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1 Executive Summary

Introduction

This project has successfully set the basis for establishing common standards for radioactivity monitoring in steel mills and the certification of the non-radioactivity of scrap metals. It has done this by producing SI-traceable reference standards and methods for radioactivity measurements, and the construction and end-user validation of two prototype detectors for radioactivity monitoring.

The Problem

Every year, more than 500 steel production sites in the EU produce 200 million tonnes of steel and on average 43% of this, is produced by recycling scrap materials. The scrap material is tested for the presence of orphan radioactive sources by passing the scrap containers under radiation detection portals. However, under certain circumstances, such as the self-shielding of sources or attenuation by other scrap pieces, this approach can fail and a radioactive source is smelted. This results in radioactive contamination, expensive clean-up costs (typically between 1 and 10 million €) and leakage of radioactive substances into the environment,Radioactivity surveillance in the European steel industry uses different detection systems (primarily scintillator/gamma detection systems) that vary in their technical approach. These dectection systems have differences in their geometry, energy resolution or sensitivity levels of detection, making it difficult to compare their performance. In addition, SI traceable methods for measuring radioactivity in scrap and industrially relevant reference materials (i.e. for composite steel, cast steel, slag and fume dust containing known activity of radionuclides considered as potential contaminants) are lacking and needed for the control/measurement of radioactivity at each stage of the smelting process (e.g. scrap loads, metal products, slag and fume dust).

The Solution

This project recommended the best methods and devices for monitoring radioactivity, as well as highlighting those able to provide optimal traceability and minimal uncertainty:

- SI traceable reference standards composite steel, cast steel, slag and fume dust containing known activity of radionuclides considered as potential contaminants were been produced and characterised by inter-laboratory intercomparisons
- Three protoype spectrometric devices (based on electrically cooled Ge detectors and on a $CeBr_3$ scintillator) for the measurement of activity in cast steel, fume dust and slag samples were developed and tested.

Impact

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

The SI traceable new reference materials and methods developed by the project for the detection and measurement of radioactivity in scrap loads, fume dusts and cast steel batches will lead to better and more consistent certification of steel batches and a reduction in the costs arising from disputes due to inconsistent data between different companies/countries. The reference materials are already in use in five steel mills from five different European Countries).

In the longer term the technical recommendations developed in the project will provide national and international standards organisation and regulatory bodies with the metrological basis to revise the current diversity of regulations and harmonise the European policies. Technical standards for certifying the non-radioactivity of cast steel will also benefit from project's results and talks have been initiated with the steel industry with a view to their use in certification of steel products.

2 Project context, rationale and objectives

Context and rationale

Each year, more than 500 steel production sites in 23 EU Member States produce 200 million tonnes of steel. On average 43% of this material is produced by recycling scrap loads, which come from various origins such us:

1. Dismantling of buildings and other structures. This is a main source for supplying scrap metal, sometimes in places far away from foundries, often outside Europe. It therefore becomes possible that a radioactive source contained in a device is unintentionally taken together with other metals.
2. Dismantling of Nuclear Facilities. Many nuclear power plants are being dismantled worldwide as they reach their working age. However, they are a main source of metallic materials and although metals from nuclear power plants are monitored in place, contaminated metals can also be added to clean, uncontaminated scrap, by error or device malfunction.

All scrap loads entering steel mills are tested for the presence of orphan radioactive by passing the scrap containers through radiation detection portals. However, under certain circumstances, such as self-shielding of sources or attenuation by other scrap pieces, this approach can fail and a radioactive source is smelted. This can result in radionuclides being incorporated into finished products and:

- radioactive contamination of the furnace, cast steel, slag, fume dust, filters and ducts, by radionuclides (e.g. ^{60}Co or ^{192}Ir, all frequently arising from radiotherapy, brachytherapy or gammagraphy sources),
- expensive clean-up costs (typically between 1 and € 10 million, but sometimes higher) for decontamination of facilities, storage and disposal of raw waste and lost production revenues,
- leaking of radioactive substances into the environment,
- external irradiation of foundries workers, or even the general public,
- trade disputes over the contamination level of steel products (which also has a financial impact on market prices),
- the loss of reputation for steel makers.

Although the frequency of radioactive incidents of this kind has been significantly reduced in the last years, recent incidents have demonstrated that these risks are still 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.

Current radio assay controls, used in foundries, do not provide the appropriate level of traceability for measurements of radioactivity in cast products. To address this, radiation detection equipment is needed throughout the lifecycle of steel products, i.e. from the recycled scrap loads to the certification of the final steel cast products. This intermediate testing includes the testing of steel by-products such as slags and fume dusts and several ‘intermediate’ gamma detection systems are currently in use. However these commercially available gamma detection systems vary in their technical approach, which results in differences in their geometry, energy resolution and the sensitivity levels of their measurements. They also do not have enough specific reference materials or calibration standards to ensure the SI traceability of their measurements.

In addition to this, a recent European Directive (2013/59) has set more demanding limits on the measurement of radioactivity in this context, thus calling for even more efficient and reliable methods.

To compound these issues there are also differences between the national standards/regulations of different EU countries on radioactivity monitoring. The consequence of this is that radiation detection from different steel mills/companies cannot be compared across the EU, which often leads to disputes and difficulties in trading. It is therefore vital to produce a European standard that can be used by all EU countries and the need for this approach has been the subject of many European and International reports and recommendations e.g.

- "Recommendations on monitoring and response procedures for radioactive scrap metal" (United Nations Economic Commission for Europe-UNECE, 2006), which recommends the "*Establishment of a voluntary international "Protocol" or "Recommendations" providing for a consistent and internationally harmonised approach to monitoring procedures"*.
- "Spanish Protocol for Collaboration on the Radiation Monitoring of Metallic Materials (MINER, Spain, 2005), which has been considered as model to follow by the UNECE report and by the U.S. Environmental Protection Agency (EPA).
- Council Regulation 333/2011 (European Union, 31 March 2011) on establishing the criteria determining when certain types of scrap metal cease to be waste under the Directive 2008/98/EC of the European Parliament and the Council, which states that the scrap producers shall have implemented a quality management system with (Article 6.2.d) *"documented procedures concerning... effectiveness of the radiation monitoring ..."* and that *"Each consignment of scrap shall be accompanied by a certificate established in accordance with national or international rules on monitoring and response procedures for radioactive scrap metal."* (Annex I and II, 1.5).
- "Control of Orphan Sources and Other Radioactive Material in the Metal Recycling and Production Industries" International Atomic Energy Agency Safety Standards, Specific Safety Guide No. SSG-17, 2012.

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.

3 Research results

3.1 The development of reliable, SI traceable methods optimised for the control/measurement of radioactivity at each stage of the smelting process

Introduction

The aim of this work was 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). To do this work focused on the following two topics:

1. The survey of the current situation in monitoring scrap loads, cast steel, slag and fume dust
2. Recomendations for the basic design of an optimised spectrometric device for measuring the activity of cast steel, slag and fume dust

Survey of the current situation

Although basic information concerning the ionising radiation metrology support in metal processing industry was available to project partners, one of the initial tasks was to supplement this general overview with more detailed information from end-users. To do this a questionnaire was prepared with questions concerning the measuring devices used at end-user facilities for monitoring of radionuclides in incoming rough (scrap) material and outgoing products and by-products. The questionnaire focused on three main areas: 1) monitoring of radioactive content in incoming material using gate monitors, 2) measurement of radioactive content in produced cast steel and slag and 3) measurement of radioactive content in generated fume dust.

The questionnaire was translated into native languages (where necessary) and distributed to 69 selected companies across Europe (in Germany and Spain this was through professional associations of metal processing companies). Information from the completed questionnaires was processed anonymously, however some end-users considered the information confidential and were unwilling to complete the questionnaire. In the end, 21 questionnaires were completed, from 8 countries (Czech Republic, Finland, Poland, Portugal, Romania, Slovakia, Slovenia and Spain) and collected for evaluation.

The data obtained from questionnaires were supplemented by additional information from Spain (CIEMAT), Belgium (JRC) and Germany (PTB).

Test procedure for gate monitors for scrap loads

From the results of the questionnaire on the monitoring of radioactive content in incoming material using gate monitors it was noted that virtually all metal processing companies follow the “Recommendations on monitoring and response procedures for radioactive scrap metal” from UNECE (2006), and have gate monitors installed. Gate monitors scan trucks or wagons bringing rough material (mostly scrap) to the steel mills. A few gate monitors have radiation monitors installed not only on gates, but also on grapples and conveyor belts at shredders, so the radiation monitoring of incoming material is performed two or three times before the melting of scrap occurs.

The vast majority of gate monitoring instruments are based on large area plastic scintillators from a few manufacturers and are mainly operated using commercial evaluation softwares provided by the equipment manufacturer. Most gate monitors are simple systems i.e. non-spectrometric detectors and cannot identify specific radionuclides, although some of them can discriminate between natural and artificial origin of the contaminants. The conclusions from the questionnaire on the monitoring of radioactive content in incoming material using gate monitors were that virtually all gate measurement systems are based on scintillation detectors. In most cases, the detector element was a NaI(Tl) crystal and in only a few other cases, CsI was used. The most widely used geometry was cylindrical, with typical sizes of 5 x 5 cm (2”x2”) or 7.5 x 7.5 cm (3”x3”). One end-user also reported a well-type detector, which allows a significant improvement in detection efficiency however this was discarded in the survey results, as large size sources from slags or fume dust

would be difficult to place inside the well. No active shielding for the gate monitor was reported by all end-users and all systems had lead shields of variable thickness, typically between 5 and 10 cm.

A testing procedure was developed by the project for gate monitors. Because of dynamic testing, the procedure focused on wagon loads, but it is also applicable for truck load measurements. Two main tests for gate monitors were specified in the test procedure: 1) detection limits for empty and scrap full wagon and 2) the percentage of false alarms. The testing procedure aimed to prove the capability of gate monitoring instruments to detect the presence of the source of ionising radiation in various positions on the loading surface of a wagon, including an empty wagon and wagon loaded with scrap metal. The testing procedure included the following test steps:

- Determination of detection limits for empty and full scrap wagons.
- Determination of the percentage of false alarms.
- Dependence of the response on the activity of the radioactive source.
- Non-overload ability of the measurement system.
- Dependency of the response on the energy of primary photons.
- Readiness of detection.
- Temperature effects.

Recommends for the basic design of an optimised spectrometric device for measuring the activity of cast steel, slag and fume dust samples

The results of the questionnaire showed that most control laboratories at steel mills use NaI(Tl) spectrometers, with only two laboratories reporting the use of more advanced systems based on CsI detectors. These systems are used to measure the radioactivity from cast steel, but only in exceptional cases are slag and fume dust radioactivity monitored with the same instrument. An solution to the problem would therefore be to have a single instrument that could be used for all measurements.

The following options were considered for the spectrometric device:

- **Sodium Iodide** detectors constitute the basic element of existing spectrometric devices. They provide a good light output, can be manufactured in a great variety of shapes and their cost is significantly lower than other detectors. However, their main limitation is energy resolution, typically 7% at 662 keV which hampers the identification of gamma lines of close energy. One positive point is that they have higher efficiency than semiconductor detectors of a similar size.

- **Cesium Iodide** crystals either doped with Thallium CsI(Tl) or Sodium CsI(Na) are an interesting alternative to NaI(Tl) because of their higher detection efficiency. Whilst the energy resolution of CsI(Na) is quite similar to that of NaI(Tl), CsI(Tl) crystals have superior energy resolution, below 5% for the 662 keV line of ^{137}Cs, however they require the use of avalanche diodes instead of conventional photomultiplier tubes. They also exhibit a worse non proportionality than NaI(Tl) detectors and have a high resistance to radiation (although this characteristic is of little interest for the application in this project).

- **Zinc Selenide** ZnSe(Te) detectors were not a suitable alternative, as they can only be used in small sizes, as they have limited transparency to their scintillation light.

- **Lanthanum Bromide**, ($LaBr_3$(Ce)) belong to a new generation of inorganic scintillations detectors used in gamma spectrometry. Their energy resolution is much better than NaI(Tl) crystals, about 3% at 662 keV. Nevertheless, their radioactive background makes them unsuitable for the application in this project, as the crystals contain some amount of ^{138}La which decays by electron capture to ^{138}Ba and by beta minus to ^{138}Ce. In addition, deexcitacion from excited states of the first nuclide produces photons with energies of 34.7 keV and 1435.8 keV while deexcitacion of ^{138}Ce gives rise to 788.7 keV photons. This all adds up to an estimated background of 1.5 Bq/cc which makes these detectors unsuitable for measurements of low levels of radioactivity.

- **Cerium bromide** ($CeBr_3$) scintillators are a promising alternative to NaI(Tl) that have recently became available in practical sizes. Their energy resolution is better than that of sodium iodide detectors for gamma ray energies over 200 keV (4% for the 662 keV gamma line of Cs-137), and their detection efficiency is comparable to that of Lanthanum Bromide detectors, but they don't have the intrinsic background typical of them. $CeBr_3$ scintillators have a good linearity, however a disadvantage is their energy resolution which at low energies is worse than that of sodium iodide.

- The advantages of **semiconductor detectors** compared to scintillation detectors are well known. The energy needed by semiconductor detectors for the production of information carriers (electron-hole versus light photon) is orders of magnitude lower than in scintillators. In gamma-ray spectrometry, Germanium (Ge) detectors are used for most applications, however, to minimise their electronic noise, they must be operated at a working temperature of 85 K, which requires a cooling system. The main drawbacks of Ge detectors are the lower detection efficiencies compared to that of scintillator crystals of the same size, although the superior resolution allows a better spectral estimation of the background under the gamma peaks. Traditional Ge detectors depend on a periodic supply of liquid nitrogen, something not currently available in the industrial environment of steel mills. However, electrical cooling of Ge detectors, is now available and has a higher degree of reliability.

Following careful consideration of all the above options, Ge, NaI(Tl) and $CeBr_3$ were chosen as the three main options for the design of an optimised spectrometric device for measuring the activity of cast steel, slag and fume dust samples. Figure 1 below shows the simulated gamma spectra of a source of ^{60}Co, for the three main options considered: i.e. Ge, NaI(Tl) and $CeBr_3$. From the simulated gamma spectra is can be seen that the energy resolution of $CeBr_3$ detectors is sufficient to separate the two emissions of ^{60}Co, but is significantly lower than that of Ge detectors.

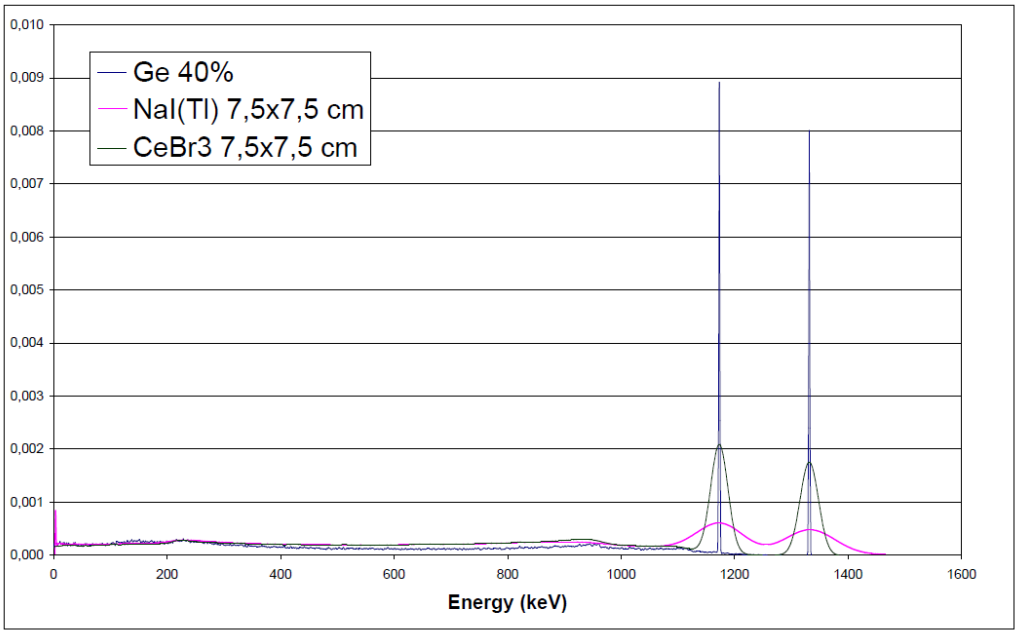


Figure 1 Simulated gamma spectra of a source of Co-60 taken with three different detectors

Detection efficiencies were calculated by Monte Carlo simulation for the three main detector options (i.e. Ge, NaI(Tl) and $CeBr_3$) and are shown in Table 2 below for the case of a slag source contaminated with a gamma-ray emitter of energy 186.2 keV, the energy corresponds to the main emission of ^{226}Ra. From the values for the full absorption peak efficiency and total efficiency it can be seen that the total and full absorption peak efficiencies are much lower for the Ge detector. This disadvantage can be compensated by the lower background and better energy resolution of Ge detectors.

Table 2 Computed detection efficiencies (Table) and simulated spectra (figure) for a slag source contaminated with a gamma-ray emitter of 186.2 keV

Detector type	Full absorption peak efficiency	Total efficiency
$CeBr_3$ (7.5x7.5 cm)	0.382	0.523
NaI(Tl) (7.5x7.5 cm)	0.375	0.514
Ge (relative eff. 40%)	0.049	0.114

Simulated spectra for slag sources of ^{226}Ra with NaI, $CeBr_3$ and Ge detectors are shown in Figure 2 below. From the simulated spectra it can be seen that the energy resolution of scintillator detectors would not allow the discrimination of the 186.1 keV emission from ^{226}Ra with that of 185.72 keV emission of ^{235}U; both are often present as a minor contaminant in Natural Occurring Radioactive Materials (NORM)

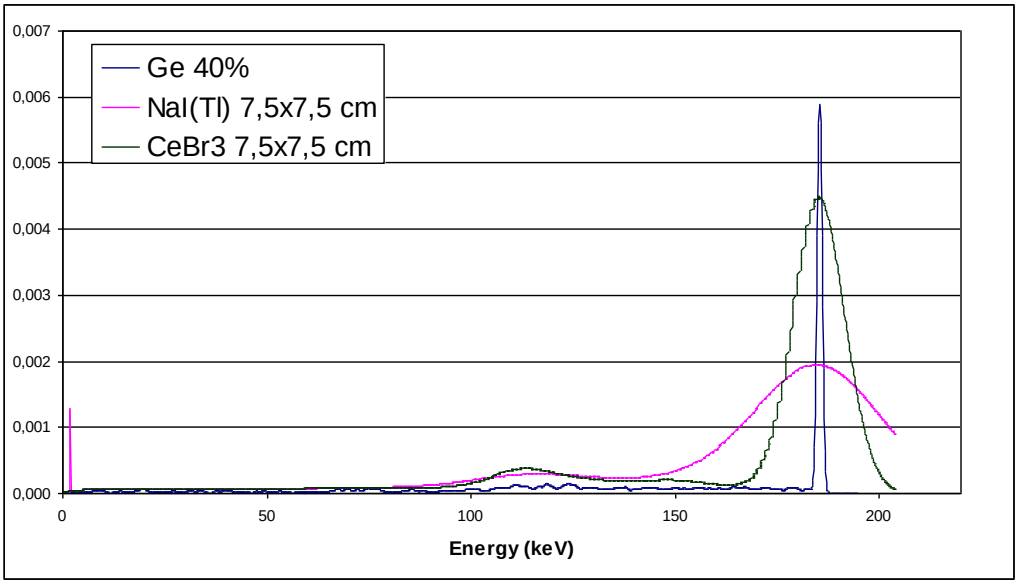


Figure 2 Simulated spectra for a slag source containing ^{226}Ra measured with NaI, $CeBr_3$ and Ge detectors. Only the region corresponding to the gamma emission of 186.1 keV is shown

Having considered Ge, NaI(Tl) and $CeBr_3$ as the three main options for the design of an optimised spectrometric device for measuring the activity of cast steel, slag and fume dust samples, the following recommendations were made:

- A spectrometric device based on a Ge detector, with a relative efficiency between 30 and 40%, surrounded by a prismatic or cylindrical lead shield of, at least, 5 cm thickness was recommended. The prototype device would be built and tested at CIEMAT and PTB and later in the industrial environment.
- Given that the detectors have to work in an industrial environment, a sample chamber must be adapted to the sensitive face of the detector in order to protect it from impact (in the industrial environment). Such a chamber should be of cylindrical shape, with an open top surface, with a minimum diameter of 120 mm and a heigh of similar dimensions. To minimise the attenuation of the gamma radiation, the chamber must also be made of low-atenuation materials.
- Finally, a configuration based on a $CeBr_3$ detector was also recommended for testing. The prototype device would be built and tested at CIEMAT.

Conclusions

From the results of a questionnaire sent to end-users across Europe, 1) test procedures and 2) the basic design of an optimised spectrometric device for measuring the activity of cast steel and slag samples were established by the project.

1. Test procedures: whilst specific (portal gate monitors) equipment is required at the entrance of end-user facilities to monitor the radioactivity of scrap loads, the project recommended the use of a common instrument to measure the radioactivity of cast steel and by-products, although the use of other instruments (i.e. measurement of fume dust by continuous monitoring) was not precluded . SI traceability was achieved by the project developing methods that could be validated using certified samples whose shape and composition were optimised for the measuring equipment proposed. Process consistency was ensured because the same technique (gamma-ray spectrometry) is used for all post-melting measurements.
2. The basic design of an optimised spectrometric device for measuring the activity of cast steel and slag samples: the recommended design for the spectrometric prototype was based on a Germanium (Ge) detector. In this way, easy identification of possible contaminants can be achieved. A configuration based on a CeBr3 detector was also recommended for testing.

3.2 The development of reference standards for cast steel (real and composite reference standards), slag and fume dust

Introduction

The aim of this work was 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 development of reference standards adapted to the needs of the metallurgical industry and at the higuest possible metrological level was one of the key aims of the project. Such an objective required the cooperation of all partners, either in the identification of possible suppliers, by taking part in the source preparation or in the many technical discussions to find the best strategy (held in the project meetings). The sources for the reference standards were:

1. Real cast steel and composite cast steel
2. Slag
3. Fume dust

Table 2 Characteristics and number of reference samples produced in the project..

Type of Standards	Nuclide	Lead Participant	Nominal Contamination level (Bq/g)	Number of standards	
				ILC certified	Partners Certification
Real cast steel	Co-60	PTB	1	14	
			10		4
		SMU	10	14	
	Ir-192	PTB	10		4
Composite cast steel	Am-241	ENEA	1		2
			10		2
	Cs-137	PTB	1		2
			10		2
	Co-60	CMI	1		2
			10		2
	Am + Cs + Co	ENVINET	1 and 10		3 x 2
Slag	Ra-226	CIEMAT	0.1		3
			1		3
			10	14	
	Co-60		10		3
	Cs-137		10		3
	Am-241		1		3
	Am-241	PTB	50		3
Fume dust	Cs-137 + Co-60	PTB	10 + 0.5	13	

Reference standards

Reference standards were prepared with potential contaminant radionuclides (e.g. ^{60}Co, ^{137}Cs, ^{192}Ir, ^{226}Ra and ^{241}Am) in 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 numbers and characteristics of the set of sources prepared are given in Table 2 below. A subset of the sources were calibrated by inter-laboratory comparisons (ILC) carried out with the participation of most partners (see section 3.3 for more details on the inter-laboratory comparisons).

Real cast steel samples with dimensions and composition similar to those used at end-user laboratories were provided by PTB and represented real contaminated material originating from a waste management facility (Siempelkamp). Those from ^{60}Co were prepared in two different activity levels: 1 and 10 Bq/g. The first activity level corresponds to material contaminated at levels that require intervention. They have cylindrical shapes and their approximate dimensions are 3.5 cm diameter and 1 cm height. Samples with 1 Bq/g are well suited to test the detection limits of equipment, while those of higher activity concentrations are more appropriate for detector calibration. The composition of the PTB cast steel samples was obtained at CIEMAT by XRF spectroscopy.

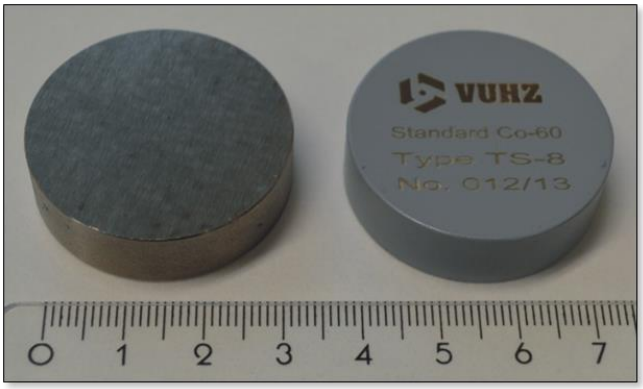


Figure 3 above shows two of the cast steel reference sources produced in the project

Another set of cast steel samples was provided by SMU and was specifically produced by a metallurgical company (VUHZ) by adding ^{60}Co to cast steel. Cast steel sources with ^{192}Ir with similar structure were also produced in a german factory following PTB specifications.

Figure 3 above shows two of the cast steel reference sources produced and standardised in the project (in total there were more than 45). The shape closely matches those of the typical probes taken from the control laboratories of end-users (see Figure 4).

Figure 4 a typical probe taken from the control laboratories of end-users

When real contaminated material is not available, as often happens for some combinations of radionuclides and matrices, composite steel sources can be used to emulate the unavailable material. They are produced, as shown in Figure 5, with a number of disks contaminated by dropping aliquots of a radioactive standard solution. In this way, sources of ^{241}Am, ^{137}Cs and ^{60}Co were produced at PTB, CMI and ENEA and composite sources containing a mixture of radionuclides (^{241}Am, ^{60}Co and ^{137}Cs) by REG(ENVINET)

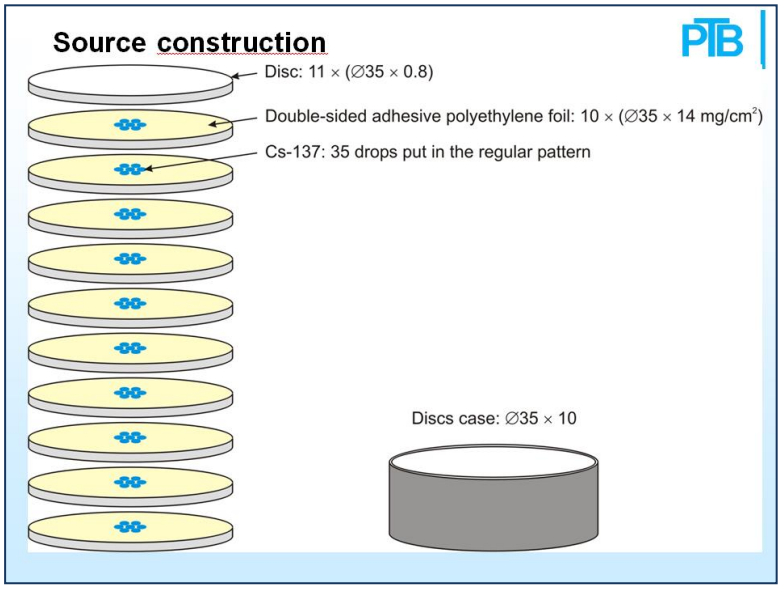
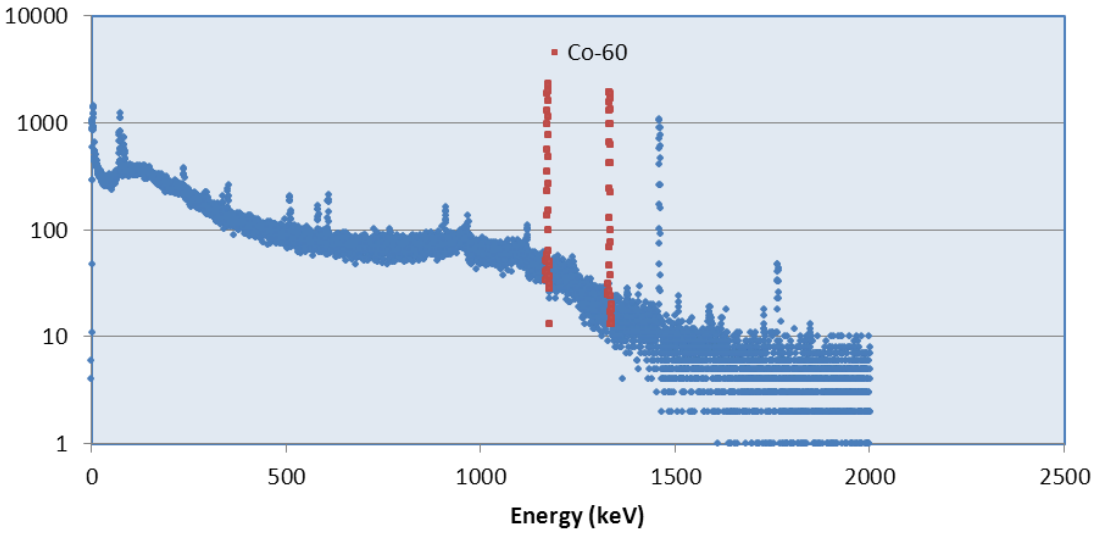
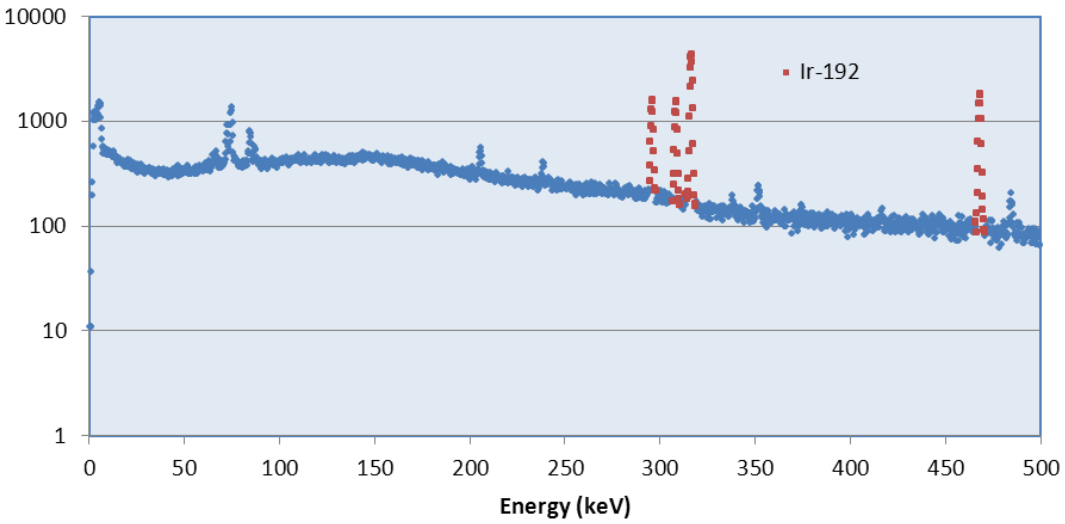


Figure 5 Internal structure of composite steel sources

These composite steel (mock) sources can be efficiently used for calibrating equipment for nuclides which have gamma emissions of medium and high energy, (above a few hundred keV) such as ^{137}Cs, ^{192}Ir and ^{60}Co. However, their use with low-energy emitters such as ^{241}Am must be done carefully, as self-attenuation of the radiation in steel requires important corrections. The sample in Figure 6 was build following this procedure and contains a mixture of three radionuclides (^{241}Am, ^{60}Co and ^{137}Cs) covering a large gamma-ray energy interval (from 26 to 1332 keV).

Figure 6 Composite steel source produced by the project containing a mixture of three radionuclides





Figures 7 & 8 Spectrum of a cast steel source contaminated with ^{60}Co and spectrum of a cast steel source contaminated with ^{192}Ir (respectively)

Slag is a by-product generated in the production of steel. Whilst the chemical composition of slag is strongly dependent on the production process (electric or conventional furnace), its physical characteristics depend on the cooling process. The difficulties in obtaining homogeneously contaminated material lead project partners to the conclusion that contaminating a well characterised slag sample would produce better results for a reference material. Therefore, four sets of slag sources were produced at CIEMAT by spiking slag samples with aliquots of well calibrated solutions of ^{60}Co, ^{137}Cs, ^{241}Am and ^{226}Ra.

Figure 9 Slag source contaminated with ^{226}Ra in a cylindrical container

Raw slag was collected from a steel foundry in Getafe, close to Madrid, Spain. It was then was dried for 48 hours at room temperature and passed over a 2 mm screen to remove rocks, large chunks of aggregated slag and other components (this is the standard size for reference soil materials). Slag contents of C, H, N and S was determined using elemental analysis. For these major elements and several trace elements it was also analysed using X-ray fluorescence spectrometry and inductively coupled plasma atomic emission analysis. It is well known that all slags exhibit some degree of contamination with natural occurring radionuclides, from the ^{238}U, ^{235}U, ^{232}Th series and ^{40}K, which must be determined before spiking. This was done by IFIN-HH and CIEMAT, and included measurements in an underground laboratory in Romania and several gamma-measuring laboratories from both institutions.

Slag material was spiked using a ^{226}Ra solution previously standardised at CIEMAT by absolute alpha counting as well as with standard solutions of the other three nuclides (^{60}Co, ^{137}Cs and ^{241}Am) also calibrated at the same lab. Slag was then put in cylindrical plastic containers with dimensions adapted to typical control instrumentation at laboratories. A second set of slag contaminated with ^{241}was prepared by PTB from a batch of real contaminated slag. Additional sources of ^{226}Ra with lower specific activities were also produced and measured at the HADES underground laboratory in Belgium as part of ESRMG2.

Figure 10 Raw black slag from a steel mill factory used to prepare reference slag sources

Fume dust reference sources were prepared using material provided by the Siempelkamp Nukleartechnik GmbH melting plant for radioactively contaminated residual substances in Krefeld, Germany The preparation of the fume dust samples and homogeneity testing was done at PTB. In order to achieve a fixed sample geometry the fume dust (50 g) was compressed to 54 % of the initial volume, using 3 inner plastic lids/place-holders in a hermetically closed polyethylene container (see Figure 13). Using this method the reproducibility of the analysis of the fixed samples, in terms of mathematical efficiency calibration, was estimated to be better than 2 %. The total number of fume dust reference samples produced was 13.

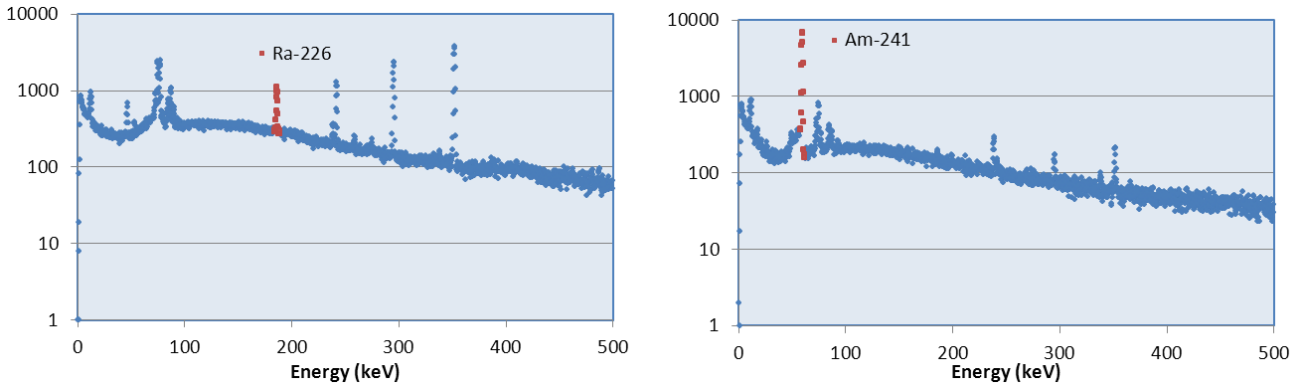


Figure 11 Spectra of slag sources contaminated with ^{226}Ra and ^{241}Am

Although the initial aim was to produce fume dust reference sources contaminated only with ^{137}Cs, a small contamination of ^{60}Co became visible during the measurement process. This therefore allowed the extension of the inter-laboratory comparison to ^{60}Co, present in small amounts (25 times lower activity) in fume dust reference sources. For the elemental composition analysis of the fume dust, X-ray Fluorescence Analysis (XRF) was applied at CIEMAT using pellets of the same material prepared at JRC-IRMM. The pressing of the pellet was applied at 250 kN using Fluxana Vaneox -25 t and 32 mm die and the mass was determined gravimetrically.

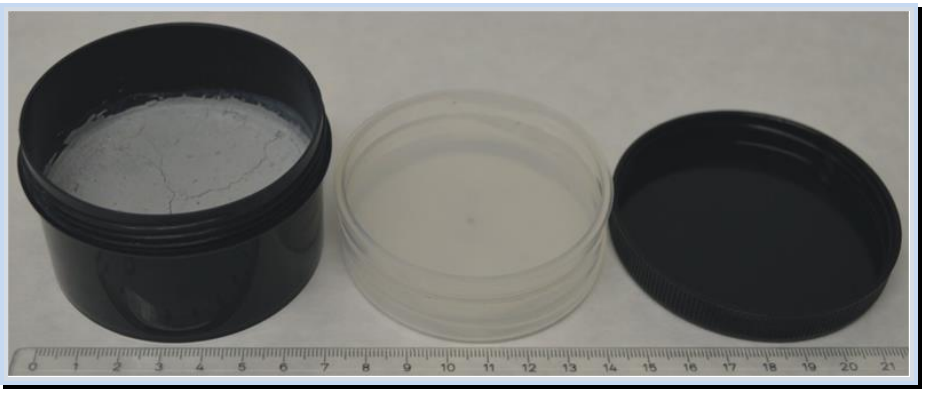


Figure 12 Reference sources of fume dust produced in the project. Internal lids are used in the in the cylindrical container to keep dust under pressure and to maintain a reproducible geometry

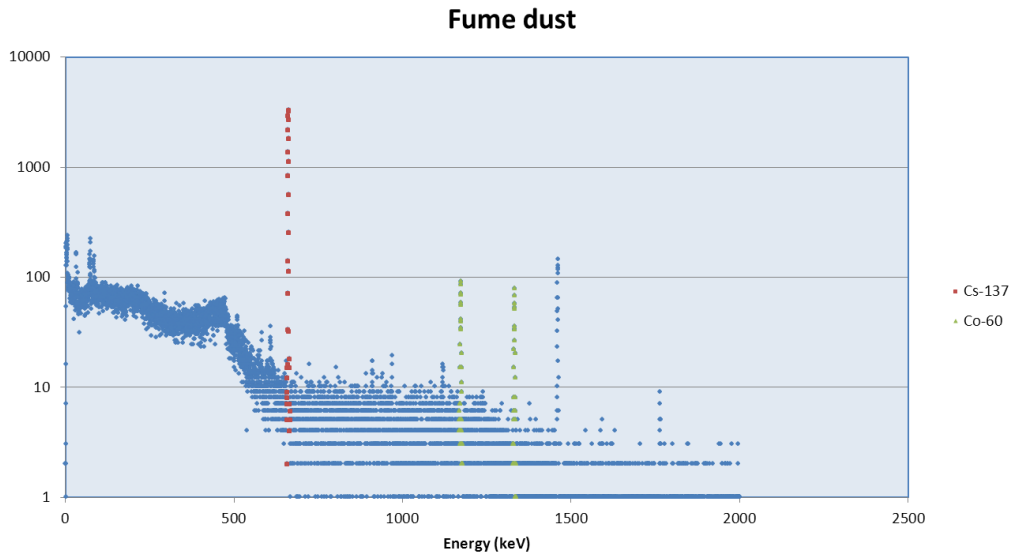


Figure 13 Spectrum of fume dust sources contaminated with ^{137}Cs and ^{60}Co

Conclusions

The project produced a set of reference standards for composite steel, real cast steel, slag and fume dust that went beyond the current state of the art as they conformed to the typical shapes, compositions and radionuclidic contents likely to be found at end-user facilities. The reference standards were certified by the joint work of a large number of National European Metrology Laboratories and are now available for use by end-users. In fact, five steel mills across Europe are already using in them.

3.3 The characterisation of the measurement methods with the reference standards produced previously, using inter-laboratory comparisons and Monte Carlo (MC) simulations

Introduction

The aim of this work was the characterisation of the measurement methods with the reference standards produced previously, using inter-laboratory comparisons and MC simulations to cover the large diversity of sample geometries, shapes, densities and elemental compositions. This objective served as a bridge between the first two objectives and the last two objectives which focussed on the construction and testing of prototype spectrometric devices. Intercomparions from numerical methods and measurements were conducted at the highest metrological level. In partcular, for the the calibration of measurement sources, the procedures established for EURAMET or CCRI(II) comparisons were followed. All partners were involved in this work and the three main areas were:

1. The development and validation of Monte Carlo models of a generic spectrometric device proposed in the first steps of the project
2. The preparation of a procedure for the calculation of true coincidence summing corrections and the calculation of such corrections for the reference materials produced in the project
3. The inter-laboratory comparisons of activity measurements of selected reference materials

Monte Carlo models

Using the recommendations for the basic design of an optimised spectrometric device for measuring the activity of cast steel, slag and fume dust samples developed as part of objective 1, MC models were created by partners in order to determine the typical perfomances that could be expected from the measurement devices and the adequacy of the calculation methods. Each partner chose the MC Package they would use as shown in Table 3.

Table 3 Monte Carlo packages used in the simulation of the measurement setups. Data concerning the cross section librarys are also included

Partner	MC code and version	Photon cross-section library	Electron cross-section library
BEV/PTP	PENELOPE 2011	EPDL	ELSEPA
CEA	PENELOPE 2012	EPDL, XCOM	ELSEPA
CIEMAT	PENELOPE 2012	EPDL, XCOM	ELSEPA
CMI	MCNPX v2.7E	ENDF/B-VI.8	el03
ENEA	GEANT v3.21	standard GEANT3	standard GEANT3
IFIN-HH	GESPECOR v4.2	XCOM	GESPECOR
IST	MCNPX v2.7.0	ENDF/B-VI.8	el03
	MCNP-CP v3.2	ENSDF2	el03
JRC	EGSnrc v4-r2.4.0	XCOM	BH
NCBJ	PENELOPE 2012	EPDL	ELSEPA
PTB	GESPECOR v4.2	based on XCOM	GESPECOR
SMU	MCNPX v2.4	mcplib02	el03
STUK	VGSL v1.2	ENDF/B-VI.8	el1

The validation of the MC models was performed through an inter-comparison of results using a common detector model, whose dimensions and composition was agreed by all partners. Inter-comparison

calculations were carried out by all partners and the results were evaluated utilising the Mandel-Paule weighted mean method. The partners performed MC simulations of full-energy peak and total detection efficiencies for the three different reference sources (i.e. cast steel, slag and fume dust) developed as part of objective 2, as well as gamma ray energies corresponding to different key radionuclides found in the metallurgical industry. The calculations were done for the same geometry in different MC codes versions. In general, the values for the full-energy peak detection efficiencies and total detection efficiencies were within ±0.5 % and ±1.0 % of the mean value, respectively, which is well within the limits of uncertainty for cross section tables (roughly 2 %). However, several results deviated outside the limits. The discrepancies were later clarified and were found to be due to limitations of the MC codes in geometry modelling or specific normalisation of the results**.** A typical chart showing the results of the intercomparison is shown in Figure 14.

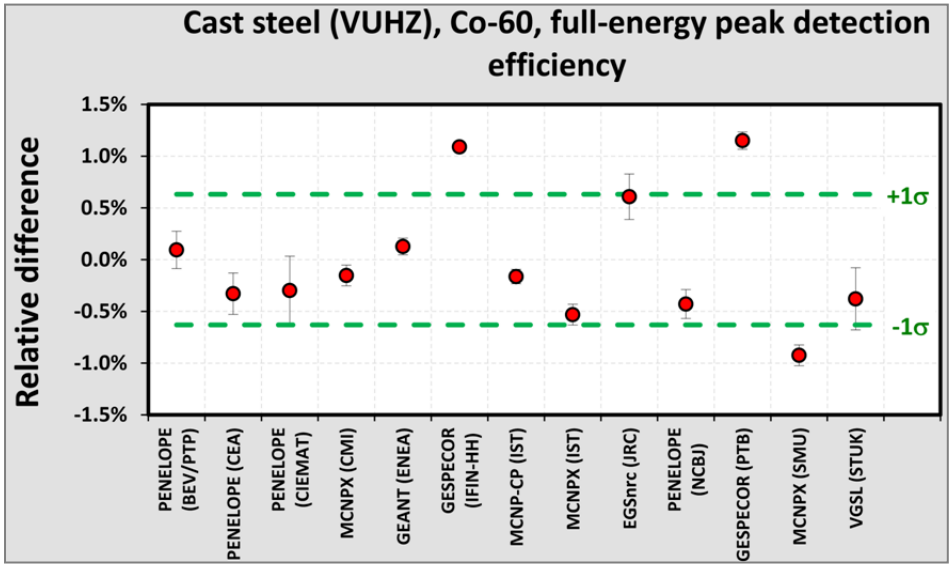


Figure 14 Results of the intercomparison of detection efficiency calculations with different Monte Carlo packages

The MC models created by the project have been made available for free download by end-users on the project's website. A subset of the MC models are shown in Figure 15

Figure 15 Graphical representation of the 5 Monte Carlo models of the Ge detector developed in this project. each one corresponding to a simulation package

Procedures for coincidence-summing corrections

True coincidence summing is caused by the coincident detection of photons (generated in a cascade during radionuclide decay) whose contributions can not be recorded separately as they correspond to a simultaneous event from the point of emission. The effect of coincidence summing must be taken into account when measuring radionuclide activity and its intensity depends on the measurement method (i.e. type of a detector), sample geometry (i.e. distance from the detector, sample shape) and self-absorption (i.e. density and thickness). The coincidence summing effect is significant especially in the close geometries used to measure samples at steel mills. In addition, in the metallurgical industry, true coincidence summing correction needs to be applied for any potential contamination with some artificial radionuclides (e.g. ^{60}Co, ^{192}Ir) and natural radionuclides (e.g. ^{214}Pb, ^{208}Tl).

True coincidence summing factors can reach up to more than 30 % in the case of a source of cast steel contaminated with ^{192}Ir. Values reported by the project partners varied in most cases within ±1.0 % around the mean value. Larger deviations found for some partners might be caused by implementation of the decay schemes in the given MC code. The next table shows typical correction factors for nuclides and matrices of interest.

Table 4 Typical coincidence-summing correction factors in the efficiency calibration of nuclides and matrices

Nuclide	Co-60		Ir-192		Bi-214	
Gamma-ray Energy (MeV)	1.17	1.33	0.316	0.468	0.609	1.24
Source matrix						
Cast steel	1.1759(26)	1.1835(21)	1.3238(13)	1.2368(11)	1.2090(23)	1.2135(27)
Slag	1.1062(19)	1.1118(14)	1.2020(25)	1.1527(16)	1.1277(29)	1.1300(22)
Fume dust	1.0955(23)	1.1018(17)	1.1813(20)	1.1391(19)	1.1188(32)	1.1170(22)

Inter-laboratory comparisons of the measurement of reference sources

The aim of the inter-laboratory comparisions (ILCs) was to test the proposed methods for measurement of radioactivity in matrices used in metallurgical industry and in particular for the three different reference sources (i.e. cast steel, slag and fume dust) developed as part of objective 2. Four ILCs were conducted one for each of the three sources plus one additional ILC for a second batch of cast steel sources:

1. **ILC_1A**: One set of standards of cast steel (from Spielemkamp origin) containing the radionuclide ^{60}Co with and activity concentration of about 1 Bq/g.
2. **ILC_1B**: One second set of standards of cast steel (VUHZ), containing the radionuclide ^{60}Co with and activity concentration of about 1 Bq/g.
3. **ILC_2**: One set of standards of slag (from CIEMAT), contaminated (spiked) with the radionuclide ^{226}Ra and activity concentration of about 10 Bq/g.
4. **ILC_3** One set of standards of fume dust (from Siempelkamp), contaminated with the radionuclide ^{137}Cs and activity concentration of about 10 Bq/g.

A total of 13 partners participated in the inter-comparisons and their contributions are shown in Table 5 (anonymity of the laboratories is not preserved as this was not a requirement of this ILC programme). However, the partner who produced the reference standard did not participate in the corresponding ILC. The ILCs were coordinated and evaluated by JRC with the help of all partners and the individual laboratory‘s performance was expressed in terms of relative deviations and En numbers.

The results of all ILC_1A, ILC_1B, ILC_2 and of ILC_3 were used to derive the reference activity concentrations of the radionuclides in question. In this way, a set of reference sources with the highest degree of metrological characterisation could be obtained.

No	partner	ILC_1A	ILC_1B	ILC_2	ILC_1A
1	CIEMAT	×	×	*	×
2	BEV/PTP	×	×		×
3	CEA	×	×	×	×
4	CMI	×	×	×	×
5	ENEA	×	×	×	×
6	IFIN-HH	×	×	×	×
7	IJS	×	×	×	×
8	ITN	×	×	×	×
9	JRC	×	×	×	×
10	POLATOM	×	×	×	×
11	PTB	×	×	×	×
12	SMU	×	×	-	-
13	STUK	×	×	×	×
Total number of participants		13	13	11	12
Number of participants reporting results		13	13	10	12

Table 5 Participation in the inter-laboratory comparison exercises. *CIEMAT prepared and characterised the material for this inter-laboratory comparison

All activity concentrations were reported in Bq/g with the associated combined standard uncertainty. This was done via EXCEL reporting templates prepared and distributed by JRC. The worksheets in these files were based on those developed at BIPM CCRI(II) for key comparisons, modified to handle the specific needs of each ILC. The reporting files also served as questionnaires and participants were asked to answer relevant questions regarding their measurement systems and the procedures that they used as well as to provide references to their data sources to facilitate evaluation of the ILCs results and ensure traceability of activity determination. As JRC participated in all four ILCs the independency of their results was ensured by JRC reporting their results first to an independent person in the partner´s institute responsible for samples preparation or by an independent person at JRC.

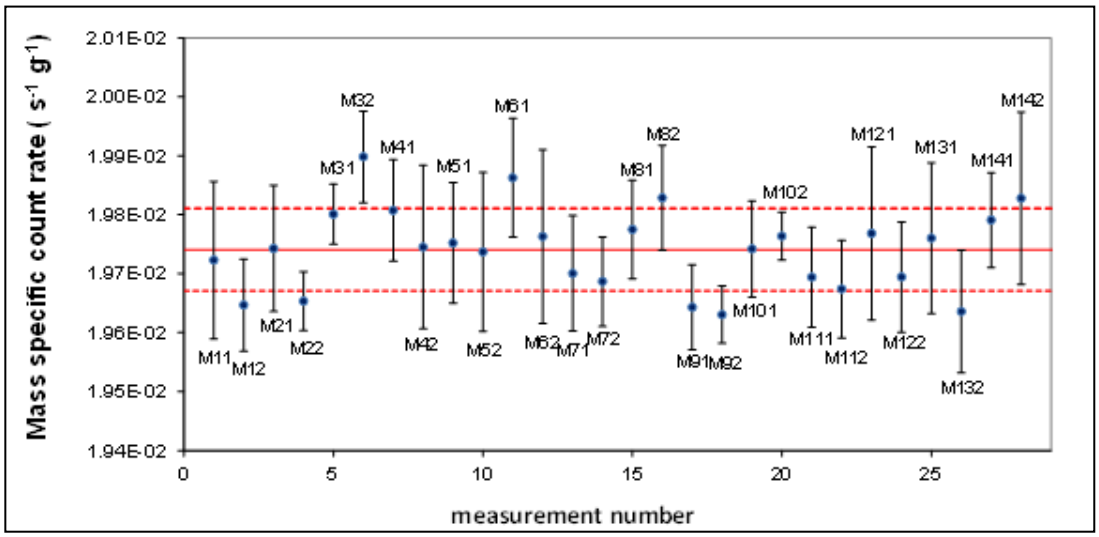
ILC_1A *and* **ILC_1B** *cast steel sources*

Two batches of cast steel samples (produced as part of objective 2) from different origins were used in the ILC1A and **ILC_1B**: one series of samples were provided by PTB and represented real contaminated material originating from a waste management facility (Siempelkamp); the other one was provided by SMU and was produced by a metallurgical company (VUHZ) by adding ^{60}Co to cast steel. The ILC was divided in two stages ILC_1A and ILC_1B corresponding to the Siempelkamp and the VUHZ cast steel batches, respectively.

The technical protocol for ILC1A and **ILC_1B** included sample information, instructions and relevant dates. Participants were instructed to analyse the sample as provided using gamma ray spectrometry and report the activity concentrations for ^{60}Co. The elemental composition of raw materials of the batches was provided in one case (ILC1B) by the partner responsible for preparation of the samples and in the other set determined by CIEMAT using elemental and XRF analysis. This information was used as a common basis for the participants conducting MC simulations for relative or absolute efficiency calibration of their gamma ray spectrometry systems.

The homogeneity testing of the first and second batches of cast steel samples was conducted by PTB and JRC, respectively. It was performed on the basis of reproducible measurements of the samples using the same Ge detector in the respective partners‘ institute. The count rates of the samples normalised over the sample mass were compared and the standard deviation of the mean was taken to represent the between samples in-homogeneity. In particular the ILC_1B samples were measured in normal and upside down orientation. The results are shown in Figure 16 and it should be noted that, for the case of ILC_1B cast steel

samples, the derived standard deviation between samples from the one way ANOVA on the two groups of data, for the different measurement orientations, was 0.21 %. Uncertainty due to instability was disregarded because this type of material was considered mechanically strong and no changes with time or during shipment were expected.



Fighure 16 Homogeneity test measurements of the VUHZ cast steel samples used in the ILC_1B exercise

ILC_2 *Slag with ^{226}Ra*

Homogeneity testing of the slag samples was conducted at CIEMAT by gamma ray spectrometry. The samples were shaken before measurements were made, as they were composed of particles of varying sizes that could slide down if left unmoved for some time. The mean (from 4 measurements each of counting time of 2500 s) net counts in the 186 keV peak ^{226}Ra, peak are shown Figure 17 for each sample. They were measured on the same position on the CIEMAT Ge detector and the standard deviation of the data was 1.1 %. The latter was considered as the uncertainty component for homogeneity and was introduced to the activity concentration in the spiked slag material.

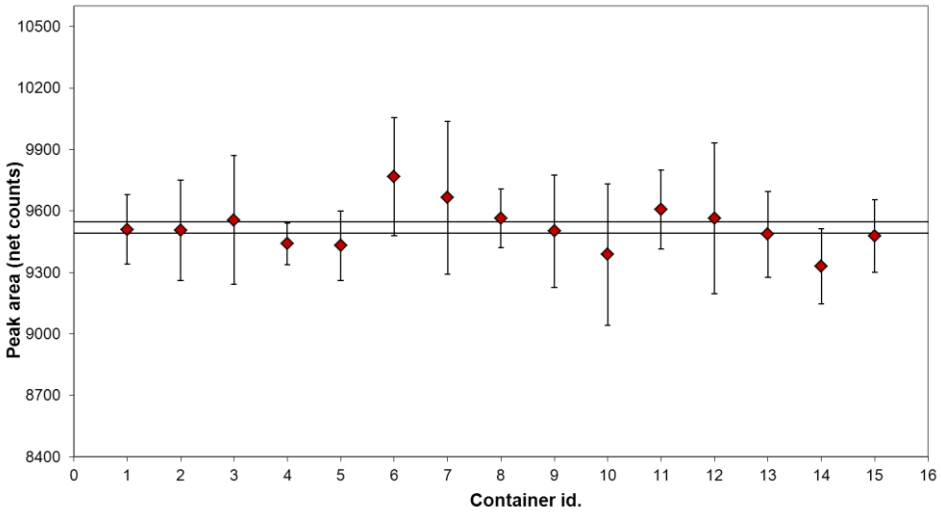


Figure 17 The mean net counts in the 186 keV peak of ^{226}Ra for each slag sample

The activity content in the raw slag was analysed using different techniques, from which the weighted mean value of (14.0 ± 0.6) Bq/kg was determined for the concentration of natural ^{226}Ra radioactivity. By multiplying this value with the mean mass of the ILC_2 samples the mean natural ^{226}Ra radioactivity in the ILC_2 samples, was calculatedto be (2.25 ± 0.10) Bq. In the estimation of the combined standard uncertainty of the latter the uncertainty components of a) the activity concentration and b) the mass (taken as the stdev of the mass values of the 12 ILC_2 samples) were included. The ILC_2 technical protocol provided the necessary information on the samples including a proposed elemental (major elements) composition by CIEMAT (producer). CIEMAT's relevant report was also provided to ILC participants.

ILC_3 *Fume dust with ^{137}C and ^{60}Co*

The homogeneity testing of fume dust sources was completed by relative gamma ray spectrometry measurements of all samples, under the same measurement conditions and position using the PTB gamma ray spectrometry detector. The results of the count rates, normalised to sample mass, for the peaks of ^{137}Cs and ^{60}Co are shown Figure 18, in which homogeneity between samples is demonstrated with respect to the uncertainties of the measurements. The standard deviations of the results for ^{137}Cs and ^{60}Co, of 0.35 % and 3.74 %, respectively, were taken as homogeneity indicators.

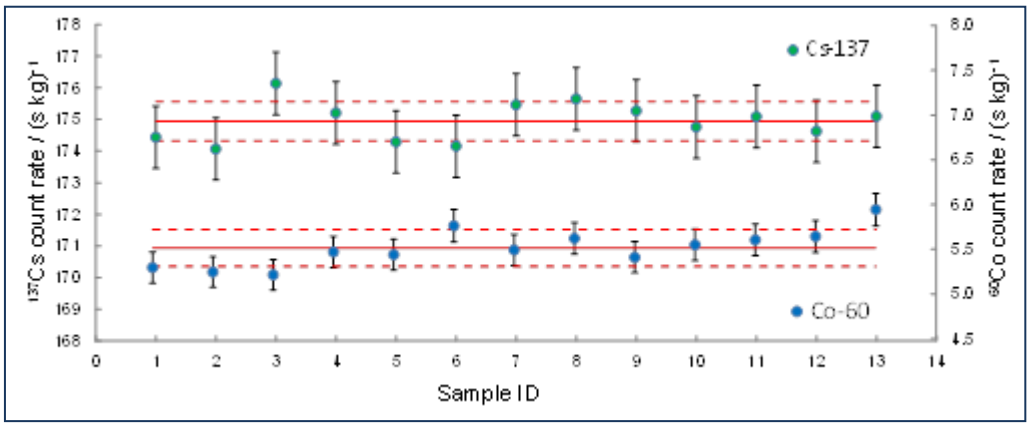


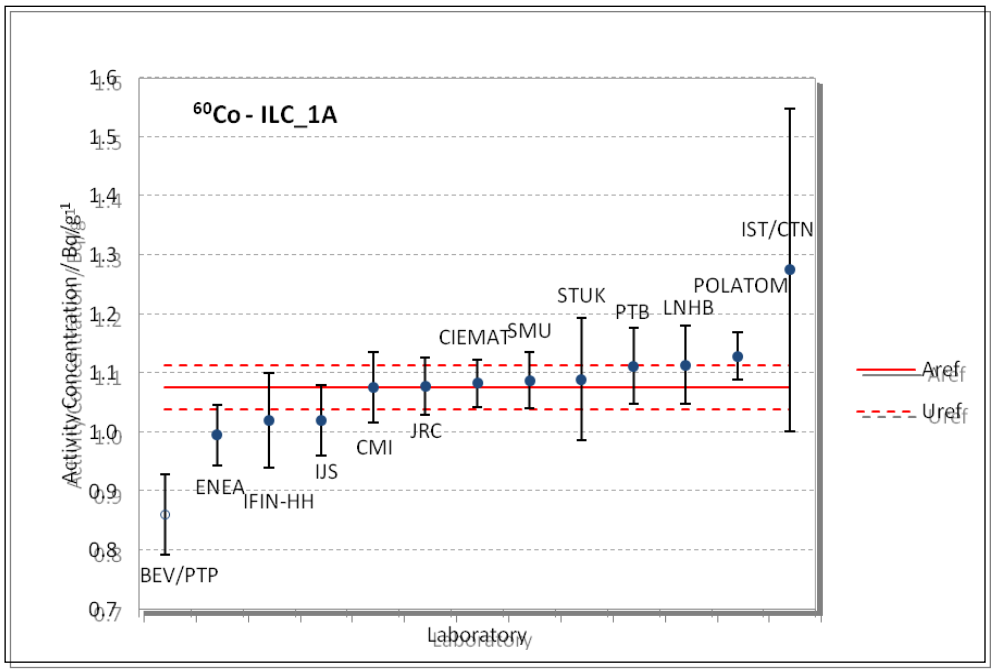
Figure 18 Homogeneity test measurements of the fume dust Cs-37 samples from the ILC_3 exercise

ILC analysis methods and results

The participants in the ILCs analysed their samples by HPGe based gamma-ray spectrometry (GS) as recommended by the ILCs technical protocols. In each participant laboratory the measurements were performed using one or more HPGe detectors and in some cases low level gamma spectrometry (LLGS). Different efficiency calibration methods (experimental, MC and methods that combine both techniques) were used. In total 7 different MC codes and 2 numerical codes were used to calculate the detection efficiencies and true coincidence summing corrections, where applicable. Two partners applied absolute efficiency calibration using their validated MC detector models and the remaining partners applied efficiency corrections to the experimental efficiencies for standard sources (i.e. the efficiency transfer approach). The distances from the detector end-cap to the bottom of the sample ranged between partners from in-contact to 17 cm. Nuclear decay data was also obtained from Monographie BIPM-5, which is identical to the data evaluated by the international collaboration DDEP (Decay Data Evaluation Project)

Results from ILC1 on cast steel sources are shown in Figure 19 and uncertainties estimated by partners were given for k=2. The consensus values A_{ref} are represented by red lines. They were calculated as the power-moderated means (pmm) of the reported activity concentrations excluding outlier results. The uncertainty of the consensus value, u_{ref}, was derived from the uncertainty from characterisation, u_{char}, and the uncertainty due to inhomogeneity, u_{hom}, using the formula:

$$u_{ref} = \sqrt{u_{char}^2 + u_{\mathrm{hom}}^2}$$



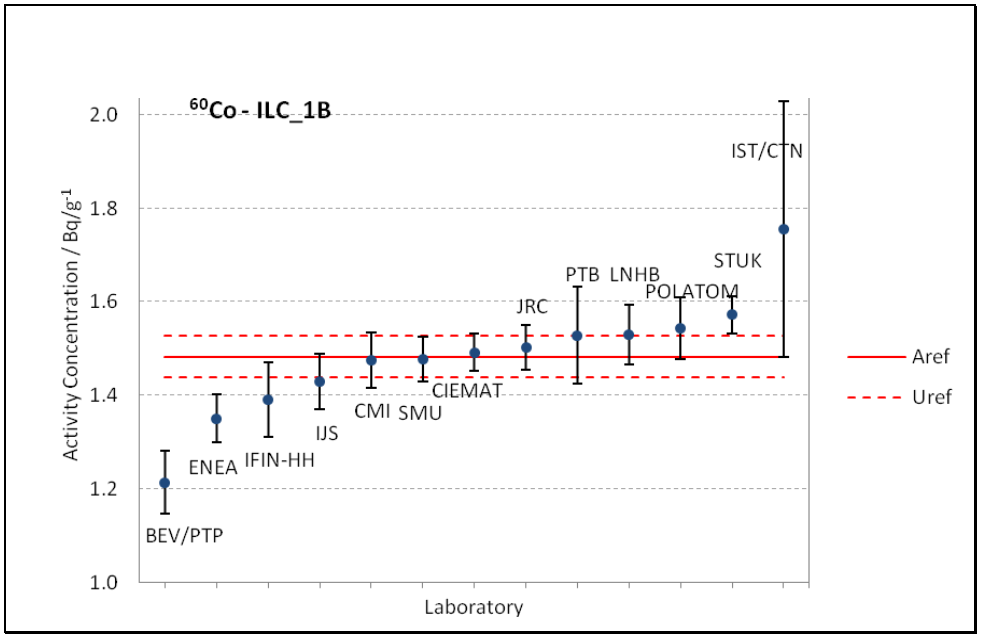


Figure 19 Laboratory results for ^{60}Co activity concentration in Siempelkamp and VUHZ cast steel samples. Error bars represent U_{lab}, calculated from the reported combined standard uncertainties, for *k* = 2 and red lines the $A_{ref} \pm U_{ref}$ (*k* = 2).

Most laboratories obtained satisfactory results for ^{60}Co in ILC_1A and ILC_B; nevertheless 2 (15 %) of each of ILC_1A and ILC_1B results deviated more than 10 % from the reference values One of these two partners, was BEV/PTP who reported lower than the reference values (by -20 % and -18 % for ILC_1A and ILC_1B, respectively), due to technical problems with their detector (resulting in count rate loses). The technical issues of BEV/PTP were reported prior to the disclosure of their ILC results. An overall comparison between the results of ILC_1A and ILC_B revealed a similar pattern due to the fact that the same radionuclide was determined in samples of similar dimensions and matrix and the same calibration was used by BEV/PTP for the analysis of the two cast steel samples.

The results of the measurements of slag sources from ILC_2 are shown in Figure 20. From the 10 reported results for ILC_2, 6 deviated by less than 5 % from the reference value, 2 in the range between 5 % and 10 % and 2 more than 10 %.

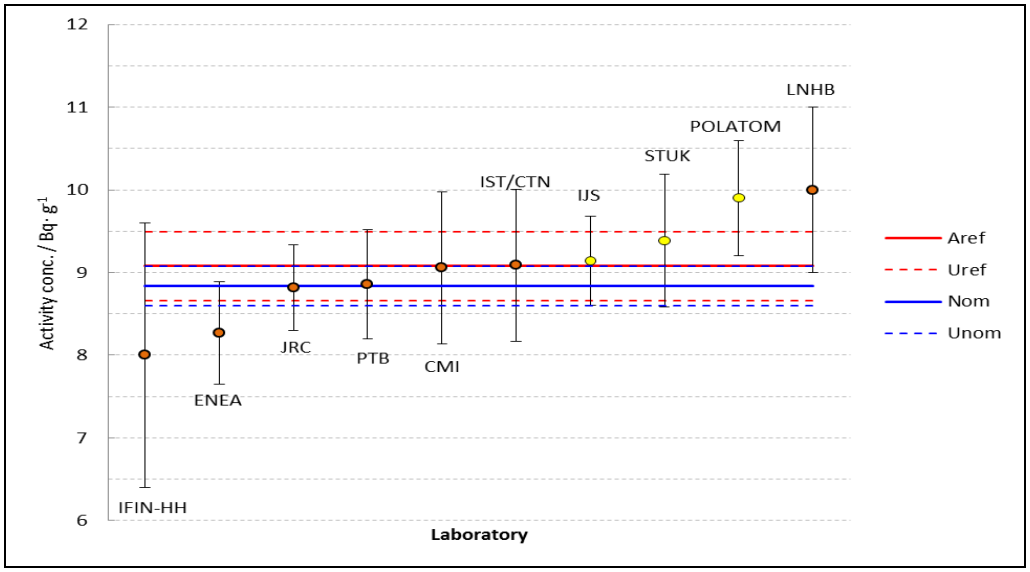


Figure 20 Laboratory results for ^{226}Ra activity concentration in CIEMAT H-Slag (orange and yellow data points represent lab results based on analysis of original and own sample geometries, respectively). Error bars are calculated from the reported combined standard uncertainties, for k = 2..The consensus value A_{ref} is calculated from all reported values. The blue solid line indicates the mean nominal activity concentration, and the dashed ones its uncertainty (k=2 including inhomogeneity).

The measurements of ^{137}Cs activity concentrations in fume dust samples were analysed and reported by partners in ILC_3 in two phases. In the second phase, some partners included several methods to account for the container curvature which in turn biased the geometry of the source. Results indicated that 10 results deviated less than 5 % from the reference value, 1 slightly more than 5 % and 1 by >10 % as shown in Figure 21. Similar results but with higher uncertainties were obtained in the measurement of the minor ^{60}Co contamination found in the fume dust samples.

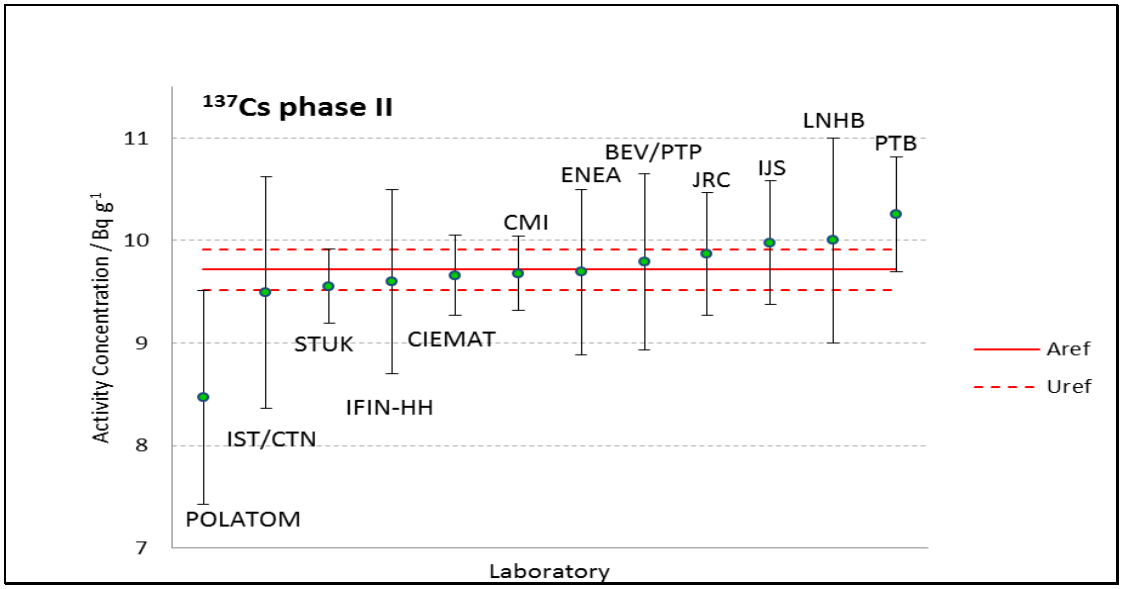


Figure 21 ILC-3 results reported for ^{137}Cs activity concentration in fume dust (ref. date 1 June 2013) in Phase II of ILC_3. Error bars represent U_{lab}, calculated from the reported combined standard uncertainties, for *k* = 2 and red lines the $A_{ref} \pm U_{ref}$ (*k* = 2). The consensus value A_{ref} is calculated from all reported values.

In addition to the materials already described in ILC1-3, slag sources contaminated with low levels of ^{226}Ra were prepared at CIEMAT and measured at JRC and at the underground laboratory HADES in Belgium as part of ESRMG2 HADES is one of the few laboratories in Europe placed deep under the ground, thus reducing the radioactive background and allowing the measurement of very low level activities. Measurements for the slag sources contaminated with low levels of ^{226}Ra were confirmed with the data obtained at CIEMAT and JRC.

Conclusions

The aim of the ILCs was not to test the proficiency of the partners, but the development of a reference procedure and standards for radioactivity monitoring in metal foundries. The ILC results demonstrated the validity of the proposed methods for the measurement of ^{60}Co in cast steel, ^{226}Ra in slag, as well as ^{137}Cs and ^{60}Co in fume dust matrices. The validated MC detector models also proved to be an important tool in this process. The ILC results were further used to determine the reference activity concentrations in the developed standards.

In conclusion, the cooperation of the project partners allowed the characterisation and validation of the reference sources (developed in section 3.2), according to standard procedures following CCRI(II) and EURAMET comparisons; thus producing a set of results with the highest metrological significance.

3.4 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

Introduction

The aim of this work was 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. This part of the work benefits from the developments carried out in sections 3.2 and 3.3

Instrument design and testing

Having completed the characterisation of the reference materials (i.e. cast steel, fume dust and slag) with the ILCs in section 3.3. and produced a basic for an optimised spectrometric device in section 3,1, the next step was the construction of two prototype spectrometric devices for the measurement of activity in cast steel, fume dust and slag samples at CIEMAT and PTB. Approximate efficiency data was calculated with the simplified MC models created in section 3.3 as well as experience gained in the production and characterisation of reference materials. This was then used to establish the basic features of the prototype spectrometric devices in order to match the requirements in terms of detectable activities and counting times. The requirements were:

- A measurement system based on a High Purity Ge detector able to measure photons in the energy interval from 10 to 2000 keV.
- Relative detection efficiencies between 35 % and 40 %
- Portability: including an electrical cooling system for the detector using pulse-tube cooler technology.
- Cylindrical shielding (made of lead) surrounding the Ge detector and with a portable size and weight
- A measurement chamber made in a low gamma-ray attenuating material that holds the source to be measured.
- Digital electronics providing high voltage bias and multichannel analyser capacity.

In order to accurately determine the final design of the prototype spectrometric devices for the measurement of activity in cast steel, fume dust and slag samples, detailed numerical models of the detector-shield-source ensemble were created. The simulations were carried out using two different MC models/codes to model the system, including with typical samples of cast steel, slag and fume dust. Full-energy peak and total detection efficiencies as well as true coincidence summing correction factors were calculated for a combination of radionuclides of interest in the metallurgical industry and with typical model probes similar to those in use at end-user facilities.

At CIEMAT calculations were based on the general-purpose MC code PENELOPE and the PENNUC tool. The latter implements the simulation of the decay scheme of the nuclide under consideration, thus avoiding the need to calculate separate coincidence-summing factors. Figure 22 shows a visual model of the prototype spectrometric device including a typical steel source. The detector element was a 35 % relative efficiency HPGe detector

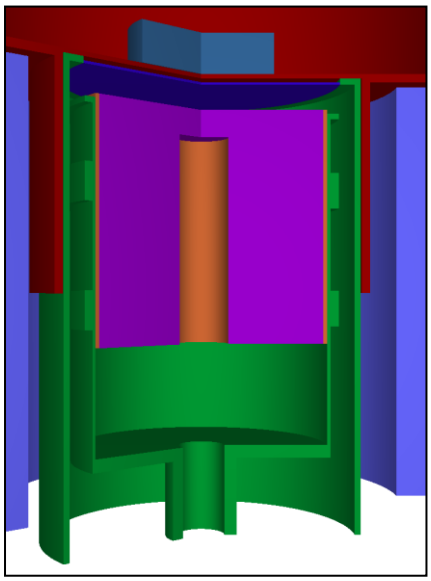
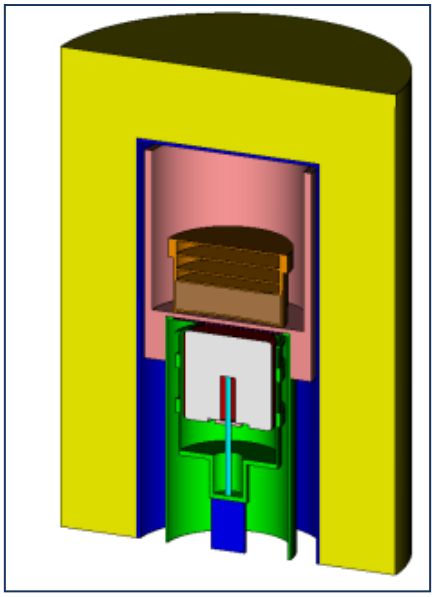


Figure 22 a visual model of the CIEMAT prototype spectrometric device including a typical steel source



At PTB, a similar detector model was created using the MCNPX MC simulation package. Figure 23 shows a typical setup with a fume dust sample in a plastic container inside the measurement chamber of the prototype spectrometric device, on top of the Ge detector, whose relative efficiency was about 40 %

The prototype spectrometric devices

Data obtained in the MC simulations confirmed that the final design of both prototype spectrometric devices fulfilled the requirements established. The main characteristics of both spectrometric devices (produced as CIEMAT and PTB) are presented in Table 6.

Figure 23 a visual model of the PTB prototype spectrometric device including a typical fume dust source

	PTB	CIEMAT
Detector type	Extended-range coaxial HPGe	
Energy range	5 keV – 10 MeV	
Entrance window	Carbon epoxy 0.6 mm	
Portable cooling unit	Pulse-tube technology (Stirling-cycle cooler)	
Relative detection efficiency	~ 40%	~ 35%
Portable shielding	50 mm Pb, 2 mm Cu,	50 mm Pb
Measurement chamber material	PMMA	Teflon
Electronics	DSA 1000	DSA-LX
Software	Genie 2000/InterWinner Specific software embedded	Genie 2000 Specific software (EXCEL)

Table 6 The main characteristics of the CIEMAT and PTB spectrometric devices

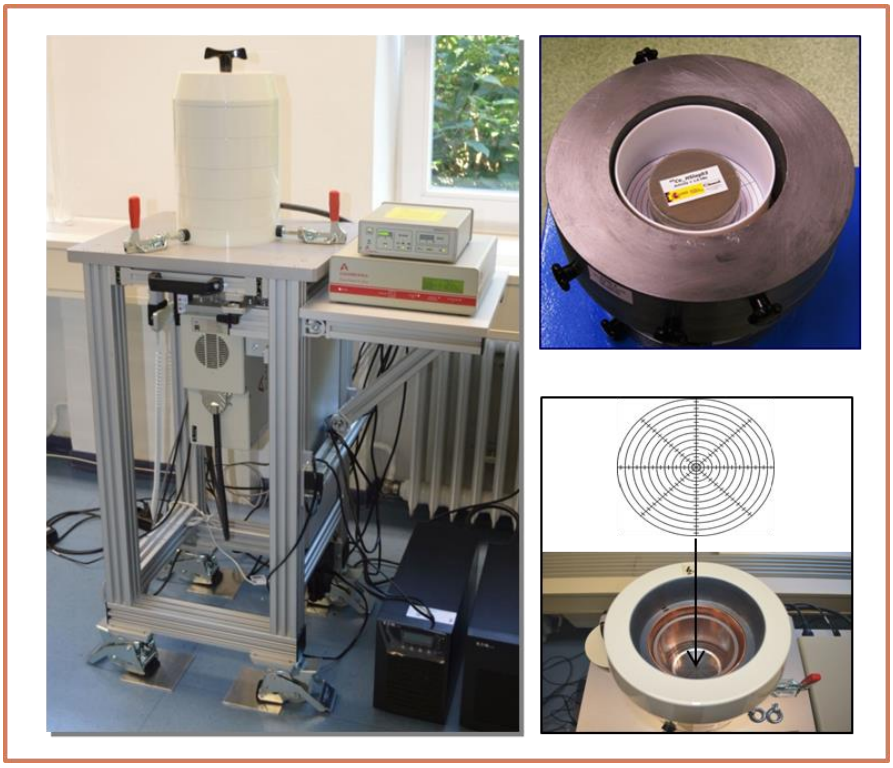


Figure 24 The prototype spectrometric device built at PTB and the measurement chambers of the CIEMAT (upper) and PTB (lower) prototype spectrometric devices

Two prototype spectrometric devices were built at CIEMAT and PTB, then were tested and characterised at both laboratories using the cast steel, fume dust and slag reference materials produced in section 3.2 and characterised in the ILCs in section 3.3. Validation of the prototype spectrometric devices was carried out by a series of measurements and calculations in which calculated and experimental values were compared. Table 7 presents the results of one of the comparisons carried out at CIEMAT in terms of measured and simulated full-energy peak detection efficiencies for each cast steel source. Relative differences between measured and simulated efficiencies varied around 1 %, except for some emissions of ^{192}Ir with energies of 296 and 316 keV for which relative differences were a little higher.

Table 7 Comparison of measured and simulated full-energy peak detection efficiencies for cast steel samples

Source	Radionuclide	Energy (keV)	Experimental Efficiency ($\varepsilon \cdot P\gamma$)	u (%)	Simulated Efficiency ($\varepsilon \cdot P\gamma$)	u (%)	Relative Difference MC/EXP -1 (%)
PTB3	Co-60	1173.23	0.0187	1.6	0.0188	1.0	0.4 ± 1.9
		1332.49	0.0168	1.6	0.0170	1.1	1.2 ± 2.0
VUHZ1	Co-60	1173.23	0.0189	1.6	0.0185	1.1	1.7 ± 2.0
		1332.49	0.0166	1.6	0.0168	1.1	1.5 ± 2.0
PTB4	Co-60	1173.23	0.0197	1.6	0.0199	1.0	0.8 ± 1.9
		1332.49	0.0179	1.6	0.0179	1.1	-0.1 ± 1.9
VUHZ4	Ir-192	295.96	0.0126	2.5	0.0121	0.9	-4.0 ± 2.5
		308.46	0.0126	2.5	0.0125	0.9	-0.5 ± 2.6
		316.51	0.0383	1.8	0.0375	0.5	-2.1 ± 1.9
		468.07	0.0170	2.1	0.0169	0.8	-0.7 ± 2.2

Similar results were obtained at CIEMAT and PTB for the comparisons between calculated and experimental efficiencies from sources of slag and fume dust. Although standard acquisition and control software was provided by the equipment manufacturer, specific code was developed to evaluate the critical aspect of the prototype spectrometric device: i.e. the detection limits, also known formerly as Minimum Detectable Activity (MDA). The determination of the instrument's characteristic limits in the detection of low levels of activity was done following the recent ISO11929 standard. The ISO11929 standard integrates systematic uncertainties into calculation and MDA calculation was done at CIEMAT on their prototype spectrometric device with specific software developed by ENEA in an EXCEL file, while algorithms were implemented at PTB on their prototype spectrometric device directly into the control software.

Typical values obtained for both prototype spectrometric devices are presented in Table 8 for several counting times. In order to enable fast screening, monitoring instruments at end-user facilities usually operate with 60 s counting time, whilst for more accurate results longer measurement times are needed. Values presented in Table 8 indicate that the system can operate in short counting times (60 s) and that the implementation of lower activity levels would require increasing counting times

Source			Counting time (s)	MDA (Bq/g)	Source			Counting time (s)	MDA (Bq/g)
Material	Mass (g)	Radionuclide			Material	Mass (g)	Radionuclide		
Cast steel	71	Co-60	60	0,19	Cast steel	74	Ir-192	60	0,13
			180	0,11				180	0,08
			300	0,07				300	0,06
			600	0,05				600	0,04
Fume dust	50,2	Cs-137	60	0,17	Black slag	160	Cs-137	60	0,21
			180	0,09				180	0,09
			300	0,08				300	0,06
			600	0,05				600	0,04
Black slag	160	Ra-226	60	1,62	Black slag	160	Am-241	60	0,21
			180	0,87				180	0,12
			300	0,67				300	0,09
			600	0,46				600	0,06

Table 8 Typical MDA values characteristics of the detector-shielding ensemble

One of the most relevant features of the new prototype spectrometric devices was their ability to discriminate between many possible contaminants in the same source and to individualise the results. Figure 25 presents a screen plot of a spectrum from a radioactuve source with multiplicity of components that can be identified and quantified in a single measurement

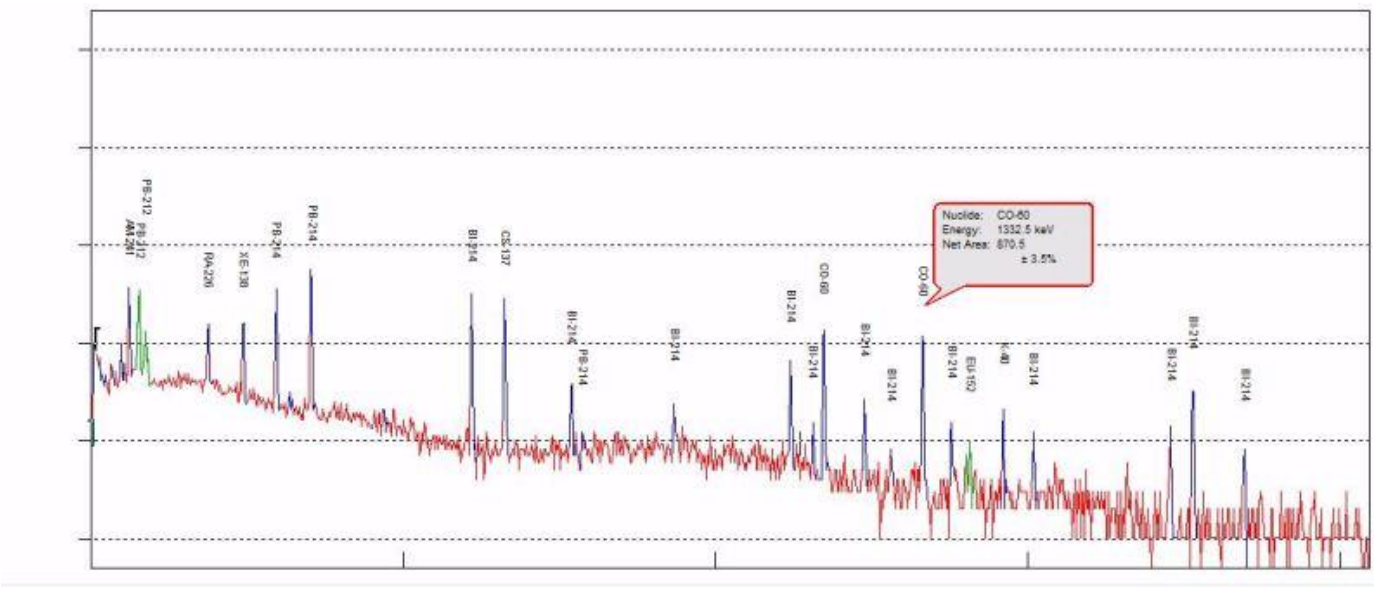


Figure 25 Spectrum of a mixed source showing the identification performance of the HPGe-based prototype spectrometric devices

In addition to the two Ge-based devices, an alternative approach was also explored at CIEMAT based on a $CeBr_3$ scintillation detector. Cerium bromide detectors are being proposed as an alternative to NaI(Tl) detectors, because they exhibit an energy resolution significantly better for energies over 100 keV and also have good counting efficiencies. The CIEMAT $CeBr_3$ experimental setup is based on a cylindrical detector with 75 mm diameter x 75 mm height. The detector was placed inside a cylindrical lead shield, 5 cm thick. The experimental setup was completed by a preamplifier and a digital Multichannel Analyzer. Reference sources produced in the project were measured with this system and the detection limits determined as a part of the ESRMG1 work. Results indicated that the detection limits are similar to those obtained with the Ge prototypes, although the energy resolution is much worse. On the other hand, scintillation detectors do not require the specific cooling systems needed to operate Ge detectors, which makes them suitable for applications were energy resolution is not critical.

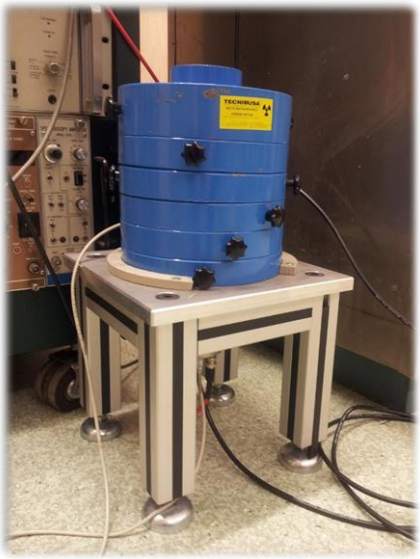


Figure 26 Testing of a prototype based on a $CeBr_3$ detector at CIEMAT

Conclusions

Comprehensive laboratory testing was completed at CIEMAT and PTB on the all three of the prototype spectrometric devices. As part of this, the prototypes were calibrated using the reference sources developed and validated in sections 3.2 and 3.3 and the detection limits of the prototype spectrometric devices were determined. The results showed that, when operating in laboratory conditions, the performance of the prototype spectrometric devices match the target values established in section 3.1 (objective 1) in terms of the detection limits achievable in short counting times (of approximately 1 s).

3.5 Evaluation and demonstration of the prototypes at end-user facilities

Introduction

The aims of this work were:

1. 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 and
2. Demonstration of the prototype spectrometric devices at selected foundries in Europe

Validation and demonstration of the prototype spectrometric devices *at end-user facilities*

Validation of the prototype spectrometric devices at end-user facilities was performed in order to verify the performance of the spectrometric prototypes in industrial conditions. At the same time, a demonstration was performed to the staff at the end-users in order to promote the uptake of the devices. The critical parameters of the prototype spectrometric devices were checked in situ at the end-user facilities and the system reliability was assayed by measuring reference sources with known activity of target radionuclides. The detection limits (MDA values) of the prototype spectrometric devices were then determined to verify that the values obtained were below the clearance levels (1 Bq/g for most nuclides, 10 Bq/g for ^{226}Ra), which is a critical proof of the spectrometer performance.

Selection of steel mills for demonstration

An initial survey indicated the willingness of end-user facilities (steel mills) in seven European countries (Austria, Czech Republic, Portugal, Romania, Slovakia, Slovenia and Spain) to host the scheduled tests in industrial environments at convenient dates. A detailed questionnaire was sent to the candidate facilities to understand the types and sizes of the probes at the steel mills to check the radioactivity contents. This was an essential requirement as in most cases the types and sizes of the probes presented small differences with reference sources and therefore correction factors had to be calculated.

The final selection of the end-user facilities was done based on the end-users capability to provide an appropriate environment, a convenient location for the measurement equipment and the willingness to prepare in advance some material for testing in accordance with standard procedures. The distance from CIEMAT and PTB as well as the time window available for demonstration both by the testing team and the factory staff was also considered. The requirement to assure the participation of end-users from several European countries was also an important additional criterion. Although only two evaluations/demonstrations were initially planned it was decided to perform a third in order to collect additional information and as a safeguard in case of test failure. Demonstrations were performed at the end-user facilities listed in Table 9 whose approximate geographical situation is shown in the map in Figure 27.

Table 9 Final list of end-user facilities selected for demonstration

Country	End-user	Prototype	Date	Testing team
Portugal	Siderurgia Nacional Seixal	CIEMAT	Aug. 2014	CIEMAT, IST-ITN, ENEA, Local staff
Spain	Gerdau Aceros Especiales Europa	CIEMAT	Sept. 2014	CIEMAT, Local staff
Czech Republic	Vítkovice Testing Center S.r.o.	PTB	Oct. 2014	CMI, PTB, Local Staff

Figure 27 Location of the 3 end-user facilities selected for demonstration

In all end-user tests, the local staff cooperated closely with project partners and prepared in advance a set of samples to be assayed with the prototype spectrometric devices. Tests were carried out in two steps: first, a number of reference samples were measured then, measurement of sources obtained on-site, was performed.

Figure 28 The CIEMAT prototype spectrometric devices in place at the laboratory of a steel mill

Reference and end-user sources

The specification of the standard sources measured by the CIEMAT and PTB prototype spectrometric devices is presented in Table 10.

Table 10 Specification of standard sources used for validation with CIEMAT and PTB prototypes

Reference standard sources used with the CIEMAT system					
Description	Material	Weight (g)	Density(g/cm^3)	height (cm)	Radius (cm)
Steel PTB3	cast steel	71.2	7.88	0.98	1.74
Steel PTB4		85.3		2.25	0.7
VUHZ4		74.4		0.998	1.75
LSlag01	slag	160.0	2.185	2.049	3.6
LSlag03		160.6			
HSlag07		160.6			
Co HSlag01		160.0			
Cs HSlag01		160.0			
Fume02	fume dust	50.2	0.707	1.87	3.475
Reference standard sources used with the PTB system					
ILC_1B	cast steel	75.230	7.88	1.004	1.753
ILC_2	slag	159.19	2.185	1.830	3.475
ILC_3	fume dust	50.210	0.707	2.055	3.475

Prior to the measurement of sources produced in the project, CIEMAT measured a number of point sources to check the energy resolution of the system. The same test was made by PTB using the sources produced in the project. This was done to confirm that both protoypes were working with the same level of performance as demonstrated in the laboratory. In addition to the reference sources, on-site obtained probes were also measured with the CIEMAT and PTB prototype spectrometric devices. This required the calculation of specific calibration factors for the dimensions and materials typical from each end-user facility. Therefore, this was done prior to the demonstration and the corresponding factors were incorporated to the analysis software. The calculation of the correction factors was not part of the demonstration, but the importance of implementing adequate efficiency transfer correction factors was highlighted at demonstrations.

Measurement of reference sources

Both prototype spectrometric devices were transported by car from CIEMAT and PTB respectively to the end-user factories, including a portable shielding and the control and processing units. Prior to the measurement of samples, the proper functioning of the CIEMAT and PTB prototype spectrometric devices was carried out by an energy resolution check. In all cases, the CIEMAT and PTB prototype spectrometric devices passed the check.

The measurement of the reference sources described in the Table 10 was the first step of the process. The results of measurements at each end-user site were collected in Tables 11, 12 and 13 where the certified and measured values of activity concentration of measured sources are presented. Also given are the detection limits (MDA) obtained for the CIEMAT and PTB prototype spectrometric devices at end-user facilities.

The ζ-score test was applied to check the agreement of measurements with reference values. It is defined by the following expression

$$\zeta = \frac{a_{REF} - a_{VAL}}{\sqrt{da_{REF}^2 + da_{VAL}^2}}$$

Where a_{REF} and a_{VAL} represent the reference value and the result of the measurement at end-user site, respectively. The corresponding uncertainties are designated by d a_{REF} and d a_{VAL} .

From Tables 11, 12 and 13 it can be concluded that the results of the measurements at end-user sites are in agreement with the reference values, which demonstrates the correct efficiency calibration of the prototype spectrometric devices. The only exception is a Cs-137 slag source in the first set of measurements at Siderurgia Nacional. Although the exact nature of this discrepancy is not well understood, it can in principle be attributed to an incorrect shaking of the sample.

Table 11 Results of the measurement of reference sources at Siderurgia Nacional. Portugal

Ref date: 17 Sept 2014					**Counting time: 60 s**			
			Reference Values (Bq/g)		**Measured values (Bq/g)**			
Nuclide	**Type**	**Source id#**	**a(ref)**	**u(aref)**	**a**	**u(a)**	**MDA**	**ζ**
Co-60	Steel	Steel PTB4	14.7	0.22	15.1	0.44	0.122	-0.81
		Steel VUHZ4	0.251	0.004	0.24	0.07	0.10	0.16
Ra-226	Slag	LSlag03	0.89	0.01	0.74	0.28	0.82	0.54
		HSlag07	8.81	0.03	8.91	1.03	0.85	-0.10
Cs-137	Slag	HSlag03	9.72	0.11	8.74	0.21	0.074	4.13
Cs-137	Fume dust	Fume02	9.30	0.13	9.41	0.40	0.22	-0.26

Table 12 Results of the measurement of reference sources at Gerdau Spain.

Ref date: 17 Oct 2014					**Counting time: 60 s**			
			Reference Values (Bq/g)		**Measured Values (Bq/g)**			
Nuclide	**Type**	**Source id#**	**a(ref)**	**u(aref)**	**A**	**u(A)**	**MDA**	**ζ**
Co-60	Steel	Steel PTB4	14.6	0.21	14.8	0.44	0.255	-0.41
		Steel VUHZ4	0.189	0.003	0.15	0.05	0.16	0.78
Ra-226	Slag	LSlag03	0.89	0.01	-	-	0.828	
		HSlag07	8.81	0.03	8.86	1.03	0.84	-0.05
Cs-137	Slag	HSlag03	9.71	0.11	9.9	0.3	0.067	-0.59
Cs-137	Fume dust	Fume02	9.28	0.13	9.1	0.38	0.228	0.45

Table 13 Results of the measurement of reference sources at Vitkovice, Czech Republic

Ref date 29 Oct 2014					Counting time: 60 s				Counting time: 600s			
			Reference values (Bq/g)		Measured Values (Bq/g)							
Nuclide	Type	Source	a(ref)	u(aref)	A	u(A)	MDA	ζ	A	u(A)	MDA	ζ
Co-60	Steel	VUHZ 012/13	1.232	0.018	1.265	0.096	0.171	-0.34	1.24	0.034	0.026	-0.21
Ra-226	Slag	HSlag02	8.99	0.19	9.419	0.924	0.911	-0.45	9.355	0.320	0.224	-0.98
Cs-137	Fume dust	SK HFD ID01	9.41	0.10	9.691	0.391	0.185	-0.70	9.546	0.126	0.037	-0.85
Co-60			0.374	0.015	0.245	0.071	0.402	1.78	0.338	0.021	0.031	1.39

Measurement of on-site probes

Both prototype spectrometric devices were also used to measure several samples prepared by end-users for the three typical matrices: cast steel, slag and fume dust. These measurements aimed to determine the detection limits (MDA values) both for 60 seconds and for 600 seconds measurement times. The results are presented in the Tables 14, 15 and 16.

Table 14 Results of measurements for on-site material at Siderurgia Nacional, Portugal.

Material	Description	Container	Nuclide	Sample mass (g)	A Bq/g	MDA Bq/g
Cast steel	Steel STN1	n/a	Co-60	75	< MDA	0.015
Slag	Slag01SN, 02SN	CIEMAT	Ra-226	250	< MDA	0.444
Fume dust	Fume dust01 SN,02 SN	PTB	Cs-137	50	< MDA	0.023

Table 15 Results of measurements for on-site material at Gerdau, Spain

Material	Description	Container	Nuclide	Sample Mass (g)	A Bq/g	MDA Bq/g
Cast steel	Steel Gerdau	n/a	Co-60	97. 4	< MDA	0.18
				103.7	< MDA	0.255
Slag	Slag Gerdau	CIEMAT	Ra-226	222.8	< MDA	0.622
Fume dust	Fume dust Gerdau	PTB	Cs-137	101.7	< MDA	0.43

Table 16 Results of measurements for on-site material at Vitkovice, Czech Republic

				1 min counting time		10 min counting time	
Material	**ID**	**Nuclide**	**Mass (g)**	**A Bq/g**	**MDA (Bq/g)**	**A Bq/g**	**MDA Bq/g**
Cast steel	1	Co-60	97.24	< MDA	0.123	< MDA	0.013
	2		96.11	< MDA	0.110	< MDA	0.017
Slag	E5/11	Ra-226	193.60	< MDA	0.555	< MDA	0.127
	E5/15		168.88	< MDA	0.748	< MDA	0.143
Fume dust	LF	Cs-137	187.52	< MDA	0.088	< MDA	0.013
	F		264.30	< MDA	0.076	< MDA	0.011

As can be observed in Tables 14, 15 and 16 , all samples taken at the steel mills had no significant activity, as expected, and MDA values achieved with the prototype spectrometric devices were lower than the clearance levels for specific radionuclides.

Conclusion

The MDA values reported in Tables 11-16 were obtained *in situ* in order to verify that the detection limits achieved were below the clearance levels required, and provides proof of the spectrometric performance of the CIEMAT and PTB prototype spectrometric devices. It can also be seen that 60 s counting times allow the measurement of MDA values below the current clearance levels of target radionuclides which amount to 1 Bq/g for most nuclides.

Further to this, it can be deduced that counting times in the order of 300 s would be enough to provide the more strict clearance levels of 0.1 Bq/g expected in the near future for most radionuclides in cast steel. Slag and fume dust measurements are usually made without the same strict time requirements associated to the control of cast steel and for the special case of Ra-226 the clearance level will remain fixed at 1 Bq/g.

It must also be pointed out that these MDA values correspond to peak detection instead of total counting analysis (more usually measured) and therefore they include the identification of contaminants as part of the fast screening process. Finally, it should be noted that fixed equipment in use at end-user factories usually has shield structures heavier than those used in the prototype spectrometric devices. Therefore, lower MDA values would likely be attainable if the same shield structure were used with the prototype spectrometric devices.

In conclusion, this indicates that the prototype spectrometric devices designed and built in this project can replace existing systems at end-user facilities with significant advantages. In addition, testing the prototypes at three end-user facilities across Europe met the project's objectives in terms of the validation of the proposed equipment and its demonstration to end-users.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Siderurgia Nacional
Lisboa, September 2014


Portugal
Sn Seixal-siderurgia Nacional Sa
Porto
Salamanca
Coimbra
Castelo Branco
Cáceres
Badajoz
Sevilla
Huelva


Gerdau Aceros Especiales Europa
España
Spain
Madrid
Portugal
Gibraltar
Alboran Sea


GERDAU, Basauri,
Spain, October 2014
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October 2014


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Chemnitz
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Györ
Szombathely
Graz
Klagenfurt am Wörthersee
Maribor

4 Actual and potential impact

The impact activities for the project were designed to meet the project's objective to develop 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

Presentations to standards, technical committees and regulatory bodies

Interaction with regulatory bodies, working groups and technical committees has been maintained throughout the project. In some cases, more than one presentation took place to the same audience. Of utmost importance were the two presentations made in November 2013 and November 2014 by one of the partners (IST-ITN) in Luxembourg to the Group of Experts established under Article 31 of the EURATOM Treaty (IST-ITN is a member of this group). . This Group of Experts is a group of independent radiation protection and public health experts attached to the European Commission to help the EU make decisions concerning radioactivity. The European Commission ask their advice every time that it updates the Basic Safety Standards Directive – safety rules for radiation in applications such as medicine and research. The members of this Group of Experts are mostly members of the regulatory authorities in their countries. The members of the group took note of the project, its purpose and results achieved and expressed satisfaction with the dissemination of the project and the presentation they had had opportunity to attend

The International Atomic Energy Agency maintains a significant activity to protect people and the environment from the harmful effects of ionizing radiation and issues frequent recommendations to members. The project coordinator and a BEV-PTP member presented the main achievements of the project to members of the Division of Radiation, Transport and Waste Safety at the IAEA Headquarters in Vienna in April 2014.

The project's results have been also presented once per year to the Technical Committee for Ionising Radiation (TC-IR) of EURAMET.

A CIEMAT staff member of the Committee 82 of AENOR, the Spanish association for standardisation and certification, has kept the "Reference materials group" informed about the results and progress in the project on the production and characterisation of the reference sources developed in the project. A written statement has also been sent to ISO/REMCO Committee on reference materials describing the activities with the aim of promoting future discussions on the convenience of establishing a standard on the types of samples to be used for the calibration of detection equipment.

In addition project partner BEV/PTP has presented the project's results at the 253 Meeting of the Austrian Standards Committee "Radiation Protection" and IST-ITN has also described the project at a meeting of the Regulatory Commission for Nuclear Installations, the Portuguese Society for Radiation Protection, the Portuguese Quality Institute, end-users, the University of Lisbon and Coimbra and the Technical University of Lisbon

The Spanish Technical Group of the Protocol on Cooperation in the Radiological Surveillance of Metallic Materials is an organisation whose membership comprises representatives from the nuclear regulatory body, the public company in charge of the radioactive wastes, the association of steel producers and individual steel producers. The organisation has been proposed in international fora as a model for cooperation in this field. CIEMAT staff has made three presentations to this group, the last of which took place at the invitation of the organisers in January 2015. In the discussion that followed the presentation, issues such as the application of samples, equipments and methods developed in the project to the certification of non-radioactivity of metals were raised.

Presentations to conferences

Oral and poster presentations have supported the dissemination of the project to a broad audience. A total of 28 conferences or meetings in 14 different countries were attended by partners to present the project's results. Amongst them was the Fourth International Proficiency Testing Conference in Rumania, the

conference “Days of Radiation Protection” in the Czech Republic and Slovakia, an International Metallurgy Meeting organised in Madrid by a collaborator (PLATEA: Spanish technological platform for steel) and the 9th International Topical Meeting on Industrial Radiation and Radioisotope Measurement Applications.

Additional presentations were given to non-European audiences, such as the 8th International Conference & Expo on Isotopes in Chicago, USA and the 5th International Metrology Conference of the African Committee of Metrology in South Africa. The world metrological community was also informed of the project’s results by partners in two conferences: the Low-Level Radioactivity Measurement Techniques Conference organised by the International Committee for Radionuclide Metrology in the Republic of Korea in 2012 and the 19th International Conference on Radionuclide Metrology and its Applications in Belgium, in 2013.

Further to this, presentations were given at seminars such as those held at project partners JRC, NCBJ and CSIC, papers were published in technical press or in specialised journals (e.g. Journal of Radioanalytical and Nuclear Chemistry, Radiation Protection Dosimetry, Radiation Physics and Chemistry) and flyers on the project aims and progress were distributed to end-users and stakeholders.

Dissemination of the project’s achievements through the project website has been an effective means of reaching a wider audience. For example, the project website has been accessed more than 1000 times from 48 countries worldwide (see Figure 29) since the project started.
Finally, detailed scientific information has been made available to the scientific community in 9 papers published in peer reviewed journals, with 9 more papers either submitted or to be submitted shortly.

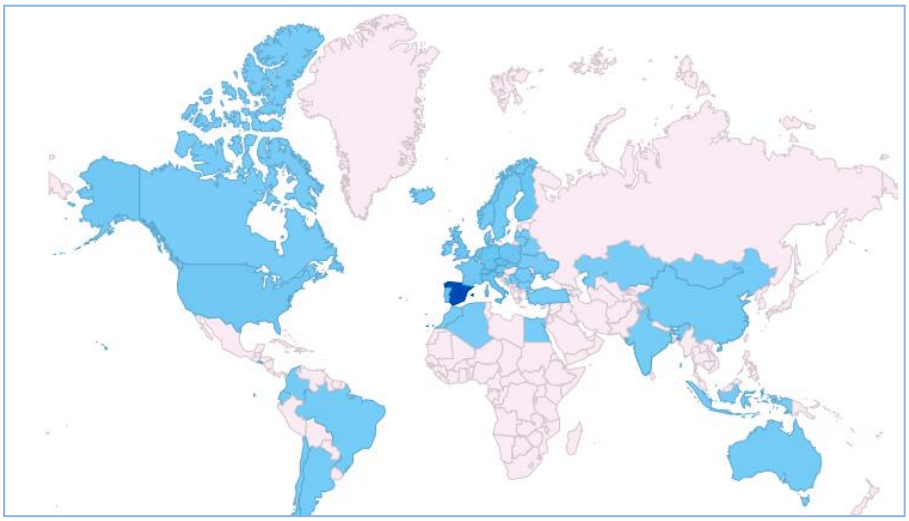

Figure 29 Geographical location of access to the project’s webpage

Workshops

Project partners have participated in six workshops which were organized in order to keep end-users informed about the project aims and its impact in the industry. Two wokrshops were organised by STUK in Finland and were open to end-users and national stakeholders. CMI organised another workshop at a steel factory in the Czech Republic, also opened to stakeholders. A project collaborator (UNESID) organised a workshop in Madrid with attendance of major stakeholders and end-users where project information was given. Finally, two more workshops were organised by IST in Lisbon and again both were open to major stakeholders. The audience for each workshop was mainly composed of industry members with typically between 25 and 50 attendees per workshop.

Cooperation between partners

Considering the strong interdependence between all project partners in most work packages, effective cooperation was a key factor for the success of the project. Frequent communications and project meetings have also allowed an effective exchange of ideas between partners. The level of cooperation can be considered as highly satisfactory and most cooperative work took place in meetings, where critical problems were discussed and future actions agreed. Of particular interest were the meetings where the testing at end-user facilities was planned and the list of potential testing places discussed. The involvement and cooperation of the partners in the project made possible the production of additional reference materials (cast steel and slag) not planned in the original protocol and that have been prepared by SMU and CIEMAT.

In addition, REG and ESRMGs participation in the project allowed us to address topics not already scheduled in the protocol such as the production and certification of samples with lower levels of activity, the development of alternative methods of gamma-ray processing and the production of mock sources of cast steel contaminated with a mixture of radionuclides. The contribution of a REG and two ESRMGs has added a significant contribution to the project.

Furthermore, testing and demonstration at end-user facilities was fostered by effective cooperation and communication between partners and end-users. The partners contacted end-users, discussed in advance the needs of testing teams and cooperated in the final phases of the process, such as IST-ITN, ENEA and CIEMAT in Lisbon, Portugal and CMI and PTB at Vitkovitce in Czech Republic

Finally, joint research has allowed the consortium to achieve goals that could not have been possible by the work of individual NMI's. The preparation and inter-comparison of reference sources, detector design and Monte Carlo evaluations are key examples of this. Both the European metrology community and end-users have been reached more effectively by a combination of the work done at national level (standardisation and technical committees) and at European/International level (conferences and workshops).

End-users, Stakeholders and collaborators engagement

Interaction with stakeholders was vital for the project and there are 38 stakeholder members associated with the project. The stakeholder composition is diverse and comprises regulatory bodies and large enterprises while collaborators are mainly industrial companies. A number of NMIs from outside Europe have also joined. Stakeholders have also actively participated in project workshops and by providing information about their needs and providing raw material for the preparation of radioactive sources, collaborators have played an essential role in the project.

Testing and evaluation of the prototypes was only made possible by the active participation of collaborators. At least 5 end-user facilities have been visited by partners in the Czech Republic, Germany, Portugal, the Slovak Republic and Spain, during the project either as small groups of partners or the whole consortium, such as the visit to US Steel Kosice, one of the largest Steel factories in central Europe. Prototype testing was done at three end-user facilities in Portugal, Spain and the Slovak Republic. In all cases, laboratory staff cooperated with project partners who highlighted the advantages of the new prototype systems. In addition to this, five end-user laboratories are already using the cast steel, slag and fume dust reference materials developed in the project.

Given the economic and social impact of any contamination problem at end-user facilities, it is likely that the project's results will be, to a major extent, incorporated into their technical procedures. The project received direct contributions from end-users, who provided detailed information about their procedures and made available raw material for source preparation. This allowed the effective exchange of information between end-users and the project and showed their interest in the project's findings. Further end-users; steel producers and members of several agencies also attended the six workshops organised by project partners and collaborators and were informed about the potential impact of the project results.

Actual and potential impact

Early Impact

Some of the projects outputs are already in use;

- 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. The reference materials can be reproduced by project partners and commercially distributed upon request from metallurgical industries. So far five end-users (steel mills from 5 different European Countries) are already using in the reference standards for composite steel, cast steel, slag and fume dust developed within the project. In most cases project partners have assayed the reference standards with equipment existing at the end-user laboratory; and in all cases sources were measured and certified by project partners. This provides the laboratories with the opportunity to

prove that their laboratories fulfil, the requirements of relevant ISO and European standards concerning quality programs.

- New Calibration and Measurement Capacities (CMCs) relevant to the metallurgical industry are now available at the project partners based on the inter-laboratory tests carried out with reference materials during the project. This will allow NMI's and DI's to offer new calibration services for the standardisation of reference samples of steel, slag and fume dust to external clients/end-users.
- Talks have been started with the steel industry with a view to developing technical standards for certifying the non-radioactivity of cast steel.

Further adoption of the project's outputs are expected:

- Input on the project's achievements has been provided by the project to the Group of Experts established under Article 31 of the EURATOM Treaty. It is hoped that the information will be used by the Group of Experts as a scientific basis for the development of new standards for radioactivity.
- A new instrument for radioactivity control of sources of cast-steel, slag and fume dust has been designed. Two prototypes have been built and tested at end-user facilities in three European Countries. The impact of the use of the new prototypes will be 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. This will be achieved by providing more efficient instruments, reference materials and methods (all developed in the project) to end-users.
- Detailed numerical models to simulate Ge detectors by Monte Carlo methods, developed in the project, have been made freely available for download in the project website. They describe in detail the detector-shielding ensemble and can be used to optimise the design of measurement systems by adapting these models to the particular needs of users. In order to make the models more applicable and useful for the wider stakeholder community, specific models have been created for the 5 most popular Monte Carlo packages (PENELOPE, MCNP-X, GEANT3, GESPECOR and EGS)

Longer-term impact

The turnover of the European steel industry is approximately 170 billion € and the number of direct employees in the sector is approximately 360 thousand. This project will impact the metallurgical industry i.e. the steel industry (directly) as well as process engineering and production technologies (indirectly). Its main potential impact is in the following areas:

- Better measurement techniques will minimise the occurrence and impact of the possible melting of orphan sources at end-user facilities, thus saving costs and contributing to a better protection against the dangers arising from exposure to ionising radiation. A reduction in the number of incidents at the steel industry will have significant cost savings due to the reduction in unnecessary shut downs of the production line. Additional financial savings should come from the implementation of reliable and traceable reference standards leading to better and more consistent certification of steel batches and a reduction in the number of disputes due to inconsistent data between different companies/countries. Certification of non-radioactivity of iron and steel will become a key aspect in the near future both from the safety and economy points of view and the procedures and prototypes developed in the project will help to implement an efficient system for this purpose.
- The project provided input into National and European standardisation and regulation bodies (such as the International Committee for Radionuclide Metrology, the International Commission for Radiation Protection, relevant European standardisation technical committees, the International Atomic Energy Agency). The technical recommendations developed in the project will provide national regulatory bodies and international institutions and committees competent in ionisation radiation measurements with the metrological basis to revise the current diversity of regulations and to harmonise current policies that affect steel trade, transport and waste management. The impact of this will be that both intra-European and external markets will benefit from a reduction in the number of trade disputes.

- A recent EURATOM directive (*Directive 2013/59/Euratom of 5 December 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation)* has modified the exemption and clearance criteria for materials for disposing, recycling or reuse. In practice, this will require the control laboratories at end-user facilities to lower the detection levels of their radioactivity monitoring system by a factor of 10 for some radionuclides. More efficient and reliable instruments and certified references will be necessary to implement these new regulations and the instruments, samples and knowledge developed in this project will support this purpose.

5 Website address and contact details

The website of the project has been updated regularly during the project and contains all basic information about its aims and results as well as a description of relevant events related to the project. It also contains freely downloadable numerical models of detectors for some of the most common Monte Carlo packages. The website is http://projects.ciemat.es/en/web/metrometal/

The main contact person for this project is the project coordinator, Dr. Eduardo García-Toraño, from CIEMAT, Spain. e.garciatorano@ciemat.es.

Additional, specific contacts are:

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Dr. Dirk Arnold, from PTB, Germany, for the development and characterisation of reference standards for the metallurgical industry, in particular for cast steel, composite steel and fume dust materials. dirk.arnold@ptb.de

Dr. Jaroslav Šolc from CMI, Czech Republic, for the characterisation of the new mesurement systems using the reference materials developed as well as Monte Carlo simulations. jsolc@cmi.cz

Dr. Virginia Peyrés from CIEMAT, Spain, for the construction of the measurement prototypes within the project. Viginia.peyres@ciemat.es

Dr. Branko Vodenik, from IJS, Slovenia, for questions concerning the evaluation of the protypes at end-user facilities. Branko.vodenik@ijs.si

6 List of publications

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