Final publishable JRP Summary for JRP IND57 MetroNORM Metrology for processing materials with high natural radioactivity

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

P1 of 7
EMRP European Metrology Research Programme Programme of EURAMET
The EMRP is jointly funded by the EMRP participating countries within EURAMET and the European Union

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.

Results and 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.

List of publications

[1]. Pöllänen R, Turunen J, Karhunen T, Peräjärvi K, Siiskonen T, Wirta M, Turunen A. Novel equipment for in-situ alpha spectrometry with good energy resolution. Health Physics 109(2015), 601–605.

[2]. Pöllänen R, Turunen J, Karhunen T, Peräjärvi K, Siiskonen T, Wirta M, Turunen A. Novel equipment for in-situ alpha spectrometry. Proceedings of the 8th International Conference on High Levels of Natural Radiation and Radon Areas, ICHLNRRA 2014. Proceedings ICHLNRRA 2014.

[3]. R. Pöllänen, T. Siiskonen. Unfolding alpha-particle energy spectrum from a membrane air filter containing radon progeny. Radiation measurements 70(2014), 15-20.

[4]. B. Sabot, P. Cassette, N. Michielsen, S. Pierre, S. Bondiguel. Development of a primary thoron activity standard for the calibration of thoron measurement instruments. Special Issue of Radiation Protection Dosimetry. Doi:10.1016/j.apradiso.2015.11.055.

[5]. Zbigniew BZOWSKI, Bogusław MICHALIK. Mineral composition and heavy metal contamination of sediments originating from radium rich formation water. Chemosphere 122(2015), 79-87.

[6]. B. Sabot, P. Cassette, N. Michielsen, S. Pierre, S. Bondiguel. A new thoron atmosphere reference measurement system. Applied Radiation and Isotopes (accepted for publication)

[7]. Mazanova M., Zdychova V., Dryak P., Bludovsky R. Development of a Reference Material for Water Industry. Journal Reference Materials (http://rmjournal.ru/english.html) (accepted for publication).

[8]. Denis Glavič-Cindro, M. Korun, B. Vodenik, B. Zorko. Activity measurements of barrels filled with radioactive waste. Journal of Radioanalytical and Nuclear Chemistry. Doi:10.1007/s10967-014-3666-0.

[9]. R. Pöllänen. Performance of an in-situ alpha spectrometer. Applied Radiation and Isotopes (accepted for publication).

[10]. Pöllänen R, Klemola S. Analysis of a liquid sample residue using in situ alpha spectrometry. Journal of Radioanalytical and Nuclear Chemistry 307(2016), 747–750.

[11]. Pöllänen R, Siiskonen T. Detection of photons and electrons using a semiconductor alpha detector. Journal of Physical Science and Application 5(2015), 319-329.

[12]. Bulanek B., Hulka J., Jilek K, Stekl I. The Portable Device for Continual Measurement of Radon Progenies on Filter using the Detector TIMEPIX. Radiation Protection Dosimetry 164,4(2015), 493–496.

[13]. F. Kabrt, A. Baumgartner, M. Stietka, F.J. Maringer. A simplified measurement method for on-site determination of the radon emanation from building materials. Radiation Measurements (submitted for publication).

[14]. M. Stietka, A. Baumgartner, F. Kabrt, F.J. Maringer. Measurement strategies for radon in indoor air of waterworks - A review. Radioprotection (in press; on-line: https://doi.org/10.1051/radiopro/2017004).

[15]. FJ Maringer et al. Advancements in NORM metrology - Results and impact of the European joint research project MetroNORM. Applied Radiation and Isotopes (in press; on-line: http://doi.org/10.1016/j.apradiso.2016.12.040).

[16]. Baumgartner A., Stietka M., Wiedner H., Maringer F.J. Study of particular problems appearing in NORM samples and recommendations for best practice gamma-ray spectrometry. Applied Radiation and Isotopes (in press; on-line: http://doi.org/10.1016/j.apradiso.2016.12.035).

[17]. Larijani C., Crespo T., Jerome S., Maringer F.J., Lutter G., Mazanova M., De Felice P. Reference materials produced for a European metrological research project focussing on measurements of NORM. Applied Radiation and Isotopes (in press; on-line: http://doi.org/10.1016/j.apradiso.2017.02.007).

[18]. Wiedner H., Peyrés V., Crespo, T., Mejuto M., García-Torano E., Maringer F.J. Application of an Artificial Neural Network for evaluation of activity concentration exemption limits in NORM industry. Applied Radiation and Isotopes (in press; on-line: http://doi.org/10.1016/j.apradiso.2016.12.044).

[19]. G. Lutter, I. Vandael Schreurs, T. Croymans, W. Schroeyers, S. Schreurs, M. Hult, G. Marissens, H. Stroh, F. Tzika. A low energy set-up for gamma-ray spectrometry of NORM tailored to the needs of a secondary smelting facility. Applied Radiation and Isotopes (in press; on-line: http://doi.org/10.1016/j.apradiso.2016.12.048).

[20]. Quinten Remijsen. Characterisation of materials suitable as NORM reference materials. University of Hasselt, June 2015.

[21]. Indy Vandael Schreurs. A set-up for in-situ measurement of Pb-210 and Po-210 at a metal production plant. Univ. Hasselt, June 2016.

[22]. Marie-Christine Lépy, Maria Marouli, Sylvie Pierre, Raf van Ammel. Photon emission intensities in the decay of U-235. Applied Radiation and Isotopes (in press; on-line: http://doi.org/10.1016/j.apradiso.2016.12.045).

[23]. Boguslaw Michalik et al. The self-absorption correction factors for ^{210}Pb concentration in mining waste and influence on environmental radiation risk assessment. Environmental & Health Studies 23(2016), 1-7.

[24]. Virginia Peyrés, Teresa Crespo, Marcos Mejuto, Eduardo García-Toraño. Measurement of NORM samples with $CeBr_3$ detectors. Applied Radiation and Isotopes (in press; on-line: http://doi.org/10.1016/j.apradiso.2017.02.012).

[25]. B. Sabot. Etalonnage des instruments de mesure de l'activit e volumique du thoron (^{220}Rn) dans l'air. PhD Thesis. Université Paris Saclay - Paris XI, HAL, 2016-01-11.

JRP start date and duration:	1 September 2013, 36 months
JRP-Coordinator: Prof. Dr. Franz Josef Maringer, BEV/PTP, Tel: +43 1 21110 6372 E-mail: franz-josef.maringer@bev.gv.at JRP website address: http://metronorm-emrp.eu/	
JRP-Partners: JRP-Partner 1: BEV/PTP; Austria JRP-Partner 2: CEA, France JRP-Partner 3: CIEMAT, Spain JRP-Partner 4: CMI, Czech Republic JRP-Partner 5: ENEA, Italy JRP-Partner 6: IJS, Slovenia	 JRP-Partner 7: IST, Portugal JRP-Partner 8: JRC, European Commission JRP-Partner 9: MKEH, Hungary JRP-Partner 10: NPL, United Kingdom JRP-Partner 11: NRPA, Norway JRP-Partner 12: STUK, Finland
REG-Researcher 1 (associated Home Organisation):	Andreas Baumgartner BOKU, Austria
REG-Researcher 2 (associated Home Organisation):	Jiri Hulka SURO, Czech Republic
REG-Researcher 3 (associated Home Organisation):	Boguslaw Michalik GIG, Poland

The EMRP is jointly funded by the EMRP participating countries within EURAMET and the European Union