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Interim guidance for the safe transport of reprocessed uranium

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INTERIM GUIDANCE FOR THE SAFE TRANSPORT OF REPROCESSED URANIUM

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FOREWORD

Increasingly reprocessed uranium is being used for the fabrication of nuclear fuel elements. This entails chemical separation of the uranium isotopes, conversion to uranium hexafluoride, re-enrichment, reconversion to uranium dioxide and fuel fabrication. Since many of these processes take place at different locations, transport of uranium material will be necessary. Because of the slightly different isotopic composition of reprocessed uranium and the presence of traces of impurities, the radiation levels are generally higher than for unirradiated uranium. These different properties cast some doubt on the presumption that packages used for the transport of unirradiated uranium are automatically suitable for the transport of reprocessed uranium compounds.

The IAEA Transport Regulations themselves are rather ambiguous on this matter, since at the time that these were drafted, transport of reprocessed uranium compounds was a rare event. The Standing Advisory Group on the Safe Transport of Radioactive Material (SAGSTRAM) recommended that the issue be reviewed by consultants and that a document be developed that would give guidance to users of the Regulations.

This TECDOC is the result of the endeavours of the experts convened at two Consultants Services meetings. It contains guidance on the provisions in the current Regulations as well as proposals for changes to the new Revised Edition whose publication is planned for 1996. This document demonstrates that under the present Transport Regulations it is possible in most cases to ship reprocessed uranium compounds in the same packages as unirradiated uranium compounds. In few cases a more stringent package type is required. It is expected that this document will assist to clear out any uncertainties on the correct interpretation of the present Transport Regulations and to ensure international acceptance on the conditions for transport of reprocessed uranium.

Further information on the transport of reprocessed uranium may be obtained from H.A. Selling, IAEA Division of Nuclear Safety.

EDITORIAL NOTE

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1. INTRODUCTION

Application of the *Regulations* (1985 Edition) to the transport of reprocessed uranium has been under examination following the decision by the 7th Meeting of the Standing Advisory Group on the Safe Transport of Radioactive Material (SAGSTRAM) of the IAEA.

It was decided at the Technical Committee Meeting for the Continuous Review of the IAEA Regulations for the Safe Transport of Radioactive Materials and the Supporting Documents (TCM-405.3, 12–16 July 1989) to enlist consultants services for examination of the following items concerning transport of reprocessed uranium:

(1) To develop proposals for provisions suitable for the incorporation of reprocessed uranium in the next edition of Safety Series No. 6 [1], especially in the definition of uranium in paras 149 and 150.

(2) To find out the radiological effect caused by the real composition of reprocessed uranium (e.g. other uranium isotopes, transuranic elements, fission products, daughter products and other impurities) taking into account practical problems like the buildup of daughter product 'heels' in UF_6 cylinders recycled many times.

Consultant services meetings (CSMs) were held in November 1989 and March 1992 to examine the above issues. This "Interim Guidance for the Safe Transport of Reprocessed Uranium" summarizes the results of these two meetings.

In this document, names of certain IAEA publications are abbreviated as follows:

(1) Safety Series No. 6 [1]; The *Regulations*.

(2) Safety Series No. 7 [2]; *SS7*.

(3) Safety Series No. 37 [3]; *SS37*.

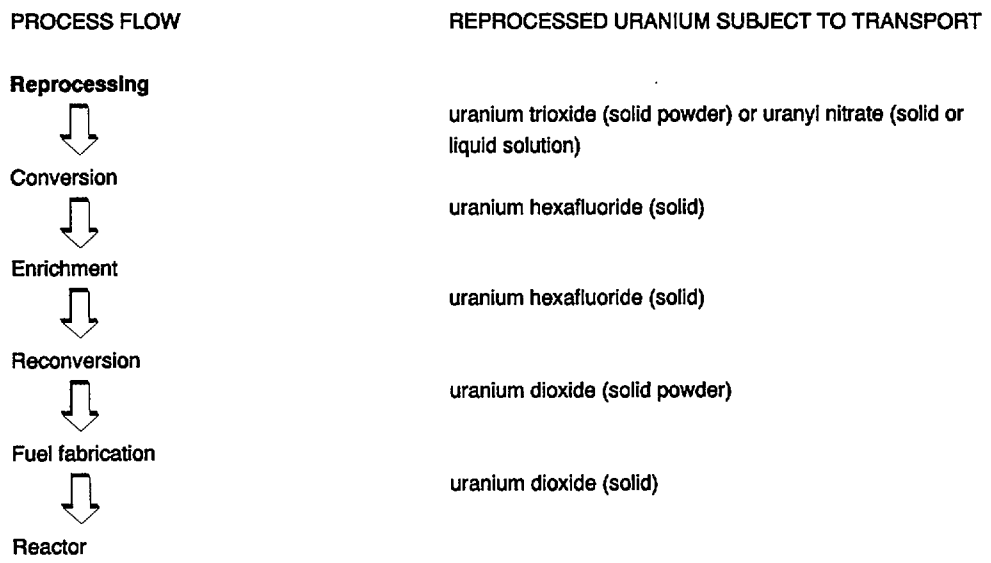
Conformity of the transport of reprocessed uranium with the *Regulations* was examined to determine whether packages used for the transport of unirradiated (enriched) uranium may also be used for reprocessed uranium transport. As a result of this scrutiny, it has been concluded that the provisions in the current *Regulations* that govern transport of natural uranium can generally be applied to reprocessed uranium compounds and that reprocessed uranium can consequently be transported as a low specific activity (LSA) material.

However, the revision of the *Regulations* themselves is under way with a view to completion in 1996. This document therefore is an interim document providing advice until the revision of the *Regulations* will have taken effect.

2. EXPLANATORY NOTE ON REPROCESSED URANIUM TRANSPORT

2.1. REPROCESSED URANIUM SUBJECT TO TRANSPORT

The process flow of reprocessed uranium is shown in Figure 1; the physical properties are shown in Table I.



Note: Because of differing process routes and locations it is conceivable that reprocessed uranium could also be transported in the form of U_3O_8 or UF_4.

FIG. 1. The process flow of reprocessed uranium.

2.2. CHARACTERISTICS OF REPROCESSED URANIUM

Reprocessed uranium is obtained by recovery of the uranium from the spent fuel from commercial nuclear power stations. The spent fuel, which is cooled for a period of time, then undergoes chemical separation and purification called 'reprocessing'. Through reprocessing, uranium is separated from plutonium and fission products (FPs) and then used as fuel again within the same fuel cycle as unirradiated uranium, i.e.: conversion–enrichment–reconversion–fabrication–reactor. Spent fuel contains FPs, transuranic elements (TRUs) and uranium isotopes, i.e. ^{232}U and ^{236}U generated only during reactor operation. Appendix I describes the radioactivity of natural and enriched uranium. Appendix II gives a full description of the nuclides that may be found in reprocessed uranium.

Through the reprocessing process, FPs such as ^{99}Tc and ^{106}Ru and TRUs such as ^{237}Np and ^{239}Pu are separated from the spent fuel. Thus the non-uranic impurity elements in reprocessed uranium are chemically removed, but the composition of uranium isotopes remains unchanged.

Although the percentage of ^{235}U in reprocessed uranium is lower than that in initial fuel, it may be either higher or lower than that in natural uranium (0.71%), depending on the degree of fuel enrichment and burnup conditions. Reprocessed uranium usually is re-enriched to augment the enrichment of ^{235}U to the prescribed degree. The concentrations of ^{232}U, ^{234}U and ^{236}U also increase during this process of re-enrichment (when the methods of enrichment utilizing differences in atomic mass between ^{235}U and ^{238}U such as gaseous diffusion and

TABLE I. THE PHYSICAL PROPERTIES OF REPROCESSED URANIUM

Chemical form (molecular formula)		Physical state during transport	Soluble/ insoluble	Fractional uranium content (w/w)	
Uranium trioxide	UO_3	solid	moderately soluble	U/UO_3	0.832
Uranyl nitrate	$UO_2(NO_3)_2$	solid/liquid	soluble	$U/UO_2(NO_3)_2.nH_2O$	0.474[a]
Uranium tetra-fluoride	UF_4	solid	moderately soluble	U/UF_4	0.758
Uranium hexafluoride	UF_6	solid	soluble	U/UF_6	0.676
Uranyl fluoride	UO_2F_2	solid	soluble	U/UO_2F_2	0.773
Uranium dioxide	UO_2	solid	insoluble	U/UO_2	0.881
Tri-uranium octoxide	U_3O_8	solid	insoluble	U/U_3O_8	0.848

[a] Generally, the water of crystallization in the solid is n = 6, and so the uranium content is 0.474 in that form. In liquid form, the uranium content depends on the concentration.

centrifuge separation are used). Attention must be paid to ^{234}U, particularly as it accounts for a large percentage of the radioactivity of reprocessed uranium.

Particular factors to be considered are:

(1) Re-enriched reprocessed uranium has higher specific activity because it contains a proportionately larger amount of ^{234}U, and the uranium isotopes ^{232}U and ^{236}U that did not exist before irradiation.

(2) Trace amounts of FP and TRU can be present as radioactive impurities.

(3) The dose rate can be higher than that of unirradiated uranium due mainly to a daughter nuclide of ^{228}Th, from the decay of ^{232}U. At higher concentrations ^{228}Th can also contribute significantly to reducing the annual limit on intake (ALI).

(4) Attention must be paid to emptied uranium hexafluoride cylinders from which UF_6 has been vaporized since the daughter nuclide ^{228}Th remains in the 'heels' in the cylinder in addition to ^{234}Th (from the decay of ^{238}U).

Appendix III discusses particular aspects of the *Regulations* that may apply to reprocessed uranium.

3. DEFINITION OF UNIRRADIATED URANIUM

The definition of unirradiated uranium given in the *Regulations* is not adequate for examining conformity of reprocessed uranium to the *Regulations* (see Appendix IV). Both reprocessed uranium and uranium contaminated by reprocessed uranium can be classified as unirradiated uranium by para. 149 of the *Regulations*. It is also necessary to make a clear distinction for reprocessed uranium because the 1990 As Amended version of the *Regulations* now includes in footnote ([d]) to Table I: "These values do not apply to reprocessed uranium".

The following definition is adopted for the purpose of this document and was submitted and approved as a change to the *Regulations* in the course of the continous review and revision process.

Unirradiated uranium

149. Unirradiated uranium shall mean uranium containing not more than 2×10^3 Bq of plutonium per gram of ^{235}U, not more than 9 MBq (0.20 mCi) of fission products per gram ^{235}U and not more than 5×10^{-3} grams ^{236}U per gram ^{235}U.

(a) The existing *Regulations* restrict plutonium by mass concentration (not more than 10^{-6} g of plutonium per gram of ^{235}U). This is unsatisfactory, insofar that it does not take account of the widely differing specific activities of plutonium isotopes. A typical plutonium isotope composition arising from reprocessing operations can have a specific activity 100 times greater than the ^{239}P referred to in para. E-149 (*SS7*). The proposed definition restricts the activity of plutonium to a rounded down value consistent with the original definition expressed as ^{239}P.

Adoption of this value limits the contribution of plutonium to 1/1000 of the specific activity of 5% enriched unirradiated uranium and to 1/2000 of the $10^{-4}A_2/g$ value for ^{239}P.

(b) The limitation of fission product activity remains unchanged.

(c) The restriction in uranium-236 content is added to provide demarcation between unirradiated and irradiated uranium. Uranium-236 is formed from uranium-235 by a neutron capture mechanism and its presence provides a technically superior means of recognizing exposure to a neutron flux. The value adopted of 5×10^{-3} g per gram uranium-235 is the upper value of the consensus standard ASTM-C-996 for enriched uranium hexafluoride. Up to this concentration the material may be considered identical to unirradiated natural uranium. It recognizes that during the conversion, enrichment and fabrication processes some accidental trace contamination by irradiated uranium can occur. This specification represents the maximum value for uranium isotopes for which composition the A_2 value can be considered to be unlimited.

Note: The footnote ([d]) of Table I of the *Regulations* then becomes "These values apply only to unirradiated uranium".

4. APPLICATION OF THE *REGULATIONS* TO THE TRANSPORT OF REPROCESSED URANIUM

4.1. THE *REGULATIONS*

SAGSTRAM has acknowledged that the current edition of the *Regulations* is unsatisfactory when applied to certain transports of reprocessed uranium. Having established a suitable definition of unirradiated uranium, it is therefore necessary to define how the *Regulations* should be applied to reprocessed uranium transport. This section provides a rationale for this, with expansions of the *Regulations* where appropriate to clarify uncertainties or inconsistencies.

4.2. APPLICATION OF THE *REGULATIONS*

4.2.1. Principles of the *Regulations*

(a) The value of A_2 for reprocessed uranium shall be determined using the composition of uranium isotopes and the amount of other radioactive impurities contained in the material, in accordance with the "formula for mixtures" stiputaled in paras 304 and 305 of the *Regulations*. Wherever the chemical form of a nuclide is known it is permissible to use the A_2 value related to its known solubility class, taking the forms appropriate to normal and accident conditions.

(b) Reprocessed uranium whose specific activity does not exceed $10^{-4}A_2/g$ shall be classified as LSA-II. A_2 may be taken as unlimited if 10 mg of the material contains less than the ALI for that mixture, and such material can also thus be classified as LSA-II.

(c) Reprocessed uranium not included in (b) above whose specific activity does not exceed $2 \times 10^{-3}A_2/g$ shall be classified as LSA-III if it can meet standards on leachability.

(d) Uranium in a powder state may be classified as LSA-III provided it can be demonstrated that there is no dispersal from a damaged package as defined in para. 564(b)(i) of the *Regulations*.

The formula for determining the value of A_2 is specified as follows in paras 304 and 305 of the *Regulations*:

$$A_2 \text{ for mixture} = \Sigma_i \frac{1}{\frac{f(i)}{A_2(i)}}$$

Where f(i) : fraction of activity of nuclide i in the mixture, and
$A_2(i)$: appropriate A_2 value for the nuclide i, e.g.

^{232}U: 3×10^8 Bq
^{234}U: 1×10^9 Bq
^{235}U: no limit
^{236}U: 1×10^9 Bq
^{238}U: no limit

For examples see Appendix VIII.

4.2.2. Reprocessed uranium classified as LSA-II

LSA-II is defined as follows in the current *Regulations*:

"131. Low specific activity (LSA) material shall mean radioactive material which by its nature has a limited specific activity, or radioactive material for which limits of estimated average specific activity apply. External shielding materials surrounding the LSA material shall not be considered in determining the estimated average specific activity.

LSA material shall be in one of three groups:

(a) LSA-I *(not considered here)*.

(b) LSA-II

(i) Water with tritium concentration up to 0.8 TBq/L (20 Ci/L); or
(ii) Other material in which the activity is distributed throughout and the estimated average specific activity does not exceed 10^{-4} A_2/g for solids and gases, and 10^{-5} A_2/g for liquids.

(c) LSA-III *(not considered here)*".

Reprocessed uranium in the solid state which meets either of the following criteria shall be classified as LSA-II:

(1) Reprocessed uranium material whose specific activity does not exceed 10^{-4} A_2/g.

(2) Reprocessed uranium material 10 mg of which does not exceed 1 ALI taking into consideration its chemical form and the appropriate inhalation classes D, W or Y of ICRP publications [4, 5] for both normal and accident situations.

According to ICRP 30 [5], uranium compounds whose inhalation class is D (water soluble) are UF_6, UO_2F_2 and uranyl nitrate; those whose inhalation class is W (moderately soluble) are UO_3 and UF_4; UO_2 is inhalation class Y. Where the likely degree of solubility or chemical form (organic or inorganic) of a particular radionuclide is known the inhalation class of the chemical form under both normal and accident conditions should be taken into account using the most appropriate value as currently recommended by ICRP. A fuller discussion of the application of LSA-II to reprocessed uranium can be found in Appendix V.

4.2.3. Reprocessed uranium classified as LSA-III

Reprocessed uranium whose specific activity exceeds $10^{-4}A_2$/g (the upper limit for LSA-II), which is at the same time an insoluble compound whose specific activity is $2 \times 10^{-3}A_2$/g or less, shall be deemed as LSA-III if it meets the requirements specified in para. 131 of the *Regulations*:

"131. Low specific activity (LSA) material shall mean radioactive material which by its nature has a limited specific activity, or radioactive material for which limits of estimated average specific activity apply. External shielding materials

surrounding the LSA material shall not be considered in determining the estimated average specific activity.

LSA material shall be in one of three groups:

(a) LSA-I (*not considered here*).

(b) LSA-II (*not considered here*).

(c) LSA-III

Solids (e.g. consolidated wastes, activated materials) in which:

(i) The radioactive material is distributed throughout a solid or a collection of solid objects, or is essentially uniformly distributed in a solid compact binding agent (such as concrete, bitumen, ceramic, etc.);
(ii) The radioactive material is relatively insoluble or it is intrinsically contained in a relatively insoluble matrix, so that, even under loss of packaging, the loss of radioactive material per package by leaching when placed in water for 7 days would not exceed 0.1 A_2; and
(iii) The estimated average specific activity of the solid, excluding any shielding material, does not exceed 2×10^{-3} A_2/g."

4.2.3.1. Uranium dioxide ceramic pellet as LSA-III material

The relevant requirements for LSA-III are summarized as follows:

(1) It must be a solid in which radioactive material is uniformly distributed.

(2) Its specified activity does not exceed $2 \times 10^{-3} A_2$/g.

(3) Radioactive material exceeding $0.1A_2$ does not leach from a package during a leaching test in water for 7 days.

The leaching tests for 7 days on uranium dioxide pellets conducted in the UK and Japan in 1990 (Appendix VI) confirmed that a value adequately lower than the standard $0.1A_2$ is obtained with this material.

Based on the definition of LSA and the results of these leaching tests, uranium dioxide pellets (including the case of a fuel assembly) satisfying these criteria can be deemed as LSA-III.

4.2.3.2. Uranium dioxide and triuranium octoxide powders as LSA-III material

The leaching tests on UO_2 and U_3O_8 powder conducted in the UK and Japan in 1990 (Appendix VI) also confirmed that these materials satisfy the standard leaching test of less than 0.1 A_2 value after immersion in water for 7 days.

However, since the reference to LSA-III in the current *Regulations* refers to non-dispersible radioactive materials, the additional requirement that there is no dispersal from a damaged package under conditions likely to be encountered during normal conditions of

transport and in an accident shall be applied to a package intended to transport such reprocessed uranium oxide powder (Appendix VII). The criteria for establishing this requirement are specified in para. 564(b)(i) of the *Regulations*.

4.2.4. Consideration of the daughter nuclides of uranium

According to para. 303 of the *Regulations*, when calculating the values of A_1 and A_2, when a decay chain in which a daughter nuclide has a half-life longer either than ten days or that of the parent nuclide, such parent nuclide and daughter nuclide are to be considered as a mixture of different nuclides. In calculating the value of A_2 of reprocessed uranium, it is therefore necessary to take ^{228}Th (daughter nuclide of ^{232}U) into account (see Appendix VIII).

Among the daughter nuclides of reprocessed uranium ^{228}Th should always be taken into account. While undisturbed the amount of ^{228}Th builds up to reach the maximum value in approximately 10 years. When calculating the A_2 value it should be assumed that ^{228}Th is in full equilibrium with ^{232}U to give the maximum figure.

4.2.5. Classification of enriched commercial grade uranium

The new definition of unirradiated uranium, which corresponds to enriched commercial grade uranium (ECGU) at ASTM, can have a specific activity very slightly greater than 10^{-4} A_2/g at 5 w/o enrichment. However, this is also true of completely naturally derived enriched uranium under some enrichment conditions, although this is always specified as 'unlimited A_2'. It is concluded therefore that ECGU differs insignificantly from this and may also be accepted as 'unlimited A_2' within the specified composition. (Appendix IX details the calculations).

4.2.6. Observations on radioactive impurities

Although trace quantities of radioactive impurities are contained in reprocessed uranium, their contribution to the A_2 value may be neglected in principle. In calculating the A_2 value of reprocessed uranium in accordance with paras 304 and 305 of the *Regulations*, it is sufficient to take uranium isotopes and their daughter nuclides as well as representative nuclides of the radioactive impurities (FP, TRU) into account.

In calculating the A_2 value of reprocessed uranium to classify it into one of the LSA categories, it is important always to take daughter nuclides of uranium isotopes and nuclides of radioactive impurities (FP, TRU) into account. It shall be confirmed whether nuclides of radioactive impurities (FP, TRU) can be neglected or not by taking their representative nuclides into calculating the A_2 value. (Refer to Appendix X).

4.2.7. Consideration of changes in dose equivalent rate with time

Since the increment of gamma ray dose-equivalent rate by ^{208}Tl increases in proportion to the build-up of ^{228}Th, which is the first daughter nuclide of ^{232}U, it is important to control the build-up period from the stage at which daughter nuclides are chemically separated from reprocessed uranium (for instance from the point at which a cylinder is filled with UF_6).

The dose equivalent rate of a package containing reprocessed uranium is higher than one containing unirradiated uranium due to differences in radiation source specifications. This

is primarily because ^{208}Tl, which emits high energy gamma rays, is produced in the decay chain of ^{232}U which is not found in unirradiated uranium.

^{208}Tl, which rapidly reaches radioactive equilibrium with ^{228}Th (half-life: 1.91 years), the first daughter nuclide of ^{232}U, shows a maximum activity value in about ten years. (See Appendix XI).

According to the results of the German study of 48Y cylinders, 'Investigations on the Transport of UF_6 in 48Y containers with Uranium from Reprocessed PWR Fuel Elements' (Appendix XII), the dose equivalent rate of a cylinder filled with uranium hexafluoride, builds up to a peak in approximately ten years, but is then still adequately below the surface dose equivalent rate limits stipulated in the *Regulations* (2 mSv/h). After emptying the UF_6, the ^{228}Th remains in the heels within the cylinder but is no longer shielded by the bulk of the uranium. Such a cylinder can then exceed the maximum allowable surface dose rate for transport, depending on the initial composition of the uranium and its storage time while full.

4.2.8. Handling of cylinders emptied of uranium hexafluoride

When a cylinder filled with reprocessed uranium hexafluoride is to be re-used, consideration must be given to removing heels by cleaning the cylinder before transport of the empty cylinder.

Taking the example of an empty 48Y type cylinder which had been filled with reprocessed uranium before enrichment, the heel specific activity exceeds the limit for LSA-II, even if the maximum permissible heel amount (22.68 kg) is assumed. This is primarily due to the contribution by ^{228}Th. Moreover, uranium hexafluoride cannot meet the insolubility criterion for LSA-III. In addition its total activity exceeds the value of A_2 for the mixture and hence it cannot be classified as Type A (Appendix XII). The contribution of ^{228}Th becomes much greater after enrichment as a result of the enrichment of ^{232}U.

It should be noted that, if heels are not removed, the contamination level inside the cylinder also exceeds the limits for an empty container, and for SCO-I, and SCO-II.

4.2.9. Additional requirement for an IP type package containing reprocessed uranium

General testing conditions for IP-2 or IP-3 packages specified in paras 519 and 520 of the *Regulations* to be replaced for transport of LSA-III powders by those prescribed by para. 564b(i) (tests for demonstrating ability to withstand normal and accidental conditions of transport).

4.2.10. Approval by the authorities and multilateral approval of reprocessed uranium transport

As indicated in Sections 4.2.1–4.2.8, the transport of reprocessed uranium, which takes account of chemical form shall be subject to approval (design) by national authorities; international transport shall be subject to multilateral approval.

4.3. MARKING OF PACKAGES AND INSPECTION BEFORE SHIPMENT

Marking, labelling and placarding concerning transport shall be in compliance with requirements specified in paras 436–445 of the *Regulations*. It is not necessary to give a special indication of 'reprocessed uranium' during transport.

Package inspection to the requirements specified in para. 402 of the *Regulations* should be made before each shipment to confirm the compliance of transported items to these requirements. Special attention should be paid to confirm the amount of radioactivity contained in the package, if reprocessed uranium is to be transported. It should be confirmed that the reprocessed uranium contained in the packaging is in compliance with the requirements on radioactivity limits prescribed for LSA-III or LSA-III materials (para. 311 of the *Regulations*).

The overall composition of reprocessed uranium depends on the type of reactor in which the original uranium has been irradiated, the burnup, the cooling history after irradiation, the decontamination efficiency of subsequent purification processes and the time that has elapsed after reprocessing.

Consequently, in the package inspection conducted before each shipment is made, the value A_2 for reprocessed uranium must be calculated on the basis of the percentage of uranium isotopes and radioactive impurities (FP/TRU, etc.) in the reprocessed uranium, using the "formula for determining the value of A_2 for a mixture" specified in the para. 304 of the *Regulations*.

After considering its chemical form under normal and accident conditions, reprocessed uranium material whose specific activity does not exceed 10^{-4} A_2/g (10^{-5} A_2/g in the case of liquid) is classified into LSA-II. Alternatively, if 10 mg of the reprocessed uranium does not exceed 1 ALI (annual limit on intake) it may also be classified as LSA-II.

Solid reprocessed uranium material whose specific activity exceeds the above value ($10^{-4}A_2$/g) but does not exceed $2 \times 10^{-3}A_2$/g may be classified as LSA-III. Powdered reprocessed uranium may be classified as LSA-III if the package meets the tests specified in para. 564(b)(i) without dispersal of the contents.

In the package inspection before the actual shipment, the compliance of the contents of the reprocessed uranium material shall be confirmed by analysis and evaluation.

5. SUMMARY

The consultants services meetings have examined the transport of reprocessed uranium in the various IAEA Safety Series documents and have established a methodology which allows full compliance into the *Regulations*. In practice the LSA category will be applicable to most such transport movements. Some clarification of the *Regulations* is needed to demonstrate this.

The existing definition of unirradiated uranium is inadequate, because it would allow reprocessed uranium to be classified as unirradiated. Consequently, it has been proposed to change the definition of unirradiated uranium such as to encompass uranium containing not more than 2×10^3 Bq Pu/g ^{235}U of Pu, not more than 9 MBq/gU of fission products and not more than 5×10^{-3}g ^{236}U/g ^{235}U.

The classification methods for reprocessed uranium material for transport are summarized as follows:

(1) Reprocessed uranium material in which the activity is distributed uniformly and which fulfills either of the following requirements 1(a) or 1(b) shall be classified as LSA-II.

(a) The specific activity of solid reprocessed uranium material does not exceed 10^{-4} A_2/g where the value of A_2 is calculated for all significant nuclides in the material. For liquid reprocessed uranium material the specific activity of the material shall not exceed 10^{-5} A_2/g.

TABLE II. LSA CLASSIFICATION OF THE MAIN REPROCESSED URANIUM MATERIALS

Form	Activity content	Classification	Comments
Uranium trioxide	less than $10^{-4}A_2$/g	LSA-II	
Uranyl nitrate	less than $10^{-5}A_2$/g	LSA-II	Relevant chemical forms are: $UO_2(NO_3)_2$, U_3O_8[a]
Uranium hexafluoride	less than $10^{-4}A_2$/g	LSA-II	Relevant chemical forms are: UF_4, UO_2F_2[a]
Uranium dioxide powder	less than $10^{-4}A_2$/g less than $2 \times 10^{-3}A_2$/g	LSA-II LSA-III	[b]
Uranium dioxide pellet OR fuel rod OR assembly / bundle	less than $10^{-4}A_2$/g	LSA-II	
	less than $2 \times 10^{-3}A_2$/g	LSA-III	

[a] The chemical forms indicated are all those considered by the consultants as being likely to arise in significant quantities from a transport accident involving uranyl nitrate or uranium hexafluoride.

[b] Package must not allow dispersal under normal and accident conditions during transport.

(b) When 10 mg of reprocessed uranium material contains in radioactivity less than 1 ALI taking account of its compound form and solubility class under normal and accident conditions.

(2) Solid reprocessed uranium material whose specific activity exceeds $10^{-4}A_2/g$ (the upper limit for LSA-II), but does not exceed $2 \times 10^{-3}A_2/g$ and which meets the leaching standard as defined in para. 131(c)(ii) of the *Regulations* shall be classified as LSA-III. Uranium material in the powder state must meet the additional performance requirements that no material is dispersed from the package under conditions of test specified in para. 564(b)(i) of the *Regulations*.

The LSA classification of the main reprocessed uranium materials is shown in Table II.

Appendix I

THE RADIOACTIVITY OF URANIUM

The total radioactivity of uranium at a given enrichment depends on a number of factors including the origin of feed material, the method of enrichment, the period of time after processing of the uranium and the percentage of other radioactive impurities.

The relation between specific activity and enrichment of uranium-235 (*SS37*, 1990) is shown in Table III. The value of specific activity of uranium includes the activity of uranium-234 enriched during the enrichment process for uranium-235 but does not include contributions by uranium daughter nuclides. The values are for uranium enriched by the gaseous diffusion method starting from natural uranium. If the origin of source material is unknown, then its specific radioactivity must be measured or calculated on the basis of data on isotopic ratios.

Since uranium fuel cycle compounds are produced through chemical processes, their equilibrium with uranium daughter nuclides may be disturbed during those processes. Whereas uranium isotopes have long half-lives, the daughter nuclides or uranium-238 and uranium-235 (thorium-234 and thorium-231, respectively) have short half-lives. Therefore equilibrium is almost reached in approximately 150 days. After this period, for instance, according to IAEA-TECDOC-608, the specific activity of UF_6 reaches nearly 3.6×10^4 Bq/g at 0.45% enrichment or 4.0×10^4 Bq/g at natural enrichment.

The enrichment process also enriches the minor isotopes of uranium (i.e. ^{232}U, ^{234}U and ^{236}U). As the ratio of ^{234}U to ^{235}U is greater in reprocessed uranium than for naturally derived uranium at the same ^{235}U enrichment, this isotope contributes significantly to the increased specific activity of enriched reprocessed uranium.

TABLE III. SPECIFIC ACTIVITY VALUES FOR URANIUM AT VARIOUS LEVELS OF ENRICHMENT

Mass percent of uranium-235 present in uranium mixture	Specific activity[a]	
	Bq/g	g/Bq
0.45	1.8×10^4	5.6×10^{-5}
0.72 (natural)	2.6×10^4	3.8×10^{-5}
1.0	2.8×10^4	3.6×10^{-5}
1.5	3.7×10^4	2.7×10^{-5}
5.0	1.0×10^5	1.0×10^{-5}
10.0	1.8×10^5	5.6×10^{-6}
20.0	3.7×10^5	2.7×10^{-6}
35.0	7.4×10^5	1.4×10^{-6}
50.0	9.3×10^5	1.1×10^{-6}
90.0	2.2×10^6	4.5×10^{-7}
93.0	2.6×10^6	3.8×10^{-7}
95.0	3.4×10^6	2.9×10^{-7}

[a] These values of the specific activity include the activity of uranium-234 which is concentrated during the enrichment process, and do not include any daughter product contribution. The values are for the material originating from natural uranium enriched by a gaseous diffusion method. If the origin of the material is not known the specific activity should be either measured or calculated using isotopic ratio data.

Appendix II

NUCLIDES COMPOSING REPROCESSED URANIUM

Uranium compounds such as UF_6 and UO_2 produced from unirradiated uranium do not include sources of radioactivity other than from uranium isotopes existing in nature, i.e. ^{238}U, ^{235}U, ^{234}U and their daughter nuclides.

Uranium materials such as UF_6 and UO_2 generated from uranium separated from spent fuel through reprocessing (reprocessed uranium) can contain the following nuclides in varying concentrations:

(1) Uranium isotopes, for instance, ^{232}U, ^{234}U[1], ^{235}U[1], ^{236}U, ^{237}U and ^{238}U[1].

(2) Transuranic isotopes (TRUs), for instance, ^{237}Np, ^{239}Pu.

(3) Fission products (FPs), for instance, ^{106}Ru, $^{95}Zr/^{95}Nb$ and ^{99}Tc.

(4) Daughter products of these nuclides, for instance, ^{233}Pa and ^{228}Th and ^{208}Tl.

The composition of nuclides contained in reprocessed uranium depends on the type of reactor in which irradiation has taken place, its burnup and subsequent cooling history, impurity decontamination efficiency in reprocessing, enrichment, and reconversion processes, and time that has elapsed after such processes have been completed. Processes which change radioactive equilibria, e.g. solvent extraction or vaporization of UF_6, are important.

Under these conditions, indices for transportation in accordance with the *Regulations* must be calculated for each transported item on the basis of specific analysis data.

The specific activity and other properties of various types of reprocessed uranium are shown in Table IV.

[1] Uranium isotopes existing in nature.

TABLE IV. CHARACTERISTICS OF REPROCESSED URANIUM PACKAGE

Item	Enriched uranium of natural source	ECGU[a]		ERU[a]		Remarks
	U	UF_6	UO_2	UF_6	UO_2	
Enrichment	5%	5%	5%	5%	5%	$A_2 = \frac{1}{\sum_i \frac{f(i)}{A_2(i)}}$
A_2 (Bq)	Unlimited	1.1×10^9	1.1×10^9	8.8×10^8	8.8×10^8	
Quantity equivalent of A_2 (kg)	Unlimited	12.2	9.2	2.3	1.8	
Activity (Bq/g)	1.0×10^5	9.0×10^4	1.2×10^5	3.9×10^5	5.0×10^5	
Activity limit of LSA-II ($10^{-4}A_2$/g)	1.1×10^5	1.1×10^5	1.1×10^5	8.8×10^4	8.8×10^4	f(i): fraction of activity of nuclide i
Activity limit of LSA-III ($2 \times 10^{-3}A_2$/g)	2.0×10^6	not applicable	2.2×10^6	not applicable	1.8×10^6	A_2: appropriate A_2 value for the nuclide i

[a] ECGU: enriched commercial grade uranium (ASTM C-996) maximum values.
ERU: enriched reprocessed uranium (ASTM C-996) maximum values.

Appendix III

IAEA REGULATIONS CONCERNING REPROCESSED URANIUM

This appendix summarizes those standards specified in the IAEA Regulations for the Safe Transport of Radioactive Material (the *Regulations*) that are especially pertinent to this document. The IAEA periodically revises the *Regulations*.

The *Regulations* have served as a model for a regulatory rule established by Member States as well as for a published rule instituted by international transport organizations. The latest revision of the *Regulations* was published in 1985. Supplements were issued in 1986 and 1988. As amended versions of the *Regulations* and its supporting documents *SS7*, *SS37* and *SS80* [6] were published in 1990.

Below are summarized the major requirements in the *Regulations* concerning UF_6, UO_2 powder and pellets, fuel rods, and other reprocessed uranium materials described in this document.

The *Regulations* consist of the following requirements on contents and packaging:

(1) The quantity of LSA material or SCO in a single industrial package Type I (IP-I), industrial package Type 2 (IP-2), industrial package Type 3 (IP-3) or object or collection of objects, if appropriate, shall be so restricted that the external radiation level at 3 m from the unshielded material or object or collection of objects does not exceed 10 mSv/h (1 rem/h) (para. 422).

(2) The radioactive material is distributed throughout the nuclear fuel or in a matrix. (para. 131(b)(ii) and (c)(i)).

(3) The specific activity of reprocessed uranium shall not exceed the standard value as follows:

(i) LSA-II:

- For distributed solids: "The estimated average specific activity does not exceed 10^{-4} A_2/g" (para. 131(b)(ii)).

- For liquids: "The estimated average specific activity dose not exceed $10^{-5}A_2$/g" (para. 131(b)(ii)).

(ii) LSA-III:

"The estimated specific activity of the solid, excluding any shielding material, shall not exceed 2×10^{-3} A_2/g" (para. 131(c)(iii)).

(4) In the case of LSA-III, even if a container is lost and if the radioactive material transported is left in water for 7 days, the amount of radioactive material leaking to water per transport unit shall not exceed 1/10th of the value of A_2 (paras 131(c)(ii) and 603).

(5) A transport containing reprocessed uranium which is both fissile and classified as LSA II/LSA III shall be subject to:

- the package requirements for LSA material and SCO (para. 426, Table V);

- the requirements for containment and shielding for IP-2/IP-3 packages (paras 518–523);

- the requirements for fissile material (paras 559–568).

(6) Total radioactivity of one transported item of LSA material shall be limited so as not to exceed the radiation level stipulated in para. 422.

In addition, the total activity per conveyance is limited (para. 427, Table VI).

(7) Transported matter in excepted packages, including transported matter containing fissile material not exceeding 15 g, and an empty cylinder for UF_6 shall meet the same requirements as natural uranium for transport and control (paras 415–421).

Appendix IV

DEFINITIONS OF UNIRRADIATED/REPROCESSED URANIUM AND THEIR IMPLICATIONS

INTRODUCTION

In the current *Regulations*, the definition of unirradiated uranium defines limits for fission products and plutonium, for the purpose of excluding any uranium which has been irradiated in a nuclear reactor. But because of technical improvements in the efficiency of reprocessing purification processes, this limitation on fission product and plutonium concentration is no longer effective.

Table I of the *Regulations* (as amended 1990) in providing A_1/A_2 values adds a footnote to the 'unlimited' A_2 values ascribed to U (natural), U (enriched 5% or less) and U (depleted) to exclude reprocessed uranium. As reprocessed uranium is not defined in the *Regulations*, the implication is that uranium which does not meet the definition unirradiated must be considered to be 'reprocessed'.

After extensively having studied alternatives the experts concluded that a clear definition of unirradiated uranium would remove any need to define a new category 'reprocessed uranium'.

The reason for defining 'unirradiated uranium' and by virtue the exclusion of 'irradiated' (reprocessed, recycled) uranium is to ensure that the special characteristics of irradiated uranium are recognized in assessing safety and the technical suitability of recommended practices for transport.

Further consideration of such a new definition of unirradiated uranium raised the practical issue as to whether the commercial category of uranium, ECGU (ASTM specification), could be accepted as 'unirradiated'. This topic is dealt with by examples in calculations presented in the attachments.

DEFINITION OF UNIRRADIATED URANIUM

In terms of the 'definition of unirradiated uranium' in para. 149 of the *Regulations*, an investigation was made into criteria for discriminating between unirradiated and reprocessed uranium.

(1) *Regulations* (1985 Edition): Pu, FP

This definition alone is unsatisfactory as Pu and FP can be removed from irradiated uranium by solvent extraction or other chemical processing technology to values below the limits of specified. This improvement in reprocessing technology could therefore allow some uranium to be designated as 'unirradiated' even after exposure in a reactor.

(2) *UK proposal*: Pu, FP, ^{236}U

Whilst chemical purification techniques can remove associated radioactive species such as fission products and transuranic elements, the purified uranium, because of its irradiation, will contain the minor isotopes uranium-232 and uranium-236. These minor isotopes are technically significant to subsequent safety considerations and fuel properties and also provide a means of identifying prior irradiation history. Because uranium-236 is produced in greater abundance and because measurement techniques are simpler for this isotope, it is the preferred identifier for irradiation.

(3) *Investigation in Japan*: Pu, FP, U (α)

Whilst measurement of U(α) activity is relatively simple and is a direct measure of the principal activity associated with uranic materials, it is significantly affected by the concentration of the naturally occurring isotope uranium-234. The ratio of uranium-234 to **total** uranium **falls** with irradiation but to a lesser degree than uranium-235 during reactor burnup. This means only **after re-enrichment** is the ratio of uranium-234/uranium-235 enhanced relative to natural products — hence the higher specific activity.

The measurement of U (α) is therefore not a satisfactory discriminator for irradiation in all circumstances. Variations in enrichment processes, e.g. gaseous diffusion or centrifuge, type of feed, and operating tails assay, can also modify the ratio uranium-234/uranium-235. This has implications for the values for specific activity ascribed to enriched (natural origin) materials.

Therefore the following definitions relating to unirradiated uranium shall be added:

Unirradiated uranium

The *Regulations*:

149. Unirradiated uranium shall mean uranium containing not more than 2×10^3 Bq of plutonium per gram of uranium-235, not more than 9 MBq (0.20 mCi) of fission products per gram uranium-235 and not more than 5×10^{-3} g uranium-236 per gram uranium-235.

Footnote [d] Table I

These values apply only to unirradiated uranium.

SS7

E-149. The term 'unirradiated uranium' is intended to exclude any uranium which has been exposed to a neutron flux so as to transform some of the uranium-238 into plutonium-239 and some of the uranium-235 into fission products. The limits of 2×10^3 Bq plutonium per gram of uranium-235 and 9 MBq (0.20 mCi) of fission products per gram uranium-235 are intended to identify the presence of irradiated uranium whilst recognizing the presence of trace amounts of plutonium and fission products in all natural uranium.

The presence of uranium-236 is a more satisfactory indication of exposure to a neutron flux. 5×10^{-3} grams uranium-236 per gram uranium-235 has been chosen as representing the consensus view of ASTM Committee C-26 in specification C-996 for enriched commercial grade uranium. This value recognizes the possibility for trace contamination by irradiated uranium but provides that the material may still be treated as unirradiated. This specification represents the maximum value for uranium isotopes for which composition the A2 value can be demonstrated to be unlimited for uranium hexafluoride. The difference in A2 for uranium dioxide is considered to be insignificant. The defined terms in paras 149 and 150 have been combined in para. 131(a)(ii) to prescribe acceptable forms of LSA-I.

Note: Changes from the 1985 version (as amended 1990) of the Safety Series documents are shaded.

Attachment 1 to Appendix IV

DISCUSSION OF DEFINITION OF UNIRRADIATED URANIUM AT 1ST CSM

(1) The definition given in para. 149 of the *Regulations* is not technically correct because it does not formally exclude all irradiated uranium.

(2) The present definition based on plutonium and fission product contents could be met by irradiated uranium after suitable reprocessing conditions and for any uranium isotope composition.

(3) The presence of uranium-236 is more satisfactory evidence for exposure to a neutron flux and it consistent with the intention of excluding irradiated material according to E-149 of *SS7*.

(4) It is not proposed to remove the present restrictions on plutonium and fission products because of the reasons provided in para. E-129 of *SS7*.

(5) In order to improve the present definition, it is proposed to add the additional criterion that the material should not contain more than 5×10^{-3} grams uranium-236 per gram uranium-235[2].

(6) This value has been chosen as representing a consensus view of ASTM Committee C-26 in specification C-996. This value will limit plutonium and fission products more stringently than the existing definition and can be readily demonstrated by measurement.

[2] The evaluation of uranium-236 is provisional and will require further evaluation.

Attachment 2 to Appendix IV

PROBLEMS OF UNIRRADIATED/REPROCESSED URANIUM FUEL CYCLES

The definitions of unirradiated and reprocessed uranium must make a distinction between four types of uranium, as indicated in Figure 2.

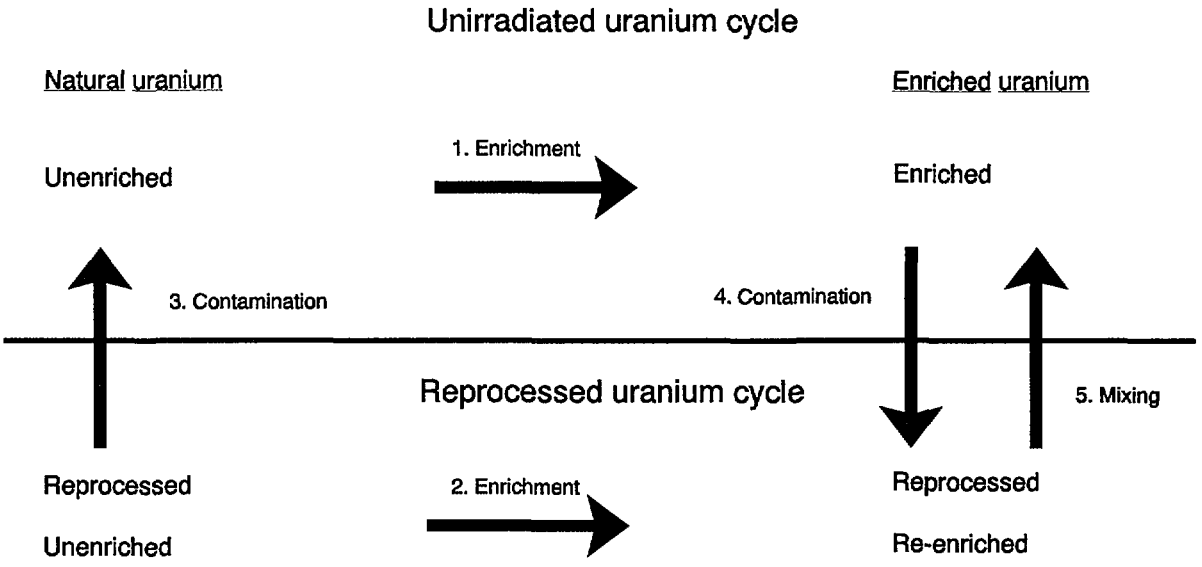


FIG. 2. Interaction zones of unirradiated/reprocessed uranium.

(a) If unirradiated and reprocessed uranium are treated at the same time in enrichment and fabrication facilities, then contamination of unirradiated uranium by reprocessed uranium may occur. Therefore it is necessary to examine the permissible extent of this contamination and establish such limits in accordance with the Q system of IAEA, taking into account ASTM and other commercial standards. (Problem arising in 3, 4 and 5 of Figure 2).

(b) It is unreasonable that what is classified as unirradiated or natural uranium could be classified otherwise after an enrichment process. In order to avoid this possibility, and taking into account existing enrichment process technology, an internationally accepted composition of "commercial natural uranium" (CNU) has been produced by ASTM Committee C-26 in the form of a specification for the feed to an enrichment process (ASTM C-787). Within this specification the maximum trace contamination by irradiated uranium is controlled by limiting the 'irradiated' uranium isotopes:

uranium-232 not greater than 0.01×10^{-9}g/g U

uranium-236 not greater than 20×10^{-6}g/g U

On enrichment to 5% uranium-235 this material will satisfy C-996 specification for "enriched commercial grade uranium" (ECGU).

The justification is that CNU and ECGU can be treated in all respects as being the same as uranium of pure natural origin.

The purpose of the definition of unirradiated uranium to "exclude uranium used in a reactor" may remain unaltered, but it must recognize the possibility of contamination of unirradiated uranium by reprocessed uranium in the light of parallel use of unirradiated and reprocessed uranium.

(c) The specific activity of ECGU to the ASTM C-996-90 standard (upper limits for specifications of 5.0% enrichment) and the domain where "10 mg is 1 ALI or less" are shown in the Figure 3, along with specific activity in relation to enrichment indicated in the 1973 *Regulations*. The specific activity of ECGU to the ASTM Standard in the form of uranium hexafluoride is below the specific activity of LSA-II, but it can, in the form of uranium dioxide, conceivably exceed the specific activity limits for LSA-II.

However, the specific activity of 5% enriched uranium arising from purely uncontaminated natural sources is dominated by the concentration of the uranium-234 isotope. The current *Regulations* do not mandate for the variability of uranium-234 in enriched products arising from commercial enrichment processes. The ratio 10 000 μg uranium-234/gram uranium-235 is not unusual in enrichment plant tolerances (ASTM C-996) and at this level the effect of the minor isotopes uranium-232 and uranium-236 as specified for ECGU is insignificant. It follows that if this unirradiated uranium to 5% uranium-235 enrichment can be classed as 'A_2 unlimited' (Table I of the *Regulations*) then ECGU at the same enrichment must also be 'unlimited A_2' (see Table X).

(d) As noted in (c), the specific activity of unirradiated enriched uranium is governed by the uranium-234 isotope. During irradiation as fuel in a reactor the concentrations of the uranium-235 and uranium-234 isotopes fall. It was a demonstrable conclusion of the first CSM in 1989 that the net specific activity of uranium decreases during irradiation with loss of uranium-234 and uranium-235 even through uranium-232 and uranium-236 increase. If the activities of daughter products and transuranic nuclides are sufficiently low after reprocessing, it is possible for reprocessed uranium (prior to further enrichment) to have a

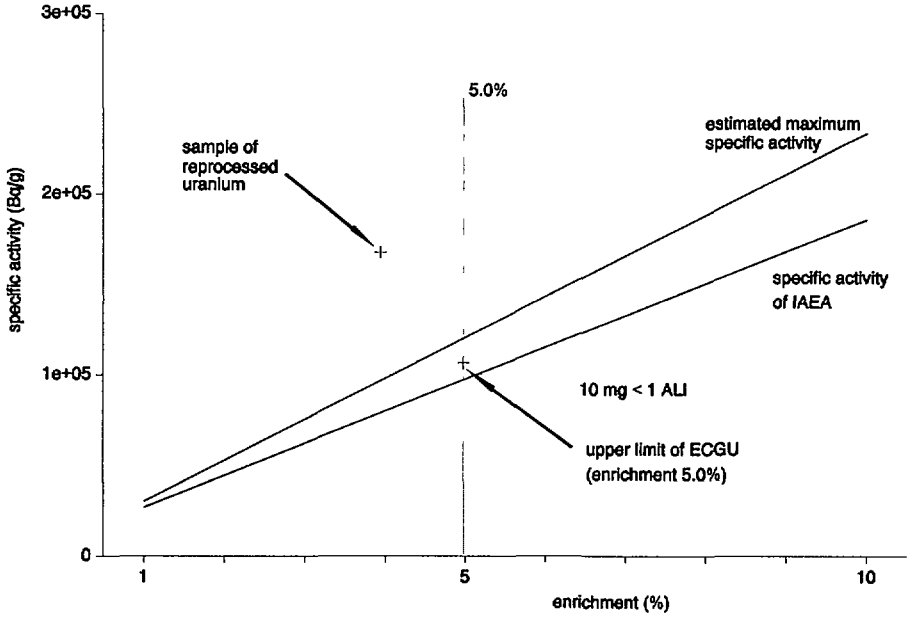


FIG. 3. Enrichment and specific activity.

lower specific activity than the starting fuel, thus placing the material in the same 'unlimited A_2' category for transport. Irradiated uranium after reprocessing but prior to further enrichment can therefore meet the existing requirements for LSA-II without any need to change the *Regulations*.

(e) Following further enrichment the ratio of uranium-234/uranium-235 is greater for irradiated uranium than for unirradiated. This relationship arises from two factors. Firstly, the loss of uranium-234 during irradiation occurs proportionately more slowly than fission of uranium-235 (burnup). Hence the ratio uranium-234/uranium-235 is increased. Secondly, during enrichment (gaseous diffusion, centrifuge) the physical processes enhance the relative concentration uranium-234/uranium-238 more rapidly than uranium-235/uranium-238, increasing the ratio uranium-234/uranium-235 in the product. As noted on previous occasions this increases the specific activity of the uranium because of the higher specific activity of uranium-234 isotope. Enrichment can also lead to increased concentrations of residual radioactive impurities such as fission products (^{106}Ru, ^{99}Tc) and transuranic nuclides (^{237}Np, ^{239}Pu, etc.).

Appendix V

REPROCESSED URANIUM CLASSIFIED AS LSA-II

Reprocessed uranium can be classified as LSA-II in the following cases:

(1) Where the specific activity of reprocessed uranium material does not exceed 10^{-4} A_2/g.

(2) Where, taking into consideration its compound form, 10 mg of reprocessed uranium material does not exceed 1 ALI under normal and accident conditions.

This criterion is based on *SS7* (1985 Edition), Second Edition. The following description is found in Section AI.5.5 of *SS7* concerning the case where the value of A_2 is judged to be 'unlimited' in the Q system.

"AI.5.5. Low specific activity materials

The 1973 Edition of the *Regulations* recognized a category of materials whose specific activities are so low that it is inconceivable that an intake could occur which would give rise to a significant radiation hazard, namely low specific activity (LSA) materials. These were defined in terms of a model where it was assumed that it is most unlikely that a person would remain in a dusty atmosphere long enough to inhale more than 10 mg of material. Under these conditions, if the specific activity of the material is such that the mass intake is equivalent to the activity intake assumed to occur for a person involved in an accident with a Type A package, namely 10^{-6} A_2, then this material should not present a greater hazard during transport than the quantities of radioactivity in Type A packages. This hypothetical model is retained within the Q system and leads to an LSA limit of $10^{-4} \times Q_C$ g^{-1}; thus the Q values for those radionuclides whose specific activity is below this level are listed as unlimited. In the cases where this criterion is satisfied the radioactivity associated with 10 mg of the nuclide is less than the appropriate ALI value recommended by the ICRP."

That is, where radioactivity from 10 mg of the material in question does not exceed the annual limit on intake (ALI) in ICRP 30 then the value of A_2 is judged to be 'unlimited' in the Q system. Consequently, if $\zeta \leq 1$, where:

$$\zeta = \Sigma_i \frac{\textit{Radioactivity 10 mg of uranium isotope } i \textit{ (Bq)}}{\textit{ALI of uranium isotope } i \textit{ (Bq)}}$$

then the value of A_2 can be judged to be 'unlimited'.

Reference to the chemical form in calculation of the value A_2 in the Q system is described as follows in *SS7*.

"AI.4.3. Q_C — Internal dose via inhalation

$$Q_C = \frac{ALI\ (Bq)}{10^6}\ TBq$$

This expression does not explicitly take account of the chemical form in which material is transported, but the use of the most restrictive of the ALI values recommended by the ICRP ensures that the most restrictive situation is considered. Where the likely degree of solubility or the chemical form (organic or inorganic) of a particular radionuclide is known, the degree of pessimism introduced by this procedure can be quantified by reference to the tabulated ALI values".

It is important that consideration is given to potential accident scenarios in which the chemical form can change through reactions with the surrounding environment and materials. The most pessimistic value for the ALI should be adopted.

In the case of enriched reprocessed uranium, such chemical forms can be specified on the basis of the fuel cycle flow as indicated in Figure 2, and UF_6 and UO_2 can be cited as materials which are frequently transported. Other possible forms include $UO_2(NO_3)_2$ UO_2F_2, UF_4 and UO_3. According to ICRP 30, among these uranium compounds, those whose inhalation class is D (water-soluble) are UF_6 and UO_2F_2, and those of inhalation class W (moderately soluble) are UO_3 and UF_4.

To provide an example calculations were applied to a composition arising from a hypothetical cycle:

Initial enrichment of unirradiated uranium	5.0% ^{235}U
Burnup	55 GW·d/t U
Uranium reprocessed and re-enriched	5.0% ^{235}U
Specific activity	5×10^5 Bq/g U
ζ (inhalation class Y)	5.4

The isotopic composition was based upon the burnup calculation code ORIGEN and the enrichment code CENTIS.

When ζ values were calculated using ALI values of uranium nuclides corresponding to inhalation classes D or W indicated in ICRP 30, the result is, as indicated in Table V (ζ = 0.46– 0.62).

For soluble or moderately soluble compounds of reprocessed uranium, by using ALI values of uranium nuclides corresponding to the inhalation class D or W indicated in ICRP 30, those uranium compounds can be classified as LSA-II.

This is also shown for uranium hexafluoride under the ASTM C-996-90 Standard (enriched reprocessed uranium: ERU) in Table VI.

TABLE V. EXAMPLE OF REPROCESSED URANIUM CLASSIFIED AS LSA-II (CHEMICAL FORM OF URANIUM IS CONSIDERED)

Uranium isotopes	Uranium specific activity[a] [Bq/g U]	Inhalation class D		Inhalation class W	
		ALI [b] [Bq]	Activity in 10 mg U/ALI	ALI [b] [Bq]	Activity in 10 mg U/ALI
^{232}U	2.24×10^4	8×10^3	0.028	1×10^4	0.022
^{228}Th	2.24×10^4 [c]	4×10^2	0.559	4×10^2	0.559
^{234}U	3.93×10^5	5×10^4	0.079	3×10^4	0.131
^{235}U	4.00×10^3	5×10^4	0.001	3×10^4	0.001
^{236}U	6.96×10^4	5×10^4	0.014	3×10^4	0.023
^{238}U	5.22×10^5	5×10^4	0.002	3×10^4	0.004
TOTAL	5.22×10^5	-	0.682 = ζ value	-	0.740 = ζ value
Remarks		Uranium compound and ζ value UF_6: 0.461 UO_2F_2: 0.527		Uranium compound and ζ value UO_3: 0.616 UF_4: 0.561	

[a] Results calculated by ORIGEN and CENTIS code.
Assumptions for calculation:
Initial enrichment: 5.0%
Reactor type: PWR
Burnup: 55 GW·d/t U
Specific power: 38 MW/t U
Enrichment of reprocessed uranium: 5.0%.

[b] Value of ICRP 30 (1977), but value of ^{228}Th is inhalation class W (minimum).

[c] In equilibrium with ^{232}U parent.

TABLE VI. EXAMPLE OF URANIUM HEXAFLUORIDE CLASSIFIED AS LSA-II (CHEMICAL FORM OF URANIUM IS CONSIDERED)

Uranium isotopes	Uranium specific activity[a] [Bq/g U]	Inhalation class D	
		ALI [b]	Activity in 10 mg UF_6/ALI
^{232}U	4.14×10^4	8×10^3	0.035
^{238}Th	4.14×10^4 [c]	4×10^2	0.700
^{234}U	4.62×10^5	5×10^4	0.062
^{235}U	4.00×10^3	5×10^4	0.001
^{236}U	8.40×10^4 [d]	5×10^4	0.011
^{238}U	1.15×10^4	5×10^4	0.002
TOTAL	6.44×10^5	-	0.811

[a] Value of enriched reprocessed uranium, enrichment 5% ASTM C-996-90 "Standard Specification for Uranium Hexafluoride Enriched to less 5% ^{235}U".

[b] Value of ICRP 30 (1977), but value of ^{228}Th is inhalation class W (minimum).

[c] In equilibrium with ^{232}U parent.

[d] Estimated value (not specified by ASTM Spec).

Appendix VI

MEASUREMENT OF LEACHING RATIO OF URANIUM OXIDES

1. PURPOSE

To measure the leaching ratio of uranium oxides into water, and to demonstrate that these can be are classified as LSA-III.

2. EXPERIMENTAL

(1) Test samples

UO_2 pellet ..... Pellet and fragments (to simulate the broken pellet in the accident condition)

UO_2 powder
U_3O_8 powder

Test samples were supplied from the fuel manufacturers (A, B, C and D) listed in Table VII. The number of samples is two in each category.

TABLE VII. TEST SAMPLES

UO_2 powder	U_3O_8 powder	UO_2 pellet	
		pellet	fragment
A	A	—	—
B	B	B	—
—	—	C	C
—	—	D	D

(2) Test method and measuring items

(a) Test method

The test method is given in para. 603 of the IAEA *Regulations*.
The detail is shown in the Attachment I to this Appendix.

(b) Measuring items

(i) Leaching water (demineralized water)

Conductivity (mS/m) and pH at 20°C.

(ii) Samples

UO_2, U_3O_8 powder	U(%), O/U^3, F(ppm), Cl (ppm) Average grain size (μm) Specific surface area (m^2/g) Specific activity (Bq/g U)
UO_2 pellet	Specific activity (Bq/g U)

(iii) Leaching ratio

- Liquor quantity.
- Sample weight.
- U density in filtrate.

3. TEST RESULTS

The test results are shown in Table VIII.

The results show that UO_2 powder, U_3O_8 powder and UO_2 pellet have lower leaching ratios than the acceptable limit of ERU (enriched reprocessed uranium) of ASTM (Attachment 2).

4. CONCLUSION

The test result demonstrates that UO_2 powder, U_3O_8 powder and UO_2 pellet meet the leaching criterion of less than $0.1A_2$ in 7 days for LSA-III material.

[3] For UO_2 only.

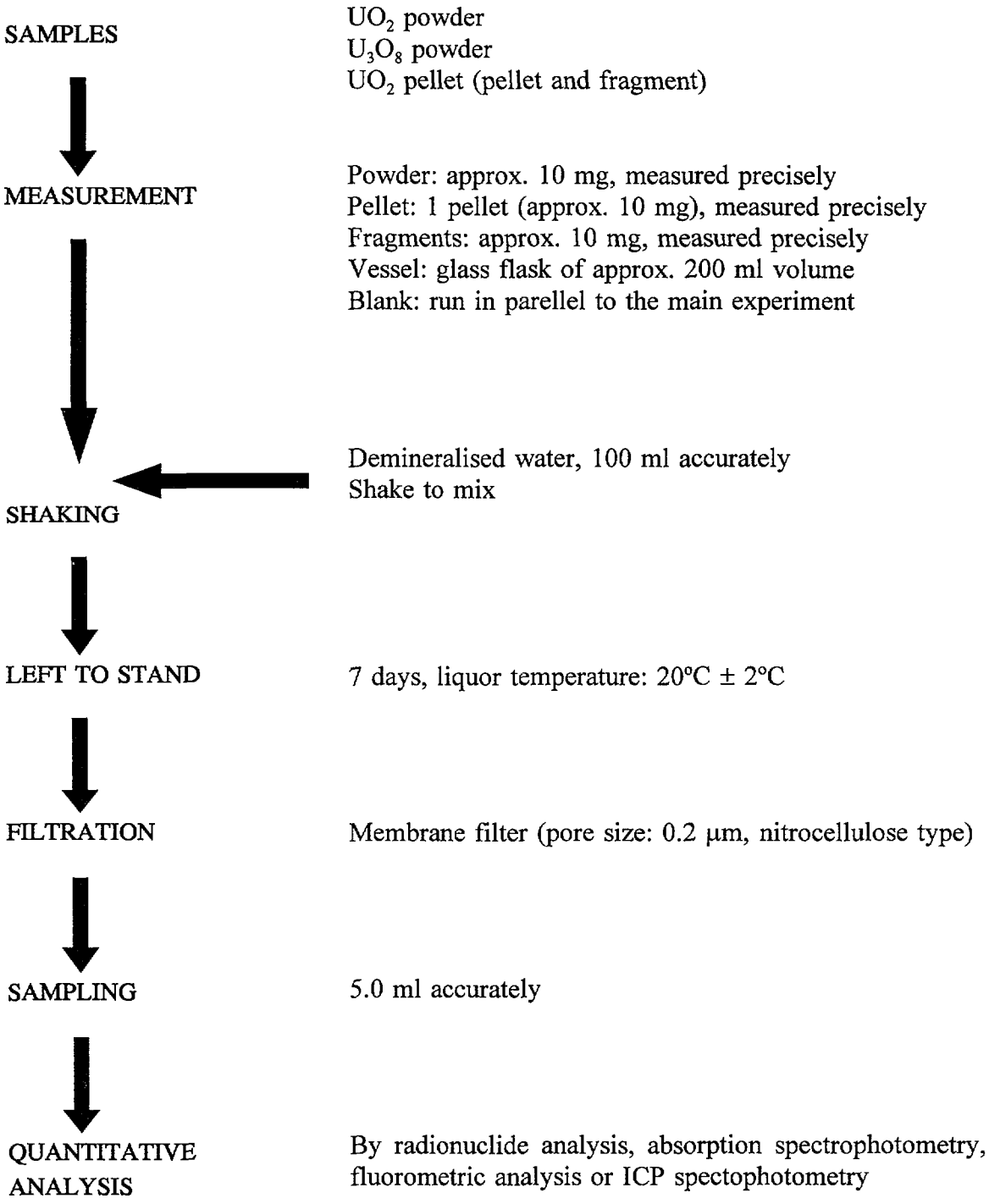
TABLE VIII. LEACHING TEST RESULTS FOR POWDER (UO_2, U_3O_8) AND PELLET

Item \ Sample	UO_2 Powder				U_3O_8 Powder				UO_2 Pellet						UO_2 Pellet Fragment			
	A_1	A_2	B_1	B_2	A_1	A_2	B_1	B_2	B_1	B_2	C_1	C_2	D_1	D_2	C_1	C_2	D_1	D_2
Weight (g)	10.416	10.116	9.947	10.022	10.083	10.091	10.058	9.868	7.838	7.846	9.197	9.153	10.063	10.093	9.012	8.798	10.221	10.527
FiltrateU ($\mu g/ml$)	19.4	20.2	38.4	40.9	14.9	14.9	2.91	3.66	7.54×10^{-3}	3.14×10^{-3}	0.5	0.5	0.106	0.121	0.7	0.6	0.197	0.212
Leached U (g)	1.94×10^{-3}	2.02×10^{-3}	3.84×10^{-3}	4.09×10^{-3}	1.49×10^{-3}	1.49×10^{-3}	2.91×10^{-4}	3.66×10^{-4}	7.54×10^{-7}	3.14×10^{-7}	5×10^{-5}	5×10^{-5}	1.06×10^{-5}	1.21×10^{-5}	7×10^{-5}	6×10^{-5}	1.97×10^{-5}	2.12×10^{-5}
Leached U (%/Sample)	0.019	0.020	0.039	0.041	0.015	0.015	2.89×10^{-3}	3.71×10^{-3}	9.62×10^{-4}	4.00×10^{-4}	5.44×10^{-4}	5.46×10^{-4}	1.05×10^{-4}	1.20×10^{-4}	7.77×10^{-4}	6.82×10^{-4}	1.93×10^{-4}	2.01×10^{-4}
Water: Conductivity (mS/m)	0.12		0.59		0.12		0.59		0.59		0.07		0.01		0.07		0.01	
Water: PH	6.4		6.6		6.4		6.6		6.6		5.0		5.7		5.0		5.7	
Water: Temp. (°C)	20		20		20		20		20		20		20		20		20	
Properties: U (%/Sample)	87.77		87.82		84.67		84.79											
Properties: O/U	2.05		2.05		—		—											
Properties: F (ppm/u)	9		32		6		< 5											
Properties: Cℓ (ppm/u)	< 2		< 5		< 2		< 5											
Properties: Mean Diameter (μm)	0.75		0.93		2.64		1.98											
Properties: Surface Area (m^2/gSample)	3.88		1.93		3.99		0.69											
Properties: Specific Activity (Bq/gU)	8.24×10^4		7.51×10^4		5.81×10^4		7.51×10^4		7.51×10^4		8.01×10^4		8.16×10^4		8.01×10^4		8.16×10^4	
Note: BNFL Data (%/Sample)	0.005 , 0.0014				6.6×10^{-5} , 4.5×10^{-5}				6×10^{-4} , 3×10^{-4}						—			
Note: Allowable Leaching Ratio* ($10^{-1}A_2$)	100 kg -UO_2 ASTH ECGU (5%) : 0.85 ERU (5%) : 0.15				100 kg-U_3O_8 ASTH ECGU (5%) : 0.85 ERU (5%) : 0.15				PWR (450 kg -UO_2 x2) ASTH ECGU (5%) : 0.094 ERU (5%) : 0.017						BWR (200 kg -UO_2 x2) ASTH ECGU (5%) : 0.21 ERU (5%) : 0.039			

*Refer to Annex II

Attachment 1 to Appendix VI

TEST METHOD OF MEASUREMENT OF LEACHING RATIO



Attachment 2 to Appendix VI

CALCULATION OF LEACHING RATIO CORRESPONDING TO "0.1A_2 VALUE"

1. Equation for leaching ratio

The equation to convert "0.1A_2" leaching criterion into leaching ratio. (L%/sample) is as follows:

$$L = 100 \times \frac{W1}{W} \ (\%)$$

where, W1: Leaching quantity

$$W1 = 0.1A_2/Q$$

Q : Specific activity (Bq/g U)

W: Weight of contents of package (g UO_2)

Then, leaching ratio (L%/sample) is:

$$L = 10 \times \frac{A_2}{W \times Q} \ (\%)$$

2. Examples of calculation

2.1. In case of 5.0% ECGU (ASTM Standard)

A_2 value:	1.1×10^9 Bq
Specific activity:	1.3×10^5 Bq/g U

(1) Package of uranium powder (W = 100 kg UO_2): L = 0.85%
(2) Package of PWR fuel bundles (W = 450 kg UO_2 × 2 bdls): L = 0.094%
(3) Package of BWR fuel bundles (W = 200 kg UO_2 × 2 bdls): L = 0.21%

2.2. In case of 5.0% ERU (ASTM Standard)

A_2 value:	8.8×10^8 Bq
Specific activity:	5.7×10^5 Bq/g U

(1) Package of uranium powder (W = 100 kg UO_2): L = 0.15%.
(2) Package of PWR fuel bundles (W = 450 kg UO_2 × 2 bdls): L = 0.017%.
(3) Package of BWR fuel bundles (W = 200 kg UO_2 × 2 bdls): L = 0.039%.

Appendix VII

TREATMENT OF URANIUM POWDERS IN THE *REGULATIONS*

It was agreed by the experts and endorsed by SAGSTRAM that the *Regulations* may be interpreted as follows with regard to insoluble material.

(1) Insoluble compounds (UO_2, U_3O_8) may be classified as LSA-III within the existing *Regulations* by demonstrating compliance with the leaching criteria, which is subject to further investigation.

(2) For dispersible materials (UO_2 and U_3O_8 powder), the application of the containment requirement for a fissile material package to prevent any loss of material under accident conditions should be considered.

A contained material classified as LSA-III is stipulated to be transported as an IP-2 or IP-3 type package in accordance with Table V and para. 426 of the *Regulations*:

> "426. LSA material and SCO, except as otherwise specified in para. 425, shall be packaged in accordance with the package integrity levels specified in Table V, in such a manner that, under conditions likely to be encountered in routine transport, there will be no escape of contents from packages, nor will there be any loss of shielding afforded by the packaging. LSA-II material, LSA-III material and SCO-II shall not be transported unpackaged.

TABLE V (of the *Regulations*). INDUSTRIAL PACKAGE REQUIREMENTS FOR LSA MATERIAL AND SCO

Contents	Industrial package type	
	Exclusive use	Not under exclusive use
LSA-I[a]		
Solid	IP-1	IP-1
Liquid	IP-1	IP-2
LSA-II		
Solid	IP-2	IP-2
Liquid and gas	IP-2	IP-3
LSA-III	IP-2	IP-3
SCO-I[a]	IP-1	IP-1
SCO-II	IP-2	IP-2

[a] Under the conditions specified in para. 425, LSA-I material and SCO-I may be transported unpackaged".

Reprocessed uranium dioxide pellets in the sintered solid state (including in a fuel assembly) falling into the category of LSA-III can be transported as a Type IP-2 or IP-3 package in accordance with the requirements and controls for transport stipulated in this section. Insoluble reprocessed uranium whose specific activity exceeds $10^{-4}A_2/g$ but does not exceed $2 \times 10^{-3}A_2/g$, in the form of uranium dioxide or triuranium octoxide powder,

requires further regulation under LSA-III due to is potential dispersibility. Applying the conclusion (2) mentioned above, the additional requirement that no material is dispersed from a package under the conditions likely to be encountered during normal conditions of transport and in an accident was added to those for Type IP-2 or IP-3 packages. Non-dispersibility is secured by the additional requirement of demonstrating no dispersal of radioactive material from a damaged package as defined in para. 564(b)(i). The transport safety of reprocessed uranium oxide powder can then be deemed as good as or better than that of LSA-III material in a Type IP-2 or IP-3 package.

Appendix VIII

A_2 OF MIXTURE

TABLE IX. EXAMPLE OF CALCULATING A_2 VALUE (MIXTURE)[a]

Uranium isotopes	Content ratio of reprocessed uranium	Specific activity of isotopes [Bq/g]	Activity of reprocessed uranium [Bq/g U]	Fraction of activity f (i)	A_2 (i) [Bq]	$\frac{f(i)}{A_2(i)}$
^{232}U	2.7×10^{-8}	8.27×10^{11}	2.24×10^{4}	0.043	3×10^{8}	1.43×10^{-10}
^{234}U	1.7×10^{-3}	2.31×10^{8}	3.93×10^{3}	0.752	1×10^{9}	7.52×10^{-10}
^{235}U	5.0×10^{-2}	8.00×10^{4}	4.00×10^{3}	0.008	unlimit	0
^{236}U	2.9×10^{-2}	2.40×10^{6}	6.96×10^{4}	0.133	1×10^{9}	1.33×10^{-10}
^{238}U	9.2×10^{-1}	1.24×10^{1}	1.14×10^{4}	0.022	unlimit	0
^{228}Th	--	3.04×10^{13}	2.24×10^{4} [b]	0.043	unlimit	1.08×10^{-10}
	1.0	--	5.00×10^{3}	1.000	--	1.14×10^{-9}
					A_2	$= 8.80 \times 10^{8}$

[a] Results calculated by ORIGEN and CENTIS codes.
Assumption for calculation:
Initial enrichment: 5%
Reactor type: PWR
Burnup: 55 GW·d/t U
Specific power: 38 MW/t U
Enrichment of reprocessed uranium: 5.0%.

[b] ^{228}Th is assumed to be equilibrium to ^{232}U.

Appendix IX

A_2 VALUE OF URANIUM WHICH IS SUSPECTED TO HAVE BEEN CONTAMINATED BY REPROCESSED URANIUM

In consideration of the possibility that reprocessed uranium may contaminate unirradiated uranium during processing, ASTM C-996-90, "Standard Specification for Uranium Hexafluoride Enriched to Less Than 5% ^{235}U" defines enriched commercial grade Uranium (ECGU) with the following limits upon uranium isotopes:

uranium-232: 0.002 $\mu g/g^{235}U$
uranium-234: 10 000 $\mu g/g^{235}U$
uranium-236: 5000 $\mu g/g^{235}U$

That is, the concentration limits of these uranium isotopes depend on the enrichment of uranium-235. The value of A_2 was calculated by the formula for calculating the A_2 value of a mixture, to examine the range of uranium enrichment for classification as LSA-II.

Table X shows the relation between uranium enrichment and A_2 values. ECGU will be transported in the form of uranium hexafluoride from enrichment facilities to reconversion facilities and in the form of uranium dioxide from reconversion facilities to fuel fabrication facilities. Based on the result in Table X, enrichment upper limits for satisfying the LSA-II classification standard (estimated mean specific activity of material is $10^{-4}A_2$ Bq/g or less) were calculated to be as follows.

Type of material	**Maximum enrichment**
UF_6	5.0%
UO_2	4.8%

As indicated in Appendix V, radioactive impurities were ignored because they make almost no contribution.

However, it must be noted that even uranium of natural origin can, in certain circumstances of enrichment, also have a ^{234}U content equal to or greater than 10 000 $\mu g/g^{235}U$ (see Table XI). For these natural circumstances the A_2 value is 1.14×10^9, almost identical to the ECGU value which includes ^{232}U, ^{228}Th and ^{236}U. However, A_2 is 'unlimited' for unirradiated uranium to 5% ^{235}U. This calculation demonstrates that ECGU may be considered to differ insignificantly and may also be accepted as 'unlimited A_2' within the specified composition.

TABLE X. THE RELATION BETWEEN URANIUM ENRICHMENT AND A_2 VALUES

Uranium isotopes	A_2(i) [Bq]	ECGU Spec[a] [µg/gU-235]	Activity[b] [Bq/g]	ENRICHMENT 4.8%			ENRICHMENT 5.0%		
				Uranium specific activity [Bq/gU]	Rate of activity f(i)	$\frac{f(i)}{A_2(i)}$	Uranium specific activity [Bq/gU]	Rate of activity f(i)	$\frac{f(i)}{A_2(i)}$
^{232}U	3×10^{8}	0.002	8.28×10^{11}	7.95×10^{1}	6.25×10^{-4}	2.08×10^{-12}	8.28×10^{1}	6.26×10^{-4}	2.09×10^{-12}
^{228}Th[c]	4×10^{8} [c]	---	3.04×10^{13}	7.95×10^{1}	6.25×10^{-4}	1.56×10^{-12}	8.28×10^{1}	6.26×10^{-1}	1.57×10^{-12}
^{234}U	1×10^{8}	10 000	2.31×10^{8}	1.11×10^{5}	8.73×10^{-1}	8.73×10^{-10}	1.16×10^{5}	8.76×10^{-1}	8.76×10^{-10}
^{235}U	∞		8.00×10^{4}	3.84×10^{3}	3.02×10^{-2}	0	4.00×10^{3}	3.02×10^{-2}	0
^{236}U	1×10^{8}	5 000	2.40×10^{8}	5.76×10^{2}	4.53×10^{-3}	4.53×10^{-12}	6.00×10^{2}	4.53×10^{-3}	4.53×10^{-12}
^{238}U	∞		1.24×10^{4}	1.16×10^{4}	9.12×10^{-2}	0	1.16×10^{4}	8.76×10^{-2}	0
^{99}Tc	9×10^{11}	0.2	6.28×10^{0}	6.03×10^{0}	4.74×10^{-6}	5.27×10^{-17}	6.08×10^{0}	4.59×10^{-3}	5.10×10^{-17}
Total		---	---	1.27×10^{5}	1.00	8.81×10^{-10}	1.32×10^{5}	1.00	8.84×10^{-10}
A_2 value	$A_2\ value = \frac{1}{\Sigma\ [f\ (i)\ /\ A_2\ (i)}$			1.14×10^{9} [Bq]			1.13×10^{9} [Bq]		
Remarks				Activity of UF_6 8.60×10^{4} [Bq/g UF_6] Activity of UO_2 1.12×10^{5} [Bq/g UO_2]			Activity of UF_6 8.95×10^{4} [Bq/gUF_6] Activity of UO_2 1.17×10^{5} [Bq/gUO_2]		

[a] ASTM C-996-90 "Standard Specification for Uranium-Hexafluoride Enriched to Less than 5% ^{235}U"

[b] IAEA *SS37* (As Amended 1990) Table AIII-1.

[c] Assumption that ^{228}Th is in radioactive equilibrium with the parent nuclide ^{232}U.

TABLE XI. VARIATION OF ^{234}U WITH ENRICHMENT FACTORS

^{235}U product assay g/100g U	^{234}U in enrichment feed $\mu g/g$ U	^{234}U in enriched product $\mu g/g^{235}U$		
		Percentage ^{235}U in tails		
		0.2	0.3	0.4
5	54	8 960	9 520	10 020
	58	9 620	10 230	10 760
4	54	8 900	9 400	9 870
	58	9 550	10 100	10 600
3	54	8 770	9 230	9 630
	58	9 430	9 900	10 330

Appendix X

CONTRIBUTION OF RADIOACTIVE IMPURITIES IN REPROCESSED URANIUM TO THE VALUE OF A_2

In calculating the value of A_2 of reprocessed uranium with the aim to classify it into one of the LSA categories, it is important to always take into account daughter nuclides of uranium isotopes and nuclides of radioactive impurities (FP, TRU). Whether the nuclides of radioactive impurities (FP, TRU) can be ignored or not is established by taking their representative nuclides into a calculation of the value of A_2 and in accordance with para. 304 of the *Regulations*.

The forms and quantities of radioactive impurities (FP and TRU) contained in reprocessed uranium are determined in each case by several factors including the decontamination factor at the time of its reprocessing. Table XII illustrates that these radioactive impurities make almost no contribution to the value of A_2 of reprocessed uranium at levels specified in national standards.

TABLE XII CONTRIBUTION OF RADIOACTIVE IMPURITIES TO THE VALUE OF A_2 (TEST CALCULATION)

Nuclides	A_2 value [Bq][a]	ERU (ASTM)		ERU (case of Japan)		ECGU (ASTM)	
		Specification[b] [Bq/g U]	Specific activity /$10^{-4}A_2$	Specification[c] [Bq/g U]	Specific activity /$10^{-4}A_2$	Specification[b] [Bq/g U]	Specific activity /$10^{-4}A_2$
^{99}Tc	9×10^{11}	3 200 (5 μg/g U)	$3\ 5 \times 10^{-5}$	2	$2\ 2 \times 10^{-6}$	6 3 (0 01 μg/g U)	$7\ 0 \times 10^{-6}$
^{106}Ru	2×10^{11}	—	—	15	$7\ 5 \times 10^{-7}$	—	—
^{237}Np	2×10^{8}	3 3 (200 dpm/g U)	$1\ 7 \times 10^{-4}$	0 01	$5\ 0 \times 10^{-7}$	0 001	$5\ 0 \times 10^{-8}$
Pu-α	2×10^{8}			0 001	$5\ 0 \times 10^{-7}$		
^{241}Pu	1×10^{10}			0 03	$3\ 0 \times 10^{-6}$	—	—
TOTAL		—	$2\ 1 \times 10^{-4}$ (="ζ" value for radioactive impurities)	—	$1\ 4 \times 10^{-4}$ (="ζ" value for radioactive impurities)	—	$1\ 2 \times 10^{-7}$ (="ζ" value for radioactive impurities)

[a] IAEA *Regulations* for the Safe Transport of Radioactive Material (1985 Edition)

[b] Specification of enriched commercial grade uranium hexafluoride (ECGU) in ASTM C 996 draft and enriched reprocessed uranium hexafluoride (ERU) Enrichment 5%

[c] Example of discussion in Japan Enrichment 3 9%

Since irradiated uranium is reprocessed and refined into reprocessed uranium, it is possible that the latter contains, in addition to representative nuclides shown in the Table XII, radioactive impurities (FP and TRU) with even smaller contributions to the value of A_2. However, as indicated in the Tables XIII (a) and (b), their impact on the value of A_2, specific activity, and the ratio of A_2 value to specific activity is so small as to be negligible.

TABLE XIII. EXTENT OF CONTRIBUTION OF RADIOACTIVE IMPURITIES AS A WHOLE TO THE VALUE OF A_2 (TEST CALCULATION WITH ENRICHED REPROCESSED URANIUM)

(a) All nuclides taken into account

	Nuiclides	Density of activib (Bq/gU)	Fraction of activity f(i)	A_2 X(i)(Bq)	f(i)/X(i)	Effect of A_2 value
Uranium isotops	U-232	2.95 E+03	1.33 E-02	3E+08	4.43 E-11	4.90 E+00
	U-234	1.64 E+05	7.39 E-01	1E+09	7.39 E-10	8.182E+01
	U-235	3.14 E+03	1.41 E-02	unlimited	0.00 E+00	0.00 E+00
	U-236	2.27 E+04	1.02 E-01	1E+09	1.02 E-10	1.13 E+01
	U-238	1.19 E+04	5.36 E-02	unlimited	0.00 E+00	0.00 E+00
	subtotal	2.047E+05	9.220E-01		8.853E-10	9.802E+01
Daughter nuclide of uranium isotopes	Ra-223	3.87 E-03	1.74 E-08	3E+10	5.80 E-19	6.41 E-08
	Ra-226	1.28 E-03	5.77 E-09	2E+10	2.89 E-19	3.20 E-08
	Ra-228	2.16 E-07	9.73 E-13	4E+08	2.43 E-21	2.69 E-10
	Ac-227	4.15 E-03	1.87 E-08	2E+07	9.35 E-16	1.04 E-04
	Ac-228	2.16 E-07	9.73 E-13	4E+11	2.43 E-24	2.69 E-13
	Th-227	3.81 E-03	1.72 E-08	1E+10	1.72 E-18	1.90 E-07
	Th-228	1.54 E+03	6.94 E-03	4E+08	1.74 E-11	1.93 E+00
	Th-230	2.96 E+00	1.33 E-05	2E+08	6.65 E-14	7.36 E-03
	Th-231	3.14 E+03	1.41 E-02	9E+11	1.57.E-14	1.74 E-03
	Th-232	2.24 E-06	1.01 E-11	unlimited	0.00 E+00	0.00 E+00
	Th-234	1.19 E+04	5.36 E-02	2E+11	2.68 E-13	2.97 E-02
	Pa-231	3.33 E-01	1.50 E-06	6E+07	2.50 E-14	2.77 E-03
	subtotal	1.658E+04	7.465E-02		1.778E-11	1.972E+00
Transuranic element (TRU)	Np-237 *	2.71 E+00	1.22 E-05	2E+08	6.10 E-14	6.75 E-03
	Pu-236	1.76 E-06	7.93 E-12	7E+08	1.13 E-20	1.25 E-09
	Pu-238 *	1.24 E-01	5.59 E-07	2E+08	2.80 E-15	3.10 E-04
	Pu-239 *	3.52 E-02	1.59 E-07	2E+08	7.95 E-16	8.80 E-05
	Pu-240	4.85 E-02	2.18 E-07	2E+08	1.09 E-15	1.21 E-04
	Pu-241 *	7.00 E+00	3.15 E-05	1E+10	3.15 E-15	3.49 E-04
	Pu-242	1.19 E-04	5.36 E+10	2E+08	2.68 E-18	2.97 E-07
	Am-241	1.86 E-11	8.38 E-17	2E+08	4.19 E-25	4.64 E-14
	Am-242 m	9.99 E-14	4.50 E-19	2E+08	2.25 E-27	2.49 E-16
	Am-243	1.48 E-13	6.66 E-19	2E+08	3.33 E-27	3.69 E-16
	subtotal	9.918E+00	4.464E-05		6.884E-14	7.618E-03
Fission Product (FP)	Se-79	5.54 E-06	2.50 E-11	2E+12	1.25 E-23	1.38 E-12
	Zr-93	6.56 E-04	2.95 E-09	2E+11	1.48 E-20	1.64 E-09
	Nb-93 m	1.57 E-04	7.07 E-10	6E+12	1.18 E-22	1.31 E-09
	Tc-99 *	7.30 E+02	3.29 E-03	9E+11	3.66 E-15	4.95 E-04
	Rh-102*	5.46 E-04	2.46 E-09	5E+11	4.92 E-21	5.45 E-10
	Ru-106*	3.26 E-00	1.47 E-05	2E+11	7.35 E-17	8.14 E-06
	Pd-107	1.46 E-04	6.58 E-10	unlimited	0.00 E+00	0.00 E+00
	Ag-110m	7.38 E-10	3.32 E-15	4E+11	8.30 E-27	9.19 E-16
	Cd-113m	3.95 E-15	1.78 E-20	9E+10	1.98 E-31	2.19 E-20
	Sn-119m	8.48 E-08	3.82 E-13	4E+13	9.55 E-27	1.06 E-15
	Sn-121m	2.39 E-06	1.08 E-11	9E+11	1.20 E-23	1.33 E-12
	Sn-123	2.74 E-11	1.23 E-16	5E+11	2.46 E-28	2.72 E-17
	Sn-126	1.00 E-05	4.50 E-11	3E+11	1.50 E-22	1.66 E-11
	Sb-125	1.76 E-02	7.93 E-08	9E+11	8.81 E-20	9.75 E-09
	Te-125m	1.49 E-09	6.71 E-15	9E+12	7.46 E-28	8.26 E-17
	Te-127m	2.30 E-10	1.04 E-15	5E+11	2.08 E-27	2.30 E-16
	I-129	4.28 E-05	1.93 E-10	unlimited	0.00 E+00	0.00 E+00
	Cs-134	4.14 E-07	1.86 E-12	5E+11	3.72 E-24	4.12 E-13
	Cs-135	5.86 E-11	2.64 E-16	9E+11	2.93 E-28	3.24 E-17
	Cs-137	1.09 E-05	4.91 E-11	5E+11	9.82 E-23	1.09 E-11
	Ce-144	1.05 E-11	4.73 E-17	2E+11	2.37 E-28	2.62 E-17
	Sm-151	5.07 E-15	2.28 E-20	4E+12	5.70 E-33	6.31 E-22
	subtotal	7.333E+02	3.305E-03		3.734E-15	5.032E-04
Total		2.222E+05	1.000E+00		9.032E-10	1.000E+02
					A_2=1.107E9	

* : Typical nuclides of radioactive impurities (FP, TRU)
Note : Example of enriched reprocessed uranium in Japan.

(b) Only representative nuclides evaluated

	Nuiclides	Density of activib (Bq/gU)	Fraction of activity f(i)	A_2 X(i)(Bq)	f(i)/X(i)	Effect of A_2 value
Uranium isotops	U-232	2.95 E+03	1.33 E-02	3E+08	4.43 E-11	4.90 E+00
	U-234	1.64 E+05	7.39 E-01	1E+09	7.39 E-10	8.182E+01
	U-235	3.14 E+03	1.41 E-02	unlimited	0.00 E+00	0.00 E+00
	U-236	2.27 E+04	1.02 E-01	1E+09	1.02 E-10	1.13 E+01
	U-238	1.19 E+04	5.36 E-02	unlimited	0.00 E+00	0.00 E+00
	subtotal	2.047E+05	9.220E-01		8.853E-10	9.802E+01
Daughter nuclide of uranium isotopes	Th-228	1.54 E+03	6.94 E-03	4E+08	1.74 E-11	1.93 E+00
	subtotal	1.658E+04	7.465E-02		1.74 E-11	1.93 E+00
Radioactive impurities nuclides	Np-237	2.71 E+00	1.22 E-05	2E+08	6.10 E-14	6.75 E-03
	Pu(α)	2.08 E-01	9.36 E-07	2E+08	4.68 E-15	4.19 E-04
	Pu-241	7.00 E+00	3.15 E-05	1E+10	3.15 E-15	3.49 E-04
	Tc-99	7.30 E+02	3.29 E-03	9E+11	3.66 E-15	4.05 E-04
	Ru-106	3.26 E+00	1.47 E-05	2E+11	7.35 E-17	8.14 E-06
	subtotal	7.431E+02	3.349E-03		7.257E-14	8.031E-03
Total		2.220E+05	1.000E+00		9.032E-10	9.998E-01
					A_2=1.107E9	

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Appendix XI

DAUGHTER NUCLIDES OF ^{228}Th AND THEIR INGROWTH

According to para. 303 of the *Regulations*, for calculation of the value of A_2, a daughter nuclide with a half-life of either more than 10 days or longer than that of its parent nuclide is to be evaluated independently. In reprocessed uranium, each of the uranium isotopes has its own decay chain in which a daughter nuclide is generated. Therefore those daughter nuclides must be considered.

TABLE XIV. CONSIDERATION OF DAUGHTER NUCLIDES FROM REPROCESSED URANIUM

Parent Nuclides	Daughter nuclides with shorter half-lives	Daughter nuclides with longer half-lives	Feature for calculation of A_2 values
^{232}U	^{228}Th	—	^{228}Th
^{233}U	^{229}Th ^{225}Ra ^{225}Ac	—	negligible contribution of daughter nuclides due to a very small amount of products
^{234}U	^{230}Th ^{226}Ra ^{210}Pb ^{210}Po	—	"
^{235}U	^{231}Pa ^{227}Ac ^{227}Th ^{223}Ra	—	"
^{236}U	^{232}Th ^{228}Ru ^{228}Th	^{232}Th	"
^{238}U	^{234}Th ^{234}U ^{230}Th ^{226}Ra ^{210}Pb ^{210}Po	—	"

Of the many daughter nuclides subject to such evaluation, according to the Table XIV, only ^{228}Th needs to be taken into account in calculating the value of A_2. It must be borne in mind that ^{228}Th builds up over several years in Table XV [7].

TABLE XV. ^{228}Th INGROWTH[a]

Years	% of equilibrium	Activity (Bq/gU)
0.25	0.087	72
0.50	0.166	141
1.0	0.304	248
2.0	0.516	431
4.0	0.766	638
8.0	0.945	787
equilibrium	1.0	828

[a] Activity of ^{228}Th [$^{232}U = 10^{-9}$g/gU (1 ppb)].

Therefore, if the effect of the daughter nuclide ^{228}Th is to be conservatively evaluated, a radioactive equilibrium with its parent nuclide ^{232}U must be assumed.

Appendix XII

CLASSIFICATION OF AN EMPTY CYLINDER CONTAINING UF_6 HEEL

The specific activity of heel in an empty cylinder for uranium hexafluoride is calculated as follows, which is shown with an example of UF_6 before enrichment.

Assumptions

Reprocessed uranium

PWR 5.0% E, 55 GW·d/t, 38 MW/t, reprocessed 4 years later, evaporated 10 years later, 30 days attenuation.

48Y cylinder

Filled amount: 9525 kg UF_6 or 6440 kg U: minimum filled amount [8].

Heel: 22.68 kg UF_6 or 15.33 kg U: maximum permissible heel amount [8].

TABLE XVI. CALCULATION OF A_2 VALUE FOR HEELS

Nuclide	Activity of heels (Bq/heel)	f(i)	A_2(i) (Bq)	f(i)/A_2(i)	A_2(Bq)
^{232}U	4.4×10^{7}	0.0011	3.0×10^{8}	3.7×10^{-12}	
^{228}Th	1.6×10^{10}	0.40	4.0×10^{8}	1.6×10^{-9}	
^{234}U	9.0×10^{8}	0.022	1.0×10^{9}	2.2×10^{-11}	
^{230}Th	3.4×10^{8}	0.00084	2.0×10^{8}	4.2×10^{-12}	9.8×10^{8}
^{235}U	1.1×10^{7}	0.00084	2.0×10^{8}	-	
^{236}U	2.7×10^{8}	0.0067	1.0×10^{9}	6.7×10^{-12}	
^{238}U	1.9×10^{8}	0.0047	1.0×10^{9}	-	
^{234}Th	2.3×10^{10}	0.57	2.0×10^{11}	2.9×10^{-12}	
TOTAL	4.0×10^{10}	1.00	-	1.02×10^{-9}	

On the basis of the above calculation, the transported item in question is classified as follows:

1. Radioactivity of heel in 48Y cylinder

$$\frac{4.04 \times 10^{10}}{22.68 \times 10^{3}} \frac{(Bq)}{(g\ UF_6)} \quad 1.78 \times 10^{6}\ (Bq/g\ UF_6)$$

2. LSA-II criterion

$$10^{-4}\ A_2/g = 10^{-4} \times 9.8 \times 10^{8} = 9.8 \times 10^{4}\ Bq/g$$

Since the specific activity under 1 is greater than that under 2 the transported item does not satisfy the standard for LSA-II. Alternatively, to qualify for a Type A transport, the total activity must be less than the A_2 value, 9.8×10^8 Bq. However, Table XVI shows the total activity to be much greater than this at 4.0×10^{10} Bq.

If an empty container is transported, its contamination level must not exceed 10^3 times the level specified for transported matter exempted from Sections 407 and 421 of the *Regulations* (0.04 Bq/cm^2 for a radioactive body). If the matter in question is deemed to be a surface contaminated object (SCO), the contamination level of its inaccessible surface must be 4×10^3 Bq/cm^2 or less to be SCO-I, and 8×10^4 Bq/cm^2 or less to be SCO-II (both for the case of a radioactive body).

Taking the example of a 48Y cylinder, its internal surface area is 1.70×10^5 cm^2, and its radioactivity is 4.04×10^{10} Bq/heel. Therefore the average contamination level is:

$$\frac{4.04 \times 10^{10}}{1.70 \times 10^5} = 2.4 \times 10^5 \ Bq/cm^2$$

Hence it does not satisfy any of those categories.

In practice the heel is not uniformly distributed following vaporization of the contents but is more concentrated in a band along the base of the cylinder. This is of significance to gamma dose measurements.

Annex

INVESTIGATIONS ON THE TRANSPORT OF UF_6 IN 48Y CONTAINERS WITH URANIUM FROM REPROCESSED PWR FUEL ELEMENTS

Abstract and selected results
prepared for BMU, Bonn

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Wuppertal, March 1990

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Abstract

The aim of the assessment is the determination of environmental dose rates of 48Y containers as used for transportation of UF_6 taking into account uranium isotope vectors to be expected for reprocessed uranium vectors and the definition of possible countermeasures to reduce dose rates. Results of some measurements of dose rates are given for some examples. Calculation of dose rates are based on the ISOSHLD-programme. The model applied in ISOSHLD-programme was fitted in a suitable manner for calculating dose rates in the environment of 48Y containers. As radiation sources four uranium vectors were used (natural uranium, uranium-composition according to ASTM-specification, reprocessed uranium from PWR-fuel elements with an burnup value of 50 000 MWd (t U)$^{-1}$, reprocessed uranium from PWR-fuel elements with a burnup value of 27 000 MWd (t U)$^{-1}$). Relevant daughter nuclides are taken into account too. The two container states "container with homogeneous maximum filling" and "container homogeneously drained (evacuated)" are considered. The results obtained show: Legal dose rate limit values for external radiation exposure are in some cases significantly exceeded. Under certain circumstances drained (evacuated) containers may produce significantly higher dose rates in the vicinity of an 48Y container compared with containers filled with natural uranium. If the 48Y container is completely filled with natural uranium, even a certain residual amount of daughter radionuclides from preceding fillings with other uranium vectors may lead to environmental dose rates below limit values. Possible countermeasures to reduce dose rates may be: Limitation of time intervals between reprocessing of fuel elements or conversion and transport, intermediate purification of containers according to the special requirements and the use of overpackings (shieldings) of 48Y containers according to special requirements.

1. DESIGN PROPERTIES OF 48Y CONTAINERS

Basic properties of the 48Y container are compiled in Table 1. A picture of this type of container is given in Figure 1. The maximum content of the 48Y container is 12.5 t UF_6.

TABLE 1. DATES OF THE 48 Y-CONTAINER

diameter	1219 mm (48 in.)
length	3810 mm (150 in)
wall thickness	16 mm (5/8 in.)
minimum wall thickness	12, 7 mm (1/2 in.)
tare weight	2359 kg (5200 lb)
net weight max.	12501 kg (27560 lb)
gross weight	14860 kg (32760 lb)
volume	4, 04 m (142, 7 ft)
material	Stahl (TTSTE 29 = ASTM A516^0 55 oder 60)
work pressure	14 bar (200 psig)
test pressure	28 bar (400 psig)
planned outer pressure	1, 52 bar (22 psig)
planned temperature	-40^{0}C bis 121, 1 ^{0}C (-40^{0}F bis 250 ^{0}F)
accumulation max.	4,5 Gew.-% U-235)
filling limit max	12501 kg (27560 lb)
"heel"-weight max.	22, 7 (50 lb)

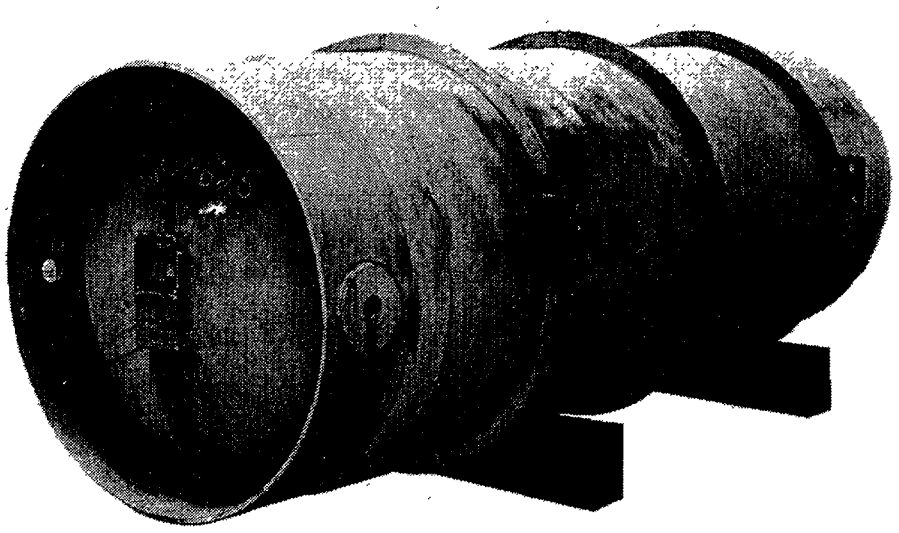

FIG. 1. UF_6 cylinder model 48Y.

2. MODELLING FOR APPLYING THE ISOSHLD-PROGRAMME

The programme ISOSHLD has been applied to a container geometry shown in Figure 2. Dose rates were calculated on the lines "1", "2" ... "7" in certain defined distances from the model surface. The programme ISOSHLD is fitted to a cylindrical geometry. However, the programme ISOSHLD does not allow to calculate dose rates at points in areas as shown in a scraped manner according to Figure 3. The geometrical model therefore had to be modified by "expanding" the given model container A by an "auxiliary" model container B under the assumption that both containers A and B may be filled with UF_6 in an equivalent manner. The dose rate at any point on the lines 5 or 6, for example, may be gained by calculating the total dose rate, caused by the radiation source A + B diminished by the dose rate caused by the source B. Dose rate data calculated in this way by applying the programme ISOSHLD on the modified model, as indicated in Figure 5, are in good agreement with measurements as demonstrated in Figure 5.

Dose rates for drained (evacuated) containers were calculated by using a model shown in Figure 6. This model is essentially based on the assumption that daughter radionuclides may be concentrated within a thin layer of 1cm thickness on the inner container surface. This assumption is used for modelling the container to calculate the spatial dose rate. The comparison of calculated values with experimental data show an acceptably good agreement of data.

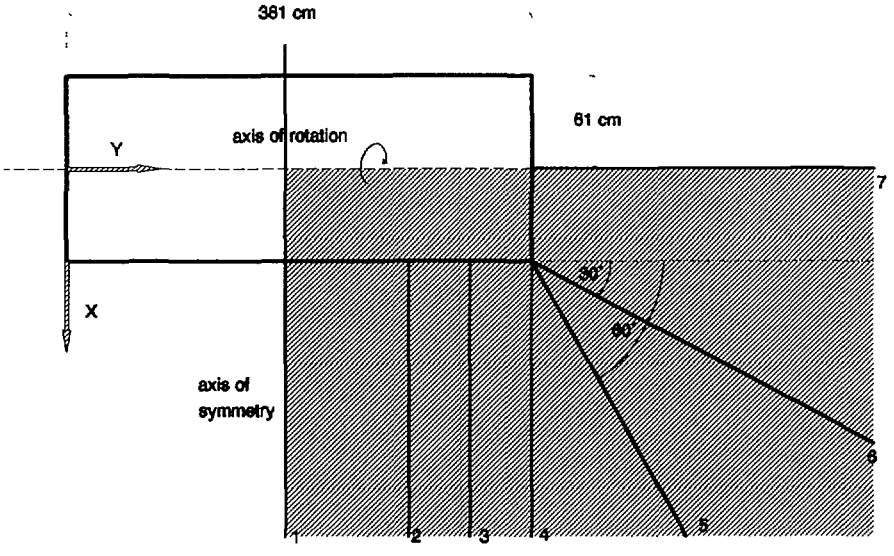

FIG. 2. Geometry of the 48Y container.

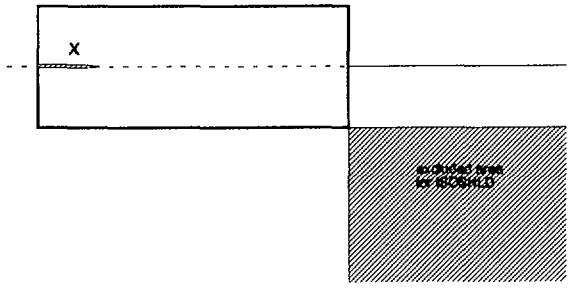

FIG. 3. Excluded areas for application of ISOSHLD.

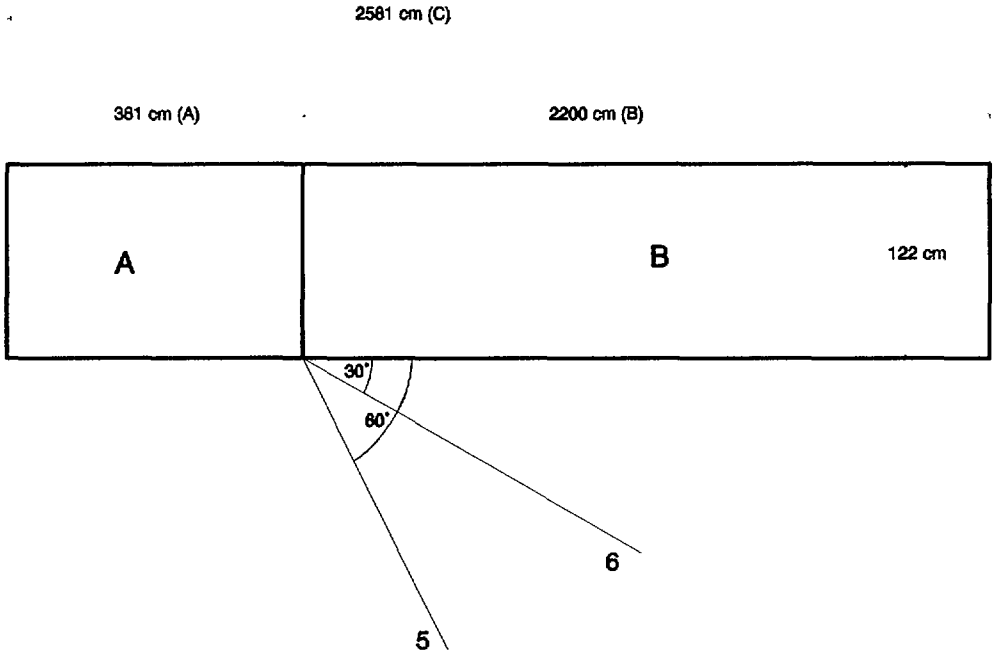

FIG. 4. Modified models for applying the ISOSHLD programme on 'excluded' areas.

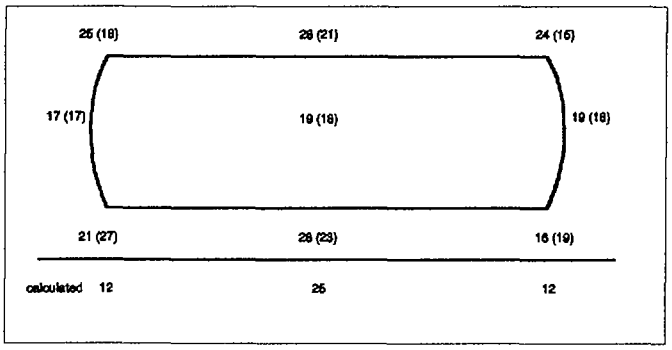

FIG. 5. Calculated and measured values of dose rates at the surface of a drained container 48Y (values in $\mu Sv\ h^{-1}$).

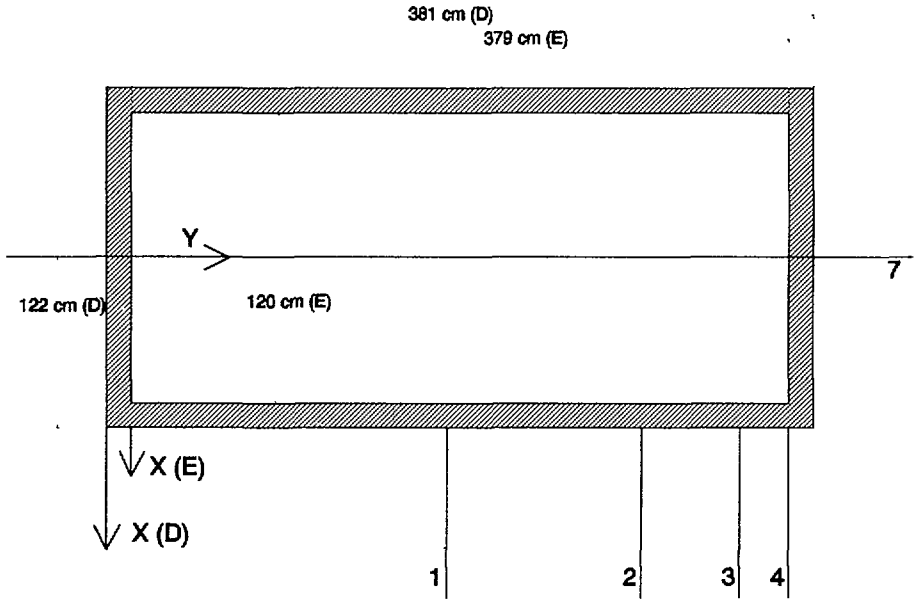

FIG. 6. Model of drained container.

3. SELECTED RESULTS

3.1. Uranium vectors

As sources four uranium vectors were defined. The isotope compositions are figured in Table 2.

3.2. Influence of time on dose rates

The concentrations of different daughter radionuclides may be built up, depending on time, resulting in corresponding time depending dose rates at points taken into account.

The dose rates in case of vector 1 do not depend on time because mother and daughter radionuclides are practically in radioactive equilibrium. The relative concentrations do not change with time either, Figure 7. In cases of uranium vectors 3 and 4 the dose rates will change with time as is demonstrated in Figure 8 and Figure 9.

TABLE 2. USED URANIUM VECTORS

Uranium vector 1 (natural uranium)

U-234	0.005	Mol.-%	=	4.916	$\times 10^{-5}$ g/gU
U-235	0.720	Mol.-%	=	7.10971	$\times 10^{-3}$ g/gU
U-238	99. 275	Mol.-%	=	9.92841	$\times 10^{-1}$ g/gU

Uranium vector 2 (average vector Uranit; Burnup: 27.000 MWd $(t.U)^{-1}$)

U-232	6.5×10^{-8}	Mol.-%	=	6.337	$\times 10^{-10}$ g/gU
U-234	0.0144	Mol.-%	=	1.416	$\times 10^{-4}$ g/gU
U-235	0.81	Mol.-%	=	7.99891	$\times 10^{-3}$ g/gU
U-236	0.31	Mol.-%	=	3.0743	$\times 10^{-3}$ g/gU
U-238	98.8656	Mol.-%	=	9.88785	$\times 10^{-1}$ g/gU

Uranium vector 3 (proposed on ASTM-specification)

U-232	5.1×10^{-6}	Mol.-%	=	5.000	$\times 10^{-8}$ g/gU
U-234	0.0488	Mol.-%	=	4.800	$\times 10^{-4}$ g/gU
U-235	1.0000	Mol.-%	=	9.8764	$\times 10^{-3}$ g/gU
U-236	0.8470	Mol.-%	=	8.400	$\times 10^{-3}$ g/gU
U-238	98.1042	Mol.-%	=	9.8124	$\times 10^{-1}$ g/gU

Uranium vector 4 (this vector has been chosen because of calculations of Uranit; Burnup: 50,000 MWd . $(t.U)^{-1}$)

U-232	3.0×10^{-8}	Mol.-%	=	2.900	$\times 10^{-10}$ g/gU
U-234	0.020	Mol.-%	=	2.000	$\times 10^{-4}$ g/gU
U-235	0.700	Mol.-%	=	6.910	$\times 10^{-3}$ g/gU
U-236	0.600	Mol.-%	=	5.950	$\times 10^{-3}$ g/gU
U-238	98.680	Mol.-%	=	98.8694	$\times 10^{-1}$ g/gU

3.3. Spatial distribution of dose rates

Lines of constant dose rates are shown in Figure 10 through Figure 12 for container fillings using uranium vectors 1, 2 and 3 respectively.

The legal dose rate values are in some cases significantly exceeded. Similar results were gained from model calculations applied to the drained (evacuated) container. Figure 13 gives results for the container previously filled with uranium of vector 1-type.

In Figure 14 and Figure 15 corresponding examples are given. In these cases limit values are partly significantly exceeded too.

3.4. Influence of uranium composition (uranium vector)

The comparison of surface dose rates for all four uranium vectors indicates, corresponding to results shown in Figure 16 and Figure 17, that dose rates will grow. Especially in case of vectors 3 and 4 the legal limit value will not be exceeded after 1 year, but it will after 4 years. The comparison between dose rates for filled and drained containers resulted in essentially higher dose rate values for drained containers as indicated in Figure 18 and Figure 19, respectively.

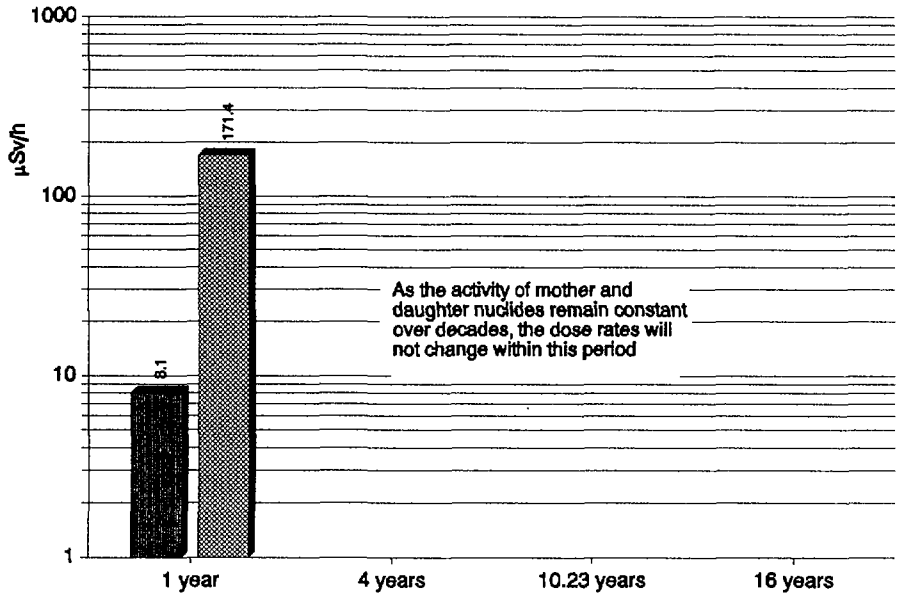

FIG. 7. Dose rates for vector 1.

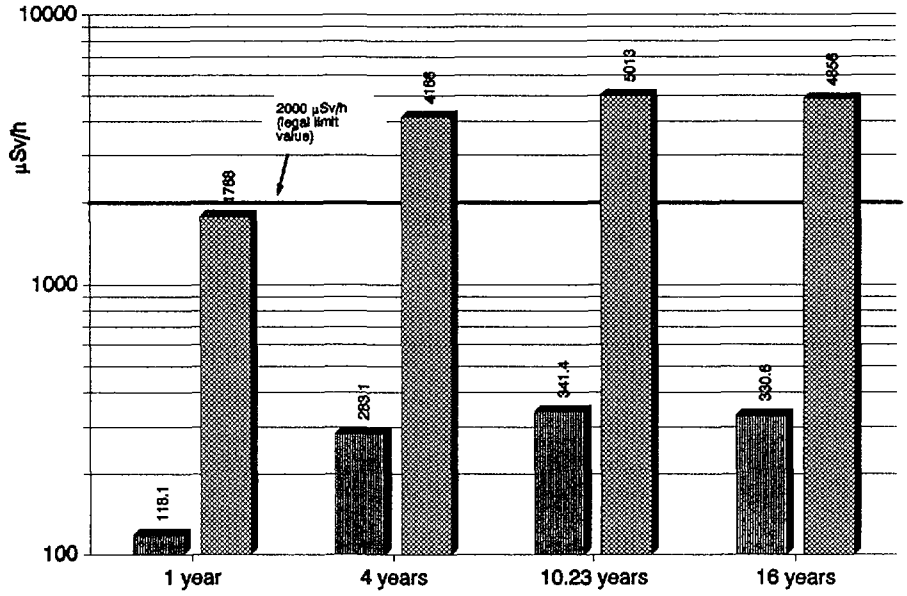

FIG. 8. Dose rates for vector 3.

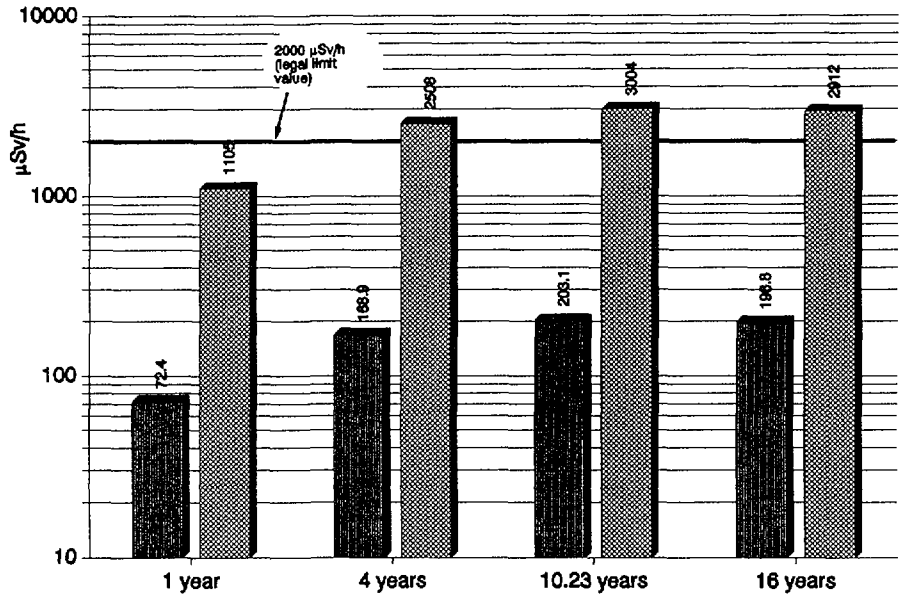

FIG. 9. Dose rates for vector 4.

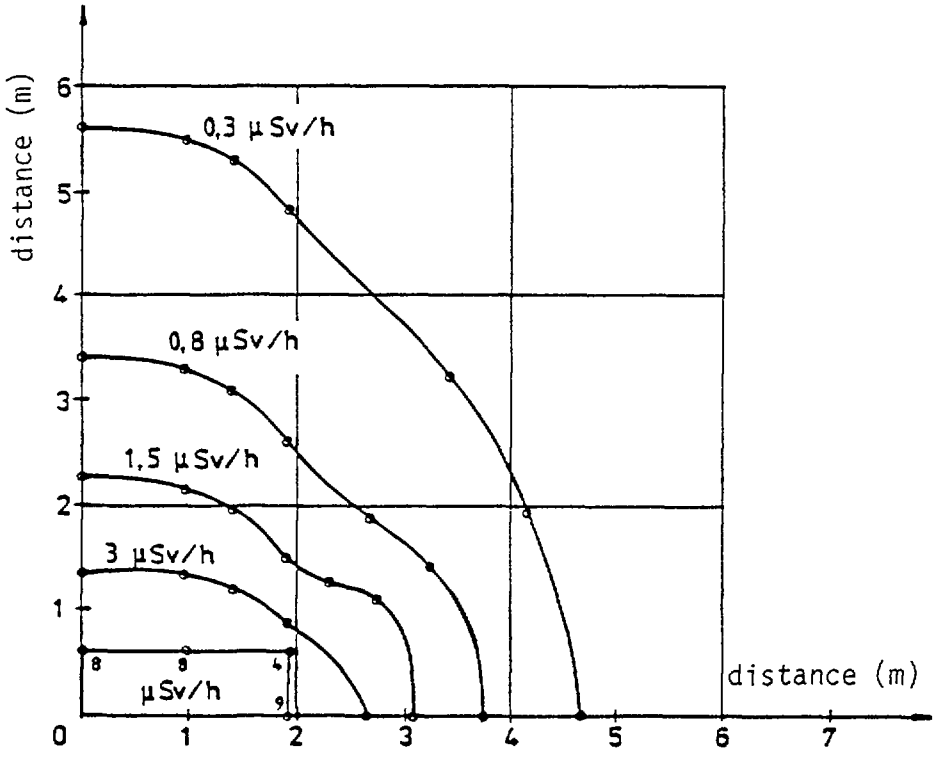

FIG. 10. Vector 1, container filled, time: 1 year.

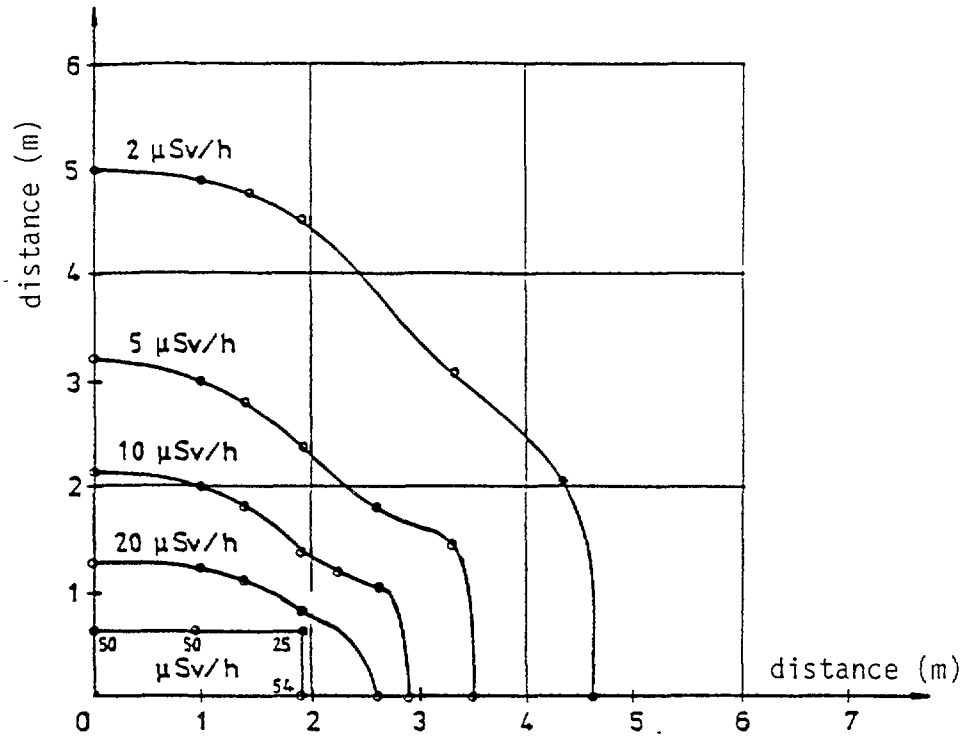

FIG. 11. Vector 2, container filled, time: 10.23 years.

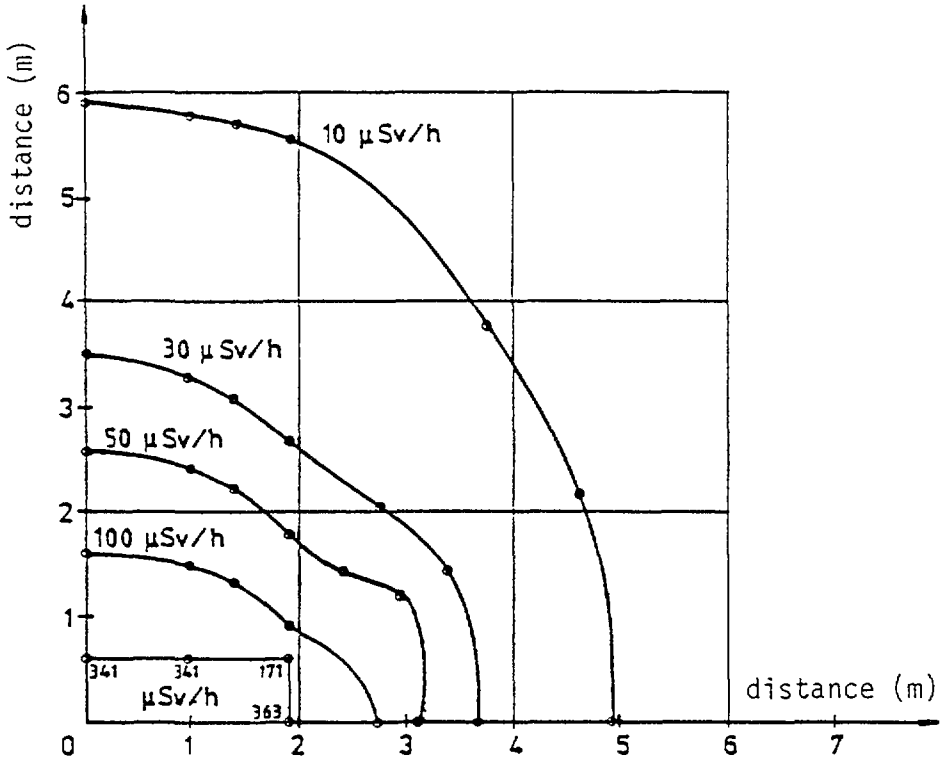

FIG. 12. Vector 3, container filled, time: 10.23 years.

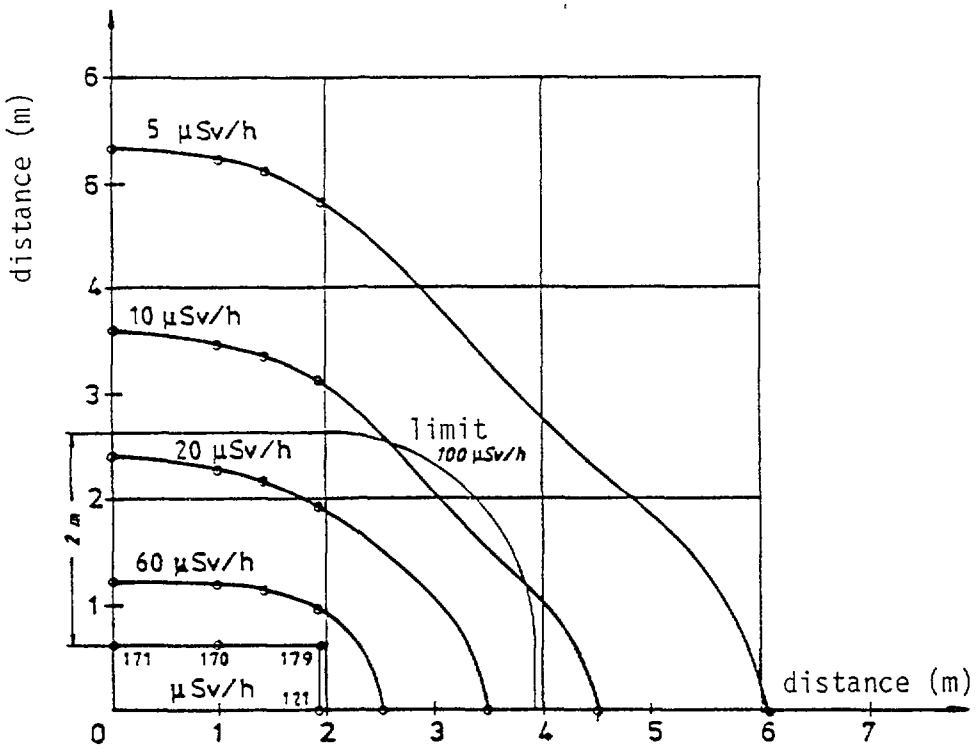

FIG. 13. Vector 1, container drained, time: 1 year.

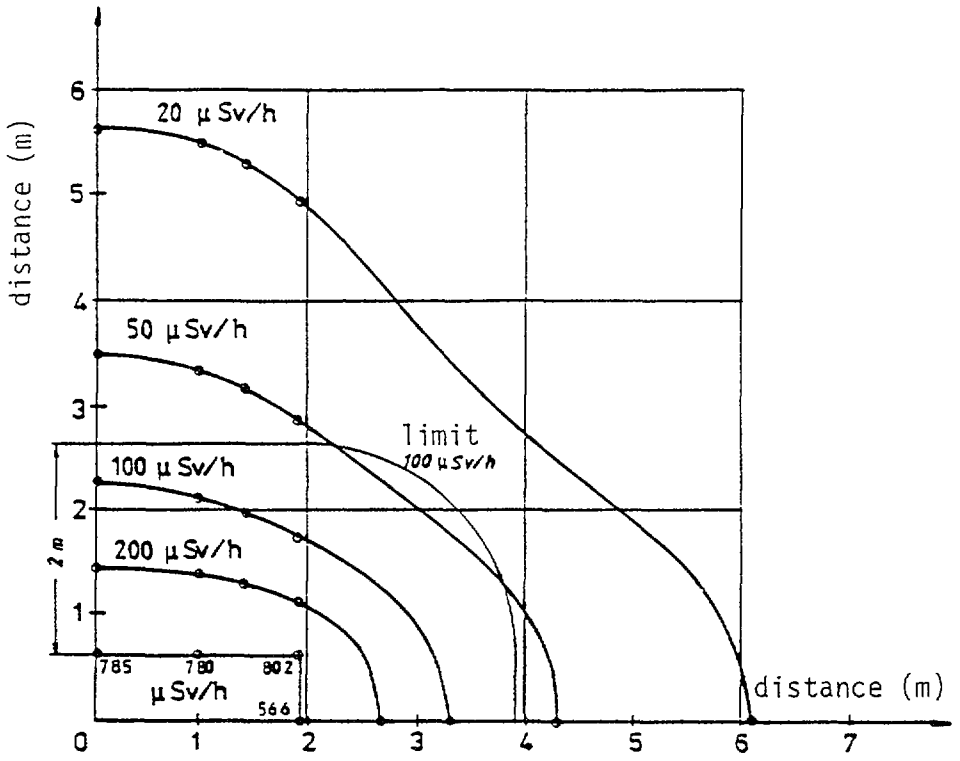

FIG. 14. Vector 2, container drained, time: 10.23 years.

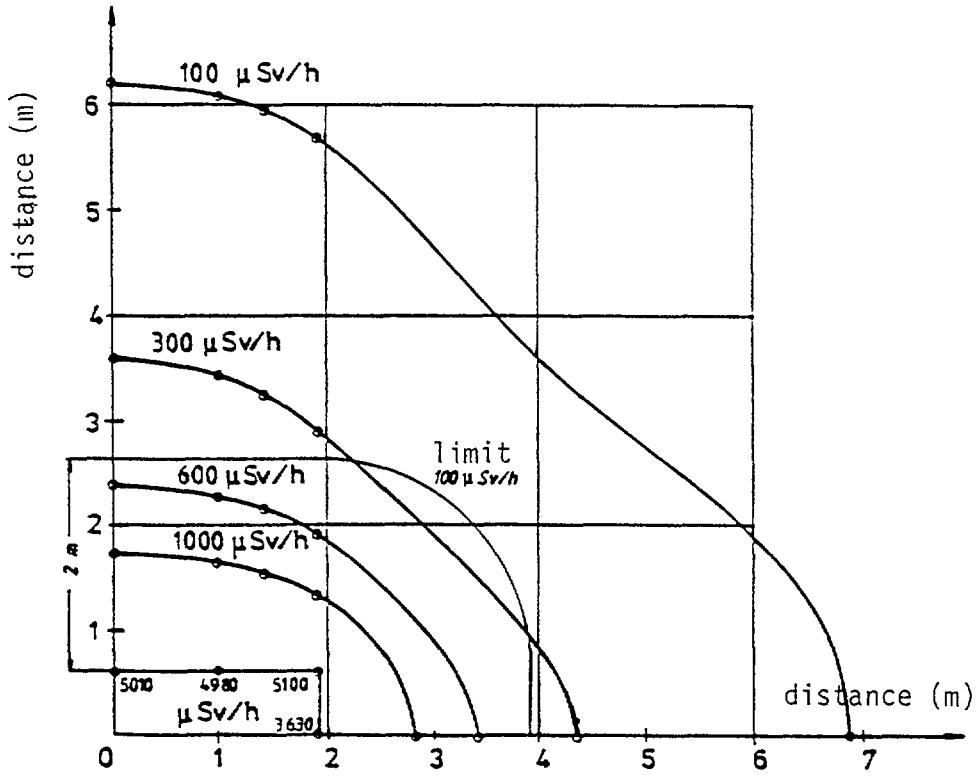

FIG. 15. Vector 3, container drained, time: 10.23 years.

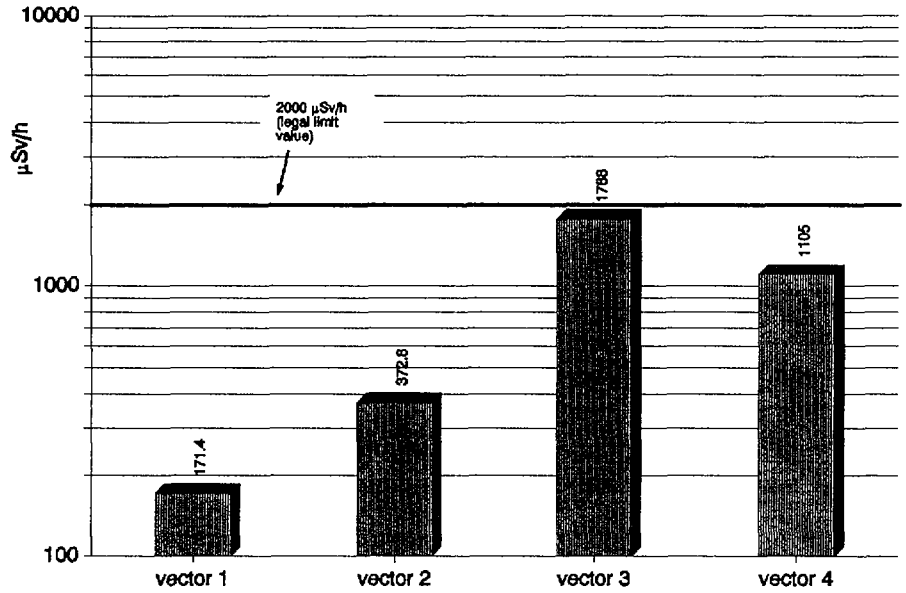

FIG. 16. Comparison of dose rates of the drained container after 1 year.

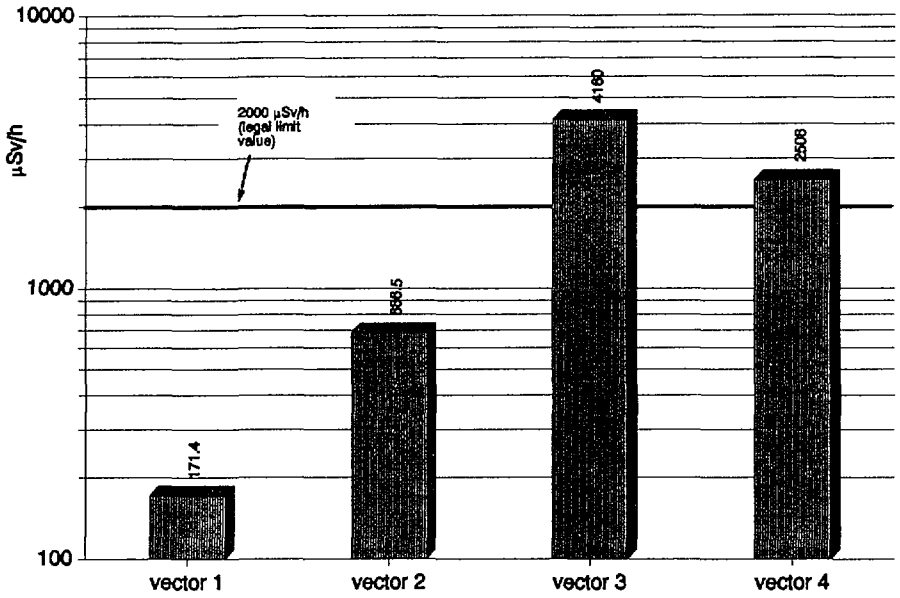

FIG. 17. Comparison of dose rates of the drained container after 4 years.

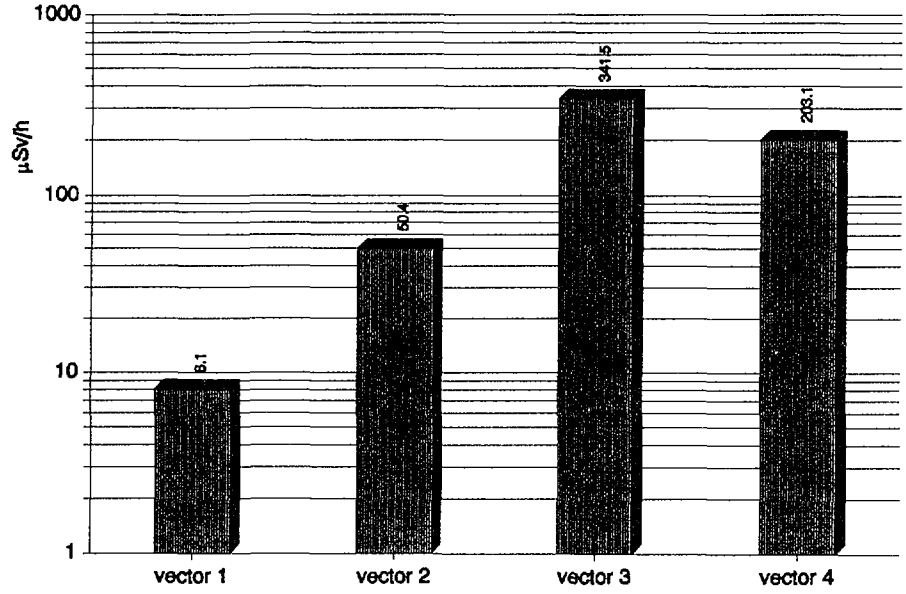

FIG. 18. Comparison of surface dose rates of the filled container after 10.23 years.

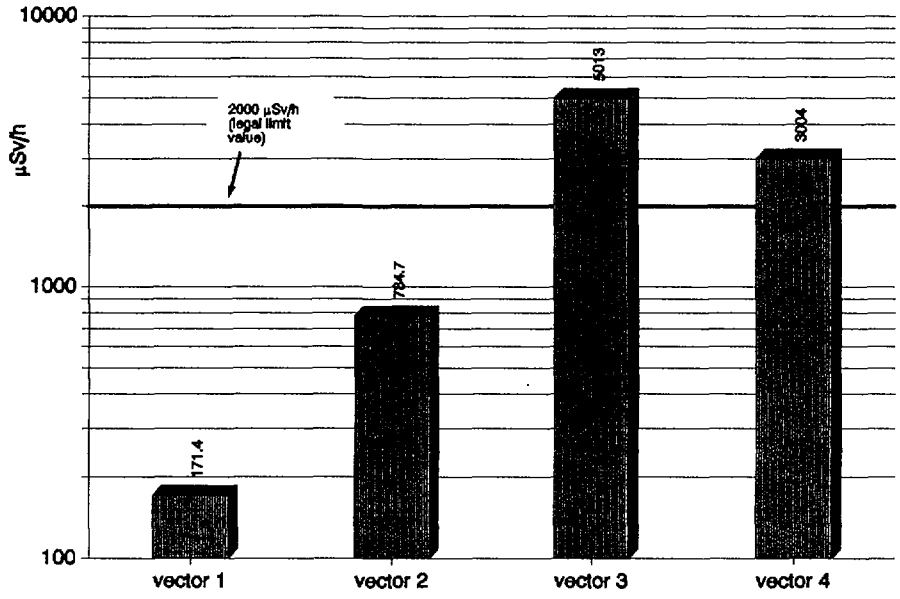

FIG. 19. Comparison of surface dose rates of the drained container after 10.23 years.

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