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Activity measurements of solid materials considered for recycling, re-use, or disposal as non-radioactive waste

Mesures d'activité de matériaux solides considérés comme déchets non radioactifs destinés à un recyclage, une réutilisation, ou une mise au rebut

Reference number
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Contents

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Foreword

ISO (the International Organization for Standardization) is a worldwide federation of national standards bodies (ISO member bodies). The work of preparing International Standards is normally carried out through ISO technical committees. Each member body interested in a subject for which a technical committee has been established has the right to be represented on that committee. International organizations, governmental and non-governmental, in liaison with ISO, also take part in the work. ISO collaborates closely with the International Electrotechnical Commission (IEC) on all matters of electrotechnical standardization.

Draft International Standards adopted by the technical committees are circulated to the member bodies for voting. Publication as an International Standard requires approval by at least 75 % of the member bodies casting a vote.

International Standard ISO 11932 was prepared by Technical Committee ISO/TC 85, *Nuclear energy*, Working Group WG2, *Radioactivity measurements*.

Annexes A and B form an integral part of this International Standard. Annex C is for information only.

Introduction

During the TC 85 Plenary Meeting in Paris (1988-10-13), it was decided to set up a new ad hoc working group "*Radioactivity Measurements*" to deal with, among other topics, the task to investigate in which areas measurements, in particular regarding low-level radioactivity, can be carried out and to what extent international standardization in this area is desirable. In view of the topics treated in other Technical Committees, for example radioactivity measurements in soil and water, a need exists for a standard on "Determination of the Decision Threshold and the Detection Limit for Ionizing Radiation Measurements". Moreover, a need for an International Standard directly related to the field of nuclear energy and, in particular, to the problem of recycling, re-use or disposal of materials from the dismantling of nuclear installations as inactive materials, was identified. Primarily, major nuclear installations for electricity production and to a lesser extent, installations like particle accelerators and reprocessing plants may be over thirty years old and therefore reaching the end of their scheduled lifetime. It is estimated that 51 commercial nuclear power plants were decommissioned in OECD countries between 1981 and 1995 and another 237 should be decommissioned during the following 15 years (reference [1] in annex C). Of the contaminated and activated metals arising from decommissioning, it has been calculated (reference [2] in annex C) that about 4 800 t of steel from a 1 000 MW(e) pressurized water reactor (PWR) would have a specific activity at, or below, 1 $Bq \cdot g^{-1}$. This specific activity limit is proposed as a possible limit for unrestricted recycling of steel scrap in the CEC. Consequently, in view of the quantities of scrap concerned and the fact that material released in one country for unrestricted use may enter other countries as ordinary scrap, international standardization of the radioactivity measurement procedures used to show compliance with release criteria is urgently needed.

Until now, the recycling or re-use of materials from nuclear installations is handled in individual countries using ad hoc criteria based on existing national legislations. However, international criteria for release of materials have been developed and are waiting to be adopted formally. For instance, a CEC Group of Experts recommended the following clearance levels for recycling of contaminated or activated steel (reference [2] in annex C) as mentioned above, and they are given here only as an example.

For beta/gamma activity:

— a specific activity limit of 1 Bq · g^{-1} averaged over a maximum mass of 1 tonne with no single piece exceeding 10 Bq · g^{-1};

— a surface activity limit of 0,4 Bq · cm^{-2} for non-fixed contamination on accessible surfaces, averaged over 300 cm^2 or over the whole area if it is less than 300 cm^2;

— for fixed contamination, the specific activity limit is assumed to be applied.

For alpha emitters:

— a surface activity limit of 0,04 Bq · cm^{-2} averaged over an area of 300 cm^2.

Other examples may be found in references [3] and [4] in annex C. In addition, release criteria are sometimes based on total activity.

This International Standard concerns radioactivity measurements in materials, to show compliance with release criteria as mentioned above or criteria of similar magnitude, as defined by national or international authorities. A review of experience in this field is given in reference [5] in annex C.

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Activity measurements of solid materials considered for recycling, re-use, or disposal as non-radioactive waste

1 Scope

This International Standard specifies basic guidance and methods for activity measurements of materials to be released for recycling, re-use or disposal as non-radioactive waste arising from the operation and decommissioning of nuclear facilities, in order to show compliance with established criteria for unrestricted release. It does not apply to ordinary radioactive waste.

2 Normative references

The following standards contain provisions which, through reference in this text, constitute provisions of this International Standard. At the time of publication, the editions indicated were valid. All standards are subject to revision, and parties to agreements based on this International Standard are encouraged to investigate the possibility of applying the most recent editions of the standards indicated below. Members of IEC and ISO maintain registers of currently valid International Standards.

ISO 4037:1979, *X and γ reference radiations for calibrating dosemeters and doseratemeters and for determining their response as a function of photon energy.*

ISO 6980:1984, *Reference beta radiations for calibrating dosemeters and doseratemeters and for determining their response as a function of beta radiation energy.*

ISO 7503-1:1988, *Evaluation of surface contamination — Part 1: Beta-emitters (maximum beta energy greater than 0,15 MeV) and alpha-emitters.*

ISO 7503-2:1988, *Evaluation of surface contamination — Part 2: Tritium surface contamination.*

ISO 7503-3:1996, *Evaluation of surface contamination — Part 3: Isomeric transition and electron capture emitters, low energy beta emitters (maximum beta energy less than 0,15 MeV).*

ISO 8769:1988, *Reference sources for the calibration of surface contamination monitors — Beta-emitters (maximum beta energy greater than 0,15 MeV) and alpha-emitters.*

ISO 11929-1:1996, *Determination of the lower limits of detection and decision for ionizing radiation measurements — Part 1: Fundamentals and applications to counting measurements without the influence of sample treatment.*

ISO 11929-2:—[1]), *Determination of the lower limits of detection and decision for ionizing radiation measurements — Part 2: Fundamentals and applications to counting measurements with the influence of sample treatment.*

ISO 11929-3:—[1]), *Determination of the lower limits of detection and decision for ionizing radiation measurements — Part 3: Fundamentals and applications to counting measurements by high-resolution gamma spectrometry without the influence of sample treatment.*

IEC 325:1981, *Alpha, beta and alpha-beta contamination meters and monitors.*

IEC 846:1989, *Beta, X and gamma radiation dose equivalent and dose equivalent rate meters for use in radiation protection.*

IEC 1017-1:1991, *Portable, transportable or installed X or gamma radiation ratemeters for environmental monitoring — Part 1: Ratemeters.*

3 Definitions

For the purposes of this International Standard, the following definitions apply.

3.1 activity (of an amount of a radionuclide in a particular energy state at a given time)**:** Quotient of the expectation value of the number of spontaneous nuclear transitions, dN, from that energy state and the time interval, dt.

The special name for the SI unit of activity is the becquerel (Bq) (1 Bq = 1 s^{-1}).

3.2 specific activity: Activity of a specified material divided by its mass.

It is expressed in becquerels grams to the power of minus one ($Bq \cdot g^{-1}$).

NOTE — The term "mass activity concentration" is used sometimes in other publications (see reference [2] in annex C) but ISO terminology is used throughout this International Standard.

3.3 surface contamination: Contamination of surfaces with radioactive substances.

3.4 surface activity: Ratio between the activity of the radionuclides present on a surface and the area of that surface.

It is expressed in becquerels centimetres to the power of minus two ($Bq \cdot cm^{-2}$).

3.5 directly measurable surface contamination: Fraction of the surface contamination available for direct measurement.

3.6 removable surface contamination: Fraction of surface contamination which is removable or transferable under normal working conditions.

3.7 indirect evaluation of removable surface contamination: Evaluation of the removable activity on a surface by means of a smear sample.

3.8 smear test: Taking of a sample of removable activity by wiping the surface with dry or wet material and the subsequent evaluation of the activity transferred to the material used to wipe the surface.

3.9 removal factor, F: Ratio of the activity removed from the surface by one smear sample to the activity of the removable surface contamination prior to the sampling.

1) To be published.

3.10 instrument efficiency, ε_i: Ratio between the instrument net reading (counts per unit time) and the surface emission rate of the source (particles or photons emitted per unit time) in a specified geometry relative to the source.

NOTE — The instrument efficiency depends on the energy of the radiations emitted by the source.

3.11 contamination source efficiency, ε_s: Ratio between the surface emission rate and the number of particles or photons of the same type created or released within the source per unit time.

NOTE — According to this definition, the efficiency of a source would be expected to be less than 0,5, since emission occurs from the front face only; however, a contribution due to back-scattered particles may enhance this value.

3.12 surface emission rate of a source: Number of particles or photons of a given type above a given energy emerging from the face of the source or its window per unit time.

4 Requirements for activity measurements related to unrestricted release

4.1 General

Radioactivity measurements related to unrestricted release of solid materials to be treated in this standard deal with

— surface contamination measurements;

— specific activity measurements;

— dose-rate measurements;

— total activity estimates.

These release criteria could be, for instance:

surface contamination:	between 0,4 $Bq \cdot cm^{-2}$ and 4,0 $Bq \cdot cm^{-2}$ for beta/gamma emitters or between 0,04 $Bq \cdot cm^{-2}$ and 0,4 $Bq \cdot cm^{-2}$ for alpha emitters (averaged over areas between 100 cm^2 and 1 m^2);
specific activity:	range from 0,1 $Bq \cdot g^{-1}$ to 10^4 $Bq \cdot g^{-1}$ (limitations set also to local and average values);
dose rate:	from 0,05 $\mu Gy \cdot h^{-1}$ to 1 $\mu Gy \cdot h^{-1}$(above local background, dose rate near surface).

Criteria for unrestricted use of solids to be released for recycling, re-use or disposal and the combination rules in their applicabilities are set by national authorities.

4.2 Surface contamination measurements

4.2.1 Radionuclides to be considered

The radionuclides of concern encountered during operation and decommissioning will depend strongly on the type of nuclear installation (e.g. power reactor, enrichment plant, accelerator, fuel fabrication plant) and will differ from one nuclear installation to another. Whatever the installation, the radionuclide mixture must be known before starting a large-scale programme of surface contamination monitoring, since the response of the survey instrument depends on the radionuclide mixture. Therefore, the mixture of contaminating radionuclides shall be determined for each part of the plant, unless it concerns an installation with one known single contaminant [e.g. natural uranium oxide (UO_2)]. Such laboratory measurements are an essential precursor to provide "fingerprint" information for field measurements.

4.2.1.1 Determination of radionuclide mixture

The composition of the radionuclide mixture can be determined by one or both of the following methods:

— High-resolution gamma and X-ray spectrometry using, for example, Ge(Li) or high-purity germanium detectors for gamma radiation and Si(Li) or high-purity planar germanium detectors for X and soft gamma radiations for the energy range of 5 keV to 50 keV.

— Radioanalysis for low-activity samples using methods such as fixation on "carriers", radiochemical separation to isolate, in particular, radionuclides which cannot be measured by gamma-ray spectrometry (see annex B).

Calculations may be used to supplement the measurements, but only when the accuracy of the calculation has been confirmed by spot measurements and only in situations where activity from bulk activation dominates the activity inventory. They cannot be used whenever contamination is important.

4.2.2 Methods to determine surface contamination

4.2.2.1 General

Surface contamination can be determined by direct or indirect measurements (see also ISO 7503-1, ISO 7503-2 and ISO 7503-3). Direct measurements are carried out with contamination meters and monitors. These detectors measure both fixed and removable surface contamination. Indirect measurements are carried out using smear tests to determine the removable surface contamination.

Direct measurements may sometimes be difficult, or impossible, if inactive solid or liquid deposits are present on the surface, or if the measurements are influenced by high background-radiation levels due to, for example, activation of the objects to be checked, or if the surface to be checked is not accessible for an instrument.

Indirect methods (smear tests) can only be used to determine the non-fixed contamination level with an uncertainty in the removal fraction. Certain exemption-criteria proposals (reference [2] in annex C) recommend, however, clearance levels for non-fixed contamination only on accessible surfaces. In this case, direct measurements may result in an overestimation and smear tests are therefore more appropriate. In many cases, a combination of both methods will lead to the most reliable results. Smear tests may be ineffective for the determination of tritium (see also 4.2.2.5).

4.2.2.2 Direct measurements of surface contamination

4.2.2.2.1 Measuring instruments

The characteristics and performance of the measuring instruments shall be in accordance with IEC 325. The instruments shall be capable of detecting activities below the level of the surface-contamination release criteria as defined by international or national regulations. Guidance on the detection limit may be obtained from ISO 11929-1, ISO 11929-2 and ISO 11929-3.

4.2.2.2.2 Detection procedure

The detector is moved as close as practicable over a surface. Once a contaminated area is detected, the detector shall be positioned over this area and held stationary for a sufficient length of time to confirm the measured value of the contamination. The speed of movement shall be commensurate with the contamination limit and the performance characteristics of the detector.

4.2.2.2.3 Measurement procedure

When making a measurement, the operating instructions relating to the measuring instrument used and the following requirements shall be complied with.

a) The background count rate shall be determined at a place representative of the area to be surveyed.

b) The background count rate shall be checked regularly.

c) The correct functioning of the instrument should be verified using a suitable check source (daily for instruments used frequently). Deviations of more than 25 % from the agreed value shall give rise to a recalibration of the instrument.

d) Removable spacers may be required to keep the distance between the detector and surface as small as possible.

e) The detector shall be kept in position for at least three times the response time of the instrument (indication 95 %).

f) The instrument efficiency for the radionuclides to be measured shall be known over the anticipated range of environmental conditions.

g) The effect of the shape of the surface of the objects to be checked on the instrument efficiency shall be evaluated, in case the surface is not flat (examples can be found in reference [6] in annex C).

h) The effect on the contamination source efficiency, ε_S, of visible layers of dirt and/or oxidation on the surface of the objects to be checked shall be taken into account, in case these layers cannot be removed. Correction factors are given in annex A for various radionuclides, as a function of surface density of the absorbent layer.

According to ISO 7503-1 the surface activity, alpha or beta, A_S, of the fixed and non-fixed contamination in $Bq \cdot cm^{-2}$ is given by the following equation:

$$A_S = \frac{(n - n_B)}{\varepsilon_i \times \varepsilon_S \times W} \qquad \ldots (1)$$

where

n is the total count rate, in reciprocal seconds;

n_B is the background count rate, in reciprocal seconds;

ε_i is the instrument efficiency for alpha or beta radiation;

W is the surface of the detector window, in square centimetres;

ε_S is the efficiency of the contamination source.

In the absence of known values, ε_S can be taken as

$\varepsilon_S =$ 0,5 [beta emitters ($E_\beta \geqslant$ 0,4 MeV)]

$\varepsilon_S =$ 0,25 [beta emitters (0,15 MeV $< E_\beta <$ 0,4 MeV) and alpha emitters]

where E_β is the maximum beta particle energy.

The possibility of underestimating the alpha contamination is discussed in ISO 7503-1.

4.2.2.3 Indirect measurements of surface contamination

4.2.2.3.1 Measuring instruments

Smear samples should preferably be measured with well-shielded fixed counting equipment, such as alpha/beta proportional counters, gamma spectroscopy systems and liquid scintillation counting systems. If portable contamination monitors and meters are used, they shall be in agreement with IEC 325. The instruments shall be able to determine the activity removed, such that surface activity can be easily determined as being below the clearance criteria for surface activity concentration, as defined by national or international regulations.

NOTE — Most instruments are capable of detecting less than 0,4 Bq for alpha contamination and less than 4 Bq for beta contamination. This implies that for a smear sample covering 100 cm^2, and assuming a removal factor $F = 0,1$, it is possible to measure a non-fixed contamination of less than 0,04 $Bq \cdot cm^{-2}$ for alpha emitters and of less than 0,4 $Bq \cdot cm^{-2}$ for beta emitters, these being, for example, the CEC proposed clearance levels for surface activity concentrations.

4.2.2.3.2 Measurement procedure

After taking a smear sample according to the guidelines given in ISO 7503-1, for a surface usually of 100 cm^2 but, where regulations permit larger areas to be smeared (e.g. the CEC proposal is 300 cm^2), the surface activity, A_S, is determined as follows:

$$A_S = \frac{(n - n_B)}{\varepsilon_i \times \varepsilon_s \times F \times S} \qquad \ldots (2)$$

where

n is the total count rate, in reciprocal seconds;

n_B is the background count rate, in reciprocal seconds;

ε_i is the detection efficiency of the instrument for alpha or beta radiation;

F is the removal factor;

S is the area smeared, in square centimetres;

ε_s is the contamination source efficiency.

F shall be determined experimentally for the variety of surfaces encountered in decommissioning activities. Otherwise, a value of $F = 0{,}1$ shall be used.

4.2.2.4 Calibration of instrument efficiency

All measuring instruments used in contamination measurements shall be calibrated using reference sources, as defined in ISO 6980 and ISO 8769, following the procedure given in ISO 7503-1.

4.2.2.5 Tritium surface contamination

Tritium contamination is usually of no concern during the decommissioning of nuclear installations. Moreover, it is of low radiotoxicity. It may be present in, for example, reinforced concrete of power reactors as an activation product, or in fusion plants. Whenever tritium surface contamination is of concern, it shall be evaluated according to the procedures specified in ISO 7503-2.

4.2.2.6 Documentation of contamination measurements results

For the purpose of unrestricted release of materials from decommissioning of nuclear facilities, contamination measurements shall be reported as follows:

a) date;

b) release criteria used;

c) location and sub-location;

d) type of surface for indirect method;

e) smear material (dry or wet);

f) wetting agent;

g) removal factor for indirect evaluation (measured or assumed);

h) instrument used.

4.3 Specific activity measurements

4.3.1 General

Specific activity measurements require knowledge of the radionuclide mixture, which can be determined by spectrometry and radiochemical analysis. It is important to determine the ratio of radionuclides difficult or impossible to measure by spectrometry, such as ^{55}Fe, ^{63}Ni or ^{241}Pu, to those radionuclides, such as ^{137}Cs or ^{60}Co, which are easy to detect. Radionuclide ratios shall be determined for each category of material to be released for recycling, re-use or disposal without restriction. Such measurements shall take place after decontamination (if applicable) and before the material leaves the site of the nuclear installation.

4.3.2 Measurement techniques

4.3.2.1 Laboratory measurements

The composition of the radionuclide mixture shall be determined for each survey unit by taking core samples for spectrometric or radiochemical methods. The detection methods chosen shall be such that all radionuclides concerned can be identified.

The size of the samples taken shall be sufficiently large to make a quantitative analysis of the radionuclide mixture with a 95 % confidence level for the total activity in the sample, for samples of specific activity at the release criterion specified by national or international regulations (e.g. 1 Bq·g^{-1} for unrestricted release of steel scrap proposed in the CEC). Corrections shall be applied for self-absorption in the sample.

Preferably, samples should be prepared such that they are measured in a geometric and spatial activity distribution corresponding to those in the calibration sources.

During sampling, care shall be taken to avoid changes in the radionuclide mixture and the activity (e.g. evaporation at the surface during flame cutting). For surface contamination, material samples shall be taken at positions where the activity has penetrated the material (e.g. welding spots).

Detection limits for spectrometric methods are derived as in ISO 11929-1, ISO 11929-2 and ISO 11929-3. Methods and detectors for gamma and alpha spectrometry are also given in ISO 11929-1, ISO 11929-2 and ISO 11929-3. Radiochemical methods to determine the common radionuclides ^{55}Fe and ^{63}Ni in embedded waste are described in annex B.

4.3.2.2 Field measurements

Field or *in situ* measurements are normally made using dose-rate and/or count-rate instruments. Portable X- and gamma-radiation ratemeters covering the range of 10 nGy·h^{-1} to 10 µGy·h^{-1} shall be used. Count-rate instruments shall cover at least the ranges given in 4.1 for alpha and beta/gamma emitters respectively. They shall fulfil the requirements laid down in IEC 846 or IEC 1017-1.

The dose rate (or count rate), after subtraction of background indicated by the instrument in contact with the item to be checked, shall be related to the specific activity measured as in 4.3.2.1, at the position where the sample was taken. The conversion factor relating net dose rate (or count rate) to specific activity shall be used to show compliance with release criteria for a sufficiently large number of measuring points (see 4.4 for the statistical base of sampling).

Independent of the specific activity release criteria, other criteria may concern dose-rate limits at a given distance from the item. Such measurements can be carried out with the same instrument as used for the specific activity measurements, provided the size of the detection subassembly is small compared to the distance specified for the release criterion.

4.3.2.3 Total gamma-activity measurements

Special measurement systems, using liquid or plastic scintillation counters with large surfaces positioned in, for example, 4π geometry, enable large volumes of material to be measured using short counting intervals. In this case, the total gamma activity is determined. Such systems shall be capable of measuring the gamma rays emitted by the material to be checked with sufficient detection probability.

The shape of the material to be checked, its positioning in the measuring volume, as well as its self-shielding effect, may influence the result of the measurement. Therefore, the material batches shall be selected according to their type (e.g. objects with the same wall thickness), such that batches with similar geometry and self-shielding characteristics are used, for which the calibration of the instrument has to be established. Once the total gamma activity has been determined from the measurement, the specific activity can be determined after measuring the mass of the batch.

WARNING — The shielding effect caused by the presence of the material batch in the measuring position may reduce the background count rate. This effect has to be determined using batches of identical non-radioactive material.

The interpretation of the data derived shall be done cautiously, since small variations in the objects and positions of contamination can have a dramatic input on the evaluation of the radiation measured, specifically where low-energy emitters are concerned.

4.3.2.4 Background requirements

All measurements designed to validate release criteria shall be made in areas where the background radiation is as low as can be achieved on the site, or where the minimum detectable specific activity is below the release criteria with 95 % confidence. For *in situ* measurements of, for example, concrete structures, the background dose rate should be taken as the pre-operational dose rate when it is applicable or, if this is not the case, it can be taken from measurements on similar structures in uncontaminated parts of the facility. Mobile items shall be brought to a shielded area with a background dose rate not exceeding 100 nGy·h^{-1}. Relations between counting time, threshold and background, as defined in ISO 11929-1, ISO 11929-2 and ISO 11929-3 can be used.

4.3.2.5 Calibration of instruments

Field measurements shall be performed with portable dose-rate meters calibrated with X and gamma reference radiations as specified in ISO 4037. Special measurement systems, as defined in 4.3.2.4, shall be calibrated separately for each batch type of material using a reference measuring batch and a gamma reference source. The reference batch may consist of a non-radioactive batch of the material concerned or a calibration phantom.

The correct functioning of the instruments shall be checked regularly (daily for instruments in frequent use) using a suitable check source.

4.3.2.6 Documentation of specific activity measurement results

For the purpose of unrestricted release of materials from decommissioning of nuclear facilities, results of specific activity measurements shall be reported as follows:

a) date;

b) release criteria used;

c) location and sub-location;

d) type of material checked;

e) instrument used;

f) detection limit;

g) calibration date and reference source used;

h) background reading (dose rate or count rate);

i) total dose rate (count rate) measured;

j) conversion factor of dose rate (count rate) versus specific activity used;

k) specific activity (including composition of radionuclide mixture);

l) if the specific activity is higher than the release criterion applied, the date at which the specific activity will fall below the release limit;

m) name and signature of the person making the measurement.

4.4 Sampling strategy

Samples collected for radionuclide analysis shall be representative of the item to be released. Small items, for example pipes etc., shall be fully checked for activation and/or for surface contamination on both inner and outer surfaces. For large objects like floors, walls and roofs inside buildings, sample points shall have a statistically sound basis such that the results demonstrate compliance with an adequate level of confidence (95 %) for the minimum number of sampling points.

This International Standard uses the stratified random sampling approach described in reference [7] in annex C. It can be shown that the number of samples required for statistical acceptability is n, where

$$n \geqslant 45 \frac{s^2}{\overline{X}^2} \qquad \ldots (3)$$

where

s is the standard deviation of the sample;

$\overline{X}$ is the mean of the chosen parameter of the n samples.

With respect to floors, walls and ceilings inside buildings, they should be marked up into grids of a size consistent with the required averaging area and then numbered. A minimum of 30 initial random samples are taken from each grid, together with a measurement of the gamma dose rate 1 m above the centre of the grid. On the surface of each survey block, a number of contact measurements of surface-specific alpha activity, beta gamma dose rates and gamma dose rates are made at uniformly spaced points in the survey blocks. The number of contact measurements depends on the dimensions of the detector compared with the grid size and the variation in radioactivity across the block.

After completion of the sample analysis, the mean $\overline{X}$, and standard deviation, s, are calculated for each parameter associated with the samples. The number of samples, n required to obtain an accurate estimate of the mean is calculated from the equation

$$n = 45 \frac{s^2}{\overline{X}^2} \qquad \ldots (4)$$

Additional samples are needed if the calculated value of n is greater than 30 (the original number of samples taken).

When a sufficient number of samples has been taken, as indicated in equation (4), the mean, $\overline{X}$, and standard errors, $\overline{s}$, defined by

$$\overline{s} = \frac{s}{\sqrt{n}} \qquad \ldots (5)$$

are calculated for each parameter of interest. If the parameter values calculated as $\overline{X} + 2\overline{s}$ are below the prescribed limits, compliance with unrestricted release limits has been achieved to a confidence level of 95 %. The same approach should be used for bulky metallic items, provided the decontamination process has removed radioactive contamination in a uniform way.

Annex A
(normative)

Figures for measurements of beta surface contamination

In this annex, the effect of absorbent layers on the surface of the object, the distance between the detector and surface to be checked and the shape of the objects to be checked are illustrated in figures A.1 and A.2.

The normalized count rate of an Ar/CH_4 proportional counter with a surface layer of 0,9 mg · cm^{-2} and a surface of 112 cm^2, exposed to a thin surface contamination source of 100 cm^2 as a function of the surface density of the absorbent layer is given in figure A.1.

The thickness of the roughness of an unpolished metal surface is about half the average grain size, i.e. an approximate maximum value of 50 µm. Polished surfaces have a roughness thickness of a few micrometres. Assuming the surface density of grease or dust layers to be 0,5 mg · cm^{-2} leads to an absorbent layer of 2,5 mg · cm^{-2} in the first case and 0,7 mg · cm^{-2} in the second case.

As can be seen from figure A.1, the count rate decreases to as little as 25 % for ^{63}Ni, for a surface density of the absorbent layer of 2,5 mg · cm^{-2}.

The count rate versus distance between source and detector surface for a gas-filled proportional counter is given in figure A.2.

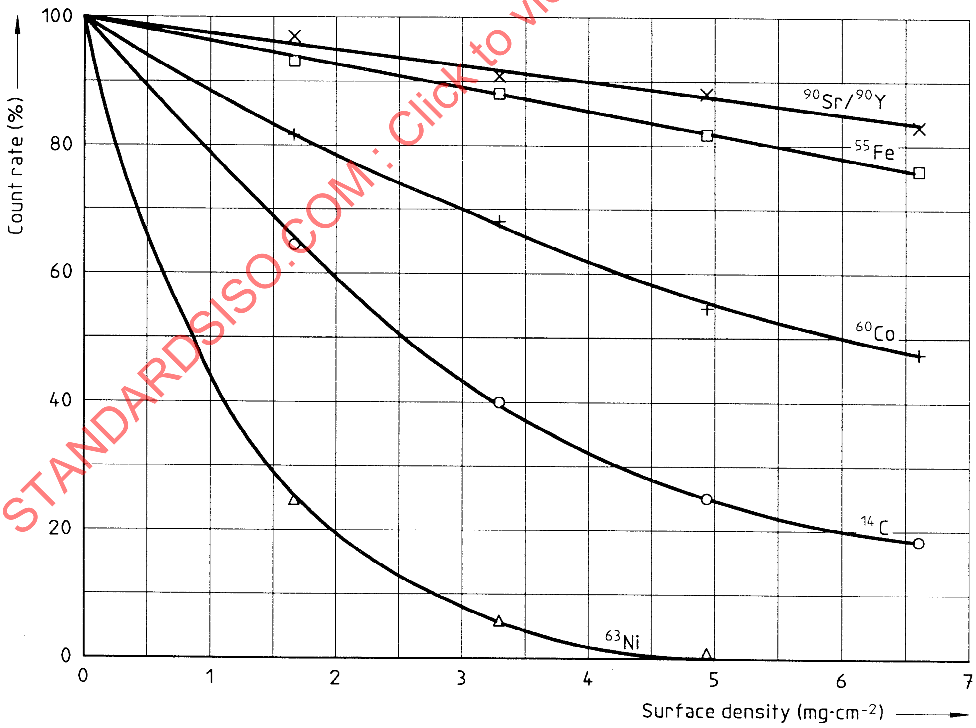
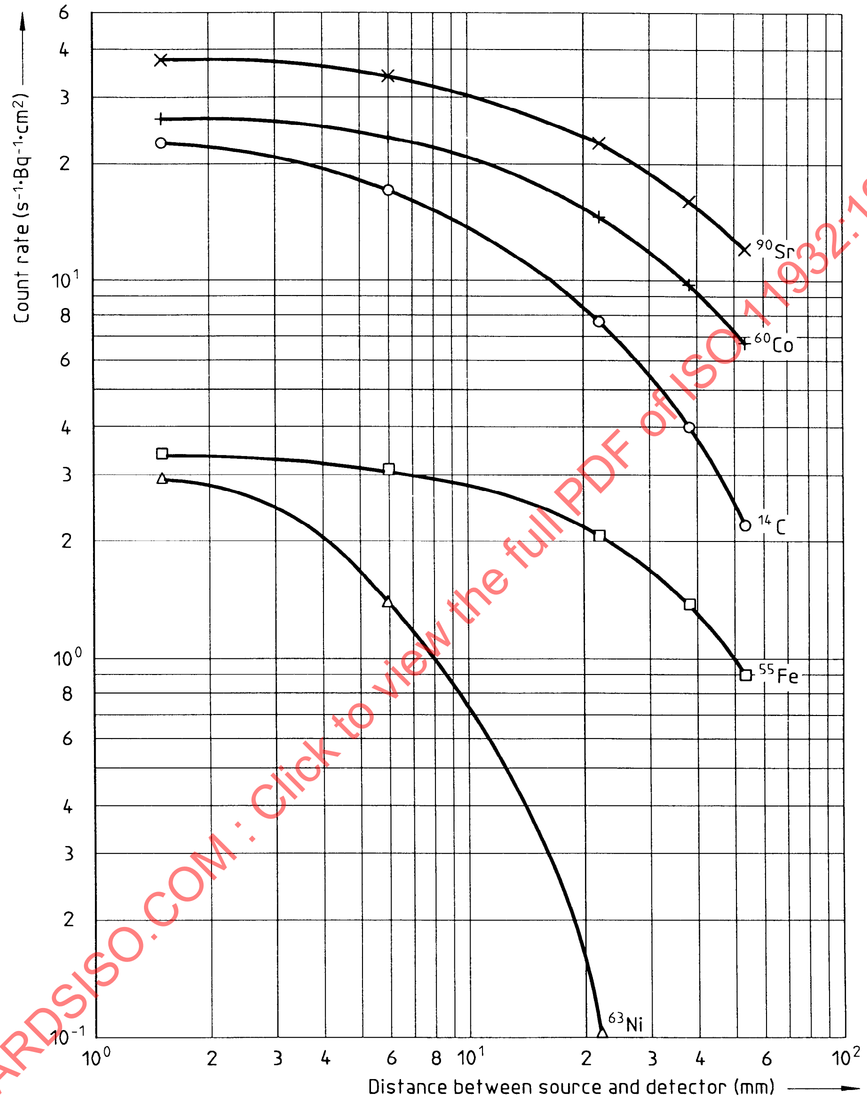


Figure A.1 — Normalized count rate versus surface density of absorbent layer



Surface calibration source 8 cm × 12,5 cm
Surface of detector 9,4 cm × 16,6 cm
Detector foil 0,3 mg · cm^{-2}
Gas Ar/CH_4

Figure A.2 — Count rate versus distance between source and detector surface

Annex B
(normative)

Low-level activity measurements of ^{55}Fe and ^{63}Ni

B.1 Introduction

This annex concerns two radionuclides which are listed as key nuclides. Their activity shall be measured if required by the safety authorities. Iron-55 is a low energy X-ray emitter (5,9 keV) and Nickel-63 is a pure beta emitter. They are not measurable directly when they are mixed with other radionuclides. Therefore, chemical extraction or isolation shall be used, so as to improve accuracy.

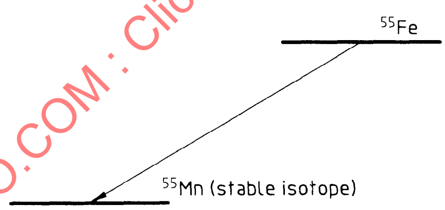
The proposed methodologies will allow low-level radioactivity measurements related to different forms of solid radioactive waste. Identical methodology used in different laboratories will allow comparisons and will improve the accuracy of the results.

B.2 Determination of iron-55

Iron is present in all the stainless steel components of nuclear reactors, which are activated by neutrons. Therefore iron-55 is commonly present in nuclear reactor waste and scrap materials.

Iron-55 has a long half-life ($T_{1/2}$ = 2,7 years).

Its decay scheme leads to electron capture (E = 232 keV) with the emission of a soft X-ray (E = 5,9 keV).



The soft X-ray emission will be used to determine the activity of this radionuclide quantitatively.

B.2.1 Reagents

B.2.1.1 Concentrated solution (10 g·l^{-1}) of Ni, Co, Ag, Cs.

B.2.1.2 Iron solution, as a hold-back carrier.

B.2.1.3 Concentrated ammonia and mineral acids, for example, sulfuric acid, nitric acid, perchloric acid, etc.

B.2.1.4 Nitric acid, 1 mol·l^{-1}.

B.2.1.5 Hydrochloric acid, 5,0 mol·l^{-1} and 0,5 mol·l^{-1}.

B.2.1.6 Organic anionic exchangers, quaternary ammonium type 1, DVB 4 %, 100 to 200 mesh.

B.2.2 Apparatus

B.2.2.1 Laboratory glassware, beakers, burettes, pipettes, etc.

B.2.2.2 pH-meter.

B.2.2.3 Centrifugal machine and/or **laboratory filtration systems.**

B.2.2.4 Atomic absorption spectrometer or **inductively coupled plasma (ICP) spectrometer.**

B.2.2.5 X-ray spectrometer or **liquid scintillation counter.**

B.2.2.6 Gamma-ray spectrometer.

B.2.3 Measurement of Iron-55

Due to its nuclear properties, iron-55 may be measured by at least two techniques which involve chemical separation, as follows.

a) X-ray spectrometry.

   This technique is mostly used for samples which are coloured or contain enough activity, i.e. more than 100 $Bq \cdot l^{-1}$ (reduced methodology).

b) Liquid scintillation counting.

   X-ray or Auger electrons act in a liquid scintillator, with photon emission, as a weak beta emitter. This technique is used for samples with an iron activity from 1 $Bq \cdot l^{-1}$ to 100 $Bq \cdot l^{-1}$, and for which transuranium emitters shall be isolated (general methodology). This methodology is always used when the sample contains cobalt, nickel, silver, caesium or tritium.

B.2.3.1 Reduced methodology

Directions for use are given in figure B.1.

Solid samples are dissolved according to their nature. In general, considerable volumes of concentrated mineral acids (B.2.1.3) are used.

The global iron concentration is measured by an atomic absorption or ICP spectrometer (B.2.2.4). The solution's iron concentration shall be about 25 $mg \cdot l^{-1}$. If the iron concentration is lower than this value, a known addition shall be made.

The ^{55}Fe separation rate is defined by determination of iron before and after separation. The eventual presence of radioactive impurities carrying over during ^{55}Fe separation are checked by a gamma-ray spectrometer (B.2.2.6).

Additions of nickel, cobalt and silver, that are not precipitated by ammonia at pH 10, are made to improve the decontamination factor of these elements. The additions shall be made until a level of 20 $mg \cdot l^{-1}$ is reached before precipitation.

Precipitate separation by centrifugation is used for the first two additions, and by both centrifugation and filtration for the third addition.

Afterwards, nitric acid (B.2.1.4) is used for the solubilization of the precipitate of iron (III) hydroxide.

The nitric acid solution is then used for chemical yield determination (> 95 %) and for specific ^{55}Fe activity determination by X-ray spectrometry.

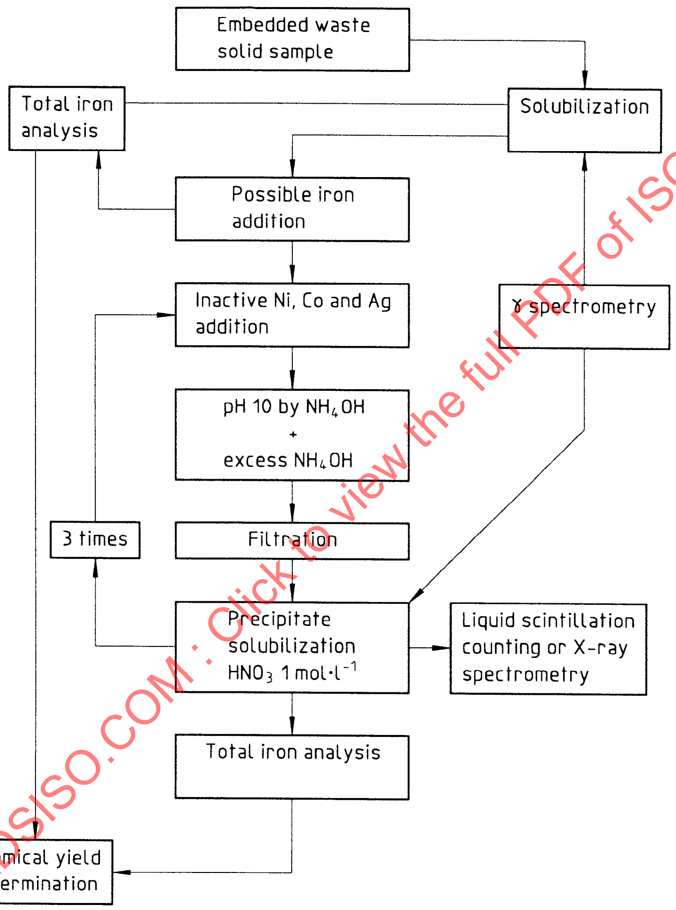


Figure B.1 — Reduced methodology for specific ^{55}Fe separation

B.2.3.2 General methodology

Directions for use are given in figure B.2.

The beginning is the same as for the reduced methodology. The third precipitate is dissolved by 5,0 mol · l−1 hydrochloric acid (B.2.1.5), in place of nitric acid (B.2.1.4), so as to obtain an anionic complex $FeCl^{-4}$.

This solution is percolated through an anionic exchanger resin and the anionic exchanger is washed with 5,0 mol · l−1 hydrochloric acid.

The iron is eluted from the resin with 0,5 mol · l−1 hydrochloric acid (B.2.1.5). The solution is evaporated to dryness and nitric acid (B.2.1.4) is used to solubilize the residue.

The iron separation yield is measured from this solution (> 95 %) and then X-ray spectrometry (see B.2.2.5) or liquid scintillation counting is used. Determination by liquid scintillation counting requires a calibration of the device.

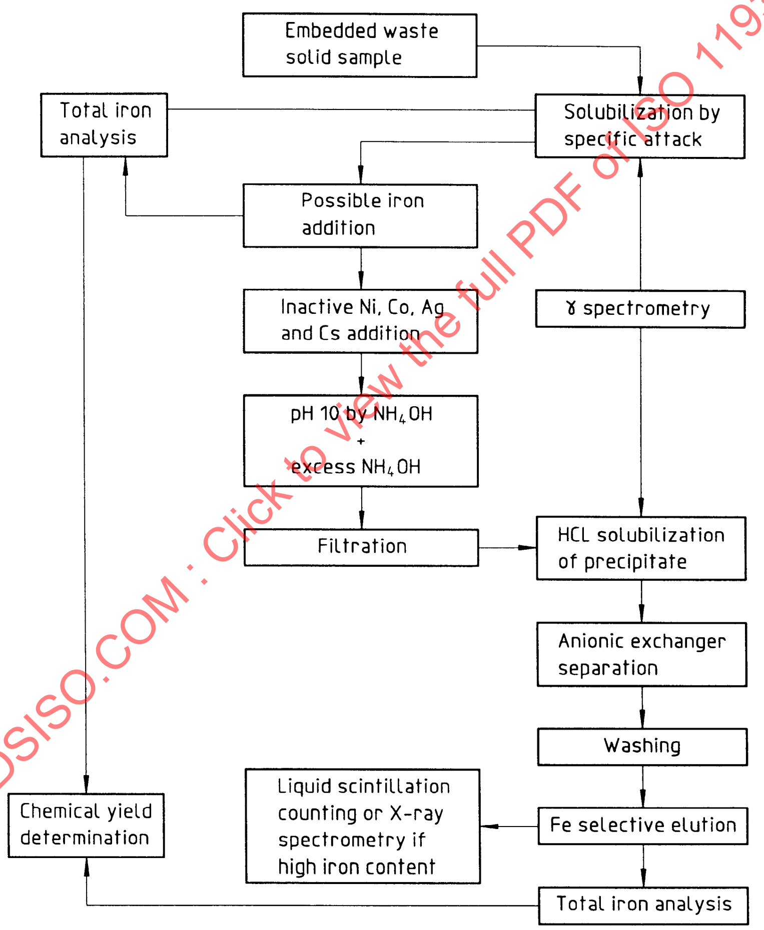


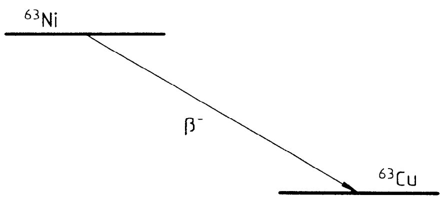
Figure B.2 — General methodology for specific ^{55}Fe separation

B.3 Determination of nickel-63

Nickel is a major component in Inconel (> 50 %) and a minor one in stainless steel (≈ 10 %). Both alloys are used under high thermal-neutron fluxes. Therefore, ^{63}Ni may be present in all nuclear waste.

Nickel-63 has a long half-life ($T_{1/2}$ = 100 years).

Nickel-63 is a pure soft beta emitter (E_{β} = 67 keV) and this beta emission will be used to quantify this element.



B.3.1 Reagents

B.3.1.1 Nickel solution, as a hold-back carrier (10 g · l^{-1}).

B.3.1.2 Dimethyl glyoxime.

B.3.1.3 Ammonium citrate solution (20 g · l^{-1}).

B.3.1.4 Chloroform.

B.3.1.5 Nitric acid, concentrated and diluted.

B.3.2 Apparatus

B.3.2.1 Laboratory glassware, beakers, pipettes, burettes, etc.

B.3.2.2 Distiller and distillation flask.

B.3.2.3 pH-meter.

B.3.2.4 Atomic absorption device.

B.3.2.5 Beta-counting device, liquid scintillation.

B.3.2.6 Gamma-ray spectrometer.

B.3.3 Measurement of nickel-63

The methodology procedure is given in figure B.3 and involves the following steps.

Step 1

Solid samples are dissolved according to their nature.

Step 2

Atomic absorption measurement of the solution's nickel concentration is carried out, and if necessary, known additions of nickel are made so as to obtain a concentration of about 10 mg · l^{-1}. A high concentration of nickel is to be avoided since this element is coloured. The colouring of a solution has a deleterious effect on quenching for liquid scintillation counting.