Final Publishable JRP Summary for IND04 MetroMetal
Ionising Radiation Metrology for the Metallurgical Industry

Overview

Steel mills across Europe use radiation detection portals to test scrap material for the presence of radioactive sources. However, such detection portals often fail in certain circumstances, leading to radioactive contamination of steel and steel by-products. To improve radiation detection capabilities this project analysed the available gamma detection systems and recommended best methods and alternative prototype devices for monitoring radioactivity. Three protoype spectrometric devices for the measurement of activity in cast steel, fume dust and slag samples were developed, two of which were tested at end-user facilities in Europe. In addition SI traceable reference standards made of typical materials encountered in steel mills were produced and characterised to support better industrial measurements. In summary, the project has successfully set the basis for establishing common standards for radioactivity monitoring in steel mills and the reliable certification of the non-radioactivity of steel and by-products of the steel industry.

Need for the project

Each year, more than 500 steel production sites in 23 EU Member States produced 200 million tonnes of steel, and an average of 43% of this material is produced by recycling scrap materials. The scrap material is tested for the presence of orphan radioactive sources from various origins (e.g. industry and medicine), by passing the scrap containers under radiation detection portals. However, under certain circumstances this approach can fail and the radioactive source is smelted. This results in:

- radioactive contamination of the furnace, its outputs and by-products with radionuclides
- expensive clean-up costs (typically between 1 and 10 million €) for decontamination of facilities, storage and disposal of waste and lost production revenues
- leakage of radioactive substances into the environment
- trade disputes over the contamination level of steel products and the loss of reputation for steel makers.

Although the frequency of radioactive incidents of this kind has been significantly reduced over the last few years, recent incidents have demonstrated that these risks are common across Europe and that as well as monitoring scrap materials before melting, additional measurements are needed after melting to ensure and certify the absence of radioactive contamination in steel, slag and fumes dust.

Prior to this project, surveillance in the European steel industry was carried out by a number of different detection systems, primarily on scintillator/gamma detection systems. However, these systems vary in their technical approach, which resulted in different measurements and made it difficult to compare detection results between steel mills. This lack of consistency could then lead to trade disputes over contamination levels and/or radioactive contamination of steel products. Therefore, improved detection devices, based on optimised spectrometric methods and alternative detection systems were needed for industry, to provide improved and more reliable spectroscopic identification and quantification of contaminants.

In addition, there were no standard methods for measuring radioactivity at all stages (i.e. scrap loads, metal products, slag and fume dust) of the melting process or relevant reference materials for the validation of measurement devices and methods. .

Further to this, national standards and regulations for radioactivity monitoring lacked uniformity between EU member states and would benefit from harmonisation.

Report Status: PU Public

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Scientific and technical objectives

The project had the following scientific and technical objectives:

1. The development of reliable, SI traceable methods optimised for the control/measurement of radioactivity at each stage of the smelting process (e.g. scrap loads, metal products, slag and fume dust).
2. The development of reference standards for cast steel (real and composite reference standards), slag and fume dust. Reference standards will be contaminated with potential contaminant radionuclides (e.g. ^{60}Co, ^{137}Cs, ^{192}Ir, ^{226}Ra) and have different geometries/matrices that correspond to the cast steel probes currently used for on-line measurements and the slag cartridges used for the calibration of radioactivity detectors.
3. The characterisation of the measurement methods with the reference standards produced previously, using inter-laboratory comparisons and Monte Carlo (MC) simulations to cover the large diversity of sample geometries, shapes, densities and elemental compositions.
4. The design of an optimised spectrometric device and the production of prototype devices for the measurement of activity in cast steel, fume dust and slag samples using the methods developed in the project and including laboratory testing of the prototype devices.
5. The design of evaluation criteria for spectrometric prototype testing at end-user facilities (i.e. foundries). They must be based on end-user needs/constraints.
6. Demonstration of the prototype spectrometric devices at selected foundries in Europe, development of technical recommendations and input into European and National Standards Committees for the standardisation of radioactivity monitoring (e.g. calibration of measurement systems, on-line monitoring of production and certification of cast steel batches), and worldwide dissemination of project results to end-users, stakeholders and the general society through journal articles, conference presentations and specialised workshops.

Results

1. *The development of reliable, SI traceable methods optimised for the control/measurement of radioactivity at each stage of the smelting process (e.g. scrap loads, metal products, slag and fumes dust).*

The project started with a review of systems currently in use for measuring the activity of radionuclides in metallurgy across Europe in end-user steel factories in 14 European countries. This was complemented with additional information on available detection systems obtained from manufacturers. This information led to recommendation that the best alternatives to the current gamma detection systems used in industry to measure the activity of cast steel and slag samples, which were gamma ray spectrometric devices using either a Germanium (Ge) detector or $BeCr_3$ scintillator. Therefore, basic designs for optimised spectrometric prototype systems, based on Ge and $BeCr_3$ were produced.

SI traceable methods were achieved by the project by developing methods for use in industry with gamma-ray spectrometry, that could be validated using certified samples whose shape and composition were optimised for the measuring equipment proposed (e.g. as part of reference standards developed in objective 2). As the same technique (gamma-ray spectrometry) is used by industry for all post-melting measurements of radioactivity, these methods should ensure consistency at each stage of the smelting process.

Improved methods for the calibration of portal monitors as well as sampling strategies were also proposed by the project for use in industry. The methods emphasised the use of SI-traceable sources/reference standards in order to improve the traceability of the calibration methods used by steel mills.

2. *The development of reference standards for cast steel (real and composite reference standards), slag and fume dust. Reference standards will be contaminated with potential contaminant radionuclides (e.g. ^{60}Co, ^{137}Cs, ^{192}Ir, ^{226}Ra) and have different geometries/matrices that correspond to the cast steel probes currently used for on-line measurements and the slag cartridges used for the calibration of radioactivity detectors.*

The project developed reference standards for the materials typically found at end-user facilities: cast steel (real and composite), slag and fume dust; for the radionuclides considered as potential contaminants; and in formats similar to those found at end-user facilities. The reference standards developed included: real cast

steel and composite cast steel standards containing several radionuclides as ^{60}Co, ^{137}Cs,^{192}Ir and ^{241}Am as well as slag standards contaminated with ^{60}Co, ^{137}Cs, ^{241}Am and ^{226}Ra and fume dust standards contaminated with ^{137}Cs.

The reference standards have been certified in activity by the joint work of a large number of National European Metrology Laboratories (objective 3) and are available for use by end-users. In fact, five steel mills across Europe are already using in them.

Figures 1 and 2 (above): Two sets of reference standards; one set from slag contaminated with Ra-226 and one set from cast steel contaminated with Co-60 (respectively)

3. *The characterisation of the measurement methods with the reference standards produced previously, using inter-laboratory comparisons and Monte Carlo (MC) simulations to cover the large diversity of sample geometries, shapes, densities and elemental compositions.*

This objective acted as a bridge between the first two of the project's objectives and the last two objectives The work included the validation of the basic designs for the Ge and $BeCr_3$ spectrometric prototype devices from objective 1 using MC models and an inter-laboratory comparison (ILC) of these MC models. It also included the validation of the reference standards produced in objective 2 via ILCs.

The basic designs for the Ge and $BeCr_3$ spectrometric prototypes using MC models were validated with an ILC using a common detector model, whose dimensions and composition was agreed by all partners. ILC calculations were carried out by all partners and the results were evaluated using the Mandel-Paule weighted mean method. As part of the ILC the partners performed MC simulations for the Ge and $BeCr_3$ spectrometric prototypes of full-energy peak and total detection efficiencies for the reference standards developed as part of objective 2, as well as gamma ray energies corresponding to different key radionuclides found in the metallurgical industry. The results of the ILC validated the basic designs for the Ge and $BeCr_3$ spectrometric prototypes (without changes), as the basic designs matched the pre-requisites established for the prototypes (as determined in objective 1).

ILCs were also used to validate the reference standards produced in objective 2. A total of 13 of the NMI/DI partners participated in the inter-comparisons and the results were used to derive the activity concentration of the radionuclides in each reference standard, resulting in a set of reference standards with the highest degree of metrological characterisation was obtained.

New Calibration and Measurement Capabilities (CMCs) relevant to the metallurgical industry have also been developed for the reference standards developed in the project. Once approved by the regional metrology organisations, the CMCs will allow users (i.e. providers of calibration standards such as National Metrology Laboratories or accredited laboratories) to provide certification of standards to be used for calibration purposes.

4. *The design of an optimised spectrometric device and the production of prototype devices for the measurement of activity in cast steel, fume dust and slag samples using the methods developed in the project and including laboratory testing of the prototype devices.*

Based on the basic designs for optimised spectrometric devices from objective 1, two portable prototype devices based on Ge detectors and a third prototype system, based on a $BeCr_3$ scintillator were produced as well as specific software for all three prototypes. The prototypes were characterised and calibrated at CIEMAT and PTB with the reference standards developed in objective 2 and using the MC models from objective 3. The reference standards from objective 2 were used to determine their detection limits. Results showed that, when operating in laboratory conditions, their performance met the target values established in objective 1 in terms of energy resolution, counting efficiency, and detection limits.

5. *The design of evaluation criteria for spectrometric prototype testing at end-user facilities (i.e. foundries) and demonstration of the prototype spectrometric devices at selected foundries in Europe.*

Following the laboratory testing, the Ge-based prototype spectrometric devices were taken to three end-user facilities (steel mills) in Portugal, Czech Republic and Spain for on-site testing. Results showed that the target detection limits, (i.e. 1 Bq/g for most nuclides) were achieved with the Ge-based devices and that a clear and unambiguous identification of contaminants in cast steel could be carried out in short counting times (i.e. in the order of a few minutes). These counting times are critical for assessing cast steel and are required by the steel industry.


Figures 3 and 4: Testing of the CIEMAT and PTB Ge-based prototype spectrometric devices at end-user facilities in Portugal (Siderurgia Nacional) and Czech Republic (Vitkovice Steel)

6. *Development of technical recommendations and input into European and National Standards Committees for the standardisation of radioactivity monitoring (e.g. calibration of measurement systems, on-line monitoring of production and certification of cast steel batches)*

The project’s results were presented to relevant technical committees related to industry in Austria, the Czech Republic, Germany, Spain and Portugal, to a Group of Experts established under Article 31 of the EURATOM Treaty, to the REMCO/ISO committee and to the Division of Radiation, Transport and Waste Safety of the International Atomic Energy Agency in Vienna. Presentations were made throughout the lifetime of the project and focused on the main achievements of the project at that time.

Further to this, to support the standardisation of radioactivity monitoring the project website includes information on the reference standards, and the recommendations for the design of optimised spectrometric devices as well as downloadable MC models of the prototype spectrometric devices.

Actual and potential impact

To ensure adoption of the project’s outputs a wide range of activities were undertaken to disseminate the project’s outputs to potential users. The dissemination activities included: 1) technical and standard committees from EURATOM, IAEA, EURAMET and industrial organisations, 2) the end-user community through six workshops and five visits to their facilities and 3) presentations to a wider community at conferences in Europe, America and Africa and publication in scientific and technical journals. The project website contains relevant information about project outputs and has been accessed from 49 countries worldwide.

As a result of the dissemination early adoption of the project's outputs is already underway:

The project has produced new SI traceable reference standards for composite steel, cast steel, slag and fume dust containing known activity of radionuclides considered as potential contaminants. So far five end-users (steel mills from 5 different European Countries) are already using these reference standards. This will lead to better detection and measurement of radioactivity in scrap loads, fume dusts and cast steel batches leading to better and more consistent certification of steel batches. Talks have been started with the steel industry with a view to developing technical standards for certifying the non-radioactivity of cast steel.

In the longer term the results of this project will help to reduce the number of 'false alarm events' at industrial sites and to minimise the impact of them occurring. This should result in significant cost savings due to a reduction in the number of shut downs of the production line at industrial sites as well as the associated clean-up costs. By improving detection methods the project should also significantly reduce the risk of accidental irradiation and radioactive material leaking into the environment.

The use of the prototype spectrometric devices, technical procedures and reference materials for the detection and measurement of radioactivity in scrap loads, fume dusts and cast steel batches by end-users will lead to better and more consistent certification of the radioactive contents of steel and by-products. Where by-products are concerned, such as the use of slags in road pavements and the concrete industry, accurate measurement will lead to a reduction in the costs arising from disputes due to inconsistent data between different companies/countries and decontamination costs.

List of publications

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3. J.Šolc, P. Dryák, H. Moser, T.Branger, E.García-Toraño, Vi Peyrés, F. Tzika, G. Lutter, M. Capogni, A. Fazio, A. Luca, B. Vodenik, C. Oliveira, A. Saraiva, L. Szucs, T. Dziel, O. Burda, D. Arnold, J.Martinkovič, T..Siiskonen, A. Mattila, 2015, *"Characterisation of a Radionuclide Specific Laboratory Detector System for the Metallurgical Industry by Monte Carlo Simulations"* . Rad. Phys. Chem. http://dx.doi.org/10.1016/j.radphyschem.2015.01.003
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8. B. Caro, F. Tzika, M. Hult, G. Lutter, M. Mejuto, M. T. Crespo (2014) *"Characterization of ^{226}Ra activity in low-level slag reference standards"*, Journal of Radioanalytical and Nuclear Chemistry DOI 10.1007/s10967-014-3851-1.
9. M. Sahagia, A. Luca, A. Antohe, R. Ioan, M. Tanase, E. Garcia-Toraño (2014) *"Comparison of analysis methods for the characterisation of the radioactive content of Metallurgical slag used within the Euramet-EMRP JRP IND04 MetroMetal"*, Romanian Reports in Physics 66,3 ,649-657.

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