

Using Nuclear Materials To Prevent Nuclear Proliferation

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Efforts to halt proliferation of nuclear weapons are threatened by vulnerability of weapons-usable material to smuggling especially in Russia. Mixing ^{232}U into highly-enriched uranium (HEU) makes it readily observable and harder to steal. Adding a proportion of ^{233}U associated with a specific storage site enables attribution to be performed on stolen HEU that has been recovered. Incorporating ^{244}Pu into plutonium does the same for this material. U.S. programs for radioactive surplus disposition could provide a source for tags. Current U.S.-Russian efforts to dispose of surplus nuclear weapons open opportunities to incorporate tags into large amounts of weapons-usable material. The ^{232}U detection tag would also enhance ongoing US-Russian efforts to detect smuggling of weapons-usable materials out of Russia.

While the end of the Cold War greatly reduced the threat of an all-out nuclear war between the United States and the Russian Federation, it created a new set of challenging national security concerns. The political and economic volatility in Russia has resulted in growing fears about the lack of fissile material security there and raised the specter of this material falling into the hands of 'rogue' nations, terrorists, and other opportunists.¹⁻³ In this report we describe a way of significantly reducing this threat by making it easier to detect smuggled fissile material and by creating a nuclear fingerprint to aid law enforcement in identifying the source of a 'leak.' We also describe a unique confluence of opportunities that make this nuclear tagging feasible at this time.

We propose uniformly mixing specific substances into fissile materials to act as intrinsic tags. The tags work in two ways. First, they brighten the radioactive signature of highly-enriched uranium (HEU) making it easier to detect. HEU emits far less radiation than plutonium and is more difficult to detect.

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The HEU tag emits a high-energy, penetrating gamma ray that would set off passive, non-intrusive monitors located at nuclear facilities, border stations, and other choke points. There is no need to intensify the plutonium signature because it is already bright and it can be detected readily by the type of monitors mentioned above. Second, the tags provide the opportunity to perform attribution on stolen plutonium or HEU. A storage-site-specific amount of the tag is added to the fissile material during chemical processing which associates the tagged material with the site; different proportions label different sites. If material is stolen and later recovered, analysis of it with readily available techniques would identify the source (the nuclear fingerprint) and enable law enforcement to cut off the leak at its source.

We propose adding a small amount (about 1 ppb) of ^{232}U , an isotope of uranium, to highly-enriched uranium (HEU) to make the material easier to detect. A sample of ^{232}U produces a high-energy (2.6 MeV), penetrating, gamma ray that is actually emitted by one of the daughter nuclei (^{208}Tl with a half-life of 3 million years) produced in the series of radioactive decays that occur before a stable, final nucleus is reached (^{208}Pb here). This gamma-ray signature can be detected by radiation monitors at Russian nuclear sites and exit points (border crossings, airports, *etc.*). The proportion of ^{232}U (1 ppb) is enough to be observed by the type of radiation detectors being installed in pedestrian portal monitors in Russia by the Department of Energy's (DOE) Second Line of Defense (SLD) program. These monitors respond to an increase of about 180 counts/s above background for the most conservative background assumptions.⁴ Once this signal is observed, an alarm goes off and the material recovered. Most HEU has some ^{232}U , but not enough to be routinely observed in these detectors (see Reference 5 and the Appendix). The ^{232}U detection tag is radioactive, but the material would still be safe to handle. For 1 ppb ^{232}U in an 8-kg sphere of ^{235}U the peak radiation is about 0.5 mrem/hr at 1 ft; below the upper limit of 200 mrem/hr for human handling. The 8-kg size is used because it has been proposed for HEU storage at the Fissile Material Storage Facility in Mayak, Siberia that is part of the US Department of Defense's Cooperative Threat Reduction program. This proportion (1 ppb) of ^{232}U is also within the American Society for Testing and Materials limits for commercial uranium (0.002 $\mu\text{g/g}$ or 2 ppb). See the Appendix and Reference 6 for more details.

To accompany the ^{232}U detection tag we propose adding about 20-100 ppm of ^{233}U to HEU as an attribution tag (a nuclear fingerprint). The presence of this material will enable investigators to determine the source of a 'leak' after the stolen material is recovered. A fixed proportion of ^{233}U will be added to the HEU and uniformly mixed throughout the volume. The exact proportion is

associated with specific storage sites or locations and would stay constant for a long time since the half-life of ^{233}U is long (159,000 years). The proportion of ^{233}U in recovered HEU can be measured with standard mass spectrometry to reveal where the material was stored. The proportion of the ^{233}U attribution is far above the typical, 20-ppb sensitivities of commercially-available, magnetic spectrometers. We note here that these amounts are above typical levels of ^{233}U in HEU.^{5,7}

Finally, we propose adding ^{244}Pu to plutonium at the 200 ppb level as an attribution tag. The proportion of ^{244}Pu is a factor of ten greater than the 20-ppb sensitivity of mass spectrometers so it is readily observable. This isotope of plutonium is rare; it is produced by five successive neutron captures on plutonium isotopes starting with ^{239}Pu . The U.S. has a stockpile of this material (about 20 grams) that was produced at the Savannah River Site after about a decade of irradiation. This isotope is difficult (and expensive) to produce so there is almost no background ^{244}Pu in weapons-usable plutonium to confuse investigators. If an adversary could produce or had a supply of this isotope, they would have little need to steal weapons-usable material. The ^{244}Pu nucleus plays the same role as ^{233}U does for the uranium case; the proportion of ^{244}Pu will tag a particular storage location. As mentioned above, the radioactive signature of plutonium is already bright so a detection tag is not necessary for plutonium.

These tags are effective only if they cannot be easily defeated even by a technically skilled person with access to the weapons-usable material. This 'insider' is considered the most likely candidate to steal weapons-usable material from a storage site.⁸ Here we consider several possible methods a would-be smuggler might try. In each case, the presence of the tags makes theft considerably more difficult. A smuggler could shield the HEU and block the gamma rays emitted by the detection tag to reach any radiation detectors at a monitoring site. This requires a lead shield with a thickness of about two inches. For the typical size of a piece of HEU (8 kg is the preferred size for storage), a box to shield the detection tag would weigh about 40 kg, would be cumbersome to move, and could show up quite easily on an x-ray monitor like the ones already in use at Russian border crossings. A smuggler can only conceal the radiation tag at the cost of making the HEU (in its container) easier to detect with x-rays.

Another strategy to defeat the detection tag would be to cut the HEU into pieces that have a smaller radioactive signature. The detection tags make this path more difficult because the smuggler has to now secretly machine the HEU (cut it up), shield it (albeit with less lead), and repeat this process several times to get the same amount of material through any portal monitors.

A technically skilled smuggler could try to remove the ^{232}U detection tag from the HEU. We have intentionally chosen the tagging materials to be the same element as the tagged material (HEU or plutonium). Different isotopes of the same element are nearly identical under most chemical processes. Hence, it requires costly and expensive equipment (i.e. gas centrifuges) to separate isotopes of the same element. These techniques are beyond the capabilities of most smugglers. This feature is a general property of all the tags we propose; none of them can be easily removed with methods available to smugglers.

A smuggler could try to remove the daughter nucleus of ^{232}U that emits the high-energy gamma ray. This gamma ray is actually emitted by the decay of one of the members of the ^{232}U decay chain, ^{208}Tl , not by the ^{232}U itself. The ^{208}Tl nucleus is not the same element as uranium so it is possible for a technically-skilled smuggler to chemically remove the ^{208}Tl and defeat the tag. However, the smuggler must move quickly after removing the ^{208}Tl to get the material through any portal monitors to a buyer. This speed is necessary because the continuing decay of the ^{232}U (which is still in the material) will produce more of the gamma-ray-emitting ^{208}Tl and will make the radioactive signature bright again. We have chosen the amount of added ^{232}U so the detection tag is observable again in about a month.

Finally, consider an attempt to mask the ^{233}U attribution tag for HEU or the ^{244}Pu attribution tag for plutonium by adding more of the tagging material. Each attribution tag is a unique material and only small quantities exist (neither element is found in nature). This means it is difficult to 'cover up' the tag because a smuggler simply can't get the ^{233}U or the ^{244}Pu to add to the stolen nuclear material. As a bonus, the uniqueness of the attribution tag material means there will be little natural background contamination to confuse investigators.

None of the tags significantly add to the radiation hazard of HEU or plutonium or disrupt possible future fuel use. The ^{232}U detection tag for HEU is added in small amounts - half the maximum of 2 ppb permitted by industry specifications. Similarly, the amount of the ^{233}U attribution tag is kept small to avoid radiation hazard from alpha emission. The ^{244}Pu attribution tag is stable and does not emit much radiation. All of the tags are added in very small quantities so they won't damage the quality of the materials if they are later incorporated into reactor fuel (a potential future use of the material).

A U.S.-Russian program to enhance weapons-usable material security with such tags could take advantage of a timely confluence of opportunities which would enable the tagging of a significant amount of material. There are three essential steps in building a program: (1) making the tags, (2) incorporating them into the weapons-usable material, and (3) building the network of

monitors to detect smuggled, weapons-usable materials. In each case there are existing or planned programs in the U.S. and Russia that could be easily extended to include tagging at a small, marginal cost.

The first step is to make the tags by processing existing material into a form suitable for blending. Adequate amounts of the tagging materials (^{232}U , ^{233}U , and ^{244}Pu) are already in U.S. stockpiles and await long-term disposition.⁹ Batches of uranium isotopes at three U.S. national laboratories contain mixtures of ^{232}U and ^{233}U that could be used to tag HEU.⁶ An adequate supply of ^{244}Pu is at United States' Savannah River Site.¹⁰ The amounts of the material in the U.S. stockpiles and the requirements for tagging are shown in Table 1 below. The first column lists the tagging isotopes and the second column lists the U.S. supply. The right-most column contains the amount of material needed to tag the Russian surplus that is the product of the tag proportion (column 3) and the amount of Russian weapons-usable material surplus (column 4).

Table 1: Supply and Demand for Nuclear Tags

Nuclide	Tag Supply (kg)	Tag Proportion	Amount to be Tagged (metric tons)	Tag Demand (kg)
^{232}U	0.3 ^a	1 ppb	500 (HEU)	0.0005
^{233}U	351 ^a	20-100 ppm	500 (HEU)	10-50
^{244}Pu	0.02 ^b	200 ppb	34 (pu)	0.007
a - Oak Ridge National Laboratory, see Reference 6. b - Savannah River Site, see Reference 10.				

The existing material containing the tag would be downblended with some other material to produce small pellets about three millimeters across. For example, the HEU attribution and detection tags could be downblended with LEU. These pellets would then be added to the weapons-usable material during processing and mixed to uniformly distribute the tag. See discussion below on taking advantage of existing programs for this processing step.

The cost of processing to make the tags is a fraction of the funds necessary for final disposition or storage of the surplus. A \$100 million dollar program in the Materials Disposition directorate of the U.S. Department of Energy (DOE) has begun processing and packaging the uranium. The long-term storage or disposition will cost even more. We estimate roughly that an additional expenditure of \$20-25 million would cover production of the detection and attribution tags for HEU.¹⁰

In the plutonium case, DOE is considering plans for the long-term disposition of the ^{244}Pu now stored at the Savannah River Site. We estimate roughly that it would cost an additional \$15-20 million to transform the ^{244}Pu into an attribution tag.¹⁰ In both the HEU and plutonium cases, a significant fraction of these funds would be spent in any circumstance to process these materials for long-term disposition or storage. It is also worth noting here that it is possible that Russia also has stockpiles of these materials. Using Russian stockpiles or facilities could be a useful incentive for persuading them to support this tagging initiative.

The second step in developing a tagging program for weapons-usable material is to incorporate the tags into the HEU and plutonium. Existing U.S.-Russian programs make this step feasible for a portion of the surplus Russian material.¹¹ The U.S. is committed to spending \$20 billion by the year 2013 to buy 500 metric tons of Russian HEU as part of the HEU Purchase Agreement.¹² This represents about 40% of the Russian stockpile.² HEU is first transformed into an oxide and then processed further to make low-enriched uranium for reactor fuel.¹² Ideally, the uranium oxide would be quickly processed into reactor fuel, but past experience reveals the HEU in oxide form is sometimes stored for a period from a few weeks to several months thereby increasing its vulnerability. To tag the HEU during the oxide-processing step one would uniformly mix small pellets of material containing the detection and attribution tags. Since the tags are the same chemical element as the HEU this step adds little to the cost and complexity of the HEU processing. In another case, the U.S. and Russia have a cooperative program costing about \$1.3 billion for the dismantling of Russian nuclear weapons and storing them in the Fissile Material Storage Facility (FMSF) at Mayak in Siberia in unclassified shapes.¹¹ The FMSF is expected to be certified for use by August 2002.

The method for dismantling the weapons is under negotiation with two techniques under consideration: melting and recasting the plutonium or HEU into an unclassified shape or turning the weapons-usable material from the weapons into an oxide. In either case one would add a step during processing to uniformly mix the tags into the uranium or plutonium. This additional step would add little to the complexity and cost of the proposed processes.

This tagging proposal would also enhance other U.S.-Russian nuclear security activities. These are programs totaling over \$265 million for FY2000 with the explicit aim of enhancing physical security of weapons-usable material in Russia (*e.g.*, DOE's Second Line of Defense program).¹¹ As part of these efforts, radiation detectors are being installed at Russian storage sites, border crossing, and exit points (*i.e.*, airports). These programs have had some success at detecting smuggled radioactive materials, but still face the difficult

problem of detecting HEU because of its dim radioactive signature.¹³ The tagging method we've described here attacks the problem from a different direction and reduces the problem by making the tagged HEU visible to existing monitoring systems. It adds a capability that would be very costly otherwise.

It is worth considering other applications of the tags. Highly-enriched uranium that is not part of the HEU Purchase Agreement should be tagged. The 500 tons of HEU that will be purchased by the U.S. under the HEU Purchase Agreement is less than half of the material in the Russian stockpile. The future disposition of the balance of the material is undecided and some of that additional HEU is vulnerable to theft.^{2,3} This material could be tagged during accelerated processing into oxide form and placed in secure storage. As time, funding, and facilities permit, it can then be downblended to low-enriched uranium that is not a proliferation risk and eventually burned as fuel. Similarly, surplus plutonium that has not been formally declared as excess should also be tagged. The 34 metric tons formally declared as excess for storage in the FMSF is only about one-quarter of the Russian stockpile.¹⁴ The remaining material should be processed in the same way as the current, declared excess, tagged, and placed in long-term storage in an expanded facility at Mayak.

Another extension of the tagging concept is to apply it to the civilian plutonium industry. Reprocessing spent reactor fuel to extract plutonium poses a threat to U.S. and international security because even 'reactor-grade' plutonium can be used to make a nuclear weapon. Britain, France and Russia all spend billions of dollars each year to process tons of plutonium. The worldwide inventory of separated, civilian plutonium is about 170 tons; rivaling military plutonium in size.^{2,15} Adding the ^{244}Pu attribution tag to civilian plutonium as the spent fuel is processed achieves the same goal of enabling investigators to identify the source of a plutonium leak after stolen material is recovered. An important caveat with this idea is that tagging the 34 tons of surplus, Russian, military plutonium uses much of the U.S. stockpile of the ^{244}Pu attribution tag. To deal with the shortfall Russian sources could be used if they have a stockpile of ^{244}Pu or by employing an existing high-neutron-flux reactor to produce the additional quantities of the ^{244}Pu attribution tag in a timely manner.

To conclude, we propose the introduction of intrinsic tags into weapons-usable material. Adding ^{232}U as a detection tag in HEU makes that material more difficult to steal. Adding ^{233}U to HEU and ^{244}Pu to plutonium creates an attribution tag or nuclear fingerprint that can be used to help investigators identify the storage site of the material if it is stolen and then recovered. The tags are robust to countermeasures and safe to handle and for any future uses. The timing of this proposal is auspicious. The U.S. now has adequate

supplies of the tagging materials and is considering long-term disposition of them. The U.S. and Russia have started processing large quantities of HEU and plutonium as part of other non-proliferation programs. These programs have created an opportunity to add the tags to weapons-usable materials at a small, marginal cost. Finally, the U.S. is supporting the construction of a radiation monitoring system in Russia to prevent smuggling of weapons-usable materials. This proposal enhances that existing program.

Much work remains to be done. We suggest a cooperative, pilot study to test the ideas discussed here using, possibly, the DOE Lab-To-Lab program to answer any technical and safety questions and enlist early, Russian involvement. Once the method is validated, the U.S. must consider how to implement the program. The U.S. and Russia should determine if such a program is best done as part of a bilateral program or if other organizations (notable the IAEA) should contribute.

Appendix: Count Rate for ^{232}U Detection Tag for HEU.

To calculate the amount of material needed to brighten the radioactive signature of HEU there are two limits to consider:

- (1) At the low radiation end, there is a minimum proportion of the ^{232}U detection tag to be added so the gamma radiation can be detected above background by a pedestrian portal monitor.
- (2) At the high radiation end we have to avoid turning the tagged HEU into a radiation hazard.

First, consider the lower limit on the proportion of ^{232}U . We start by finding the count rate needed to set off a pedestrian portal monitor and then determine the amount of ^{232}U to mix into HEU to reach that threshold. Consider the minimum radioactivity of the tagged HEU that will set off the radiation detectors used at Russian nuclear sites and exit points. The DOE's Second Line of Defense (SLD) pedestrian portal monitors require about 180 counts/s to set off an alarm.⁴ Call this minimum rate R_{min} . We need to calculate the rate of gamma rays coming off the material that will generate 180 counts/s in the SLD detectors. Let $e_1 = 0.3$ be the efficiency of the SLD detectors for detecting the gamma rays and $e_2 = 0.10$ be the fraction of the solid angle covered by the SLD detectors.⁴ The minimum emission rate T_{min} of gamma rays from the HEU needed to set off the SLD alarms is the following.

$$T_{min} = \frac{R_{min}}{e_1 e_2} = \frac{180 \text{ cts/s}}{0.3 \times 0.1} = 6300 \text{ cts/s}$$

Next, we must add enough of the ^{232}U detection tag to the HEU so it emits T_{min} gamma rays per second in all directions. The primary source of radiation from ^{232}U is not the uranium nucleus itself, but one of the daughter nuclei ^{208}Tl which emits the deeply-penetrating, 2.6-MeV gamma ray. The half-life of ^{208}Tl is 3 million years. The decay scheme of ^{232}U is shown in Figure 1 below.

We have to determine the proportion of ^{232}U at equilibrium needed to produce this count rate for the gamma ray (from ^{208}Tl) in an 8-kg sphere of HEU (the preferred size for storage). The specific activity of Tl is $S_{\text{Tl}} = 2.97 \times 10^{11}$ gammas/s-g.¹⁶ Most of the gammas emitted by ^{208}Tl would actually get blocked by the HEU metal composing the sample (self-attenuation). A fraction of about $e_3 = 0.15$ actually escapes the HEU sphere. Thus, the mass of the ^{232}U isotope m_{232} in an 8-kg sphere of HEU that will produce the minimum necessary count rate in the SLD monitors is

$$m_{232} = \frac{T_{min}}{S_{\text{Tl}}e_3} = \frac{6300 \text{ cts/s}}{(2.97 \times 10^{11} \text{ gammas/s-g}) \times 0.15} = 1.3 \times 10^{-7} \text{ g}$$

so the fraction f_{232} of ^{232}U in an 8-kg sphere of HEU is the following.

$$f_{232} = \frac{m_{232}}{m_{\text{HEU}}} = \frac{1.3 \times 10^{-7} \text{ g}}{8000 \text{ g}} = 1.5 \times 10^{-11}$$

This fraction f_{232} is the minimum proportion of ^{232}U necessary to set off the SLD alarms at a pedestrian portal monitor.

The decay of nuclei in a material is a dynamic process. The proportions of different species change with time as some isotopes are created by decays higher up the chain and others disappear as they themselves decay. We now consider the time dependence of a sample of ^{233}U and ^{232}U . Figure 2 is from reference 6 and shows the gamma ray exposure rate from a $^{233}\text{U}/^{232}\text{U}$ mixture as a function of time. In that figure the dose rate reaches a peak of 11 Rad/Hr at a distance of one foot for a 1-kg sample after about 10 years. We require the ingrowth of ^{208}Tl to be detectable in the SLD pedestrian portal monitors after about a month (0.1 years on the plot). The dose at 0.1 year is about 1/40 the maximum rate which the sample reaches after 10 years. In determining f_{232} above, we used the maximum specific activity. In order for the SLD detectors to see the tagged HEU after 0.1 years the HEU must have 40 times the proportion of the ^{232}U detection tag f_{232} calculated above which was after 10 years of ingrowth of the ^{208}Tl . In other words, we have to increase the fraction of ^{232}U in the 8-kg HEU spheres by a factor of 40 so the new value is $f'_{232} = 40 \times f_{232} = 6 \times 10^{-10}$. This modified fraction f'_{232} is still small and is the

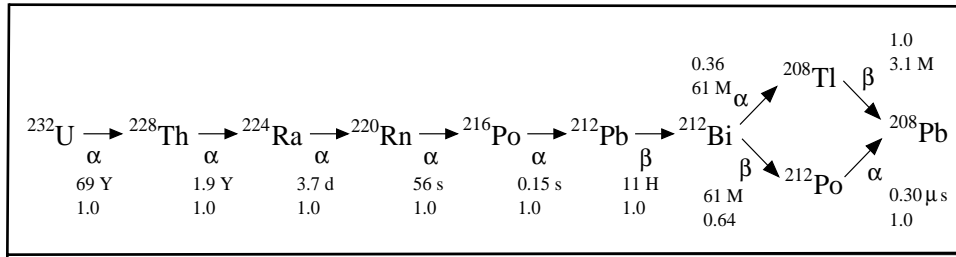


Figure 1: Major decay pathways from ^{232}U . The labels next to each arrow are (1) the type of radioactive decay in that step (α or β), the half-life, and the branching ratio. The β decay of ^{208}Tl to ^{208}Pb is followed by emission of a 2.6-MeV gamma ray from the de-excitation of ^{208}Pb .

basis for the calculation of the tag demand shown in Table 1 where we round off the proportion to 10^{-9} or 1 ppb. Note that by adding the ^{232}U detection tag at the f'_{232} proportion, the gamma-ray signal remains above the detection threshold for almost a million years (see Figure 2).

The next requirement the ^{232}U detection tag must satisfy is that it cannot create a radiation hazard after it is added to the weapons-usable material. Consider the upper limit on the amount of ^{232}U that can be added. This upper limit on the radioactivity of tagged HEU is 200 mrem/hr. Dose rates above this value require robots to handle the material so we must be far below this upper limit.⁴ Above, we calculated the lower limit of the proportion of ^{232}U that would be detectable with the SLD pedestrian portal monitors. We now show this lower limit is also below the upper limit determined by safety considerations (the 200 mrem/hr level). The peak gamma-ray dose rate in reference 6 is 11 Rad/hr at one foot for a 1.0-kg sample of ^{233}U with 100 ppm ^{232}U or a proportion of ^{232}U of $f_{100} = 10^{-4}$. Using ratios we can calculate the dose for the 8-kg HEU sphere R_{tag} with $f'_{232} = 6 \times 10^{-10}$.

$$R_{tag} = 11 \text{ Rad/hr} \times \frac{f'_{232}}{f_{100}} \times m_{HEU} \sim 0.5 \text{ mrem/hr}$$

This last dose rate R_{tag} is a factor of 400 below the radiation hazard limit discussed above. We also note that a 1ppb proportion of ^{232}U is within the commercial specification for ^{232}U (0.002 $\mu\text{g/g}$).

To summarize, we can add enough ^{232}U to HEU so that it is easily detect-

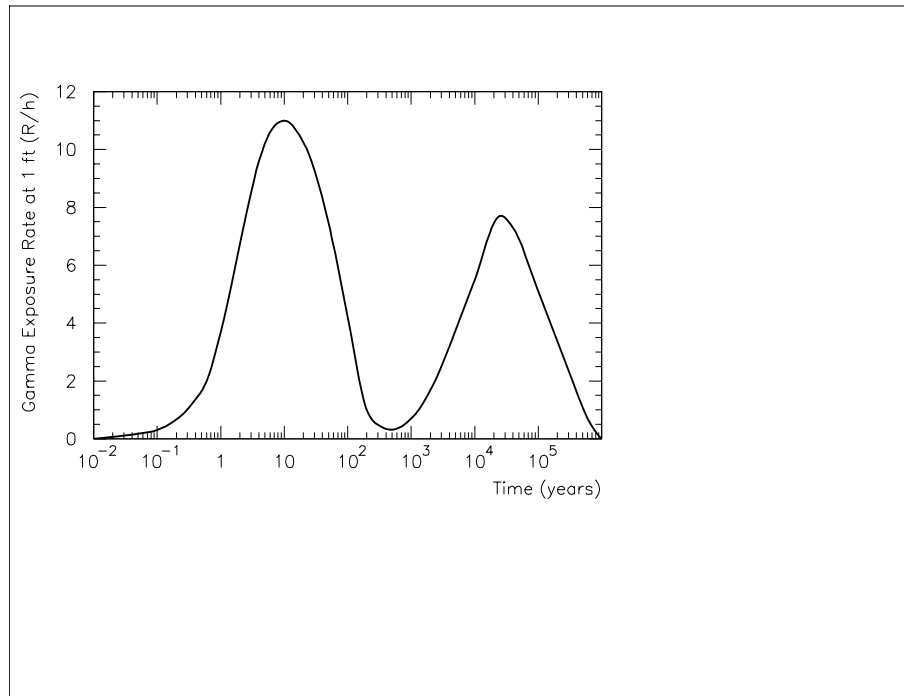


Figure 2: Gamma exposure for 1 kg of ^{233}U with 100 ppm ^{232}U at 1 ft (4). The mixture consists only of uranium at the start time and other elements appear later as the radioactive decays proceed. Both peaks are due mostly to the emission of a 2.6-keV gamma-ray from ^{208}Tl . The time dependence is not a simple one because of the complexities of the decay chain.

able with pedestrian portal monitors. The nucleus that emits the penetrating, 2.6-MeV gamma ray, ^{208}Tl , grows to detectable levels in about a month for a sample starting with only uranium isotopes. The radiation hazard associated with the tagged material is about a factor of 400 below the maximum for human handling and is within commercial specifications.

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