

## Potential for Production of Proliferation Sensitive Materials in Research Reactors

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### ABSTRACT

The issue of misuse of research reactors for the production of proliferation sensitive materials has been a matter of longstanding interest to the IAEA. Most attention has focussed on the potential for unreported production of plutonium via the irradiation of uranium targets. However, the potential for the production of other proliferation sensitive materials such as tritium and polonium-210 is also of concern and is currently being examined by the IAEA. This paper looks at the potential for the undeclared production of tritium and Po-210 in research reactors and implications in terms of the IAEA's safeguards approaches.

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### INTRODUCTION

For more than a decade the IAEA has had measures in place for the detection of unreported production of fissile material at research reactors capable of thermal outputs greater than 25 MWth. The Australian Safeguards Support Program (ASSP) was a key contributor to the research on the physical limits to plutonium production at research reactors. The results of this research led the IAEA to the conclusion that, in practice, it was not feasible to produce *significant quantities* (8 kilograms or more) of plutonium in a year in research reactors smaller than the 25MWth limit<sup>2</sup>.

In the period from the 1970s to the early 1990s the IAEA safeguards system (traditional safeguards) devoted all of its effort to determining the accuracy of declared inventories of nuclear material. With the advent of the strengthened safeguards system the IAEA is seeking to look beyond declared inventories to undeclared materials, facilities and activities. States seeking to develop a clandestine nuclear weapons capability are likely to pursue research along many different pathways to their goal, often running such efforts in parallel to each other. One of the key challenges of the strengthened safeguards system is to identify as many of these pathways as possible and to take steps to identify possible physical indicators of developments of concern.

Attempts to develop the capabilities to produce nuclear weapons involve more than just the development of the fissile material components of the weapons. They will be accompanied by research into the effective operation of a wide range of ancillary measures that will be required to make the weapon militarily useful. Such measures include efforts to ensure reliable initiation of the weapon and measures to enhance its yield in addition to the more conventional research into high explosive detonation and delivery systems.

Two materials of interest to proliferators that could be included in a possible weapons program could potentially be produced by the misuse of declared reactor facilities – these materials are Po-210 and tritium.

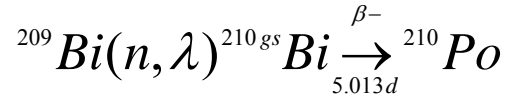
### POLONIUM-210

Historically Po-210 has been used in early generation nuclear weapons as a key element of a very basic initiator design. In the IAEA's ongoing investigation of Iran's non-compliance with its

safeguards obligations, one of the more troubling discoveries was that Iran had investigated the production of Po-210 by the irradiation of Bi-209 targets. Iran's explanations for this research have generally been considered unconvincing.

Po-210 is generated by the thermal neutron irradiation of Bi-209 as follows:

**Equation 1 – Process for the production of  $^{210}\text{Po}$**



In preparing this paper the authors found that using two different version of the ORIGEN isotope generation and depletion code<sup>3</sup> for modelling the rate of production of Po-210 via this reaction produced results that differed from each other by roughly 50%. It was not clear whether the differences in results arose from differences in the underlying calculation modules of the two versions of the program or whether differences in the user interface meant that the input data were being interpreted differently by the programs. The simplest method of resolving this was to examine the problem from first principles.

Po-210 has a 138 day half-life and Bi-210 has a 5 day half life. To a first approximation the production of Po-210 can be modelled as a one step process using the standard formula for the production of a radioisotope by neutron irradiation<sup>4</sup>:

**Equation 2 – Simplified formula for production of  $^{210}\text{Po}$**

$$N(^{210}\text{Po}) = N(^{209}\text{Bi})\sigma_{act}\phi(1 - e^{(-\lambda_p T)})e^{(-\lambda_p t)}$$

Where:

$N(^{210}\text{Po})$  = number of Po-210 atoms

$N(^{209}\text{Bi})$  = number of Bi-209 atoms

$\sigma_{act}$  = neutron activation cross-section (barns)

$\phi$  = the neutron flux (n/cm<sup>2</sup>-s)

$\lambda_p$  = the decay constant for Po-210 (sec<sup>-1</sup>)

T = time for the irradiation (seconds)

t = cooling time until separation (seconds).

A more accurate version of this equation would take into account the two step process with the five day half-life of Bi-210 included and would result in the following two step formula<sup>5</sup>:

**Equation 3 – Two step formula for production of  $^{210}\text{Po}$**

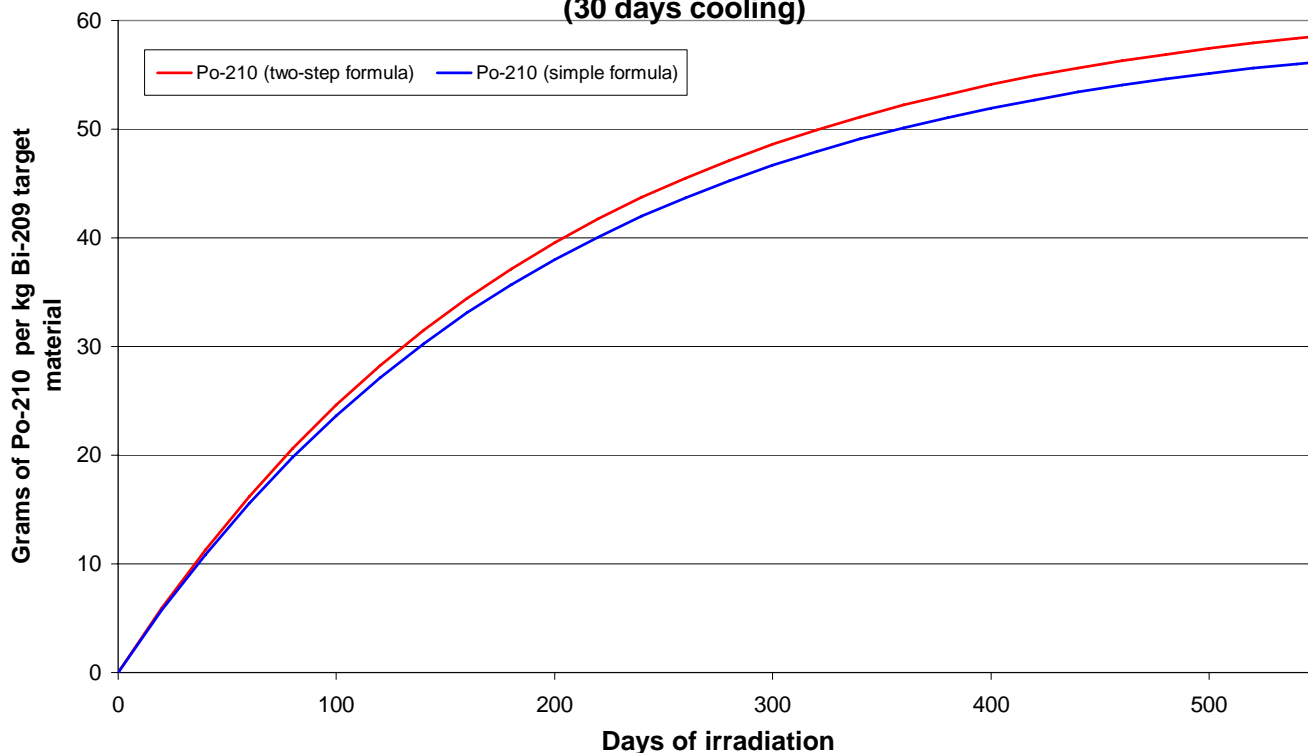
$$N(^{210}\text{Po}) = \frac{N(^{209}\text{Bi})\sigma_{act}\phi}{\lambda_B - \lambda_p} \{ \lambda_B (1 - e^{(-\lambda_p T)}) e^{(-\lambda_p t)} - \lambda_p (1 - e^{(-\lambda_B T)}) e^{(-\lambda_B t)} \}$$

Where the terms in equation 3 have the same meanings as for Equation 2 and:

$\lambda_B$  = the decay constant for Bi-210 (sec<sup>-1</sup>)

As the following graph (figure 1) shows, the two different formulas do not result in substantially different results in practical terms in this case:

**Figure 1 - Production of Po-210 from Bi-209 after N days irradiation (30 days cooling)**



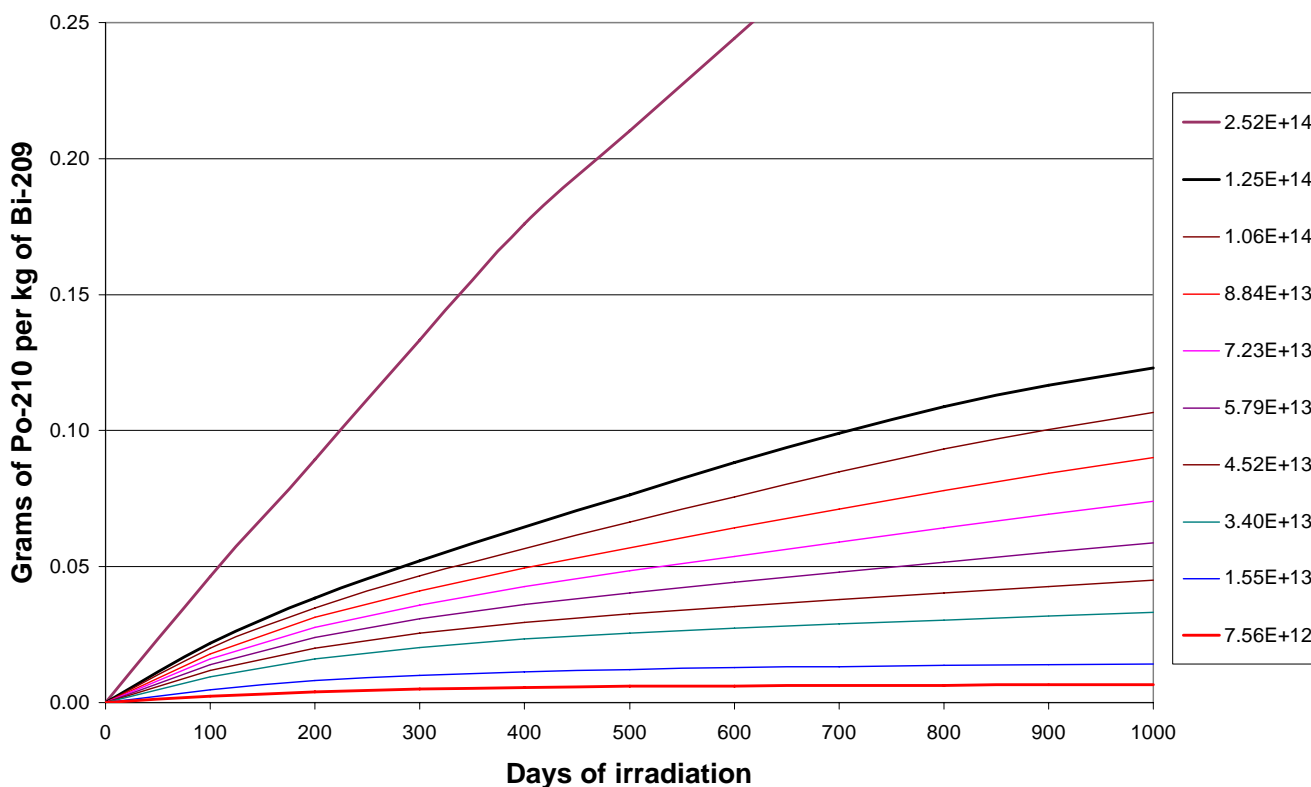
After performing the first principles calculations and comparing the results to the ORIGEN output it became clear that the two versions of the program were using different values for the  $^{209}\text{Bi}(n,\gamma)^{210\text{gs}}\text{Bi}$  reaction cross-section. A literature search on this issue led to an interesting discussion of this discrepancy by Letourneau et al<sup>6</sup>. Letourneau notes that the ENDF<sup>7</sup> figure for the reaction cross-section of 24.2 mb is approximately 50% higher than the value of earlier determinations (the paper argues that the real value for the relevant cross-section is 17.9 mb). Using the ENDF figure for the  $^{209}\text{Bi}(n,\gamma)^{210\text{gs}}\text{Bi}$  reaction cross-section of 24.2mb we can examine the potential for Po-210 production in research reactors of a variety of possible power levels.

In a given research reactor there is a well defined limit on the concentration level of Po-210 that can be reached in a given quantity of Bi-209 from a given period of irradiation, and that concentration limit is linearly dependent upon the average level of the available thermal neutron flux. The available thermal flux levels for different research reactors are well documented and are available from a variety of sources. As Figure 2 indicates, research reactors with flux levels of the order of  $10^{12}$  n/cm<sup>2</sup>-s will produce Po-210 concentrations in the parts per million range and relatively large amounts of target material would need to be processed to recover milligram quantities of Po-210.

While production of Po-210 at microgram or milligram levels would be very useful for a wide range of basic research on the physical and chemical properties of the material, including research into initiator design, it is unlikely that such production levels would be viewed as a satisfactory basis for an ongoing weapons program.

In considering research reactors as a source for the possible production of Po-210 it is also important to note that the regions within the core that are able to produce sustained thermal flux at the levels indicated will have relatively limited volumes. In a reactor with peak thermal neutron flux of order of  $10^{12}$  n/cm<sup>2</sup>-s (typical of many research reactors with thermal power in the range of a few hundred kilowatts) it would be necessary to process more than 100kg of metallic Bi-209 target material to extract 1g of Po-210. Physical limitations on irradiation ports and target positions are likely to place an upper limit on potential target size in the sub-kilogram range.

**Figure 2 - Production of Po-210 by irradiation of Bi-209 at a variety of flux levels**



The small number of larger research reactors (generally those operating with powers greater than 25 MWth) will have access to much higher levels of thermal flux, and will also have larger irradiation volumes over which this flux can be achieved. Working with a research reactor capable of sustained thermal flux levels of  $1.25 \times 10^{14}$  n/cm<sup>2</sup>-s or greater would allow for the recovery 1g of Po-210 from approximately 20kg of bismuth target material. The actual targets themselves would need to be contained in some form of cladding material, such as zirconium, and the overall weight of target would be approx 30% greater than the weight of the contained bismuth target material.

Physical indicators of this form of misuse of a research reactor would be the presence of target material in beam ports or sample tubes and hot-cell processing of target material. Due to the low

reaction cross-section, the irradiation of bismuth for this purpose will not necessarily make a readily observable change in the fuel consumption or heat production of a reactor misused in this way.

The major observable indicators would be related to the processing of Bi-209 targets. Po-210 is very difficult to contain and it tends to readily spread beyond its intended location. The spread of Po-210 would give rise to physical indicators detectable by existing environmental sampling techniques.

## TRITIUM

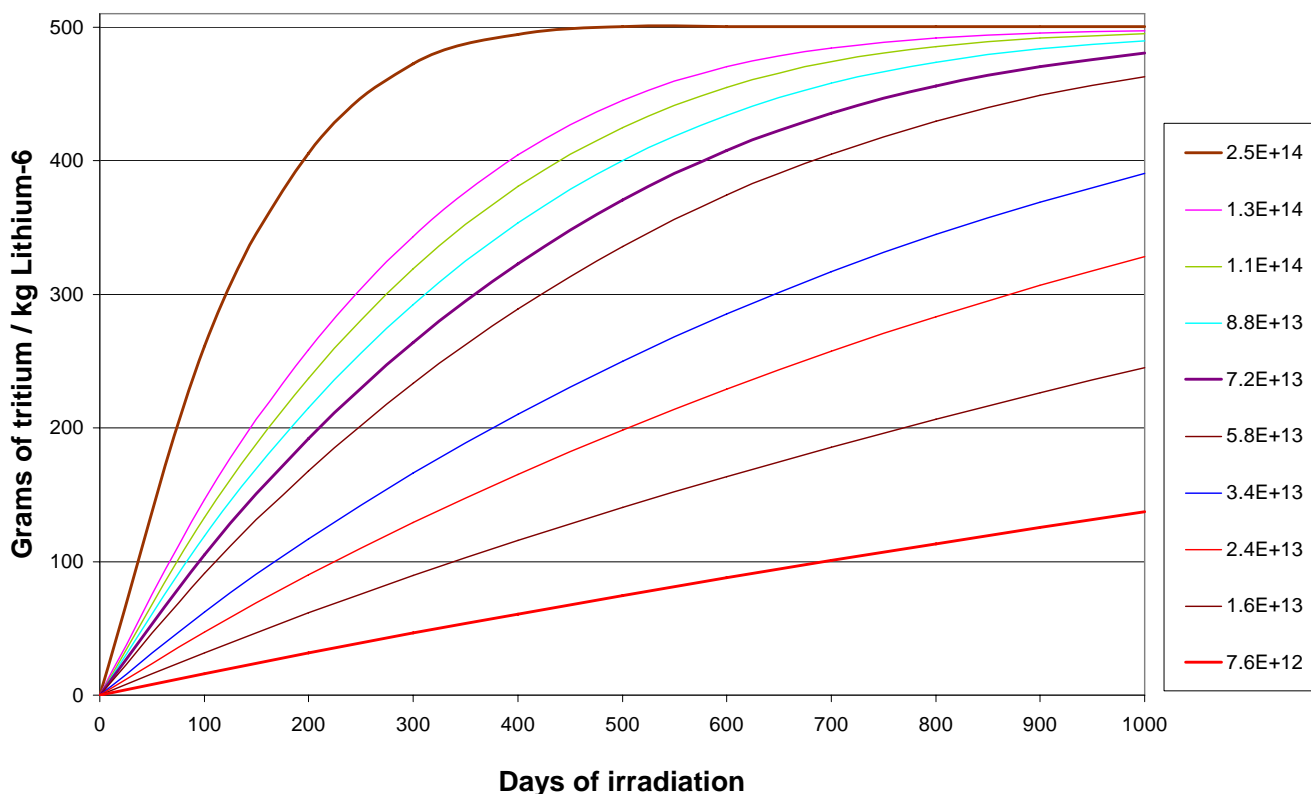
Another material of proliferation concern that could be produced in a research reactor is tritium. Tritium can be used to boost the explosive yield of a basic implosion design for a fission weapon and also has a role in ensuring that weapons have a stable and predictable yield.

There are two major possibilities for the production of tritium at research reactors:

1. recovery from tritiated heavy water;
2. irradiation of Li-6 targets.

For heavy-water moderated research reactors, tritium can be recovered from the moderator. Discussion of the processing of tritiated heavy water is beyond the scope of this paper.

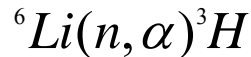
**Figure 3 - Production of Tritium by irradiating Li-6 at different flux levels**



Irradiation of lithium enriched in the isotope Li-6 with thermal neutrons is possible for all research reactors regardless of the type of moderator used by the reactor. A mathematically similar relationship

is observed between available thermal flux and concentration of tritium in the target material as was noted for production of Po-210. Lithium metal can be used as target material, but if conventional acids are used as part of the process (e.g. to dissolve the lithium target material to recover the tritium) they will result in tritium at low concentration in stable hydrogen. Various schema exist in the literature for using stable, refractory compounds of lithium such as Li<sub>2</sub>O and LiF as targets.

**Equation 4 - Production of tritium from <sup>6</sup>Li**



Due to the low densities and correspondingly high volumes of the targets, it would be necessary to process large volumes of target material to produce substantial quantities of tritium in low powered research reactors. Cladding for the lithium targets would more than double their mass (with only a marginal increase in their volume). Even in high-powered reactors the volumes of target material that would need to be processed would be considerable.

If lithium enriched in Li-6 is not available it would be possible to use natural lithium, but as the natural abundance of Li-6 is only of the order of 7.5% it would increase the volume of material that would need to be processed by a factor of 13.3 – taking it from tens to hundreds of litres of target material and correspondingly larger volumes of processing waste.

In addition to the very large volumes of material that would need to be processed, the production of tritium would have a substantial affect on the normal operation of the reactor. Due to the comparatively high (n,α) reaction cross-section of Li-6 (940 barns for Li-6 compared to 568 barns for the fission cross-section for U-235), the irradiation of Li-6 targets has a substantial effect on the number of available neutrons in the reactor. If we assume that 0.8 neutrons per U-235 fission is available for irradiation<sup>8</sup>, then the production of 1g of tritium will result in the consumption of an additional 90-100 grams of U-235, and also result in the evolution of 110-120 megawatt days of additional heat. This will require substantial changes in the fuel consumption patterns of the reactor and the operation of heat removal systems and cooling towers.

To place the fuel consumption changes in perspective, for a 1 megawatt research reactor operating with a 95% availability factor, undeclared production of 1g of tritium per year would result in a 32% increase in the amount of fuel used by the reactor and a 35% increase in the amount of heat that would have to be dealt with by the reactor's heat exchange system. The equivalent figures for a 10 megawatt research reactor would be 3.2% and 3.5%.

The changes in fuel consumption patterns associated with significant ongoing production of tritium would be readily observable by IAEA inspectors and the fact that there has been an increase in fuel consumption would be difficult to conceal. The changes that would be necessary in usage of heat removal systems would be readily observable via satellite imagery.

The volumes of target material that would need to be processed would require the development of extensive storage facilities for solid and liquid waste, and these additional facilities would also be readily observable via satellite imagery.

## CONCLUSION

Research reactors with available flux thermal levels below  $1.25 \times 10^{14}$  n/cm<sup>2</sup>-s could be used to produce small quantities (milligram or sub-milligram) of both tritium and Po-210. These quantities would be useful in basic research, but could not really serve as the basis of an ongoing weapons program.

High-powered research reactors (with available flux thermal levels above  $1.25 \times 10^{14}$  n/cm<sup>2</sup>-s) could be used to produce militarily significant quantities of either Po-210 or tritium.

Production of Po-210 by irradiating Bi-209 at research reactors would give rise to few observable indicators during normal reactor operation, but would involve the generation of large volumes of waste, and it is the waste that would tend to give rise to indicators observable by safeguards inspectors. Environmental sampling techniques would be useful in detecting this form of reactor misuse.

Production of tritium by the irradiation of Li-6 at research reactors would require substantial changes in the mode of operation of the reactor and would give rise to a wide range of observable indicators that would tend to make such efforts very difficult to conceal.

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1. The authors do not have access to any form of restricted data on the utility of various materials for nuclear weapons purposes. All calculations and related conclusions are on the basis of publicly available data and basic physics.
  2. This was the case even if such a reactor is operated well outside its normal safe operational limits, with significant increases in the consumption levels of reactor fuel and significant changes in the usage of heat removal systems (such as cooling towers).
  3. ORIGEN – Oak Ridge Isotope Depletion and Generation Code, distributed by the “Radiation Safety Information Computational Center” - RSICC at Oak Ridge National Laboratory, Oak Ridge, TN, USA. Calculations made use of ORIGEN-ARP included as part of the RSICC SCALE5 package.
  4. Nuclear Engineering Handbook, McGraw-Hill Book Company, USA, 1958.
  5. A. Letourneau, G. Fioni, F. Marie, D. Ridikas “*Measurement of the <sup>210</sup>Po production induced by thermal neutron capture on <sup>209</sup>Bi*” Annals of Nuclear Energy 9 November 2005.
  6. Ibid.
  7. ENDF - Evaluated Nuclear Data Files, National Nuclear Data Centre, Brookhaven National Laboratory.
  8. Each U-235 fission gives rise to an average of 2.5 neutrons – 1 neutron is needed to sustain criticality – 0.7 neutrons on average are lost to leakage, moderation and absorption and this leaves 0.8 neutrons on average per fission for irradiation.