European Commission

JRC TECHNICAL REPORTS

The certification of the massic activities of the radionuclide ^{60}Co in stainless steel disks EURM 800 and 801

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2019

Joint Research Centre

EUR 29790 EN

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EU Science Hub
https://ec.europa.eu/jrc

JRC116895

EUR 29790 EN

PDF ISBN 978-92-76-08764-9 ISSN 1831-9424 doi: 10.2760/90491

Luxembourg: Publications Office of the European Union, 2019

How to cite this report: Van Ammel R. et al., *The certification of the massic activities of the radionuclide 60Co in stainless steel disks EURM 800 and 801*, EUR 29790 EN, Publications Office of the European Union, Luxembourg, 2019, ISBN 978-92-76-08764-9, doi:10.2760/90491, JRC116895.

Contents

Foreword

The decommissioning of a nuclear installation such as a power plant or a research reactor is the final step in its lifecycle. It involves all activities from shutdown and removal of nuclear material to the environmental restoration of the site. The whole process can extend over a long period of time, up to 30 years.

By 2025, it is estimated that over one third of the EU's currently operational reactors will have reached the end of their lifecycle and will be shut down.

The European Commission helps to address the funding of nuclear decommissioning through a group of experts known as the Decommissioning Funding Group (DFG). These experts:

- provide up-to-date knowledge on decommissioning costs and the management of funding
- explore ways to further co-operation and harmonisation of nuclear decommissioning at European level.

Through other ways the Joint Research Centre of the European Commission (JRC) is also supporting these activities by e.g. giving training courses for people working in decommissioning and by developing calibration standards that can be used in nuclear decommissioning facilities.

Acknowledgements

The authors would like to acknowledge the support received from V. Michenka, head of Laboratories and testing shops at VÚHŽ, for the input he provided to come to a good starting product.

Furthermore, we would like to acknowledge the support of R. Jakopic, Y. Aregbe, J. Paepen, K. Sobiech-Matura, M. Hult, W. Mondelaers and A. Plompen for their support and exchange of ideas during the development of the certified reference material.

The report was also reviewed by C. Venchiarutti. The authors would like to thank her for the excellent review and the constructive discussion to improve the report.

After the internal review the report was reviewed by the certification advisory panel. The authors would like to thank the panel members M. Crozet, D. Roudil and S. D. Balsley for the effort they put in reviewing the report and the fruitful discussions we had during and after the panel meeting.

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Abstract

This report describes the production of EURM 800 and EURM 801, stainless steel disks containing a certified value for the massic activity of 60Co. The material was produced following ISO 17034 [1].

The material was produced at a contractor site. After the production the material was shipped to the JRC in Geel where compliance with the technical specifications was checked. The material was found to meet the technical specifications and characterisation was carried out.

The material was characterised by an intercomparison among six laboratories of demonstrated traceability to national or international standards. The different results of the material characterisation were combined into the certified value in compliance with the standard ISO 17034 and ISO Guide 35 [2].

The uncertainties on the certified values were calculated in compliance with the Guide to the Expression of Uncertainty in Measurement (GUM) [3]. It includes uncertainties related to possible inhomogeneity, instability and characterisation.

These materials are intended for the assessment of method performance and quality control. As with any reference materials, these materials can also be used for control charts or validation studies. The Certified Reference Materials (CRM) are available in the shape of a stainless steel disk. The disk shall be used as such (minimum sample intake shall be one disk) and shall not be treated with any chemicals.

The following values were assigned (at reference date 1 November 2017, 00h00 CET):

	Massic activity at reference date 1 November 2017, 00h00 CET	
	Certified value [1) 2)] [Bq/kg]	Uncertainty [1) 3)] [Bq/kg]
^{60}Co batch 1 EURM 800	177	6
^{60}Co batch 2 EURM 801	1301	35

1) Certified values and uncertainty at time of measurement (decay correction) to be calculated by using only the Recommended decay data: http://www.lnhb.fr/nuclear-data/nuclear-data-table/.

2) Unweighted mean value of the means of accepted data sets, each set being obtained in a different laboratory. The certified value and its uncertainty are traceable to the International System of Units (SI) via the certified activities of the calibration source used.

3) The uncertainty is the expanded uncertainty with a coverage factor k = 2.57 corresponding to a level of confidence of about 95 % estimated in accordance with ISO/IEC Guide 98-3, Guide to the Expression of Uncertainty in Measurement (GUM:1995), ISO, 2008.

Disclaimer

Certain commercial equipment, instruments, materials and software codes are identified in this paper to specify adequately the experimental procedure. In no case does such identification imply recommendation or endorsement by the European Commission, nor does it imply that the material or equipment is necessarily the best available for the purpose.

1 Introduction

1.1 Background

In Europe and even globally decommissioning old nuclear power plants and other nuclear installations is challenging. Decommissioning old nuclear power plants starts after the removal of the nuclear fuel. The remaining materials need to be managed, and perhaps even cleared from regulatory control, in a safe and reliable way. The majority of remaining radioactivity is contained in the primary circuit of the reactor. However, radioactivity is also encountered in the surroundings of the primary circuit. The levels of radioactivity in the surroundings are much lower but the amount of the material to be managed is much larger.

The two most important streams of waste are concrete and metals. As an example, a Pressurised Water Reactor (PWR) built in the 1970s, contains about 75000 m^3 of concrete and 36000 metric tons of steel and iron [4]. The majority of this material needs to be characterised before it can be safely disposed or free released.

Taking into consideration the large amount of material and the high cost of radioactive waste disposal, the European Commission issued Council Directive 2011/70/Euratom [5] stating that: "Generation of radioactive waste shall be kept to a minimum which is reasonably practicable in volume and activity by means of appropriate design measures and of operating and decommissioning practices including the recycling and reuse of materials."

The massic activity (expressed in Bq/kg) values for free release of materials which can be applied by default to any amount and to any type of solid material, are laid down in Council Directive 2013/59/Euratom [6]. These values are set per individual radionuclide.

In order to comply with the legislation numerous measurements need to be carried out for the segregation, characterisation and/or free release of all the materials present at a nuclear site. To validate and calibrate the various systems and set-ups used to perform such measurements, SI-traceable activity standards are needed. Such standards need to represent, as much as possible, the geometries, matrices and radionuclide content of interest. The routine free release measurements normally involve measurements of bulk quantities of materials (often placed in euro pallet sized containers or in 200 litre drums) of three types: metal, concrete and light materials. It is therefore critical to have suitable calibration standards of similar types available. The need for a matrix matching reference standard is even more pronounced for metals due to the strong effects of self-attenuation of radiation in the matrix. Such a standard should also match the geometrical configuration of the measured metal waste as much as possible.

1.2 Choice of the material

A nuclear power plant consists of many metal parts. In order to fill the gap in the availability of metal standards to the European decommissioning community, JRC-Geel has included in its work programme the development of a standard consisting of metal disks containing ^{60}Co. These disks can be used as a calibration standard for the characterisation of radioactive waste items. An even more important application is the use as a calibration standard in melting facilities. In these facilities metal waste released from nuclear facilities can be recycled by melting and feeding it into the production processes. Samples of batches containing recycled material are analysed to assess their radioactive content. One of the most important radionuclides present in metal waste is ^{60}Co. It is often used as a scaling factor to correlate more difficult to measure radionuclides with easier to measure radionuclides.

The feasibility of the production and characterisation of this material has been proven by previous work led by JRC-Geel. This work had been carried out in the context of the EURAMET, ENV09, Metrology for Radioactive Waste project [7] [8].

1.3 Design of the project

The massic activity of ^{60}Co in stainless steel disks is characterised using a laboratory intercomparison approach. Six designated institutes with experience in the field of measurement of radionuclides reported results for the massic activity of ^{60}Co of the two different batches. All participating laboratories are in their daily work familiar with and adhering to the concepts of metrological traceability and measurement uncertainty estimation.

2 Participants

2.1 Project management and evaluation

European Commission, Joint Research Centre, Geel, Belgium

2.2 Production

VÚHŽ, Laboratories and testing shops, Dobrá, Czech Republic

European Commission, Joint Research Centre, Geel, Belgium

2.3 Homogeneity and stability study

European Commission, Joint Research Centre, Geel, Belgium

Following the requirements of ISO 17034 and ISO Guide 35.

2.4 Characterisation

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International Atomic Energy Agency (IAEA), Department of Nuclear Sciences and Applications, Marine Environment Laboratory, Monaco, Monte Carlo

Jožef Stefan Institute, JSI Ljubljana, Slovenia, Slovenia

European Commission, Joint Research Centre (JRC) Geel, Belgium

3 Material

3.1 Requirements of the material

The technical requirements of the disks were formulated to most suitably cover the requirements of analytical laboratories as well as taking into consideration the legal levels for free release. The technical specifications of cast steel disks are summarised in Table 1.

Table 1. Technical specification of cast steel disks containing ^{60}Co

Parameter	Unit	Value	Tolerance[1]	Comments
Number of batches		2		
Number of disks	per batch	300		
Number of blanks		10		
Base material		EN steel No 1.4301/ SAE grade 304		Content in other artificial radionuclides at massic activity levels <1/1000 of the foreseen ^{60}Co massic activity
Shape	-	Disk		"right angle circular cylinder"
Shape or Squareness Tolerances	-		-	Tolerance according ISO 2768-1, class mK [9]
Diameter	mm	35	± 0.1	Tolerance according ISO 2768-1, class mK [7]
Thickness	mm	10	± 0.1	Tolerance according ISO 2768-1, class mK [9]
Mass	g	75	± 4	
Massic activity ^{60}Co (n.a. to blanks)	Bq/g	Low massic activity (batch1): 0.25 High massic activity (batch2): 1.00	± 25 %	
Coating		NONE		
Mass variation between disks in one batch	%	≤ 1		Relative standard deviation (RSD) between units
Density homogeneity in one batch	%	≤ 2		Relative standard deviation (RSD) between units
Homogeneity in massic activity (per activity level)	%	< 2		Relative standard deviation (RSD) Random stratified sampling (≥10 disks per batch)

1 Units as in the respective column unless otherwise indicated

3.2 Technical specifications

3.2.1 General

JRC-Geel set up a contract with an industrial melting facility to produce two batches of steel disks (EN steel No. 1.4301, SAE grade 304). Each disk has a diameter of 35 mm and a thickness of 10 mm. In total more than 550 disks were produced in two batches. Each batch contained a different massic activity level. The disks were made from an identical base material. Before adding the ^{60}Co to the base material, ten blank disks were produced. The radioactivity in these blank samples is below the detection limit as determined by gamma-ray spectrometry (= 0.24 Bq/kg for ^{60}Co). No radionuclides could be detected in the blank samples. In the reference materials no radionuclides but ^{60}Co could be detected.

3.2.2 Massic activity

The first batch should have a massic activity of 0.25 Bq/g (± 25 %) and the second batch should have a massic activity of 1 Bq/g (± 25 %).

The ^{60}Co shall be homogeneously distributed within each disk and between the disks of each batch. For each batch, the overall variation in ^{60}Co massic activity expressed as relative standard deviation should be below 2 %.

3.2.3 Identification

All the disks shall be uniquely numbered by an engraved three digit number. The disks of the first batch shall be numbered from "JRC-Geel Co-60 Nr.001" till "JRC-Geel Co-60 Nr.300" and the ones of the second batch from "JRC-Geel Co-60 Nr.301" till "JRC-Geel Co-60 Nr.600".

3.2.4 Storage and transport

The storage (dry storage at ambient temperature) and transport conditions shall not compromise the physical or chemical properties of the disks.

3.2.5 Documentation

The production procedure was mutually agreed upon between the contractor and JRC-Geel before signing the order. After production a report containing detailed information on the production process was issued.

4 Production/Processing

4.1 General principle of the production

The addition of selected radionuclides to melted iron alloy can be used for the manufacturing of homogeneously radioactive containing metallic materials [7] [8]. During the melting process, the radionuclides pass into the metal cast and are homogeneously mixed in the solution. The homogeneity depends on the chemical nature of the introduced radionuclide(s). The added radionuclide(s) must be in a suitable chemical and physical form. In this case the ^{60}Co was in an iron based master alloy. An electric induction furnace is used to fuse the iron master alloys because of homogenisation of the metal solution occurs by inductive stirring.

Induction furnaces also enable small quantities of melt to be prepared with good output control. After finalising the molten metal – radionuclide mixture, it is tapped off from the furnace into a ladle and transferred to a cast and eventually into molds. The casting may be performed by a gravitational casting technology.

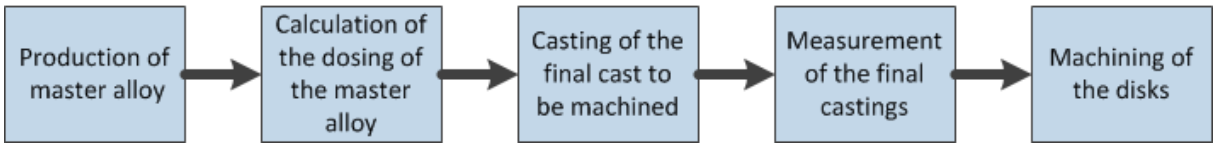
After the melting, casting and solidification of the master alloy, it is cut into fragments. Selected fragments of the master alloy are subsequently diluted in the inactive base material to obtain the desired massic activity. In this way the dosing can be done with a high accuracy and a good solubility of the materials is realised. This final cast can be poured in the molds.

After solidification and cooling, the castings are cut and machined to their final dimensions. The ^{60}Co is a suitable radionuclide for this technique as it forms a homogeneous solution in the cast [7] [8].

4.2 Production method used

The production flow was mutually agreed on before production. The flowchart describing the production process is given in Figure 1. Each of the steps is described in more detail in the next sections.

Figure 1. Flowchart of the production process



4.2.1 Production of the master alloy

A new master alloy with a composition according to EN steel No 1.4301/SAE grade 304 has been produced. The master alloy contained ten times higher ^{60}Co massic activity compared to the highest required massic activity, therefore the master alloy is an intermediate step between the ^{60}Co sample and the final castings.

4.2.2 Calculation of dosing of the master alloy

The master alloy was machined into a sample that could be measured directly by gamma-ray spectrometry. The massic activity of ^{60}Co was 12.76 Bq/g (Reference date 22 May 2017). Based on this value a dosing mass of the final casting was calculated to obtain the desired final massic activities. The applied dosing was 25% higher to compensate for possible loss.

4.2.3 Casting of bars to be machined

A basic melt of blank steel was made according to the specifications of EN steel No 1.4301/SAE grade 304. This melt was sampled to check compliance with the technical

specifications. To this melt a known amount of master alloy was added to obtain melts having the desired final massic activity. Two different amounts were added to the two different batches of the melt to obtain the two different massic activities of ^{60}Co. Some pictures (Figure 2, Figure 3, Figure 4 and **Error! Reference source not found.**) were taken during the production of the final castings.

Figure 2. Prepared steel molds (heated prior to casting)

Figure 3. Pouring of the melt into the casting pan

Figure 4. Semi-automatic casting into the steel molds

Figure 5. Slow cooling of the casting in the molds

4.2.4 Machining of the final disks

The final disks were machined from the stripped castings according to the required dimensions. The machining was carried out by a CNC lathe using cooling fluid to avoid any heat affection on the final product. The individual disks were engraved by cold laser technology. No surface treatment was performed on them.

4.2.5 Measurement of the final casting

The massic activity of the final castings was assessed by the producer. From each batch five randomly selected disks were measured and the average ^{60}Co massic activity was calculated. The results (without uncertainty) of the measurements of the producer are given in Table 2. The producer notified JRC-Geel in written that the actual values are slightly out of the technical specifications. JRC-Geel could agree on it as the massic activity of the different batches was not considered a critical parameter.

Table 2. 60Co massic activity of the final castings given by the producer

	Nominal activity Bq/g	**Actual activity Bq/g**
Batch 1	0.25	0.18
Batch 2	1.00	1.35

4.2.6 Storage and dispatch

Before transport the disks were stored in a wooden box in a dry place at ambient temperature. Filler material was used to avoid mechanical damage. The disks were transported to JRC-Geel via courier.

4.3 Assesment of the disks

Due to internal metallurgical defects in certain disks, detected at the producer site, only 288 disks from batch 1 and 278 disks from batch 2 were transported to JRC-Geel.

4.3.1 Physical dimensions

The mass of the disks was determined by weighing each of them using a yearly calibrated analytical balance Sartorius® MSX D09-09-015.

The diameter and thickness of each disk were determined using Mahr® 16EW callipers. The trueness of the measured results obtained by the callipers was checked by measuring class 0 gauge blocks with it. Gauge blocks in the same range as the thickness and the diameter of the disks were measured and the dimensions of the blocks could be exactly read out from the callipers.

The summary of the results of the different measurements of the individual disks for the two different batches are given in Table 3 and Table 4. The densities in the table were calculated from the mass and the volume of the individual disks.

Table 3. Summary of the physical properties of the disks of batch 1

	Batch 1 (288 Items)			
	Mass (g)	**Diameter (mm)**	**Thickness (mm)**	**Density (g/cm^3)**
Average	76.0290	35.03	10.02	7.87
SD	0.2421	0.03	0.02	0.02
RSD	0.32%	0.09%	0.20%	0.30%
Min	74.8481	34.88	9.96	7.78
Max	76.4975	35.21	10.12	7.91
Δ (Max- Min)	1.6494	0.33	0.16	0.13

Table 4. Summary of the physical properties of the disks of batch 2

	Batch 2 (278 Items)			
	Mass (g)	**Diameter (mm)**	**Thickness (mm)**	**Density (g/cm^3)**
Average	75.9948	35.05	10.01	7.87
SD	0.4032	0.03	0.05	0.02
RSD	0.53%	0.10%	0.48%	0.28%
Min	72.9192	34.98	9.57	7.78
Max	76.5896	35.25	10.11	7.99
Δ (Max- Min)	3.6704	0.27	0.54	0.21

A total of 15 disks originating from both batches were found to be out of the set specifications. Their diameter was too big (11 disks) or their thickness was too small (4 disks).

Homogeneity

A key requirement for any reference material (RM) is the equivalence between the various units. In this respect, it is relevant whether the variation between units is significant compared to the uncertainty of the certified value. In contrast to that it is not relevant if this variation between units is significant compared to the analytical variation. Consequently, ISO 17034 requires RM producers to quantify the between-unit variation. This aspect is covered in between-unit homogeneity studies.

The within-unit inhomogeneity is formally not assessed on the material as it shall be used as one unit, not being split up. Splitting up the samples would involve heavy mechanical manipulations compromising the certified values. Nevertheless some information on the within-unit inhomogeneity was collected.

4.4 Between-unit homogeneity

The between-unit homogeneity was evaluated to ensure that the certified values of the CRM are valid for all units of the material, within the stated uncertainty.

Ten units were selected, which is larger than the cubic root of the total number of produced units (6.7 units for n = 300). The 10 units per batch were selected using a random stratified sampling scheme covering the whole batch for the between-unit homogeneity test. The 10 samples from each batch were analysed by gamma-ray spectrometry and their massic activity was calculated. Each sample was measured twice (Annex 1).

The analytical procedure applied for the analysis of ^{60}Co in the disk was gamma-ray spectrometry. The spectrometric measurements were done directly on the disks, not involving any source preparation. The efficiency calibration was performed by measurement of traceable calibration standards in the shape of point sources applying geometry, density and true coincidence corrections. The code used to apply these corrections is EGSnrc [10] .

The measurement procedure was applied under repeatability conditions and in a randomized manner in order to separate a potential analytical drift from a trend in the production sequence. Only relative measurement results were used. This eliminated a number of uncertainty contributions for absolute values, such as the contribution from calibration. This approach resulted in a small measurement uncertainty (Table 5) so that small differences between the sample units (heterogeneity) were more easily detected. Moreover, corrections for the small differences in dimensions of the disk for gamma-ray spectrometry were applied for each individual sample by Monte Carlo simulation.

Table 5. Standard uncertainty (%) components for homogeneity measurements

Uncertainty component	**^{60}Co [%]**
Sample dimensions detection efficiency	0.5
Weighing	0.01
Counting statistics (incl. background)	0.2
Sample positioning	0.1
Combined relative standard uncertainty, $u_{meas,rel}$	0.55

4.5 Within-unit homogeneity and minimum sample intake

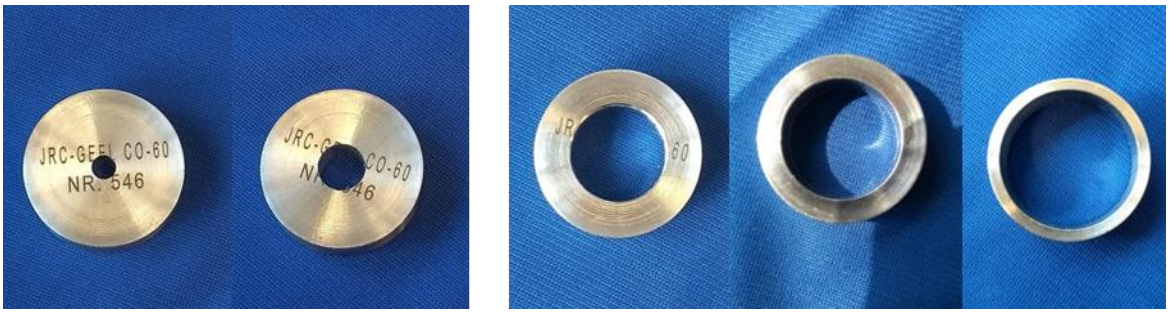
The within-unit homogeneity is not quantified as the disks shall be used as one piece and shall not be split up in subsamples. The minimum sample intake shall be one disk.

Nevertheless some tests were performed to check for possible inhomogeous distribution of ^{60}Co in the disks. Two possible ways of inhomogeneity were assessed: axial and radial.

To assess potential axial inhomogeneity as a within-unit inhomogeneity, many disks were measured rightside up and upside down on the same detector and in the same geometry. The counting efficiency was the same for both measurements of the same disk. The count rates from the ^{60}Co did not show any variation between the two measurements of the same disk. The difference can be entirely attributed to the measurement uncertainty and random scatter. The results are shown in Figure 9 of Annex 1.

To assess potential radial inhomogeneity one disk was measured entirely and re-measured after removing material starting from the middle in steps of 5 mm of material (Figure 6). After each removal of 5 mm, the disk was re-weighed and re-measured on the same detector at the same distance. The detection efficiency was recalculated for the different geometries and the relative ^{60}Co massic activity was determined. No significant differences between the massic activities of the disks of the different geometries were observed. Results of the different measurements are presented in Figure 10 of Annex 1.

Figure 6. Pictures of different samples used to assess the radial activity distribution



In conclusion, the tests did not reveal any inhomogeneity in either the axial or radial dimension. A possible inhomogeneity was too small to be quantified.

4.6 Uncertainty assessment

For each batch, the dataset obtained from the between-unit homogeneity study was tested for consistency using Grubbs outlier tests. On a confidence level of 95%, no outlying individual results were detected.

Quantification of between-unit inhomogeneity was undertaken by analysis of variance (ANOVA), which separates the between-unit standard deviation (s_{bb}) from the within-unit standard deviation (s_{wb}). The latter is at least equivalent to the method repeatability. The two measurements per disk performed in the between-unit homogeneity study, were used to estimate as well possible within unit inhomogeneity.

Evaluation by ANOVA requires mean values per unit, which follow at least a unimodal distribution and the results for each unit that follow unimodal distributions with approximately the same standard deviations. It was checked visually whether all individual data follow a unimodal distribution using histograms and normal probability plots. The 20 measurement results do not allow detecting any deviation from a normal distribution.

It should be noted that s_{bb} and s_{wb} are estimates of the true standard deviations and are therefore subject to random fluctuations. Therefore, the mean square between groups

($MS_{between}$) can be smaller than the mean squares within groups (MS_{within}), resulting in negative arguments under the square root used for the estimation of the between-unit variation, whereas the true variation cannot be lower than zero. In this case, u^*_{bb}, the maximum inhomogeneity that could be hidden by method repeatability, was calculated as described by Linsinger et al. [12]. u^*_{bb} is comparable to the limit of detection of an analytical method, yielding the maximum inhomogeneity that might be undetected by the given study setup.

Method repeatability, within-unit standard deviation ($s_{wb,rel}$), between-unit standard deviation ($s_{bb,rel}$) and $u^*_{bb,rel}$ were calculated as:

$$s_{wb,rel} = \frac{\sqrt{MS_{within}}}{\bar{y}} \qquad \textbf{Equation 1}$$

$$s_{bb,rel} = \frac{\sqrt{\frac{MS_{between} - MS_{within}}{n}}}{\bar{y}} \qquad \textbf{Equation 2}$$

$$u^*_{bb,rel} = \frac{\sqrt{\frac{MS_{within}}{n}}\sqrt[4]{\frac{2}{v_{MSwithin}}}}{\bar{y}} \qquad \textbf{Equation 3}$$

MS_{within} mean of squares within-unit from an ANOVA

$MS_{between}$ mean of squares between-unit from an ANOVA

$\bar{y}$ mean of all results of the homogeneity study

n mean number of replicates per unit

$v_{MSwithin}$ degrees of freedom of MS_{within}

The results of this evaluation of the between-unit variation are summarised in Table 6. The resulting values from the above equation were converted into relative uncertainties.

Table 6. Results of the between unit homogeneity study

	$s_{wb,rel}$ [%]	$s_{bb,rel}$ [%]	$u^*_{bb,rel}$ [%]	$u_{bb,rel}$ [%]
Batch 1	0.6	0.3	0.3	0.30
Batch 2	0.5	0.28	0.23	0.28

In summary, the homogeneity study showed no outlying results, trends in the production or analytical sequence. S_{bb} was found above u^*_{bb}, the limit to detect inhomogeneity. Therefore, S_{bb} the between-unit standard deviation is used as estimate of u_{bb}.

5 Stability

The base material was selected to ensure that stability should not be an issue. Stainless steel (EN steel No 1.4301/SAE grade 304) forms a homogenous and stable mixture with cobalt. Time and temperature were regarded as the possible influencing factors on the stability of the material.

Stability testing is necessary to establish conditions for dispatch to the customers (short-term stability) as well as conditions for storage (long-term stability). During transport, the material may be exposed to temperatures from 4°C up to 60°C for a relatively short time. Stability under these conditions shall be demonstrated as transport will be done at ambient temperature.

It should be noted that the term stability in the context of a CRM does not cover the radioactive decay. It is evident that the radionuclides are decaying according to their half-lives, and their decay is quantitatively predictable using the formula:

$$A_t = A_0 \times e^{-\lambda(t-t_0)} \qquad \textbf{Equation 4}$$

Where

t is the time at which the activity is calculated

t_0 is the reference time as indicated on the certificate

A_t is the activity at time t

A_0 is the activity at the reference time t_0

λ is the decay constant defined as $\ln(2)/T_{1/2}$

with $T_{1/2}$ the half live of ^{60}Co (5.2711(8)a) (k=1) [11]

The uncertainty on the half-life was not taken into account in the calculation of the uncertainty on the final material, as the uncertainty is negligible compared to the other uncertainty components. When using the material, the decay corrected activity to the reference date on the certificate shall be calculated and compared to certified values.

5.1 Short-term stability study

For the short-term stability study, samples were stored at 4°C or 60°C for 1, 2, 3 and 4 weeks. Two units per batch, one for each storage temperature, were selected using a random stratified sampling scheme. The samples were analysed before and after storage for 1, 2, 3 or 4 weeks at 4 or 60°C. The analysis was performed in the same measurement conditions for both measurements. The massic activity of ^{60}Co was determined using gamma-ray spectrometry. The analytical procedure applied was the same as described in section 5 for the homogeneity study.

The samples were measured under repeatability conditions using relative measurement results only, thereby eliminating a number of uncertainty contributions for absolute values such as the contribution from calibration. As the physical dimensions of the measured samples remain unchanged, only the position of the sample with respect to the detector may change between the measurements.

Table 7 gives an overview of the uncertainty components on the massic activity determination as for the short-term stability study of batch 1. Note that the uncertainty originating from the counting statistics is higher for this study compared to the homogeneity study due to poorer counting statistics caused by shorter measurement times. As a conservative approach the result of the short-term stability of batch 1 is taken into account. The uncertainty component for the short-term stability for batch 1 is slightly higher than the one for batch 2.

To be able to separate a potential analytical drift from a trend over storage time a reference source was measured in between the samples for the stability study. No analytical drift of the measurement set-up could be observed during the short-term stability study.

Table 7. Standard uncertainty (%) components on the massic activity for short-term stability measurements of batch 1

Uncertainty component	**^{60}Co [%]**
Weighing	0.01
Counting statistics (incl. background)	1.0
Stability of set-up	0.4
Sample positioning	0.2
Combined relative standard uncertainty, $u_{meas,rel}$	1.1

The obtained data were evaluated individually for each temperature. The results were screened for outliers using the single and double Grubbs test. No outlying individual results were found at 95% confidence level.

Furthermore, the data were evaluated against storage time and regression lines of the normalised net counting rate versus time were calculated using the SoftCRM software. The slopes of the regression lines (loss/increase due to simulated shipping conditions) were tested for statistical significance. The slopes of the regression lines were not significantly different from zero (at 95 % confidence level) at 4°C and at 60°C. The details of the regression analysis can be found in Table 8

Table 8. Results of the regression analysis of the short-term stability at 4°C and 60°C as calculated by SoftCRM

	Slope	**u_{slope}**	**Intercept**	**$u_{intercept}$**	**$u_{sts,rel}$ [%]**
4°C	-0.002	0.003	1.001	0.006	0.3
60°C	-0.001	0.002	1.007	0.004	0.2

The detailed results of the individual measurements are shown in Annex 2.

No significant change in the ^{60}Co massic activity at 4ºC and at 60ºC was observed, assuming that the shipping period of the material normally would not take more than 1 week.

The material can be dispatched without further precautions under ambient conditions.

5.2 Long-term stability study

As the ^{60}Co forms a very stable alloy with the stainless steel basic material, the long-term stability is expected be excellent. Stainless steel is very resistant to corrosion, oxidation, heat and cold due to its high content of chromium. It is used in many applications because of these properties.

To prove the long-term stability, similar samples that were produced in the frame of the EURAMET, IND04, ionising Radiation Metrology for the metallurgic industry project [7] were re-measured. In this project stainless steel disks containing ^{60}Co were produced and characterised in 2013. These samples have been stored for more than five years in normal laboratory conditions.

The decay corrected count rates of two samples were calculated from measurements performed in June 2013. The two samples were remeasured in June 2018 in exactly the same conditions (same sample, same detector, same sample holder, same distance between detector and sample). The corresponding decay corrected count rates were calculated. The count rates of both measurements were compared and the difference can be attributed entirely to the measurement uncertainty. The data were introduced in the SoftCRM software that showed no significant instability over a five year period Details of the regression analysis can be found in Table 9 .

Table 9. Results of the regression analysis of the long-term stability at room temperature as calculated by SoftCRM

	Slope	**u_{slope}**	**Intercept**	**$u_{intercept}$**	**$u_{sts,rel}$ [%]**
Room temp	0.000	0.000	1.000	0.002	0.6

The detailed results of the measurements are shown in Annex 3.

The measurement results show an excellent long-term stability of ^{60}Co in stainless steel under normal storage conditions. The long-term stability shall be further assessed by yearly analysing one unit of each batch in storage. These stability post-monitoring measurements can be relatively easy performed as the used analytical technique, gamma spectrometry is non-destructive.

5.3 Estimation of uncertainties

Due to the intrinsic variation of measurement results, no study can rule out degradation of materials completely, even in the absence of statistically significant trends. It is therefore necessary to quantify the potential degradation that could be hidden by the method repeatability, i.e. to estimate the uncertainty of stability. This means, even under ideal conditions, the outcome of a stability study can only be "degradation is 0 ± x % per time".

Uncertainties of stability during dispatch and storage were estimated as described. For this approach, the uncertainty of the linear regression line with a slope of zero is calculated for the short-term stability. For the short-term stability the data from the study at 4ºC were used as a conservative approach. (the short-term stability for 60ºC is slightly better) The uncertainty contributions u_{sts} and u_{lts} are calculated as the product of the chosen transport time/shelf life and the uncertainty of the regression lines as:

$$u_{sts,rel} = \frac{RSD}{\sqrt{\sum\left(t_i - \bar{t}\right)^2}} \cdot t_{tt}$$ **Equation 5**

$$u_{lts,rel} = \frac{RSD}{\sqrt{\sum\left(t_i - \bar{t}\right)^2}} \cdot t_{sl}$$ **Equation 6**

RSD relative standard deviation of all results of the stability study at relevant temperature

t_i time point *i* of measurement

$\bar{t}$ mean value of all time points

t_{tt} chosen transport time (conservative approach of 1 week at 4ºC)

t_{sl} chosen shelf life (this study 5 years at room temperature)

The following uncertainties were estimated:

- $u_{sts,rel}$, the uncertainty of degradation during dispatch. This was estimated from the 4°C studies. A possible degradation of 0.3 % per week was observed. The resulting uncertainty describes the possible change during a dispatch at 4°C lasting for one week.
- $u_{lts,rel}$, this stability uncertainty corresponds to possible degradation during storage. This uncertainty contribution was estimated from the study of the ^{60}Co steel disks produced in 2013 and found to be 0.6 % over a 5 years storage period at room temperature.

The results of these evaluations are summarised in Table 10.

Table 10. Uncertainties of stability during dispatch and storage. $u_{sts,rel}$ was calculated for a temperature of 4 °C and t_{tt} = 1 week; $u_{lts,rel}$ was calculated for a storage at room temperature and t_{sl} = 60 months

Analyte	$u_{sts,rel}$ [%]	$u_{lts,rel}$ [%]
^{60}Co	0.30	0.6

A very limited degradation during hypothetical dispatch conditions at 4°C was observed. The possible degradation combined with the uncertainty of the regression line result in a small total short-term stability compared to the uncertainty of the reference values. The material can be transported at ambient conditions without special precautions.

Since no trends were detected in the long-term stability study, the material can be stored at room temperature.

After the certification campaign, the material will be subject to the JRC-Geel stability monitoring programme according to ISO17034 and ISO Guide 35 to control its further stability.

6 Characterisation

The material characterisation is the process of determining the property values of a reference material.

The material characterisation was established by an inter-laboratory comparison of expert laboratories. The ^{60}Co massic activity of the material was determined in different laboratories. Due to the nature of the analyte all participants used gamma-ray spectrometry as analytical method for the determination of the massic activity of the ^{60}Co in the disks. Nevertheless, differing approaches to calibrate the same type of measurement equipment were used. This approach aims at randomisation of laboratory bias, which reduces the combined uncertainty.

6.1 Selection of participants

Six laboratories were selected based on criteria that comprised: proven technical competence and quality management aspects. All selected laboratories belong either to a National Metrology Institute or a designated Institute for radioactivity measurements of their country or an international organisation. The calibration of their measurement set-up is done with standard sources traceable to national standards. Having a formal accreditation was not mandatory, but the laboratories followed the technical requirements of ISO/IEC 17025.

6.2 Study setup

Each laboratory received two disks of each batch (four samples in total). They were requested to provide four independent results, one for each disk. The disks for material characterisation were selected using a random stratified sampling scheme and covered the whole batch. Each laboratory was free to choose an analysis method, with the requirement to ensure traceability of the measurement results to the International System of Units (SI). For each batch, the measurements had to be spread over at least two days to ensure intermediate precision conditions. Laboratories were also requested to estimate the combined standard uncertainties of each individual result. No specific approach for the estimation was prescribed, i.e. top-down and bottom-up were regarded as equally valid procedures.

6.3 Method used

The analytical method used by all the participating laboratories in the determination of the massic activity of the samples was gamma-ray spectrometry. The measurements were done directly on the disks, not involving any source preparation. The methods used for efficiency calibration, the traceability of calibration standards, and the methods used for applying corrections are described in Annex 4.

The challenge and major source of uncertainty in gamma-ray spectrometry of volume sources is to preserve the traceability link when transferring efficiency values from standard solutions or standard point sources to the measurement parameters of the volume sources (differences in geometry and density). Since the participants had a free choice of methods, different methods were applied to establish the counting efficiency for the measured disks:

- One laboratory measured an aqueous reference standard in the same geometry as the disk and made a correction for self-attenuation by dedicated software.

- Five laboratories combined an experimental efficiency calibration obtained from point standard sources and/or standard solutions in different measurement geometry with simulations for efficiency transfer in density (and geometry) obtained by dedicated software.

The combination of results from calibration methods based on different approaches to efficiency calibration, using different calibration sources and correction principles

mitigates undetected bias. Traceability of the used standards was either established in-house by primary standardisation of solutions, or by using solutions traceable to primary-standardised solutions from another NMI.

6.4 Evaluation of results

The characterisation campaign resulted in six datasets, which were evaluated according to ISO Guide 35. All individual results of the participants are displayed in tabular and graphical form in Annex 5.

It should be borne in mind that the method used in the characterisation is routinely applied for measuring ^{60}Co in different kind of matrices.

6.4.1 Technical evaluation

The obtained data were first checked for compliance with the requested analysis protocol and for their validity based on technical reasons. The following criteria were considered during the evaluation:

- appropriate validation of the measurement procedure
- compliance with the analysis protocol: measurements performed on at least two days and on two different samples
- absence of value given as below limit of detection or below limit of quantification
- analytical method used is selective for ^{60}Co and can produce results fulfilling the traceability criteria (Section 9.1)

Based on the above criteria, supported by a visual inspection of the data, all datasets could be included in the evaluation of the results.

6.4.2 Statistical evaluation

The results accepted based on the technical evaluation were tested for normality of dataset means using kurtosis and skewness tests and were tested for outlying means using the Grubbs test and using the Cochran test for outlying standard deviations (both at a 99 % confidence level). No outlying results could be detected so, all the datasets could be included in the evaluation. Standard deviations (SD_{between}) of the laboratory means were calculated. The results of these evaluations are shown in Table 11.

Table 11. Statistical evaluation of the technically accepted results, with p the number of technical valid datasets

^{60}Co	***p***	**Outliers**		**Normally distributed**
		Means	**Variances**	
Batch 1	6	none	none	yes
Batch 2	6	none	none	yes

There is no evidence that the laboratory means do not follow a normal distribution. As all measurement methods were found technically sound, all results were retained.

The individual uncertainty budgets are not taken into account for uncertainty related to the characterisation. The uncertainty related to the characterisation u_{char} is estimated, by taking the standard deviation of the laboratory means divided by the square root of the number of laboratories (p) (i.e. the standard error of the mean of the lab means). Since

no common calibrants were used, and the analysis procedures were different due to the largely varying approaches to efficiency calibration in gamma-ray spectrometry the results of all laboratories can be considered independent of each other.

$$u_{char} = \frac{SD\ between}{\sqrt{p}}$$ **Equation 7**

The final results for the massic activities at the reference date of 1st of November 2017 00h00 CET, standard deviations and combined standard uncertainties of the characterisation study are given in Table 12. The calculations were repeated by calculating the weighed mean and the power moderated mean. These calculations gave comparable results than the arithmetic mean. As the results are from experts laboratories using the same analytical technique having the same uncertainty components, it was opted to use results from the arithmetic mean calculations, giving an equal weight to all results used in the calculations.

Table 12. Results of the characterisation at reference date 1 November 2017, 00h00 CET

^{60}Co	***p***	**Mean [Bq kg^{-1}]**	***SD*** **[Bq kg^{-1}]**	**u_{char} [Bq kg^{-1}]**	**$u_{char,rel}$ [%]**
Batch 1	6	177	4	2	1.1
Batch 2	6	1301	24	10	0.77

7 Value Assignment

Certified values were assigned.

Certified values are values that fulfil the highest standards of accuracy. Procedures at JRC-Geel require pooling of not less than six datasets to assign certified values. Full uncertainty budgets in accordance with the 'Guide to the Expression of Uncertainty in Measurement' were established.

7.1 Certified values and their uncertainties

The unweighted mean of the means of the accepted datasets as shown in Table 12 was assigned as certified value for each parameter.

The assigned uncertainty consists of uncertainties related to characterisation, u_{char} (Section 6), potential between-unit inhomogeneity, u_{bb} (Section 5) and potential degradation during transport (u_{sts}) and long-term storage, u_{lts} (Section 5). The within-unit inhomogeneity is not taken into account as an entire disk shall be used for the measurements. The different contributions were combined to estimate the expanded, uncertainty of the certified value ($U_{CRM,}$) with a coverage factor k as:

$$U_{CRM} = k\sqrt{u_{char}^2 + u_{bb}^2 + u_{sts}^2 + u_{lts}^2} \qquad \textbf{Equation 8}$$

- u_{char} was estimated as described in Section 7
- u_{bb} was estimated as described in Section 5
- u_{sts} was estimated as described in Section 6
- u_{lts} was estimated as described in Section 6

Due to the limited degrees of freedom of the different uncertainty contributions to the determination of the massic activity of ^{60}Co, a coverage factor k of 2.57 was applied to obtain the expanded uncertainties corresponding to a level of confidence of about 95 % (for a conservative estimate of 5 degrees of freedom). The certified values and their uncertainties are summarised in Table 13.

Table 13. Certified values and their uncertainty component values and expanded uncertainty at reference time 1 November 2017, 00h00 CET

	Certified value [Bq kg^{-1}]	u_{char} [Bq kg^{-1}]	u_{bb} [Bq kg^{-1}]	u_{sts} [Bq kg^{-1}]	u_{lts} [Bq kg^{-1}]	u_{CRM} (k=1) [Bq kg^{-1}]	U_{CRM} (k=2.57) [Bq kg^{-1}]	$U_{CRM,rel}$ (k=2.57) [%]
Batch 1 EURM 800	177	2	0.5	0.5	1.1	2.3	5.9	3.3
Batch 2 EURM 801	1301	10	3.6	3.9	7.8	13.8	35	2.6

8 Metrological traceability and commutability

8.1 Metrological traceability

8.1.1 Identity

^{60}Co is a chemically and physically clearly defined analyte. The participants did not have to perform sample preparation for the final determination. These materials are structurally defined.

8.1.2 Quantity value

Only validated methods were used for the determination of the assigned values. Different methods and different calibrants of specified traceability of their assigned values were used and all relevant input parameters were calibrated. The individual results are therefore traceable to the SI. This is confirmed by the agreement among the technically accepted datasets. As the assigned values are combinations of agreeing results individually traceable to the SI, the assigned quantity values themselves are traceable to the SI as well.

8.2 Commutability

Many measurement procedures include one or more steps, which are selecting specific analytes (or specific groups of analytes) from the sample for the subsequent steps of the whole measurement process. Often the complete identity of these 'intermediate analytes' is not fully known or taken into account. Therefore, it is difficult to mimic all the analytically relevant properties of real samples within a CRM. The degree of equivalence in the analytical behaviour of real samples and a CRM with respect to various measurement procedures (methods) is summarised in a concept called 'commutability of a reference material'. There are various definitions expressing this concept. For instance, the CLSI Guideline C-53A [13] recommends the use of the following definition for the term *commutability*:

> "The equivalence of the mathematical relationships among the results of different measurement procedures for an RM and for representative samples of the type intended to be measured."

The commutability of a CRM defines its fitness for use and, thus, is a crucial characteristic in case of the application of different measurement methods. When commutability of a CRM is not established in such cases, the results from routinely used methods cannot be legitimately compared with the certified value to determine whether a bias does not exist in calibration, nor can the CRM be used as a calibrant. For instance, CRMs intended to be used to establish or verify metrological traceability of routine radionuclide measurement procedures must be commutable for the routine radionuclide measurement procedures for which they are intended to be used.

The CRM was produced meeting well know technical specifications in controlled conditions. Since the method used in the characterisation of this CRM (gamma-ray spectrometry) is a method routinely applied for measuring ^{60}Co, the agreement of results demonstrates that the processing did not affect any properties relevant for the method and that the analytical behaviour will be the same as for a routine sample of metal or steel.

Nevertheless in gamma-ray spectrometry, sample density and composition influence the gamma-ray transmission and consequently the detection efficiency. These differences can be corrected for by calculating the correction factors and applying them. These corrections are well known and are implemented in different software packages. For other steel or metal having totally different constituents than this CRM the commutability in gamma-ray spectrometry shall be assessed.

9 Instructions for use

9.1 Safety information

The usual laboratory safety measures apply.

The activity of ^{60}Co in these material are below the exemption levels so the material can be transported freely and handled in the laboratory safely without radiological concerns. Nevertheless, since the material is a radioactive sample, external exposure should be kept to a minimum. The sample shall be used as one piece, thus all mechanical impact, transformations or manipulations of the material shall be avoided.

9.2 Storage conditions

The materials shall be stored at room temperature 18°C ± 5°C in normal laboratory conditions, in a closed container.

Please note that the European Commission JRC-Geel cannot be held responsible for changes that happen during storage of the material at the customer's premises.

9.3 Preparation and use of the material

No special preparation of the material is required. The material can be used as from the shelf.

9.4 Minimum sample intake

The units shall be used as one piece and not split into parts.

9.5 Use of the certified value

The main purpose of the material is to assess method performance, i.e. for checking accuracy of analytical results/calibration or to use it as a calibrant. As any reference material, it can also be used for control charts or validation studies.

Use as a calibrant

The disks can be used as a calibrant for gamma-ray spectrometry. The uncertainty of the certified value shall be taken into account in the estimation of the measurement uncertainty.

Comparing an analytical result with the certified value

A result is unbiased if the combined standard uncertainty of measurement and certified value covers the difference between the certified value and the measurement result.

For assessing the method performance, the measured values of the CRMs are compared with the certified values. The procedure is described here in brief:

- Calculate the absolute difference between the mean measured value and the certified value (Δ_{meas}).
- Combine the measurement uncertainty (u_{meas}) with the uncertainty of the certified value (u_{CRM}): $u_{\Delta} = \sqrt{u_{meas}^2 + u_{CRM}^2}$
- Calculate the expanded uncertainty (U_{Δ}) from the combined uncertainty ($u_{\Delta,}$) using an appropriate coverage factor, corresponding to a level of confidence of approximately 95 %
- If $\Delta_{meas} \leq U_{\Delta}$ no significant difference between the measurement result and the certified value exists, at a confidence level of about 95 %.

Note that for comparing the measurement results with the certified values the measurement results shall be corrected for decay to the reference date on the certificate (1st of November 2017 00h00 CET) as described in section six of the report.

Use in quality control charts

The materials can be used for quality control charts. Different CRM-units will give the same result as inhomogeneity was included in the uncertainties of the certified values.

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List of abbreviations and definitions

A	Activity of a radionuclide
A_t	Activity at time t
A_0	Activity at the reference time t_0
ANOVA	Analysis of variance
Bq	Becquerel
CET	Central European Time
cm	Centimeter
CMI	Czech Metrology Institute
CNC	Computer Numerical Control
Co	Cobalt
CRM	Certified reference material
CLSI	Clinical and Laboratory Standards Institute
ºC	Degrees Celsius
DFG	Decommissioning Funding Group
EU	European Union
EN	European Norm
g	Gram
GUM	Guide to the Expression of Uncertainty in Measurements
IAEA	International Atomic Energy Agency
IEC	International Electrotechnical Commission
ISO	International Organization for Standardization
JSI	Jožef Stefan Institute (Slovenia)
JRC	Joint Research Centre of the European Commission
k	Coverage factor
kg	Kilogram
λ	decay constant defined as $\ln(2)/T_{1/2}$
LEA	Laboratoire d'Etalons d'Activité
LNE-LNHB	Laboratoire National de métrologie et d'Essais – Laboratoire National Henri Becquerel (France)
LTST	Index denoting homogeneity results using data of the long-term stability study
m	Mass
Max	Maximum
Min	Minimum
mm	Milimeter
$MS_{between}$	Mean of squares between-unit from an ANOVA

MS_{within}	Mean of squares within-unit from an ANOVA
n	mean number of replicates per unit
$\nu_{MSwithin}$	degrees of freedom of MS_{within}
N	Number of laboratories participating in the characterisation study
n.a.	Not applicable
NIST	National Institute for standards and technology
NMI	National Metrology Institute
NPL	National Physics Laboratory
p	Number of technically valid data sets
PTB	Physikalisch-Technische Bundesanstalt
PWR	Pressurised water reactor
rel	Index denoting relative figures (uncertainties etc.)
RM	Reference material
RSD	Relative standard deviation
SAE	Society of Automotive Engineers
S_{bb}	Between-unit standard deviation; an additional index "rel" is added when appropriate
$S_{between}$	Standard deviation between groups as obtained from ANOVA; an additional index "rel" is added as appropriate
SCK•CEN	Studiecentrum voor kernenergie, Centre d'étude de l'énergy nucléaire (Belgium)
SD	Standard Deviation
SE	Standard Error of regression analysis
SI	International System of Units
STST	Index denoting homogeneity results using data of the short-term stability study
S_{wb}	Within-unit standard deviation; an additional index "rel" is added when appropriate
$T_{1/2}$	Half-life (in years)
t	Time
$\bar{t}$	Mean value of all time points
t_0	reference time as indicated on the certificate
t_i	time point *i* of measurement
t_{tt}	chosen transport time
t_{sl}	Chosen shelf life
u	Standard uncertainty
U	Expanded uncertainty
u_{bb}	Standard uncertainty related to a possible between-unit inhomogeneity; an additional index "rel" is added as appropriate
u_c	Combined standard uncertainty; an additional index "rel" is added as

	appropriate
u_{char}	Standard uncertainty of the material characterisation; an additional index "rel" is added as appropriate
u_{CRM}	Combined standard uncertainty of the certified value; an additional index "rel" is added as appropriate
U_{CRM}	Expanded uncertainty of the certified value; an additional index "rel" is added as appropriate
u_{Δ}	Combined standard uncertainty of measurement result and certified value
u_{lts}	Standard uncertainty of the long-term stability; an additional index "rel" is added as appropriate
u_{meas}	Standard measurement uncertainty, an additional index "rel" is added as appropriate
u_{sts}	Standard uncertainty of the short-term stability; an additional index "rel" is added as appropriate
$\bar{y}$	mean of all results of the homogeneity study

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Annexes

Annex 1: Results of the homogeneity measurements

The analytical procedure is described in section 5, using relative measurements results only, thereby eliminating the uncertainty contributions for absolute values and calibrations. The decay-corrected (to the reference time 1 November 2017, 00h00 CET) and mass-normalised net count rate from the ^{60}Co is used. The counting rate is corrected for differences in the physical dimensions of the disks using Monte Carlo simulations.

The units were analysed per batch and each unit was measured twice following the randomised scheme c a b a f e b g j d h c h I j f d I g e. The same scheme was followed for both batches. The count rates relative to the average count rate of all the measurements of the samples of the same batch are calculated and given in Table 14.

To prove the stability of the measurement set-up, a check source was measured daily in between the measurements of the samples. The 28 measurements of the check source have standard deviation of 0.53%. This standard deviation is comparable to the standard deviation from the count rates of the samples of the individual batches.

Table 14. Measurement results of the homogeneity study, the values are expressed as decay corrected and mass normalised ^{60}Co count rates

Sample code	sample number	Batch 1 Relative count rate	sample number	Batch 2 Relative count rate
c	67	1.001	367	1.012
a	5	0.994	305	1.006
b	44	1.001	344	0.999
a	5	0.998	305	0.997
f	160	0.998	460	0.997
e	136	1.009	436	1.002
b	44	0.990	344	1.003
g	180	1.004	480	1.001
j	269	0.998	569	0.996
d	113	0.994	413	0.996
h	214	1.000	514	0.998
c	67	0.998	367	1.003
h	214	1.010	514	0.985
i	233	1.001	533	1.004
j	269	0.996	569	0.996
f	160	1.010	460	1.000
d	113	0.999	413	1.001
i	233	0.998	533	0.996
g	180	0.986	480	1.007
e	136	1.015	436	1.001
average		1.000		1.000
SD		0.70%		0.55%

Figure 7 and Figure 8 show mass-normalised and decay-corrected count rates of ^{60}Co in the units of both batches. The results show the absence of any trend in both measurement and production sequence.

Figure 7. No evidence of trend in measurement sequence. The uncertainties are the combined uncertainties as calculated in Table 5.

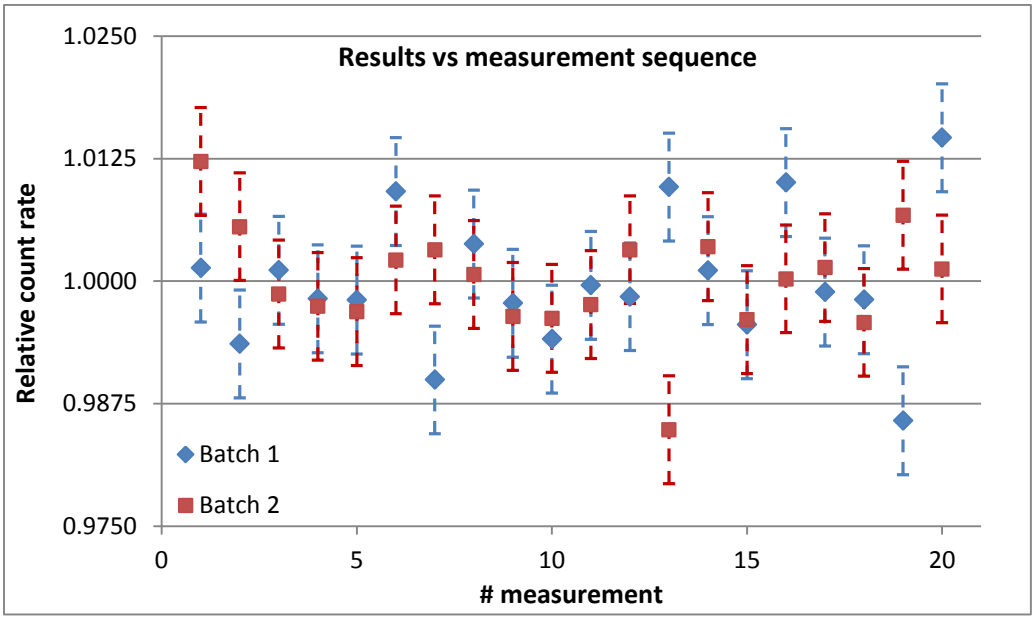
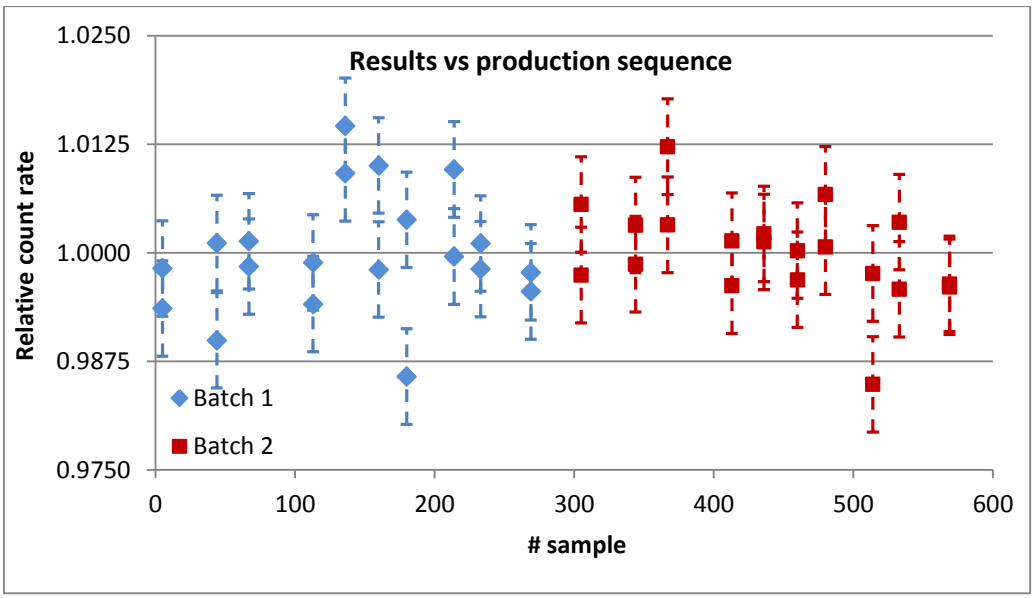
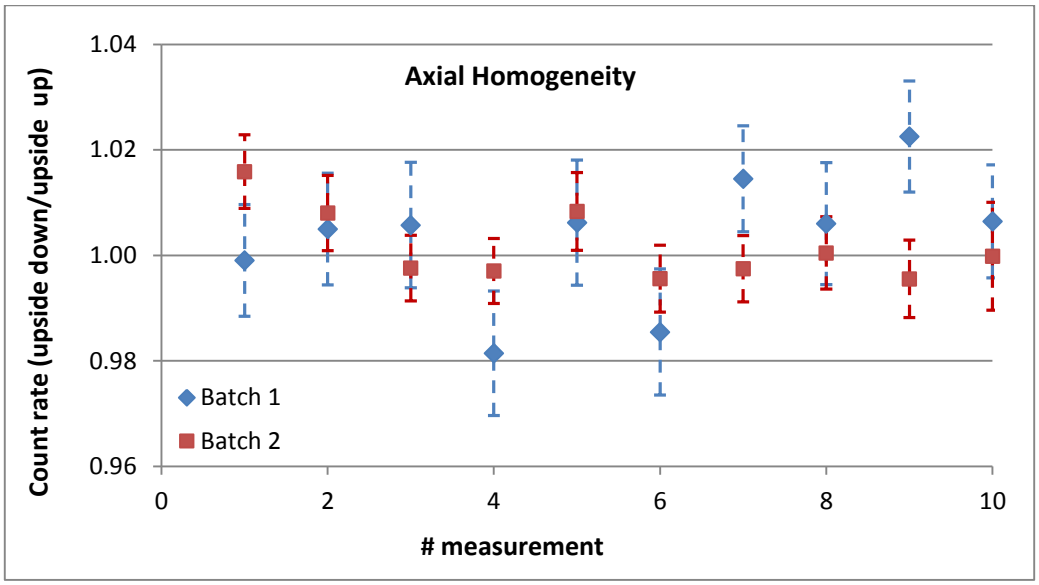


Figure 8. No evidence of trend in the production sequence. The uncertainties are the combined uncertainties as calculated in Table 5. For each sample the individual measurements are displayed.



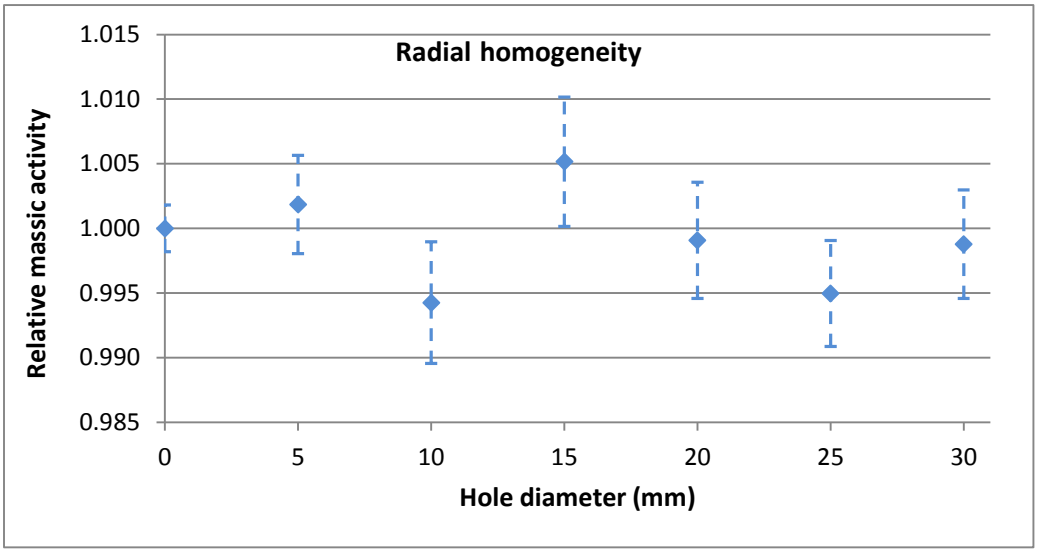
The axial homogeneity was assessed by measuring 10 disks upside up and upside down. Figure 9 displays the results, showing that there is no significant difference between the measurements upside up and upside down. The spread of data for batch 1 is bigger than for batch 2 due to poorer counting statistics.

Figure 9. Relative count rates of ^{60}Co of 10 different disks from each batch. The dashed lines correspond only to the uncertainty originating from the counting statistics (k=1)



The possible radial inhomogeneity was also investigated. One disk was measured entirely and remeasured after mechanically removing material in steps of 5mm starting from the middle of the disk (see Section 5.2). After each removal of 5 mm the disk was remeasured on the same detector at the same distance. The detection efficiency was recalculated for the different geometries and the massic activity of ^{60}Co was calculated. The results are given in Figure 10. No significant differences between the calculated activities of the disks of the different geometries could be observed.

Figure 10. Relative massic activities of 1 disk with an increasing hole made in the middle. The dashed lines correspond only to the uncertainty originating from the counting statistics (k=1)



Annex 2: Results of the short-term stability measurements

The analytical procedure applied was the same as for the characterisation study, but using relative measurements results only, thereby eliminating the uncertainty contributions for absolute values and calibrations. The decay corrected and mass-normalised net count rates in the ^{60}Co peaks are calculated for the different measurements. The short-term stability is assessed at a temperature of 4 and 60°C for a period of 1, 2, 3 and 4 weeks. For each temperature, one sample per batch and per period of storage time is measured before and after storage at the defined temperature and the count rates of ^{60}Co of both measurements is compared. The results of the short-term stability are displayed in Figure 11 and Figure 12.

Figure 11. Results of the short-term stability at 60°C. The dashed lines correspond only to the uncertainty originating from the counting statistics (k=1)

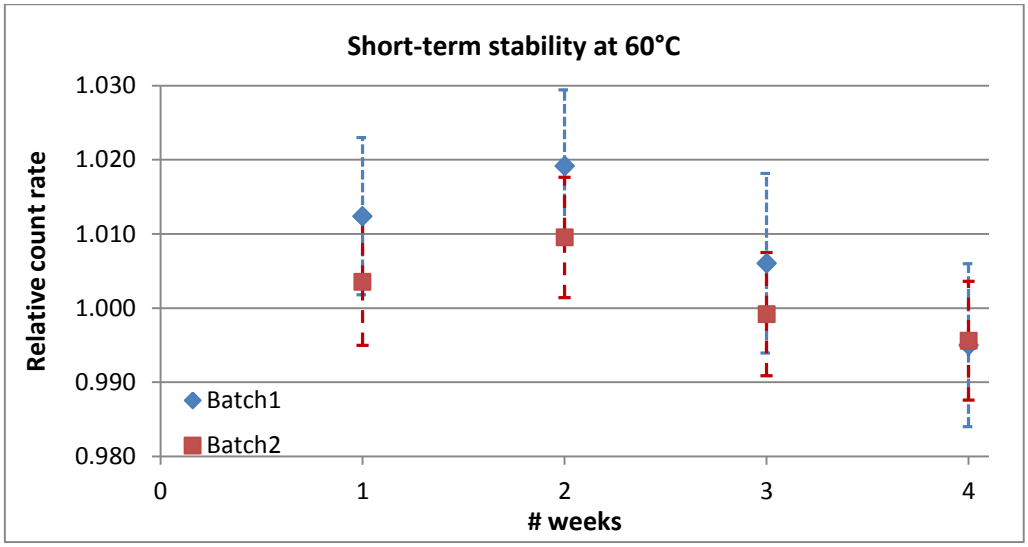
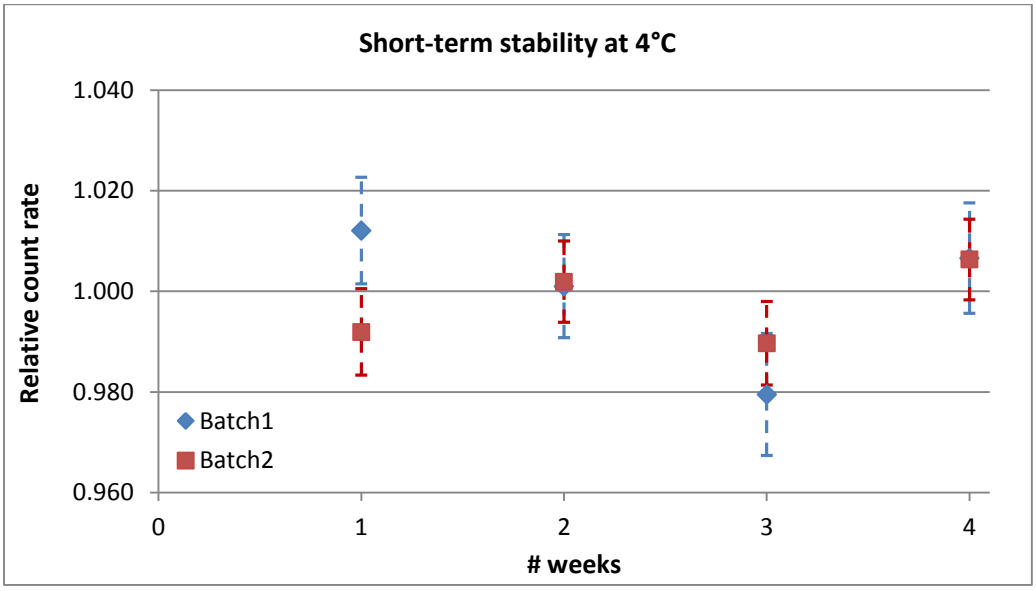


Figure 12. Results of the short-term stability at 4°C. The dashed lines correspond only to the uncertainty originating from the counting statistics (k=1)



Annex 3: Results of the long-term stability measurements

The long-term stability of stainless steel disks at normal shelf conditions is excellent as can be found in literature (Matweb.com, Azom.com). Moreover, stainless steel is used in many applications due to its excellent properties concerning stability.

To prove the long-term stability, two stainless steel disks measured in 2013 were re-measured in 2018. The decay corrected (to the reference date of 1 June 2013 00:00 CET) and mass normalised net count rates in the ^{60}Co peaks are calculated for the different measurements. The results of the long-term stability measurements are given in Table 15. They show excellent stability over a long term of five years as could be expected from literature.

Table 15. Results of the long-term stability measurements

	Disk 1	**Disk 2**
Count rate ^{60}Co June 2013	2.968	2.931
Count rate ^{60}Co June 2018	2.971	2.947
Ratio of count rates	1.001	1.006
Standard uncertainty (k=1)	0.006	0.006

Annex 4: Summary of methods used in the characterisation study

In Table 16 an overview of the different methods used for efficiency calibration and transfer in the gamma-spectrometric determination of ^{60}Co in stainless steel disks.

Table 16. Overview of the methods used for the analysis

Laboratory code	Method for efficiency calibration and transfer	Radionuclides used for calibration	Traceability	Corrections applied	Codes used for applied corrections
L01	Point and volume sources, efficiency transfer for geometry and density	^{60}Co	CMI and PTB standards traceable to SI via primary standards	True coincidence, Density, Geometry	EGSnrc
L02	volume and point sources	Multiple-nuclide containing ^{60}Co	Solutions traceable to SI via Deutschen Kalibrierdienst	True coincidence, Density, Geometry	GammaESP In house made software
L03	Volume source	Multiple- nuclide containing ^{60}Co	Primary standardisation of solutions in house	True coincidence, Density, Geometry	Efftran
L04	Calibration curve from 3 different volume sources and point sources	Radionuclides with 141 energy points containing ^{60}Co	Primary standard methods from LNE-LNHB	True coincidence, Density (with attenuation measurement) , Geometry	ETNA, Interwinner 7.0, Colegram 3.1
L05	Volume sources	Multiple- nuclide containing ^{60}Co	Solutions traceable to SI via Deutschen Kalibrierdienst	True coincidence, Density,	GESPECOR
L06	Volume and Point sources	Multiple- nuclide containing ^{60}Co	NPL and LEA standards traceable to SI via primary standards	True coincidence, Density, Geometry	GESPECOR LABSOCS

Annex 5: Results of the characterisation measurements.

Table 17 and Table 18 present the individual results, mean values and their combined standard measurement uncertainty (k=1) reported by the laboratories. All results are decay corrected to the same reference date of 1st November 2017.

Figure 13 and

Figure 14 are showing the individual results of the two replicates analysed by each laboratory with their standard uncertainties and the certified value with its standard uncertainty. The data of the different laboratories are considered to be independent as no common calibrants were used.

Table 17. Individual results of the laboratories for the samples from batch 1 (EURM 800). All analytical data and standard uncertainties (u) are expressed in Bq/kg

Batch 1 EURM 800						
Laboratory code	**Replicate 1**	**u**	**Replicate 2**	**u**	**Mean**	**u**
L01	179	2	180	1	180	2
L02	180	3	176	3	178	3
L03	169	3	168	3	169	3
L04	179	2	180	2	180	2
L05	178	4	180	4	179	4
L06	173	5	177	5	175	5

Figure 13. Laboratory mean values for batch 1 and the certified value (thick line) with their respective standard uncertainty (dashed line).

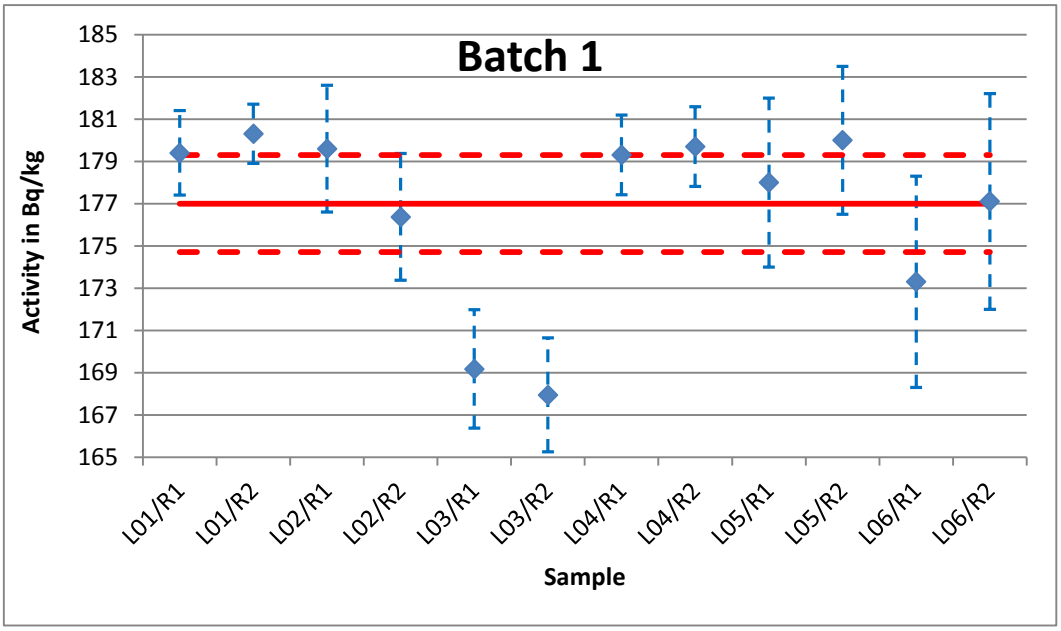
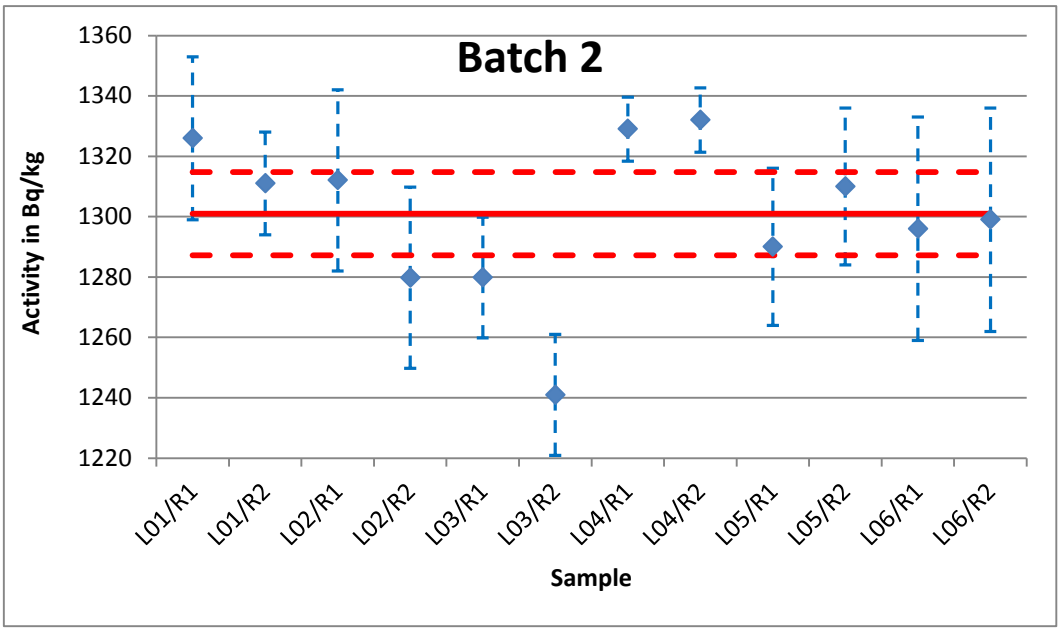


Table 18. Individual results of the laboratories for the samples from batch 2 (EURM 801). All analytical data and standard uncertainties (u) are expressed in Bq/kg

Batch 2						
Laboratory code	**Replicate 1**	**u**	**Replicate 2**	**u**	**Mean**	**u**
L01	1326	27	1311	17	1318	27
L02	1312	30	1280	30	1295	30
L03	1280	20	1241	20	1260	20
L04	1329	11	1332	11	1330	11
L05	1290	26	1310	26	1300	26
L06	1296	37	1299	37	1297	37

Figure 14. Laboratory mean values for batch 2 and the certified value (thick line) with their respective standard uncertainty (dashed line)



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KJ-NA-29790-EN-N

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doi:10.2760/90491

ISBN 978-92-76-08764-9