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Information Paper

Calculating Activity for Natural Uranium

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NB

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The information presented in the information paper is valid as per November 2019.

01

Purpose

This Information Paper has been prepared by the World Nuclear Transport Institute (WNTI) industry members to assist and guide Consignors in order to ensure compliance with International Regulations when calculating the activity for transport of uranium products such as natural uranium, uranium ore concentrate, natural uranium hexafluoride, natural uranium hexafluoride heels and natural uranium dioxide.

The Guidance given here is based upon the requirements of the International Atomic Energy Agency (IAEA) in their publication (SSR-6, Rev.1) titled: *Regulations for the Safe Transport of Radioactive Material*. This publication is the basis of many international and national regulations for the safe transport of radioactive materials.

Compliance with SSR-6 does not necessarily imply that State regulations will be complied with. It is the Consignor's duty to ensure that the method used to calculate the activity of the radioactive material transported is compliant with the State regulations of each country that the material is being transported through or into.

This Information Paper describes the industry agreed method to calculate the activity of various natural uranium products for transport. The Information Paper does not relieve the reader from their responsibility to ensure compliance with specific State based regulations applicable to the countries through which the natural uranium products are being shipped. This Information Paper does not address the calculations of activity for shipments involved in the back end of the fuel cycle.

02

IAEA Regulations Requirements

The IAEA includes a requirement within their regulations for transport to list the activity of the material transported on the shipping document and the category labels. The IAEA Regulations for the Safe Transport of Radioactive Material, 2018 Edition, No. SSR-6 (Rev.1) [1] indicates the following:

Labelling (Category I-WHITE, II-YELLOW or III-YELLOW)

- § 540 (a): contents: the names of nuclides as taken from Table 2, using the symbols prescribed therein. For mixtures of radionuclides, the most restrictive nuclides must be listed to the extent the space on the line permits.

- § 540 (b): activity: the maximum activity of the radioactive contents during the transport expressed in units of becquerels (Bq) with the appropriate SI prefix symbol.

Particulars of consignment (transport document, DGD)

- § 546 (e): the name or symbol of each radionuclide or, for mixtures of radionuclides, an appropriate general description or a list of the most restrictive nuclides.

- § 546 (g): the maximum activity of the radioactive contents during the transport expressed in units of becquerels (Bq) with the appropriate SI prefix symbol.

  Furthermore the IAEA Advisory Material SSG-26 [2] states:

Labelling (Category I-WHITE, II-YELLOW or III-YELLOW)

- § 540.1. In addition to identifying the radioactive properties of the contents, the labels also carry more specific information regarding the contents (i.e. the name of the nuclide, or the most restrictive nuclides in the case of a mixture of radionuclides, and the activity). [...].

  This information is important in the event of an incident or accident where contents information may be needed to evaluate the hazard. The more specific information regarding the contents is not required for LSA-I material, because of the low radiation hazard associated with such material.

- § 540.3. In the identification of the most restrictive radionuclides for the purpose of identifying a mixture of radionuclides as the contents on a label, consideration should be given not only to the lowest A1 or A2 values, but also to the relative quantities of radionuclides involved. For example, a way to identify the most restrictive radionuclide is by determining, for the various radionuclides, the value of:

  $$fi/Ai$$

  where fi is the activity of radionuclide i, and Ai = A_1 or A_2 for radionuclide i as applicable. The highest value represents the most restrictive radionuclide.

Particulars of consignment (transport document, DGD)

- § 546.6. The maximum activity of the contents during transport is required to be specified in the transport documents (para. 546(g)). In some cases, the activity may increase as a result of the buildup of daughter nuclides during transport. In such cases, a proper correction should be applied in order to determine the maximum activity.

- § 546.7. Advice on the identification of the most restrictive nuclides is given in para. 540.3. Appropriate general descriptions may include, when relevant, irradiated (or spent) nuclear fuel or specified types of radioactive waste.

03

Activity Calculations

Natural Uranium Ores (Raw Ores)

Natural uranium ore is in the form of broken rocks that have simply been 'crushed and ground' and as such have not been 'processed.' Natural uranium ore includes the progeny in secular equilibrium. The nuclides which are present in natural uranium ore are the following:

- *U-238 + Th-234 + Pa-234m + U-234 + Th-230 + Ra-226 + Rn-222 + Po-218 + Pb-214 + Bi-214 + Po-214 + Pb-210 + Bi-210 + Po-210*
- *U-235 + Th-231 + Pa-231 + Ac-227 + Th-227 + Ra-223 + Rn-219 + Po-215 + Pb-211 + Bi-211 + Tl-207*

Notes:

- Immediately after the ore is mined radon and its decay products (Rn-222 + Po-218 + Pb-214 + Bi-214 + Po-214 + Pb-210) will be out of equilibrium with the rest of the nuclides due to radon emanation from the ore.

  Except for Pb-210 all these nuclides have half-lives of less than 10 days and are covered by a parent nuclide according to footnote (a) in Table 2 of the IAEA SSR-6 Regulations.

- For completeness all the nuclides in U235 series are listed, although the series represents less than 4.6% of the U238 series activity in a natural form (given 0.711% U235 by mass).

- The nuclides in the U238 series, U-238, Th-234, U-234, Th-230, Ra-226, Pb-210 and Po-210 are listed in the Table 2 of SSR-6 and are not considered covered by a parent nuclide as per footnote (a) under Table 2.

- The nuclides in the U235 series, U-235, Pa-231, Ac-227, Th-227, Ra-223 are listed in the Table 2 of SSR-6 and are not considered covered by a parent nuclide as per footnote (a) under Table 2.

Total Specific Activity of Natural Uranium Ore with Half-lives of More Than 10 Days

Based on the notes above and assuming the nuclides are in equilibrium with U238, the total specific activity for natural uranium ore with half-lives of more than 10 days is 89.5 kBq/g.

99.289% x (U238 series 12.46 kBq/g x 7) + 0.711% x (U235 series 80.14 kBq/g x 5) = 86.60 kBq/g + 2.85 kBq/g = 89.5 kBq/g

Note:

- The specific values used in the calculation above were obtained from the IAEA Advisory Material SSG-26 [2].

- This value is calculated for reference only.

Total Specific Activity of Natural Uranium Ore

Assuming equilibrium, the total specific activity for natural uranium ore considering all nuclides is calculated as 179.5 kBq/g.

99.289% x (U238 series 12.46 kBq/g x 14) + 0.711% x (U235 series 80.14 kBq/g x 11) = 173.2 kBq/g + 6.3 kBq/g = 179.5 kBq/g

Note:

- The specific values used in the calculation above were obtained from the IAEA Advisory Material SSG-26 [2].

The total specific activity of natural uranium ore, 180 kBq/g, is the effective maximum activity during transport.

This value should be multiplied by the fraction of U to account for the mineralisation of the ore. Only the uranium in mineralised ore counts towards the specific activity. The table below present some uranium minerals [3]. The most common mineralisation is in the form of UO_2.

Primary Uranium Minerals

NAME	CHEMICAL FORMULA
Uraninite or pitchblende	UO_2
Coffinite	$UN(SiO_4)_{1-x}(OH)_{4X}$
brannerite	UTi_2O_6
davidlite	$(REE)(Y,U)(Ti, Fe^{3+})_{20}O_{38}$
thucholite	Uranium-bearing pyrobitumen

Secondary Uranium Minerals

NAME	CHEMICAL FORMULA
autunite	$Ca(UO_2)_2(PO_4)_2$ x 8-12 H_2O
carnotite	$K_2(UO_2)_2(PO_4)_2$ x 1–3 H_2O
gummite	gum like amorphous mixture of various uranium minerals
saleeite	$Mg(UO_2)_2(PO_4)_2$ x 10 H_2O
torbernite	$Cu(UO_2)_2(PO_4)_2$ x 12 H_2O
tyuyamunite	$Ca(UO_2)_2(VO_4)_2$ x 5-8 H_2O
uranocircite	$Ba(UO_2)_2(PO_4)_2$ x 8-10 H_2O
uranophane	$Ca(UO_2)_2(HSiO_4)_2$ x 5 H_2O
zeunerite	$Cu(UO_2)_2(AsO_4)_2$ x 8-10 H_2O

The specific activity value for a uranium can be calculated using the formula below:

$$SA = SA_{UNat} * R_U$$

Where

- SA is the specific activity
- R_U is the percent of U in the product

Hence for UO_2 mineralisation:

$$SA = 180.7 \text{ kBq/g} * 0.881 = 159.2 \text{ kBq/g}$$

Note:

- Based on atomic mass of U238 and O16, U / UO2 = 238.03 / (238.03 + 2 x 15.999) = 0.8815 [4]

It is important to note that this value (159.2 kBq/g) is for 100% grade uranium. As such this value is not constant and can't be used for natural uranium ore in general, the grade must be accounted for to make this applicable to a shipment.

Therefore natural uranium ore can be considered as 159.2 kBq/g per % uranium (UO2) grade. For example 2% natural uranium ore would be 3.2 kBq/g.

Regardless of how it's written, the critical point is this value must be calculated for each shipment based on grade. It cannot be used as a constant like UNat (all natural uranium ore concentrate "yellowcake" is the same specific activity).

Exemption Quantities of Natural Uranium ores (raw ores)

It is also important to note that the activity to be considered when looking at the exemption level in table 2 of the IAEA SSR-6 (rev.1) transport regulations, is that of parent only, that is U-238.

Natural Uranium Compounds

Producing natural uranium compounds (typically referred to as Uranium Ore Concentrate) removes the majority of Uranium decay series nuclides during chemical processes such as leaching and solvent extraction. Natural Uranium compounds will typically contain 3 main nuclides (U-238, U-235 and U-234) plus some impurities.

Just after concentration only the Uranium nuclides are present, but after 3 to 4 months ageing, the activities of Th-234 and Pa-234m reach equilibrium with that of U-238. The nuclides which are then present in natural uranium concentrate in secular equilibrium are the following:

- *U-238 + Th-234 + Pa-234m*
- *U-235 + Th-231*
- *U-234*


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Total Total Specific Activity of Natural Uranium Ore Concentrate with Half-lives of More Than 10 Days

Assuming the nuclides are in equilibrium with U238, the total specific activity for natural uranium ore concentrate with half-lives of more than 10 days is 38 kBq/g.

- *99.289% x (U238 series 12.46 kBq/g x 3) + 0.711% x (U235 series 80.14 kBq/g x 1)= 37.11 kBq/g + 0.57 kBq/g = 37.68 kBq/g*

Notes:

- The specific values used in the calculation above were obtained from the IAEA Advisory Material SSG-26 [2].
- This value is calculated for reference only.

Total Specific Activity of Natural Uranium Ore Concentrate in Secular Equilibrium

Assuming equilibrium, the total specific activity for natural uranium ore concentrate considering all nuclides in the two series is calculated as 51 kBq/g.

- *99.289% x (U238 series 12.46 kBq/g x 4) + 0.711% x (U235 series 80.14 kBq/g x 2) = 49.49 kBq/g + 1.14 kBq/g = 50.63 kBq/g*

Notes:

- The specific values used in the calculation above were obtained from the IAEA Advisory Material SSG-26 [2].
- This value is calculated for reference only.

Total Specific Activity of Natural Uranium Concentrate per IAEA Advisory Material [2]

The IAEA Advisory Material SSG-26 provides the specific activity based only on Uranium nuclides (U-238, U-235, U-234) in Table II.3 as 26 kBq/g. This value is also in accordance with the ASTM Specification C787 which indicates the maximum specific activity as 26 kBq/g because the U234 content could be higher than 55 μg/g U (natural) (up to 62 μg/g U).

WNTI members have been following the IAEA Advisory Material SSG-26, using the parent isotope alone when reporting the maximum activity on labels and shipping documents for natural uranium compounds.

To calculate the specific activity value for a uranium product the Unat value should be multiplied by the fraction of U in the uranium compound using the formula below:

$$SA = SA_{UNat} * R_U$$

Where

- SA is the specific activity
- R_U is the percent of U in the product

Below is a table providing examples of various specific activities for different uranium compounds. The specific activity will vary depending on the percentage of uranium in the product (R_U)

Note:
Ratios based on the following atomic mass [4]:

- U = 238.03
- O = 15.999
- H = 1.008
- F = 18.998

URANIUM COMPOUND	R_U	SPECIFIC ACTIVITY BASED ON IAEA ADVISORY MATERIAL 26 KBQ/G
U_3O_8	0.848	22.1 kBq/g
$UO_4x2(H_2O)$	0.7041	18.3 kBq/g
$UO_4x2(H_2O)$	0.6363	16.5 kBq/g
UO_3	0.832	21.6 kBq/g
UO_2	0.881	22.9 kBq/g
Natural UF_6	0.676	17.6 kBq/g
Natural UF_4	0.758	19.7 kBq/g
Natural UO_2F_2	0.773	20.1 kBq/g

Natural heeled UF_6

When calculating the activity for natural Uranium Hexafluoride (UF_6) heels it is assumed that all the Uranium Hexafluoride was drawn out of the cylinder and all that is remaining are the progenies. During the production process of UF_6, all isotopes of uranium are chemically separated from their decay products / progenies and bound with fluorine.

However two of the immediate decay products of U238 isotope (Th234 and Pa234m) will grow-in within a reasonably short time (with a build-up half-life of 24.1 days). Assuming that the time between the production of UF_6 and the time of shipment of UF_6 cylinder is greater than 100 days, then the activity of these short half-life progenies will be reaching equilibrium with their parent, U238. Therefore their activity in the heel can be determined by initially calculating the activity of the parent (U238) in the full cylinder.

The shipping limit for 48Y is 12,500 kg of UF_6, therefore;

- *This represents 12,500 x 0.676 = 8,450 kg of U nat;*
- *This represents 8,450 x 99.28% = 8,390 kg of U238;*
- *Activity of U238 in full 48Y = 8,390 kg x 12.46 MBq/kg = 104.5 GBq;*

Hence, the activity of Th234 and Pa234m are in equilibrium with U238 = 104.5 GBq

- *This represents 104.5 GBq / 857,900 GBq/g = 0.122 milligrams of Th234*

Th234 will decay into Pa234m and U234 on atom per atom basis, growth of radioactive product can be calculated using the following linear differential equation [5]:

B. Growth Of Radioactive Products

General Equation. In chapter 1 we considered briefly a special case in which a radioactive daughter substance was formed in the decay of the parent. Let us take up the general case for the decay of a radioactive species, denoted by subscript 1, to produce another radioactive species, denoted by subscript 2. The behavior of N_1, is just as has been derived; that is, $-(dN_1/dt) = \lambda_1 N_1$ and $N_1 = N_1^0 e^{-\lambda_1 t}$, where we use the symbol N_1^0 to represent the value of N_1, at $t=0$. Now the second species is formed at the rate at which the first decays, $\lambda_1 N_1$ and itself decays at the rate $\lambda_2 N_2$. Thus

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2$$

or

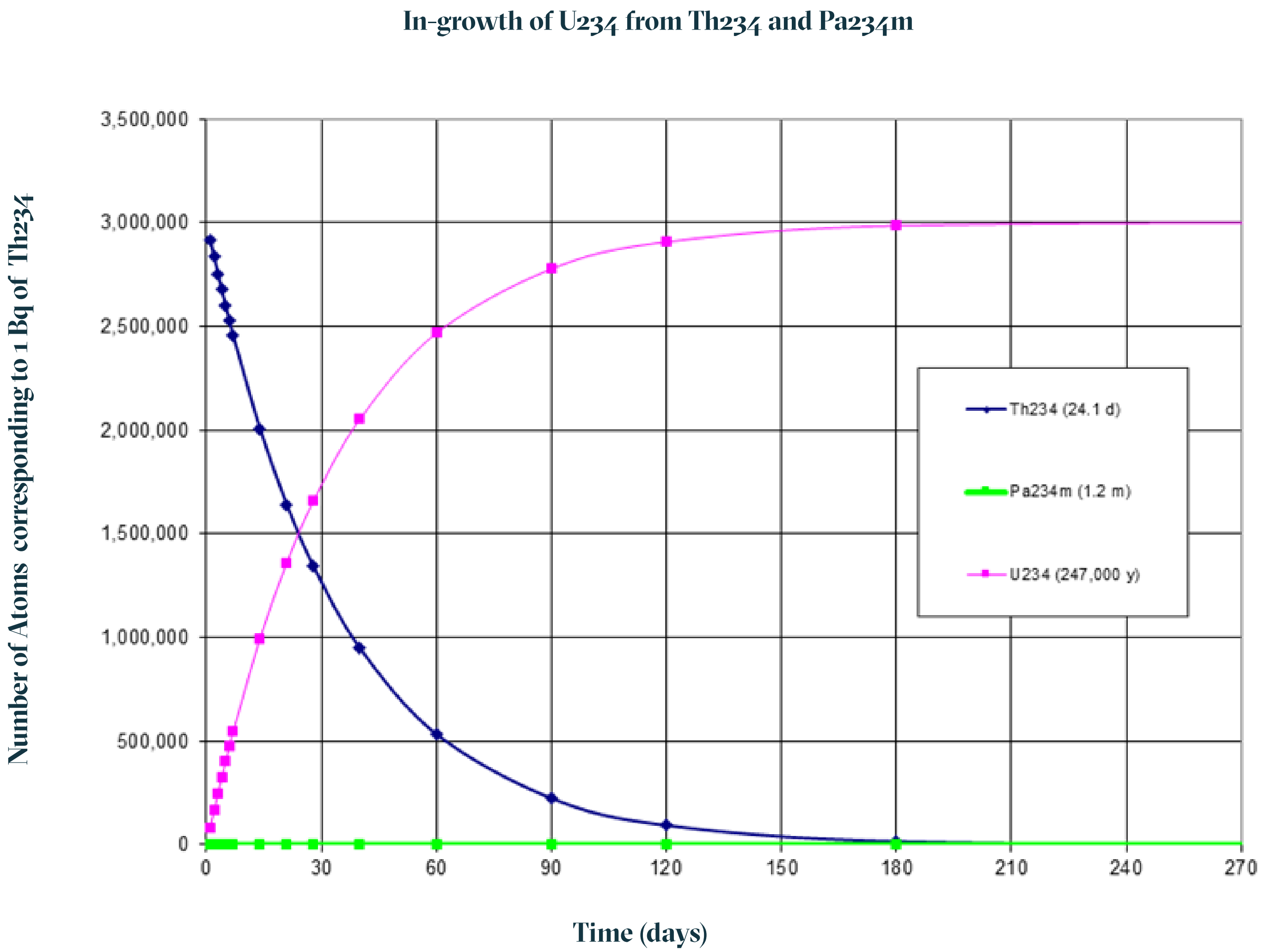
$$\frac{dN_2}{dt} + \lambda_2 N_2 - \lambda_1 N_1^0 e^{-\lambda_1 t} = 0. \qquad \textbf{(5-1)}$$

The solution of this linear differential equation of the first order may be obtained by standard methods and gives

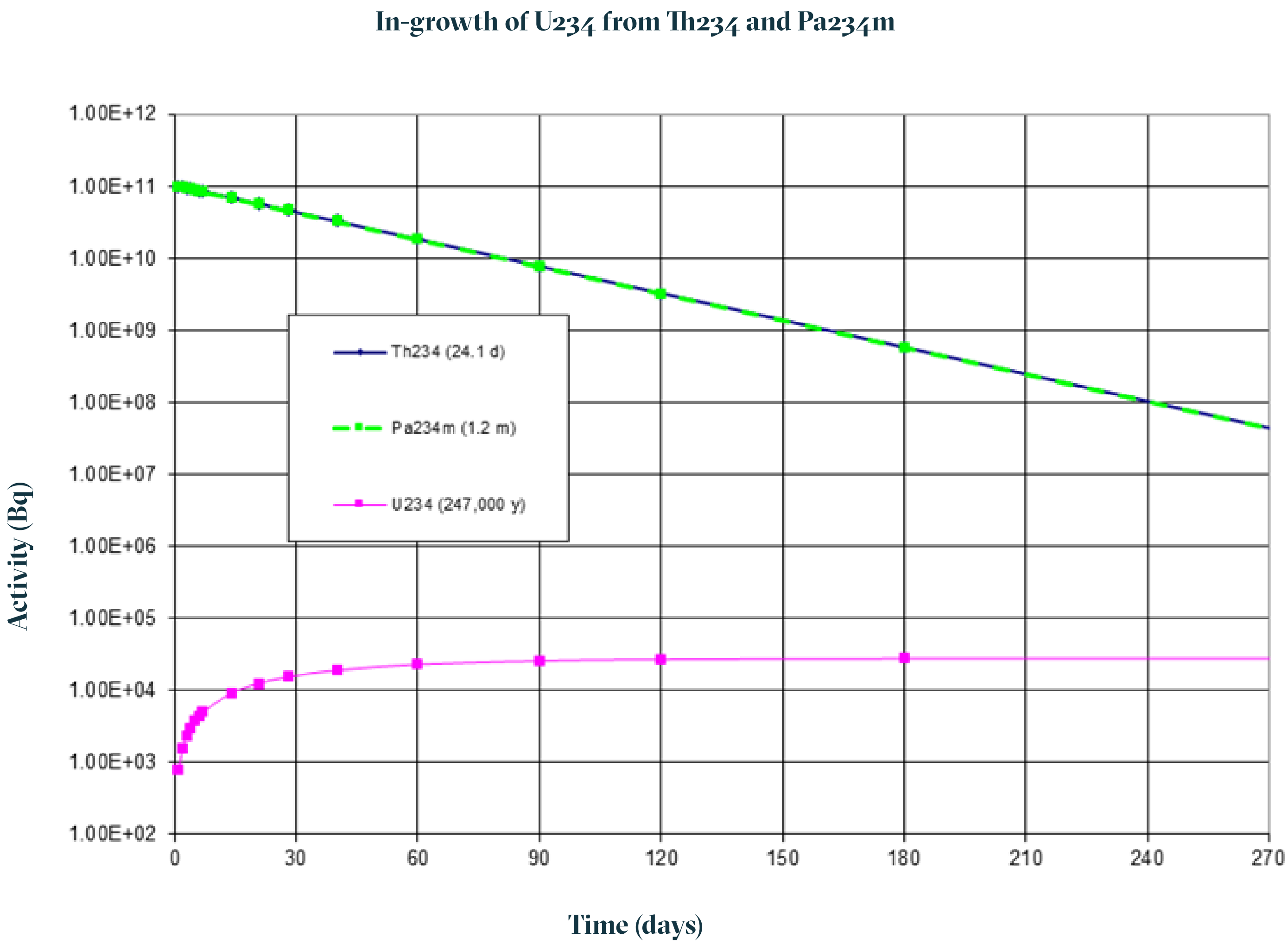
$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 \left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) + N_2^0 e^{-\lambda_2 t}, \qquad \textbf{(5-2)}$$

Using the equation above (on the previous page), growth of U234 from Th234 and Pa234m was calculated.

The figure below shows transformation of atoms from Th234 to Pa234m and U234 over a 9 month period (atoms of Pa234m decay almost instantly into U234 due its short half-life).



The figure below shows the activity growth for U234. Initial activity of Th234 of 104.5 GBq will transform into the maximum activity of U234 of 28 kBq yielding the ratio of 1/3,700,000 (note the logarithmic scale in the chart);



The U-234 activity of a 48Y natual uranium hexafluoride heel can be conservatively estimated as 28 kBq. The U-234 only represents a small fraction (about 0.122 milligrams) of the heels in a cylinder. The remaining heel amount can be assumed to be UF_6. The activity of the heel cylinder can be calculated using the specific activity of 17.6 kBq/g for UF_6. Adding the U-234 activity will have very little impact on the total activity.

04

Conclusion

WNTI members have been following the IAEA Advisory Material SSG-26 and using the parent isotope alone when reporting the maximum activity on labels and shipping documents for natural uranium compounds.

The industry is using the following specific activities to calculate the activity on the shipping documents and the transport labels.

URANIUM COMPOUND	SPECIFIC ACTIVITY (kBq/g)
U_3O_8	22.1
$UO_4x2(H_2O)$	18.3
$UO_4x4(H_2O)$	16.5
UO_3	21.6
UO_2	22.9
Natural UF_6	17.6
Natural heeled UF_6	17.6
Natural UF_4	19.7
Natural UO_2F_2	20.1

05

References

1. IAEA Safety Standards, Regulations for the Safe Transport of Radioactive Material, 2018 Edition, Specific Safety Requirements No. SSR-6 (Rev.1).

2. IAEA Safety Standards, Advisory Material for the IAEA Regulations for the Safe Transport of Radioactive Material (2012 Edition), Specific Safety Guide No. SSG-26.

3. Wikipedia: Uranium Ore; https://en.wikipedia.org/wiki/Uranium_ore

4. Dynamic Periodic Table; https://www.ptable.com/

5. G. Friedlander, J. W. Kennedy, E.S. Macias, J. M. Miller: Nuclear and Radiochemistry, 3rd Edition, John Wiley & Sons, 1981, Chapter 5: Equations of Radioactive Decay and Growth.

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