrial branches related to processing NORM and/or generating TENORM, which include:

- Extraction of rare earths

  Rare earths are used in catalysts, ceramics, refractory and metallurgical processes, magnets, and in low-temperature superconductor technology; ores of rare earth elements with natural radionuclides (e.g. ^{238}U, ^{226}Ra, ^{232}Th, ^{228}Ra, ^{138}La).

- Niobium/tantalum ore processing

  Niobium minerals usually contain both niobium and tantalum. The radionuclides occurring in niobium ore are those of the ^{238}U and ^{232}Th decay chains.

- TiO_2 pigment production

Ores from which titanium is obtained can contain concentrations of uranium and thorium in the range of 0.2 to 0.8 Bq/g. Total radium in sludge from titanium processes can have concentrations as high as 3 Bq/g.

- Phosphate industry – thermal phosphorous production, phosphoric acid production, production of phosphate fertilisers

Uranium in phosphate ores ranges in concentration from 0.26 to 3.7 Bq/g, thorium is present in background amounts, ~ 4 to 22 mBq/g. Phosphogypsum and phosphate slag as principal by-products contain uranium and thorium and their progeny; in general, about 80 % of the ^{226}Ra, 30% of the ^{232}Th and 14 % of the ^{238}U is left in phosphogypsum. Uranium and thorium become enriched in phosphate fertiliser to about 150 % of their original value.

- Building material industry, cement production

Building materials can contain elevated levels of radionuclides including ^{226}Ra, ^{232}Th and ^{40}K, either of natural origin in raw material (e.g. natural stone), or due to addition of industrial products (e.g. zircon sand), intermediates or by-products (e.g. coal ash, phosphogypsum and furnace slags).

- Tin foundries, tin/lead/copper smelting

Amang is a general term for the by-products obtained when tin tailings are processed into concentrated ores. ^{226}Ra and ^{232}Th activities in amang have been reported to range from 16 to 18 Bq/g and 43 to 330 Bq/g, respectively.

- Waterworks, ground water filtration, drinking water production

Water treatment includes passing the water through various types of filters. If water containing radionuclides is treated, radioactive wastes are generated such as radium, uranium and their progeny.

- Recycling industry

Coal ash is used as an additive in concrete, roofing materials, land reclamation, paint and undercoatings, and various other products such as structural fill for road construction. About 30 % of coal ash is reused. Since coal contains uranium and thorium, large quantities of coal ash present a potential radiological risk.

- Oil and natural gas industry

Many oil-field waters are particularly rich in chloride, and this enhances the solubility of other elements including the natural radionuclide radium (^{226}Ra, ^{228}Ra). Pipes and tanks that handle large volumes of this "produced water" can become coated with scale deposits that contain radium. Radium bearing scale is the specific type of NORM waste that occurs in the oil and natural gas industry.

Industries dealing with NORM materials and TENORM waste processing will benefit from the methods, devices and reference materials developed within this project. Efficient selection of input materials with low concentration of natural radionuclides as well as reliable measurements of radioactivity content of waste and by-products will help to lower production costs, diminish the number of trade disputes over the contamination levels of input materials and end products, and significantly reduce negative environmental impacts. Therefore, the project's results supports key economic and environmental factors for sustainability and competitiveness of the European NORM industry.

Industries working with raw materials containing naturally occurring radioactive materials (NORM industries) produce large amounts of waste. These waste materials, generated from current and past activities, constitute a huge economic and ecological burden if they are not properly disposed of or re-used as input materials for the industry. The recycling and re-use of waste material support the use of "cleaner technologies" and hence cost savings. The results from this project have established traceable, accurate, and standardised measurement methods and systems, in particular for in-situ applications, to help decide on the re-use of waste materials, without increasing costs and whilst avoiding contamination of the environment and radiation exposure to the public.

Activity measurement in the recycling industry currently focuses on artificial radionuclides. Natural radionuclides are often taken as part of the natural background, regardless of their concentration. The

reliable measurement of natural radionuclides supported by the project results will help to ensure that raw materials brought into the industrial process will not cause, as far as possible, enhanced activity levels in final products and waste.

Impact on standards

The results of the project have been disseminated to European and national standardisation bodies and working groups including CEN/TC 351 Construction products: Assessment of release of dangerous substances WG3 on radiation form construction products; CEN/TC 45 Nuclear Instrumentation; the ASI (Austrian Standards Institute) and the EC Group of Experts established under Article 31 of the EURATOM Treaty. The results on NORM activity metrology and measurement have been, and will be, considered in the radiation protection and dose modelling standards for construction products, environmental measurement methods, radiation protection instrumentation and radon / thoron measurement and dose assessment methods.

Dissemination of results

The project held regular workshops for stakeholders and industrial end-users (including NORM industry facilities, nuclear regulators, standardisation bodies and measurement device and calibration source manufacturers) which covered the new measurement techniques, best practices, reference materials, decay data, on-site demonstrations, eLearning NORM and the NORM situation in EU member states. The project also established an Advisory Board of experts from the IAEA, International Committee for Radionuclide Metrology (ICRM), European Cooperation in Science and Technology (COST), European ALARA Network for NORM (EAN-NORM) and NORM industries.

The results of the project have been presented to metrologists, stakeholders, regulators and end-users at international conferences. The project has submitted / presented 47 conference presentations and 25 scientific journal papers. A joint workshop was also held with the EU COST network NORM4BUILDINGS.

Training materials for a post-graduate course is available within a NPL e-learning course. These include:

- the hazards and risks associated with work involving NORM raw materials, by-products, residues and wastes,
- national and international requirements and recommendations,
- best practices and state-of-art measurement systems regarding NORM monitoring at industry,
- measurement systems, standard sources and reference materials to perform traceable NORM analysis.

An open access NPL e-learning course on NORM metrology and measurement has been established (http://www.npl.co.uk/commercial-services/products-and-services/training/e-learning/naturally-occurring-radioactive-materials/).

Potential impact

The results of the project (traceable, accurate and standardised measurement methods and systems, in particular for in-situ applications) will help the NORM industry to decide on the use of raw material and the re- use of waste materials without increasing costs, contaminating the environment or exposing workers or the public to harm.

The improved measurement of natural radionuclides ensures that the raw materials brought into the industrial process will not cause, as far as possible, enhanced radioactivity levels in the final products and in waste. The outcomes of the project will support better characterised of NORM raw material and waste, and identification and precisely measurement using traceable standardised methods. The project will also reduce costs in the NORM industry sector and increase safety in NORM production.

5 Website address and contact details

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6 List of publications

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