



Australian Government

Australian Safeguards Support Program

**POTENTIAL FOR PRODUCTION
OF
PROLIFERATION SENSITIVE
MATERIALS IN
RESEARCH REACTORS**

**Task AUL C 01208: Re-Examination of Basic Safeguards
Implementation Parameters**

Russell LESLIE

**Australian Safeguards and Non-Proliferation Office (ASNO)
RG Casey Bldg, John McEwen Crescent, Barton ACT 0221 Australia**

March 2008

AUL Report 2008-01

PAGE INTENTIONALLY LEFT BLANK

POTENTIAL FOR PRODUCTION OF PROLIFERATION SENSITIVE MATERIALS IN RESEARCH REACTORS

1. 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 limit¹.

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. In addition to the more conventional research into delivery systems and high explosive detonation, it is reasonable to assume that the potential proliferator's program will also include efforts to ensure reliable initiation of weapons and measures to enhance the yield of any weapons produced.

Two of the materials of interest to proliferators that could potentially be produced by the misuse of research reactor facilities are Po-210 and tritium. This report will examine this issue, presenting the standard equations for the decay and growth of radioactive substances and adapting these to produce equations suitable for the examination of the production of Po-210 and tritium by neutron irradiation in research reactors.

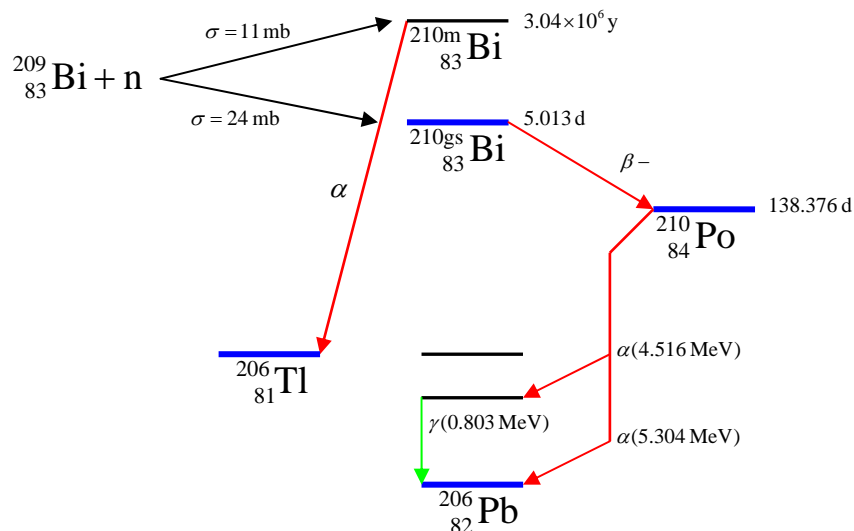
In the interests of simplicity the results of all of the forms of the equations used in this report will be, where possible, either in the form of simple atom ratios between the parent and daughter isotopes or in unit mass of daughter per unit mass of parent (in grams/kilogram). It is more common in the literature to express these results in terms of activity levels for the daughter isotope, but atom ratios and mass ratios are being used in this report as they avoid consideration of issues such as target size and cross sectional area when discussing neutron activation. Results are presented relative to the original target unless explicitly stated otherwise.

TABLE OF CONTENTS

1. INTRODUCTION.....	3
2. EXAMINATION OF POLONIUM-210	5
3. EPITHERMAL NEUTRON FLUX	10
4. SILICON IRRADIATION RIGS.....	11
5. COLD NEUTRON FLUX.....	12
6. TRITIUM	13
7. CONCLUSIONS	17
ANNEX 1. METHODOLOGY – BATEMAN EQUATIONS	19
ANNEX 2. TERMINOLOGY FOR DESCRIBING NEUTRON FLUX.....	25
ANNEX 3. REACTOR AVAILABILITY FACTORS	31
ANNEX 4. TARGET ACTIVATION SOLUTIONS VIA SOFTWARE	32
ANNEX 5. VARIATION IN CROSS SECTION VALUES FROM DIFFERING SOURCES	35
ENDNOTES – INCLUDING BIBLIOGRAPHY	37

2. EXAMINATION OF POLONIUM-210

Figure 1 - Bi-209 (n,γ) formation and decay scheme



Declassified historical information from the US military indicates that Po-210 was used in early generation nuclear weapons as a key element of a very basic “initiator” design². An initiator is a component of a nuclear weapon that is designed to deliver a high-intensity, short rise-time pulse of neutrons to the fissile components of a nuclear weapon to initiate the chain reaction. The principle design constraint upon an initiator is that the short, sharp neutron pulse it produces has to arrive at the centre of the fissile mass as close as possible in time to the point at which these fissile components reach their maximum compression.

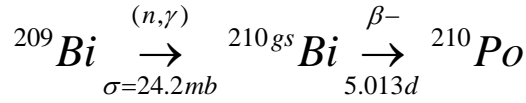
Po-210 is a highly active, short half-life isotope that is essentially a pure α emitter. When powdered Po-210 is combined with powdered beryllium, it is a high intensity neutron source due to an (α, n) reaction with beryllium. Typically a high intensity (α, n) source will produce 30 neutrons for every 10^6 α emitted¹⁰. An initiator would be expected to deliver 10^3 to 10^4 neutrons rapidly (i.e. over a period of the order of 10^{-7} seconds²). For a Po-Be initiator to emit 3×10^3 neutrons in a period of the order of 10^{-7} seconds would require an activity level for Po-210 of 10^{15} Bq. Using a Po-210 decay constant value of $\approx 6 \times 10^{-8}$ this implies that an effective initiator would need something of the order of 1-10 milligrams of Po-210.

If the mixing of the Po-210 and beryllium can be controlled in such a way that the two materials remain separated until the moment of maximum compression of a weapon’s fissile components, the sudden burst of neutrons can initiate the chain reaction and lead to the explosive release of energy. Po-210 does have legitimate industrial and scientific uses (in quantities many orders of magnitude lower than those needed for an initiator), but any program of research into the production of Po-210 has the potential to contribute to the proliferation of nuclear weapons.

The methodology used in this paper, the meanings of all variables and the form of the equations can be found in Annex 1.

The part of the process in Figure 1 that is of interest to this discussion of Po-210 is given by Equation 1:

Equation 1 – Process for the production of ^{210}Po



As noted in Figure 1 Bi-210 has a ≈ 5 day half life and Po-210 has a ≈ 138 day half-life. To a first approximation the production of Po-210 can be modelled as if it is a one step process (ignoring the intermediate step of Bi-210 production) using an adapted form of the Parent \rightarrow Daughter decay/activation equation (Equation 17, see Annex 1) for the production of a radioisotope by neutron irradiation. The adapted form is given as Equation 2 below:

Equation 2 – Simplified saturation model formula for production of Po-210

$$\left(\frac{N_T(\text{Po-210})}{N_0(\text{Bi-209})} \right) = \left(\frac{\sigma_{\text{Bi}}\phi}{\lambda_{\text{Po}}} \right) (1 - e^{(-\lambda_{\text{Po}}T)}) e^{(-\lambda_{\text{Po}}t)}$$

Where:

$N_T(\text{Po-210})$ = number of Po-210 atoms at time T

$N_0(\text{Bi-209})$ = initial number of Bi-209 atoms

λ_{Po} = the decay constant for Po-210 (sec^{-1})

T = period of irradiation in time units (e.g. sec)

t = cooling period since end of irradiation in time units

Note that this simplified form is only possible if the saturation model of the Bateman equations is valid (see Annex 1 for details).

A more accurate version of this equation would take into account the two step process with the ≈ 5 day half-life of Bi-210 included and would require a recast of Equation 19 given below as Equation 3:

Equation 3 – Two step saturation model formula for production of Po-210

$$\left(\frac{N_T(\text{Po-210})}{N_0(\text{Bi-209})} \right) = \left(\frac{\sigma_{\text{Bi}}\phi}{\lambda_{\text{Po}}(\lambda_{\text{Bi}} - \lambda_{\text{Po}})} \right) (\lambda_{\text{Bi}} (1 - e^{(-\lambda_{\text{Po}}T)}) e^{(-\lambda_{\text{Po}}t)} - \lambda_{\text{Po}} (1 - e^{(-\lambda_{\text{Bi}}T)}) e^{(-\lambda_{\text{Bi}}t)})$$

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

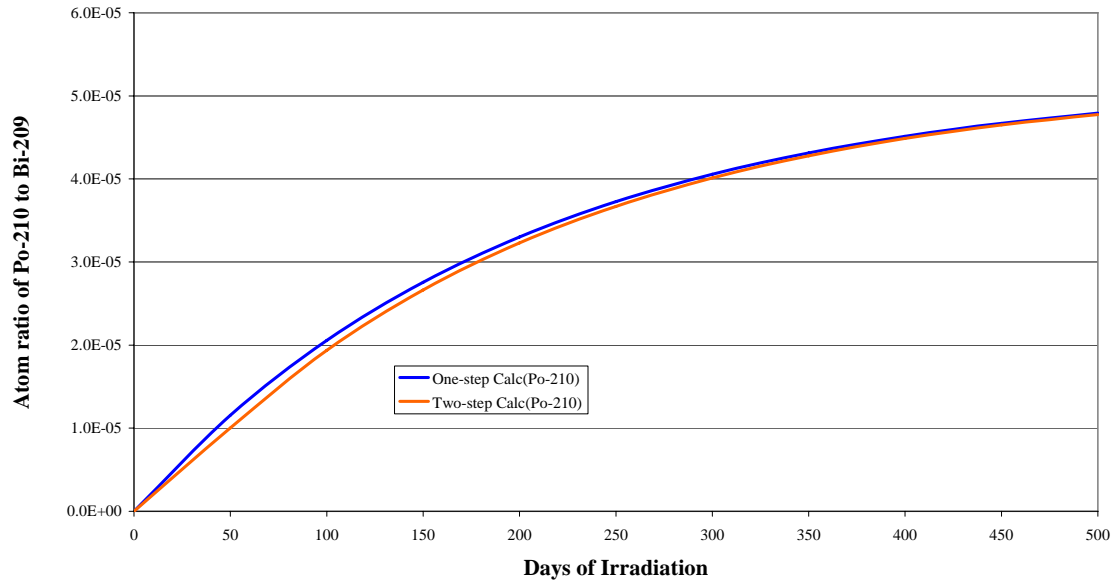
σ_{Bi} = neutron activation cross section for Bi-209 (barns)

ϕ = the neutron flux ($\text{n cm}^{-2} \text{s}^{-1}$)

λ_{Bi} = the decay constant for Bi-210 (s^{-1})

As the following chart (Figure 2) shows, the two different formulas do not result in substantially different results in practical terms in this case:

Figure 2 - Comparison of the results of Equation 2 and Equation 3 for thermal neutrons⁴ for a flux of $1.25 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$



Both Equation 2 and Equation 3 make the assumption that the saturation model¹⁰ is valid (i.e. that depletion of the target can safely be ignored). This assumption holds because the value of the activation cross section for Bi-209 is very low (in the millibarn range) and, at the flux-levels attainable in conventional reactors³, << 1% of the target material will be consumed during irradiation (see Annex 2 for details).

Figure 3 - Long-term irradiation of Bi-209 in thermal neutron flux of varying intensity⁴

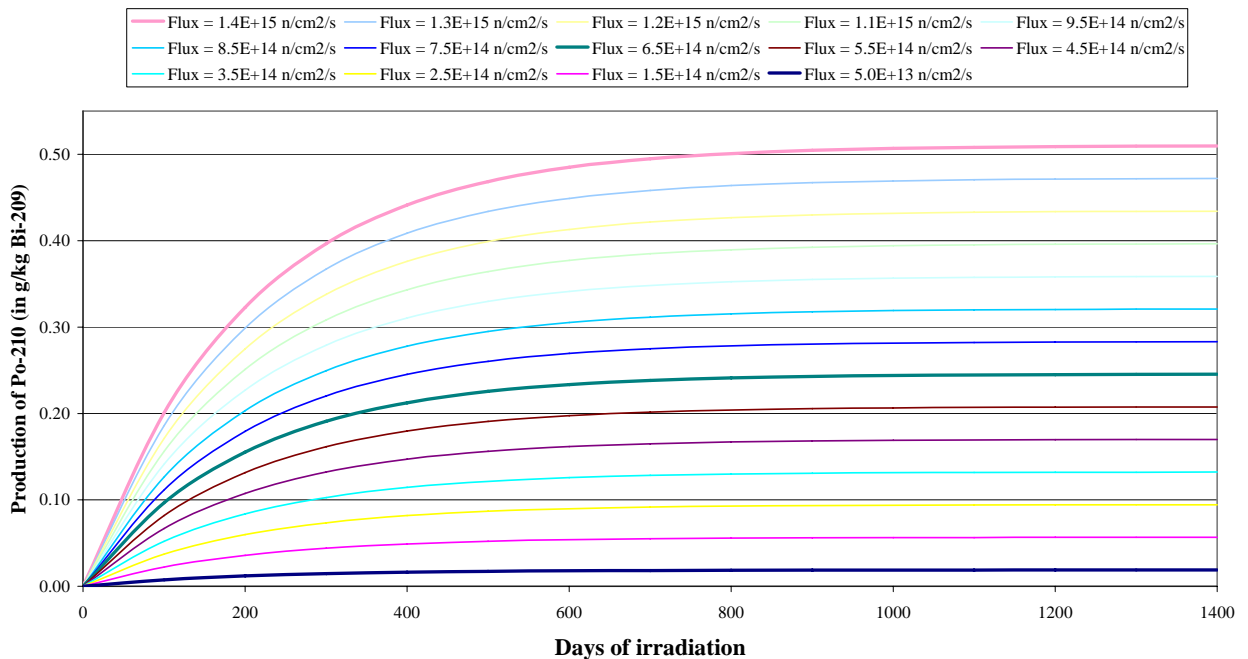
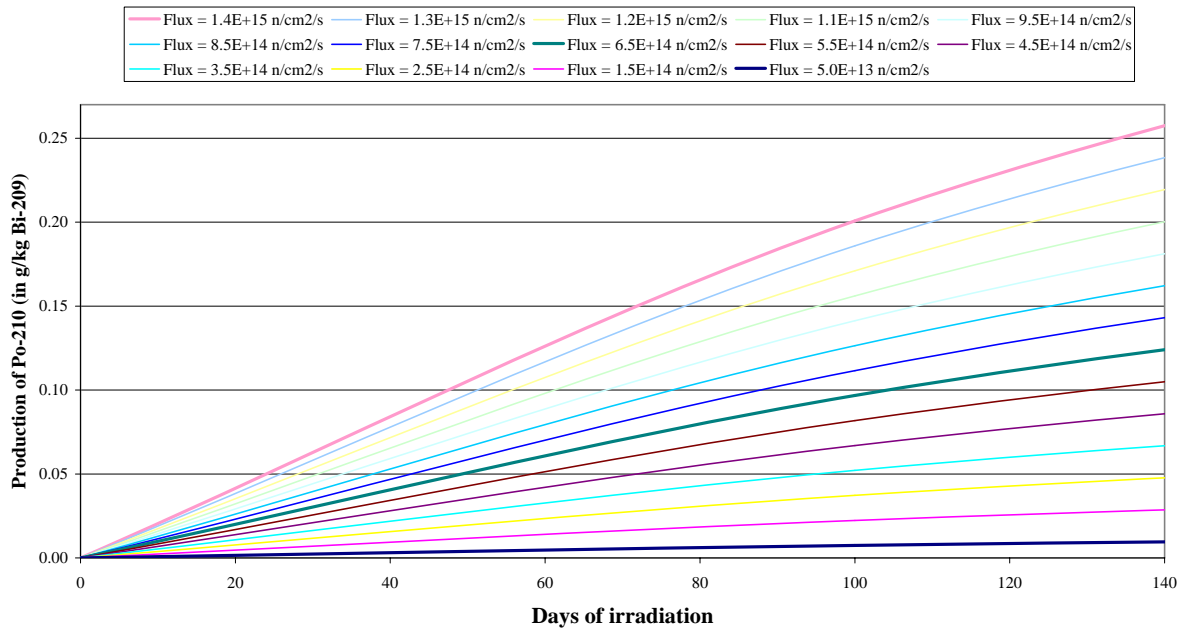


Figure 4 - 140 day detailed view of irradiation cases in Figure 3



The implication arising from Equation 2 and Equation 3 and as illustrated in Figure 3 and Figure 4 is that, 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 (see Table 3, Annex 2 for details). There is also, at each flux level, an associated “saturation value” at which further irradiation will not result in appreciably higher concentration levels of Po-210. The peak thermal flux levels for different research reactors are well documented (see Table 3) and are available from a variety of sources¹⁷.

As Figure 3 and Figure 4 indicate, research reactors with flux levels of the order of $1 \times 10^{13} \text{ n cm}^{-2} \text{ s}^{-1}$ would only be able to produce Po-210 concentrations in the parts per million range and relatively large amounts of target material would need to be processed to recover microgram 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. The relatively short half-life of Po-210 means that any initiator based on this isotope would have a relatively short shelf-life (likely to be of the order of four half-lives, roughly 20 months) and would lead to an ongoing need to continuously irradiate Bi-209 to maintain a stockpile of available Po-210.

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 and its surrounds that are able to produce sustained thermal flux at the levels indicated in the peak flux listings will have relatively limited volumes.

For lower flux levels, in reactors with peak thermal neutron flux of the order of

$1 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$ (typical of many research reactors with thermal power in the range of a few hundred kilowatts) production levels would be in the parts per billion range and it would be necessary to process more than 100kg of metallic Bi-209 target material to extract 10mg of Po-210. Physical limitations on irradiation ports and target positions are likely to place an upper limit on potential target size for these relatively small, low-powered reactors in the sub-kilogram range.

The small number of larger research reactors (see Table 3 and Figure 14) will have access to much higher levels of thermal flux, and will also have larger irradiation volumes over which this flux can be achieved⁷. Working with a research reactor capable of sustained thermal flux levels of $1 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$ or greater would allow for the recovery 10mg of Po-210 from approximately 1kg of bismuth target material after 70 days of irradiation. As Po-210 has a strong tendency to escape from immobilising matrices and contaminate its surrounds, the actual targets themselves would need to be contained in some form of cladding material, such as zirconium, and for relatively small bismuth loadings, the overall weight of the target could be expected to be up to 30% greater than the weight of the contained bismuth.

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.

3. EPITHERMAL NEUTRON FLUX

The resonance integral for the production of Bi-210 by the reaction in Figure 1 is in the range 85-103 mb (which is four to five times higher than the thermal reaction cross section)²². The higher cross section indicates that epithermal neutrons are more efficient than thermal neutrons in the production Po-210 by irradiation of Bi-209.

While epithermal neutrons are more efficient for this reaction, typical flux-levels of epithermal neutrons in major research reactors are no more than 5-10% of the thermal flux levels⁵. Most of the existing literature on the availability of epithermal neutrons in reactor flux is devoted to epithermal neutron activation analysis (ENAA)⁵. In the ENAA literature flux levels are generally quoted in terms of their “cadmium ratio”. Cadmium heavily attenuates thermal neutrons below 0.4 eV but has little effect on neutrons of higher energy. The ratio between neutron measurements made with and without cadmium filters provide an effective numeric comparison between the total flux levels and the epithermal flux levels. Cadmium ratios for a given reactor are not constant (either temporally or spatially) and will generally have to be examined and regularly re-determined in each irradiation facility in the reactor. Typical, practical cadmium ratios, quoted in the ENAA literature⁵, vary between values of 10 and 20.

ENAA irradiation positions within reactors tend to be much smaller than the isotope production target irradiation positions which generally use fully thermalised flux.

It is straightforward to calculate the amount of additional epithermal activation of target material if the resonance integral and the cadmium ratio for the irradiation position are known. The epithermal activation is simply added to both the thermal activation level and the saturation level calculated for the known thermal flux level. For Bi-209 – a cadmium ratio of 10 will increase the saturation level and the rate of Po-210 production by 50% while a cadmium ratio of 20 results in only a 15% increase in the same levels.

When the cadmium ratio is >40 the correction for additional epithermal activation for Bi-209 would be expected to smaller than other uncertainties in the Po-210 production rate.

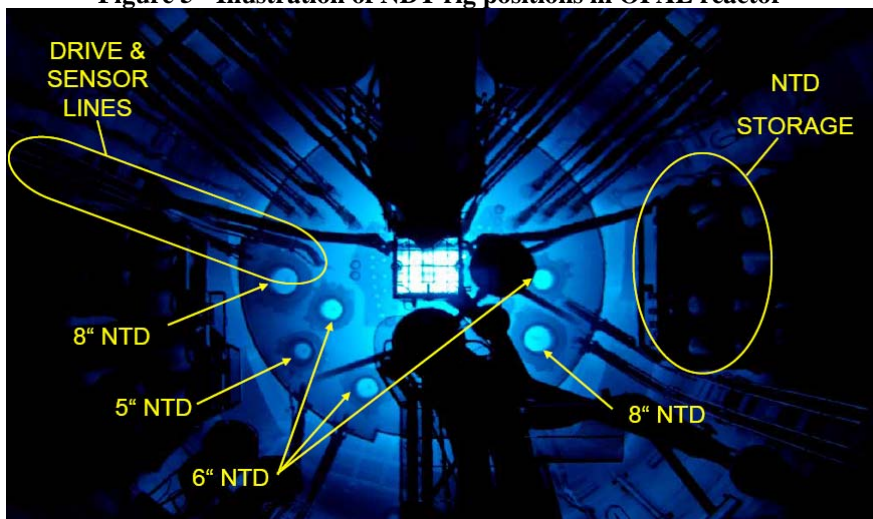
4. SILICON IRRADIATION RIGS

An example of a class of large and effective irradiation positions that could be misused for the production of Po-210 is the irradiation rig positions that are used for nuclear transmutation doping (NTD) of silicon⁶. These rigs are designed to expose high-purity cylindrical silicon ingots of diameters up to 200 mm and lengths of 600 mm to very high, uniform, fully thermalised neutron flux (cadmium ratio⁵ >400)⁷. The ingots are continuously, axially rotated to ensure uniformity of irradiation, though additional physical measures are also used to ensure flux uniformity and thermalisation⁶.

There is a large industrial demand for NTD silicon and conducting high quality irradiations of this type is highly profitable for reactor operators. The semiconductor industry pays substantial premiums for irradiations that are precise, accurate, uniform and reproducible. NTD rigs are becoming an increasingly common design feature in high flux research reactors.

With an appropriate allowance for canning materials it would be feasible to irradiate batches of up to 150kg of bismuth in a 200mm irradiation rig – though it would probably be more realistic to assume a maximum batch size of the order of 40-45kg. The axial rotation mechanisms of the NTD rigs are designed for materials that match silicon's density and they are unlikely to be able to function effectively with weight that greatly exceeds their design capacity. Use of the rig positions without axial rotation would be possible, but self-shielding within the targets may become an issue.

Figure 5 - Illustration of NTD rig positions in OPAL reactor⁷



5. COLD NEUTRON FLUX

In the thermal (1/v) range (see Annex 2 for details) there is a simple relationship between reaction cross sections at different neutron temperatures or energies (Equation 4 for temperature and Equation 5 for energy⁸).

Equation 4 - Temperature dependence of σ in the thermal range

$$\sigma = \sigma_0 \left(\frac{T_0}{T} \right)^{1/2}$$

Equation 5 - Energy dependence of σ in the thermal range

$$\sigma = \sigma_0 \left(\frac{E_0}{E} \right)^{1/2}$$

It is also possible to compare a series of neutron activation calculations (making use of Equation 18, Equation 19 or Equation 20) that produce different results and work back to determine what value for neutron temperature would produce equivalent results (Equation 6).

Equation 6 - Temperature dependence of σ in the thermal range

$$T = T_0 \left(\frac{\sigma}{\sigma_0} \right)^{-2}$$

Many large, high-flux reactors have “cold” neutron sources which are designed to thermalise a portion of the neutron flux to temperatures in the range 2-20 °K. Using Equation 6 it is simple to calculate that the effective cross section for neutrons with a temperature of 20 °K will be increased by a factor ~4 over the 293 °K value while for neutrons thermalised to a temperature of 2 °K the effective cross section will be increased by a factor of ~12.

Cold flux levels are typically an order of magnitude lower than peak thermal flux levels and the drop in flux is likely to approximately counter-balance the increase in cross section. Overall production rates using cold neutrons are unlikely to be higher than production rates using thermal neutrons.

6. 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:

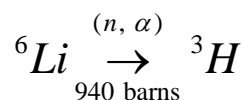
1. recovery from tritiated heavy water;
2. irradiation of Li-6 targets.

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

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. Lithium metal can be used as the target material for irradiation, but if conventional acids are used as part of the recovery 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 Li₂O and LiF as targets (with recovery of tritium by mechanical means e.g. heating and crushing).

The activation cross section for Li-6 is 940 barns and the resonance integral is 422 barns.

Equation 7 - Production of tritium from ⁶Li



Due to the comparatively high thermal neutron activation cross section for Li-6 it is necessary use the depletion model form of the Bateman equations following the model given in Equation 20 (see Annex 1 for details). The required form is given as Equation 8 below:

Equation 8 – Li-6 irradiation version of Equation 20

$$\left(\frac{N_T(\text{H-3})}{N_o(\text{Li-6})} \right) = \left(e^{(-\phi T \sigma_{\text{Li}})} - e^{(-\lambda_{\text{H3}} T)} \right) e^{(-\lambda_{\text{H3}} t)}$$

If lithium enriched in Li-6 is not available it is possible to use natural lithium as target material, but as the natural abundance of Li-6 is only ≈7.5% it would increase the volume of material that would need to be processed by a factor of ≈13.

Figure 6 - Long-term irradiation of Li-6 in thermal neutron flux of varying intensity⁴

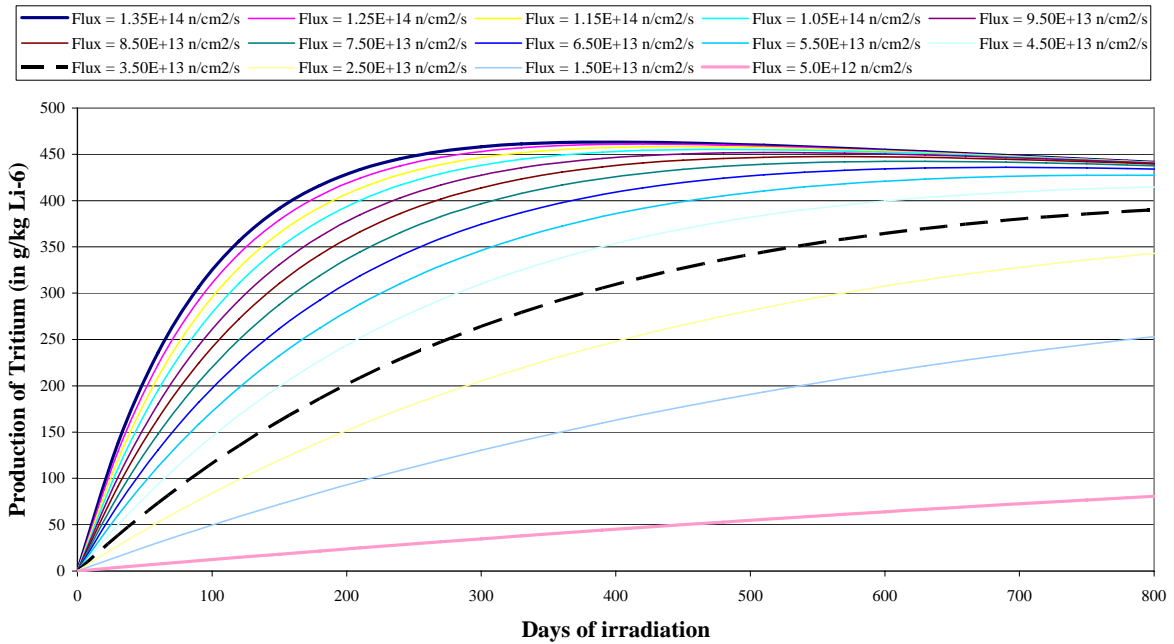
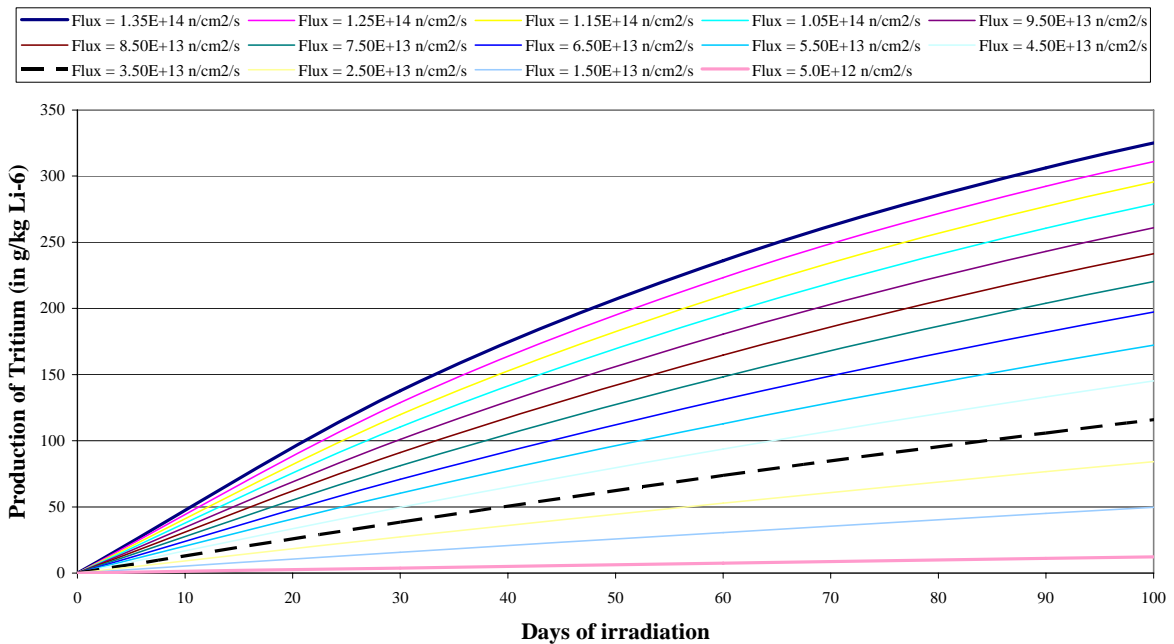


Figure 7 - 100 day detailed view of irradiation cases in Figure 6



While Figure 6 and Figure 7 show that production of tritium would be possible in a reactor of almost any power level, there are additional considerations relating to Li-6 irradiation that limit the feasibility of tritium production. 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. Li-6 is a very effective burnable neutron poison

Irradiation of Li-6 has a substantial affect on the normal operation of any nuclear reactor. It requires substantial changes in the fuel consumption patterns of the reactor and in the operation of heat removal systems and cooling towers.

Noting that:

Atomic mass Tritium ≈ 3.0 atomic mass units (amu)

Atomic mass of U-235 ≈ 235 amu

Avogadro's number $\approx 6.0 \times 10^{23}$ atoms per g-atom

One Megawatt day $\approx 5.4 \times 10^{23}$ MeV

Avg No. of neutrons / U-235 fissions ≈ 2.5 neutrons

Avg energy release / U-235 fission ≈ 200 MeV

Avg No. of free neutrons per U-235 fission in research reactor ≈ 0.8 neutrons

An approximate value for the energy release when generating one gram of tritium by neutron irradiation in a thermal reactor is detailed in Table 1 as follows:

Table 1 – Indicative calculation of energy release in generating one gram of tritium

<p>Atoms in 1 gram Tritium \equiv Neutrons absorbed to make 1 gram tritium $= \left(\frac{\text{Avogadro's number}}{\text{Atomic mass Tritium}} \right)$ $\approx 2 \times 10^{23}$</p>
<p>$\left(\frac{\text{Avg energy release}}{\text{Fission of 1 gram U-235}} \right)$ $= (\text{No. atoms in 1 gram U-235}) \times \left(\frac{\text{Avg energy release}}{\text{U-235 fission}} \right)$ $= \left(\frac{\text{Avogadro's Number}}{\text{Atomic Mass U-235}} \right) \times 200 \text{ MeV}$ $\approx 5 \times 10^{23} \text{ MeV}$</p>
<p>Energy released in producing 1 atom Tritium $= \left(\frac{(\text{Avg energy release / fission})}{(\text{Free neutrons / fission})} \right)$ $\approx 250 \text{ MeV}$</p>
<p>U-235 atoms fissioned for 1 gram tritium $= \left(\frac{(\text{Neutrons to produce 1 gram Tritium})}{(\text{Free neutrons / fission})} \right)$ $\approx 2.5 \times 10^{23} \text{ atoms}$</p>
<p>U-235 grams fissioned for 1 gram tritium $= \left(\frac{(\text{U-235 Atomic mass}) \times (\text{No. of U-235 atoms / gram})}{(\text{Avogadro's number})} \right)$ $\approx 100 \text{ g U-235}$</p>

Additional heat generated in producing 1 gram tritium

= (U-235 atoms fissioned for 1 gram tritium) × (Avg energy release/U-235 fission)

$$\approx 6.3 \times 10^{25} \text{ MeV}$$

$$\approx 120 \text{ MWd}$$

To place the fuel consumption changes in perspective, for a 1 megawatt research reactor operating with a 90% availability factor, production of 1g of tritium per year would result in a 32% increase in the amount of fuel used by the reactor and a 38% 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 3.8%.

The changes in fuel consumption patterns associated with significant ongoing production of tritium would be readily observable and the fact that there had 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.

7. CONCLUSIONS

1. Research reactors with available flux thermal levels below $1 \times 10^{13} \text{ n cm}^{-2} \text{ s}^{-1}$ could be used to produce small quantities (milligram or sub-milligram) of Po-210. These quantities would be useful in basic research, but could not really serve as the basis of an ongoing weapons program.
2. High-flux research reactors (with available flux thermal levels above $1 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$) could be used to produce militarily significant quantities of either Po-210 or tritium.
3. 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 due to the need to continually produce fresh supplies of Po-210.
4. 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.
5. In the case of the production of Po-210 by irradiating Bi-209, epithermal flux is more efficient than thermal flux and so care must be taken, when calculating production rates, to ensure that an appropriate adjustment is made for the cadmium ratio of the neutron flux.
6. The use of NTD silicon irradiation rigs at large research reactors could potentially be an effective means of producing Po-210 on a significant scale.
7. The use of “cold” neutron sources for the production of Po-210 is unlikely to provide a significant advantage as the increase in activation cross-section is, with current technology, effectively counter-balanced by a drop in available flux levels.
8. The production of tritium by the irradiation of Li-6 would give rise to substantial changes in the fuel consumption and heat disposition patterns of small research reactors and would require substantial changes in the modes of operation of the reactors. Such changes would give rise to a wide range of observable indicators that would tend to make such efforts very difficult to conceal.
9. Large spallation neutron sources (with available flux thermal levels above $1 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$) could potentially be used to produce militarily significant quantities of either Po-210 or tritium (see Annex 2). The IAEA does not currently have routine access to such facilities. In considering State Level Approaches (SLA) for states with large spallation neutron sources, the potential for production of proliferation sensitive materials at these facilities should not be overlooked. However, where (as is usual in the operation of these large-scale facilities) there is a high level of international collaboration, the potential for misuse of spallation source facilities for proliferation purposes is very low.
10. The Bateman methodology is directly applicable to studies of neutron activation of target material in research reactors. In applying the Bateman methodology care must be taken to determine whether the saturation model for activation is appropriate before it is relied

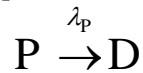
upon. In cases where there is significant depletion of the target material it is essential to make use of calculations that explicitly account for the depletion factor.

11. In making neutron activation calculations, the uncertainties in resonance integral values can be a significant source of systematic error in the calculations. The differences between various data sources' values of the resonance integral of Bi-209 were found to be greater than the quoted uncertainties on those values. Comparisons of multiple, independent data sources are a useful means of gaining perspective on underlying variation in values. In making calculations drawing upon these cross sections and resonance integrals the underlying uncertainties in the cross section values need to be factored into the overall uncertainty in the final result.
12. While isotope depletion and generation calculation tools such as ORIGEN are extremely valuable, their applicability to any particular irradiation case needs to be checked carefully rather than simply assumed. Such codes should not be treated as a "black box". The ability to perform reality check calculations to evaluate results will always be valuable (though it must be recognised that cross-checking complex irradiation cases can be very time-consuming).
13. It is important to view published thermal flux levels for research reactors with a substantial degree of caution. The thermal neutron flux in any large reactor facility will vary over orders of magnitude depending on the location within the reactor core or its surrounds. Calculations of production rates via neutron activation need to draw upon realistic values for time averaged thermal flux for the target position rather than just the published flux value for the reactor.
14. Published flux figures for research reactors are quoted in $n\text{ cm}^{-2}\text{ s}^{-1}$, but extrapolating from per second figures to long term irradiations lasting days, months or years essentially assumes that the reactor operates with a 100% availability factor. In fact it is rare for reactor operators to aim for availability factors above 90%. Many research reactors are significantly under utilised, with planned availability factors below 20% common.

Annex 1. METHODOLOGY – BATEMAN EQUATIONS

The exponential laws that govern nuclear decay and growth of radioactive substances are usually referred to as Bateman equations⁹. These were developed in 1910 by Harold Bateman, expanding upon earlier work by Earnest Rutherford. Using the Bateman formalism, the simplest form of radioactive decay is that represented by Equation 9 below

Equation 9 - Description of Parent / Daughter decay



Where:

- P** = parent isotope
D = daughter isotope
 λ_p = the decay constant of the P in units of reciprocal time (e.g. s⁻¹)

Where the decay constant “represents the probability that any particular atom of a radioactive substance containing a large number $N(t)$ of identical radioactive atoms will decay in the time interval (t) .”¹⁰

The half-life for P with decay constant λ_p is given by Equation 10 below:

Equation 10 – Relation between decay constant and half-life

$$t_{1/2}(P) = \left(\frac{\ln(2)}{\lambda_p} \right)$$

Figure 8 and Figure 9 (below) make use of arbitrary time units $t_{1/2}(T)$ where:

- $t_{1/2}(0) = 0$
 $t_{1/2}(1) = 1$
 $t_{1/2}(2) = 2 \times t_{1/2}(1) = 2$ and so on.

The ratio of the number of atoms of the parent and daughter isotopes as a function of time is given by the two forms of Bateman equations given as Equation 11 and Equation 12 below.

Equation 11 - Simple form of equation for decay of P

$$\left(\frac{N_t(P)}{N_0(P)} \right) = e^{(-\lambda_p t)}$$

Equation 12 - Simple form of equation for production of D

$$\left(\frac{N_t(D)}{N_0(P)} \right) = 1 - e^{(-\lambda_p t)}$$

Where λ_p is as defined in Equation 9 and:

- $N_t(P)$ = the number of atoms of parent isotope P as a function of time
 $N_t(D)$ = the number of atoms of daughter isotope D as a function of time

$N_0(P)$ = the number of atoms of parent isotope P at initial time

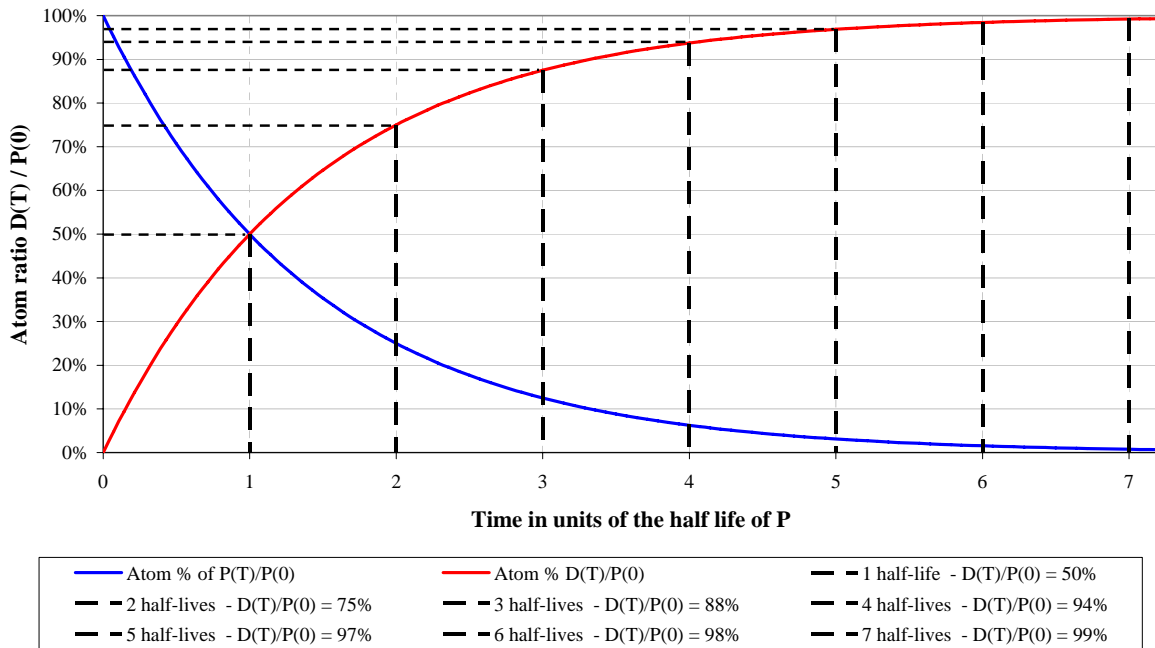
t = time of decay in time units (e.g. s)

Assuming that:

$N_0(D) = 0$

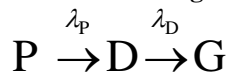
D is a stable isotope (as illustrated by Figure 8).

Figure 8 - Exponential decay of P and build-up of stable D



A more complex form of radioactive decay “Parent → Daughter → Granddaughter” is represented by Equation 13 below

Equation 13 - Description of Parent / Daughter / Granddaughter decay



Where the terms in common have the same meaning as in Equation 9 above and:

G = granddaughter isotope

λ_D = the decay constant of the daughter isotope in units of reciprocal time (e.g. s^{-1})

For this situation the ratio between the number of atoms of D and the number of atoms of P as a function of time (for $\lambda_D > \lambda_P$) is given by the form of the Bateman equations given as Equation 14 below:

Equation 14 – Formula for calculating the atom ratio of D to P

$$\left(\frac{N_t(D)}{N_0(P)} \right) = \left(\frac{\lambda_P}{\lambda_D - \lambda_P} \right) \left(e^{(-\lambda_P t)} - e^{(-\lambda_D t)} \right)$$

The ratio between the number of atoms of G and the number of atoms of P as a function of time is given by the form of the Bateman equations given as Equation 15 and illustrated by Figure 9 below

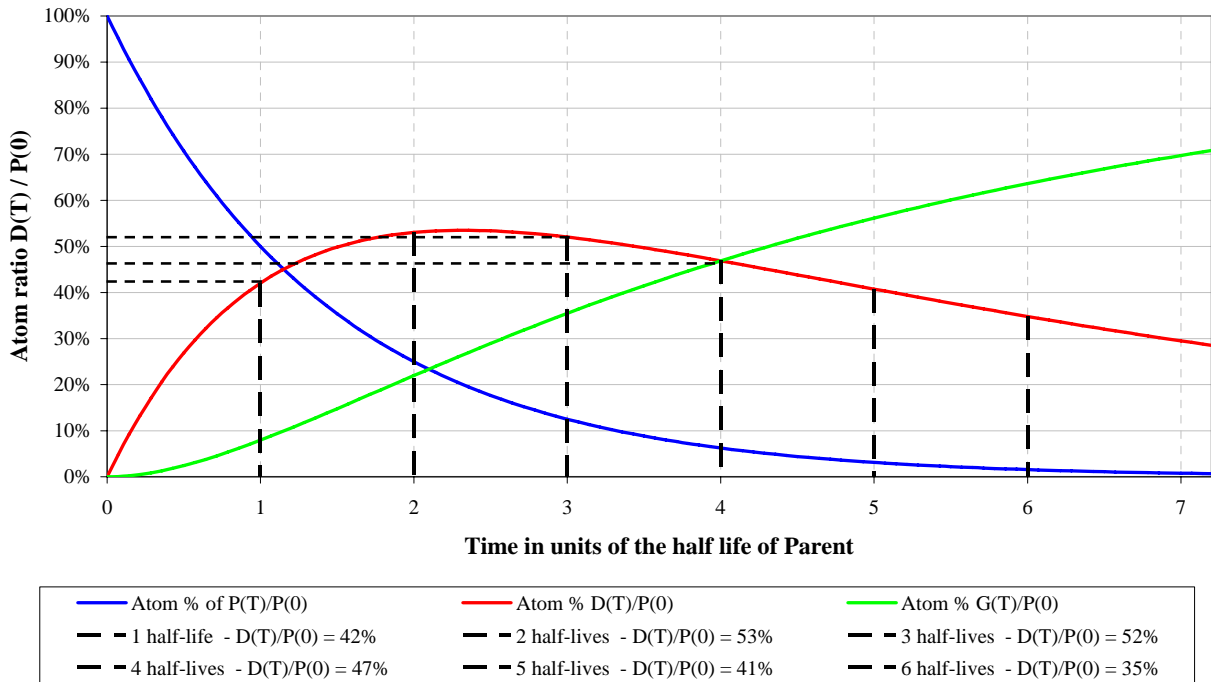
Equation 15 – Formula for calculating the atom ratio of G to P

$$\left(\frac{N_t(G)}{N_0(P)} \right) = \left(\frac{\lambda_P}{\lambda_G (\lambda_D - \lambda_G)} \right) \left(\lambda_D (1 - e^{(-\lambda_G t)}) - \lambda_G (1 - e^{(-\lambda_D t)}) \right)$$

Where the terms in common have the same meaning as in Equation 9 above and:

λ_g = the decay constant of the granddaughter isotope in units of reciprocal time

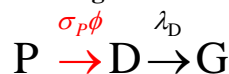
Figure 9 - Exponential decay of P and D and build-up of G - $\lambda(G) = \lambda(D) \times 4$



In Figure 9 the percentage value of D(T)/P(0) remains relatively unchanged (at 52-53%) for a period starting at two half-lives and extending past three half-lives. This type of stability is referred to as “transient equilibrium”¹⁰ and equilibria of this type are common in P→D→G decay chains¹⁰. In cases in which the $\lambda_D \gg \lambda_P$ the rate of decay of D is limited by its rate of production and its concentration level will stay stable for times comparable to the half-life of P. This form of stability is referred to as “secular equilibrium”¹⁰ and is of relevance to the discussion of neutron activation below¹¹.

The Bateman formalism can readily be applied to the generation of radioisotopes by neutron activation of stable target material¹⁰ — with only a few relatively minor modifications required as detailed in Equation 16 below:

Equation 16 - Description of Parent / Daughter / Granddaughter with neutron activation



Where:

$\sigma_P \phi$ replaces λ_P and

σ_P = the activation cross section of the parent isotope in barns (i.e. 10^{-24}cm^{-2})
 ϕ = the neutron flux in $\text{n cm}^{-2} \text{s}^{-1}$
 T = period of irradiation in time units (e.g. s)
 t = cooling period since end of irradiation in time units

In the figures that follow, Figure 10, Figure 11 and Figure 12, arbitrary time units are used, based on $\sigma_P \phi (T)$. Where:

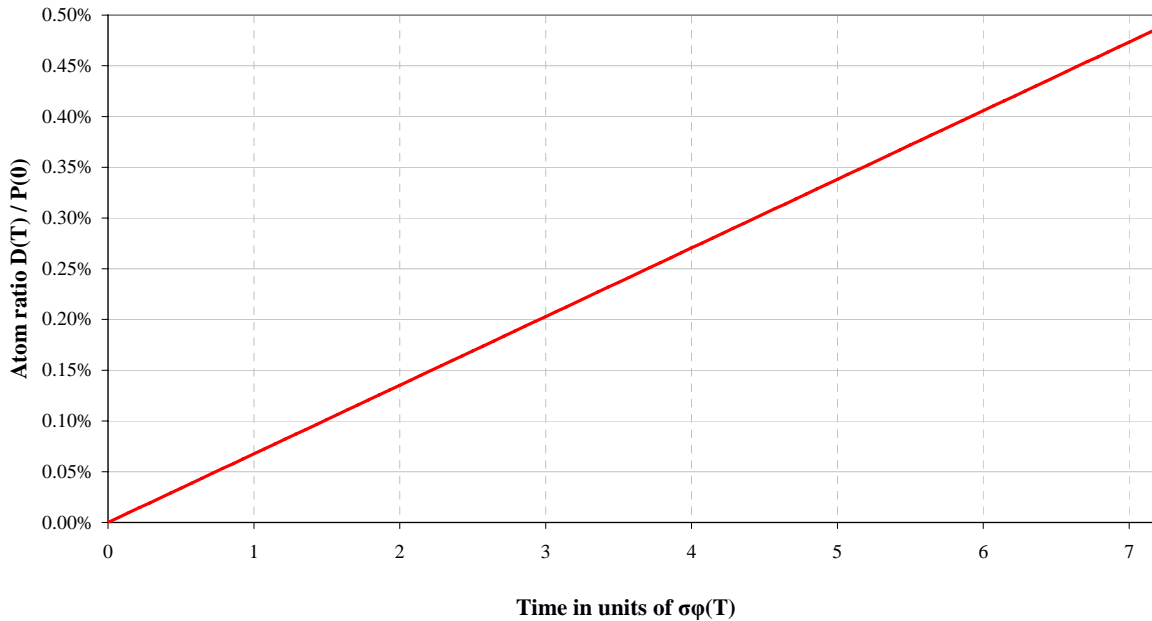
$\sigma_P \phi (0) = 0,$
 $\sigma_P \phi (1) = 1,$
 $\sigma_P \phi (2) = 2 \times \sigma_P \phi (1) = 2$ and so on

Taking this modification into account, in the limiting case where the number of atoms of the parent isotope is assumed to remain effectively unchanged throughout the period of irradiation (referred to as the “saturation model”), Equation 12 can be re-expressed in modified form as Equation 17 (for the trivial case of D stable) and Equation 18 (for the more general case of D with decay constant λ_D) below:

Equation 17 - Production of stable D by activation of P – saturation model

$$\left(\frac{N_T(\text{D})}{N_0(\text{P})} \right) = \sigma_P \phi T$$

Figure 10 – Linear production of stable D by activation of P – saturation model



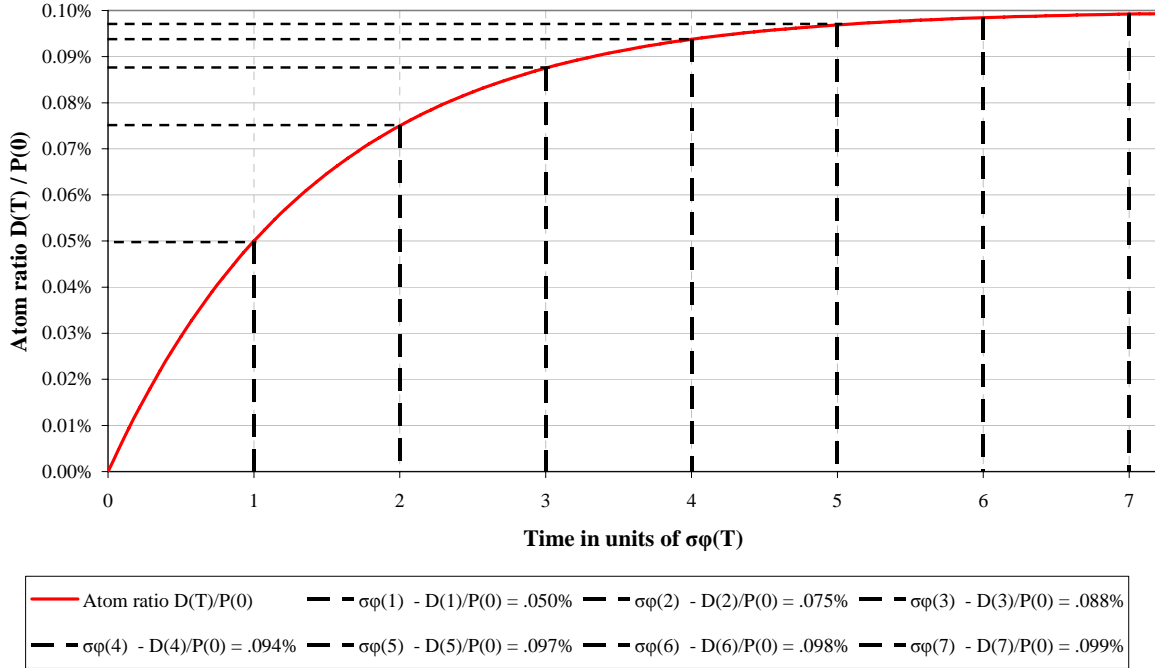
Equation 18 - Neutron activation of stable P, production and decay of D – saturation model

$$\left(\frac{N_T(\text{D})}{N_0(\text{P})} \right) = \left(\frac{\sigma_P \phi}{\lambda_D} \right) \left(1 - e^{(-\lambda_D T)} \right) e^{(-\lambda_D t)}$$

Equation 19 – Neutron activation of stable P, production and decay of D and G – saturation model

$$\left(\frac{N_t(G)}{N_0(P)} \right) = \left(\frac{\sigma_P \phi}{\lambda_G (\lambda_D - \lambda_G)} \right) \left(\lambda_D (1 - e^{(-\lambda_G T)}) e^{(-\lambda_G t)} - \lambda_G (1 - e^{(-\lambda_D T)}) e^{(-\lambda_D t)} \right)$$

Figure 11 - Exponential production of radioactive D by activation of P – saturation model



The saturation model will be valid if the depletion of P is $\ll 1\%$ and it may be useful if the change is of the order of 1% depending upon the application. If the depletion of P is in the range 1% \rightarrow 10% the validity of the results will have to be examined closely.

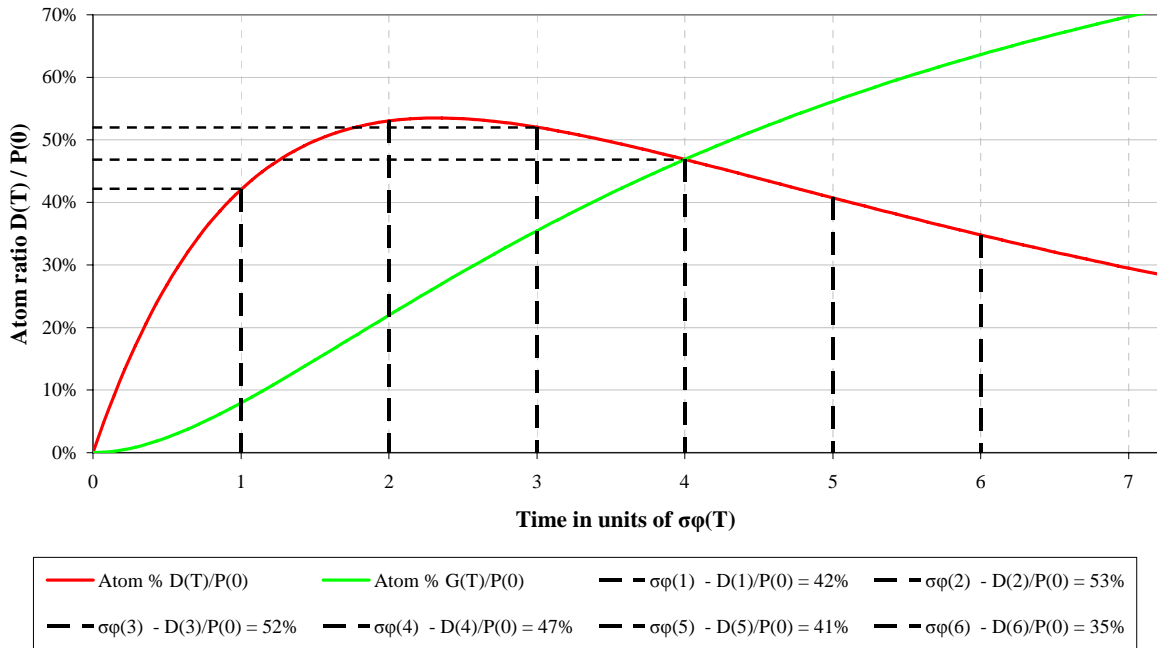
If the number of atoms of the parent isotope is significantly depleted during irradiation ($\gg 10\%$) it will not be possible to use the saturation form of the Bateman equations and it will be necessary to use a form that explicitly recognises this depletion (referred to as the “depletion model”)¹¹. The depletion model form of Equation 12 is given as Equation 20 below

Equation 20 - Depletion model version of Equation 12 for neutron activation

$$\left(\frac{N_t(D)}{N_0(P)} \right) = \left(e^{(-\sigma_P \phi T)} - e^{(-\lambda_D T)} \right) e^{(-\lambda_D t)}$$

Equation 20 is the only form of the depletion model that is needed for this report. Figure 12 illustrates Equation 20

Figure 12 - Exponential activation and decay of P, decay of D and build-up of G - depletion model
 $(\sigma\phi = \lambda(D)\times 4)$

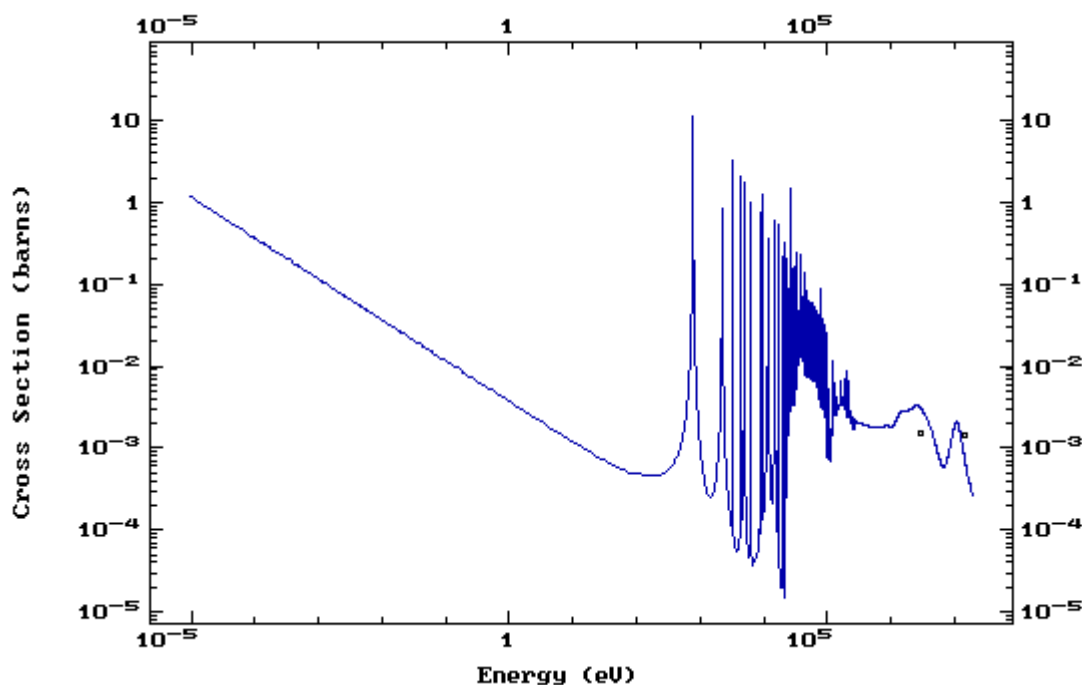


Annex 2. TERMINOLOGY FOR DESCRIBING NEUTRON FLUX

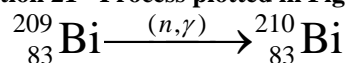
Neutron flux represents the rate of “flow” of neutrons, i.e. the number of neutrons passing through a unit area in unit time. Neutron flux can be considered in terms of the attributes of the neutrons making up the flow (neutron quality) and/or their number (neutron quantity).

When dealing with the quality of neutron flux, different terms and descriptions are considered important by different interest groups. Reactor operators tend to discuss the quality of the neutron flux in terms of the elements that contribute to the calculations of the level of reactivity in the reactor¹². Primarily the descriptive terms that are of direct relevance to reactor operators when discussing neutron quality are “fast”, “epithermal” (or resonance region) and “thermal”.

Figure 13 - Energy dependence of cross section¹³



Equation 21 - Process plotted in Figure 13



One of the reasons that it is important to know the energy-level of neutrons is that, for a considerable number of nuclides of moderately high (or high) mass numbers, the absorption cross section has three definite and distinctive regions on a curve of absorption cross section vs. neutron energy¹⁴.

First, the cross section decreases steadily with increasing neutron energy in a low energy region, which includes the thermal range ($E < 1$ eV). In this region the absorption cross section is inversely proportional to the velocity (v). This region is frequently referred to as the “ $1/v$ region” because the absorption cross section is proportional to the reciprocal of neutron velocity ($1/v$).

Next we have the epithermal or “resonance” region in which the cross section rises sharply to high values called “resonance peaks” for neutrons of certain energies, and then fall again. These energies are called “resonance energies”. Resonance energies occur when the incident neutrons’ energies closely match the quantum energy levels of the target atom.

For higher neutron energies, the absorption cross section steadily decreases as the energy of the neutron increases. This is called the “fast” neutron region. In this region the absorption cross sections are usually less than 10 barns.

In the production of isotopes it is common to refer to neutrons either in terms of their temperature, their speed or their energy. So, for example, it appears to be equally common to refer to thermal neutrons (at 293°K) as having a temperature of 293°K (or 20°C) and a corresponding most probable energy of 0.0253 eV or a most probable speed of ≈2,200 m/s.

In neutron diffraction it is common to refer to neutrons by their de Broglie wavelength, in which case, thermal neutrons are described as having a wavelength¹⁵ (λ) of ≈1.85Å.

It is important to be able to freely interpret information that is given in any one of the forms listed above in order to ensure that all comparisons are done on the basis of like with like.

The simplest and most easily reproducible physical measurement of relevance to the discussion of neutron flux is the “temperature” of the neutrons. Neutrons behave as an ideal gas which (for thermal neutrons) is in thermal equilibrium with the moderator material. Determining the temperature of the moderator determines the temperature of the neutrons. It is common to use units of temperature °K or °C (though some reference works provide values in °R, for the Rankin scale).

If the neutron temperature is known – it is straightforward to determine the most probable energy, speed, wavelength and frequency of the neutrons by using the relationships given in Equation 22, Equation 23, Equation 24 and Equation 25.

Equation 22 - Mean energy of neutrons at function of temperature in Kelvin

$$E = kT$$

Equation 23 - Mean neutron speed at a temperature T

$$v = \left(\frac{2kT}{m} \right)^{1/2}$$

Equation 24 - De Broglie wavelength of neutrons as function of temperature in Kelvin

$$\lambda = h(2kT)^{-1/2}$$

Equation 25 - De Broglie frequency of neutrons as function of temperature in Kelvin

$$f = \frac{kT}{h}$$

Where:

k = Boltzmann constant = 8.6173×10^{-5} eV K⁻¹

h = Planck’s constant = 4.13566×10^{-15} eV s

Table 2 - Indicative table of equivalences between neutron descriptive values in eV, °K, Å and m/s

E (in eV)	Temp (in °K)	De Broglie λ (in Å)	Speed (in m/s)
0.0002	2	22.35	180
0.002	20	7.07	570
0.017	200	2.24	1,820
0.0253	293	1.85	2,200
0.026	300	1.83	2,230
0.035	400	1.58	2,570
0.086	1000	1.00	4,070
1	11,500	0.29	13,830
10	115,000	0.09	43,740
100	1,150,000	0.03	135,000
1 keV	11,500,000	0.01	427,000
10 keV	115,000,000	0.00	1,300,000
100 keV	1,150,000,000	0.00	4,200,000
1 MeV	11,500,000,000	0.00	13,000,000
2 MeV	23,000,000,000	0.00	19,000,000

Temperature equivalents for neutron energies of 1 eV and above have been greyed out in Table 2 as they are above a level in which it is physically meaningful to describe neutron energy in terms of temperature. Thermal flux equivalents have been highlighted.

The next important factor in considering neutron activation is the available neutron flux (neutron quantity). This aspect of neutron flux is normally described in units of numbers of neutrons per square centimetre per second ($n\text{ cm}^{-2}\text{ s}^{-1}$). Table 3 and Figure 14 provide information on a representative sampling of “high-flux” research reactors.

It is important to view published thermal flux levels with a substantial degree of caution. The thermal neutron flux in any large reactor facility varies over orders of magnitude depending on the location within the reactor core or its surrounds¹⁶. Even when discussing peak levels there is substantial variation between published values as some information sources refer to the “perturbed” values while others quote the “unperturbed” value. The unperturbed value is only ever an estimate. What is meant by “unperturbed” is the value that would be measured if there were no irradiation facilities or beam tubes or sources (hot or cold) in the reflector. The presence of these facilities greatly reduces the peak “perturbed” flux.

With that caveat, the flux-level of research reactors is important when discussing neutron activation because it is actual flux-level at the target position (as opposed to its notional value) that, when combined with the cross section for a particular reaction, determines the overall rate of the reaction.

Figure 14 – Published thermal and fast flux levels for a range of "high-flux" research reactors

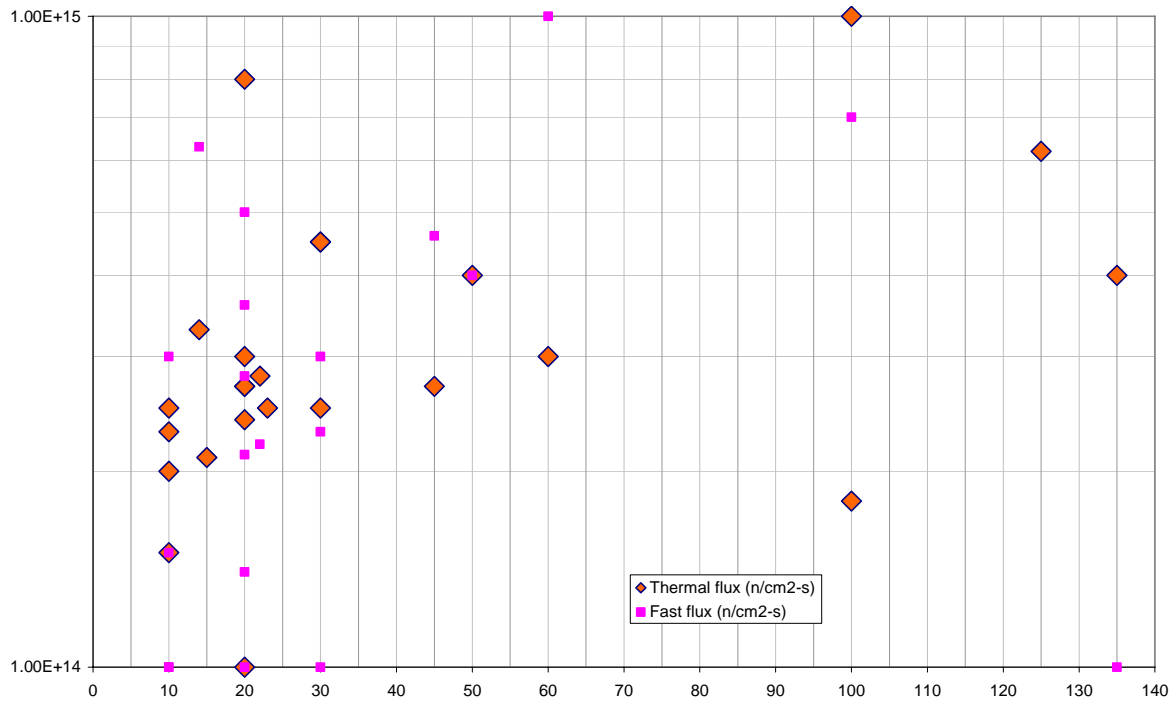


Table 3 - Published figures on “high flux” Research Reactor power and flux levels¹⁷

Country	Name	Thermal Power (MWth)	Thermal flux (n cm ⁻² s ⁻¹)	Fast flux (n cm ⁻² s ⁻¹)
Canada	NRU	135	4.00E+14	1.00E+14
China	HFETR	125	6.20E+14	1.70E+15
Belgium	BR-2	100	1.00E+15	7.00E+14
India	DHRUVA	100	1.80E+14	4.50E+13
Kazakhstan	EWG 1	60	3.00E+14	1.00E+15
Japan	JMTR	50	4.00E+14	4.00E+14
Netherlands	HFR	45	2.70E+14	4.60E+14
Indonesia	GA S MPR	30	2.50E+14	2.30E+14
Korea	HANARO	30	4.50E+14	3.00E+14
Poland	MARIA	30	4.50E+14	1.00E+14
Germany	FRJ-2 (DIDO)	23	2.50E+14	5.00E+13
Egypt	ETR-2	22	2.80E+14	2.20E+14
Australia	OPAL	20	3.00E+14	2.12E+14
Germany	FRM II	20	8.00E+14	5.00E+14
Japan	JRR-3M	20	2.70E+14	1.40E+14
Norway	HBWR	20	1.00E+14	1.00E+14
South Africa	SAFARI-1	20	2.40E+14	2.80E+14

Taiwan	TRR-II	20	2.70E+14	3.60E+14
Algeria,	ES-SALAM	15	2.10E+14	4.20E+12
Romania	TRIGA II	14	3.30E+14	6.30E+14
Czech Republic	LVR-15 REZ	10	1.50E+14	3.00E+14
Hungary	BUDAPEST R.R	10	2.50E+14	1.00E+14
Libya	IRT-1	10	2.00E+14	1.50E+14
Uzbekistan	WWR-CM	10	2.30E+14	1.00E+14

It can be seen in Figure 14 that reactors with the same notional thermal power level can have a very wide variation in the published steady-state level of available neutron flux. This is most obvious when examining the six different research reactors in Table 3 which have a notional power level of 20MWth and for which the published thermal flux levels vary from $1.0 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$ to $8.0 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$.

Large scale spallation sources, due to their high peak neutron flux levels, are a potential location for the undeclared production of polonium and tritium (and plutonium via the undeclared irradiation of depleted uranium). As these locations would not normally be declared as having nuclear material (except in the form of depleted uranium shielding) the IAEA would not normally have routine access to neutron sources of this type. However, for the reasons discussed here, these facilities would not usually be considered of proliferation significance.

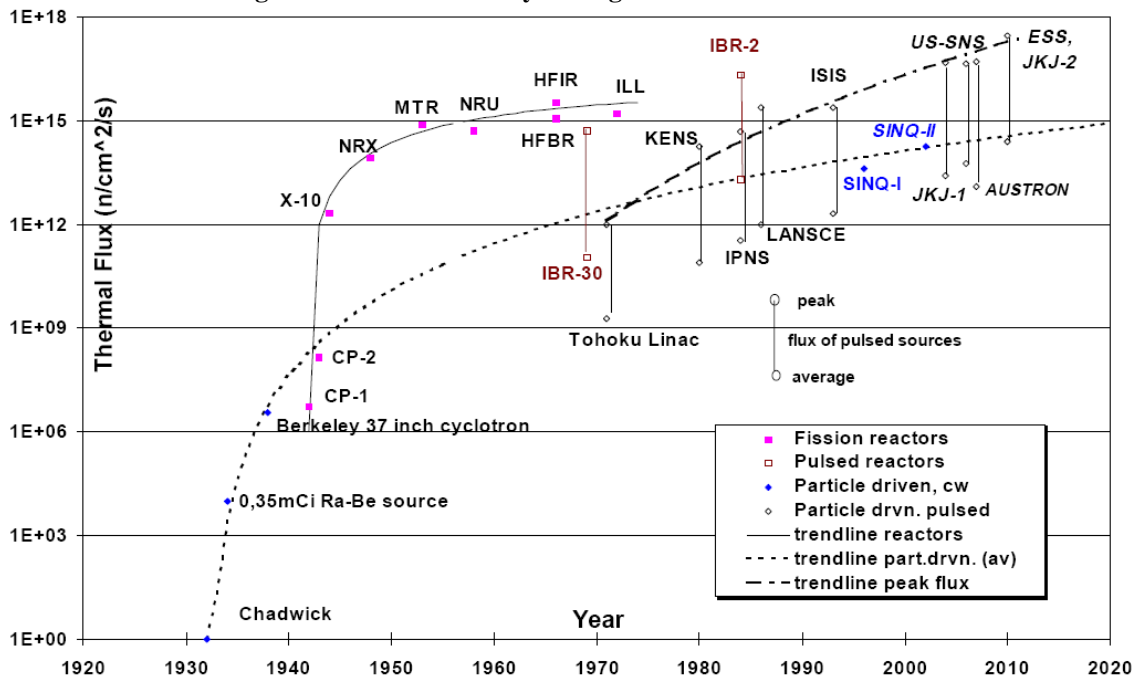
Accelerator driven sources (such as the spallation sources listed below) have considerable advantages over research reactors as high intensity sources of neutrons. Each U-235 fission gives rise to an average of ≈ 2.5 neutrons – 1 neutron is needed to sustain criticality – and, in a high-flux research reactor, ≈ 0.7 neutrons on average are lost to leakage, moderation and absorption and this leaves ≈ 0.8 neutrons on average per fission for irradiation. When operating a reactor as a neutron source it has to be taken into account that each U-235 fission also gives rise to ≈ 200 MeV of energy that must be dissipated by the reactor's cooling system. This leads to the consequence that, for every neutron available for irradiation, ≈ 250 MeV of heat must be dealt with in some way. For a particular reactor design and core configuration, any order of magnitude increase in neutron flux will result in an order of magnitude increase in heat generation.

Spallation sources generate neutrons which generally have energies in the range of 2-20 MeV. These energetic neutrons can be slowed down to thermal temperatures in a moderator system. A 1 GeV proton interacting with a heavy element produces an average of 30 thermal neutrons after moderation at an energy cost of 30 MeV per neutron. This gives spallation an 8-fold heat-loading advantage over reactors as a peak source of thermal neutrons (30 MeV/n vs. 250 MeV/n). Total integral flux levels for spallation sources are much lower than for research reactors and in practical terms a very large spallation source is probably comparable in its potential to produce Po-210 or tritium to a small or medium sized research reactor.

The key factor as regards the potential for misuse of a spallation source is the high level of international collaboration that is usual in the operation of these large-scale facilities. It would be very difficult to conceal abnormal experiments from other researchers involved with

the facility. Accordingly, the potential for misuse of spallation source facilities for proliferation purposes would usually be very low.

Figure 15 - Historic survey of "high flux" neutron sources¹⁸



Annex 3. REACTOR AVAILABILITY FACTORS

Published flux figures for research reactors are quoted in neutrons per centimetre squared per second, but extrapolating from per second figures to long term irradiations lasting days, months or years essentially assumes that the reactor operates with a 100% availability factor. In fact it is rare for reactor operators to aim for availability factors above 90%. High flux reactors are generally taken off line regularly to allow for maintenance and exchanges of fuel. Regular refuellings are necessary to maintain flux levels and ensure a flat flux distribution. Indicative operating cycles for large reactors are four weeks (24-25 days operation, 3-4 days shutdown) or five weeks (31-32 days operation, 3-4 days shutdown)¹⁶ – with extended shutdowns occurring annually or biannually.

Table 4 - Planned availability factors for various high-flux reactors

Country	Name	Thermal Power (MWth)	Planned Availability
Canada	NRU	135	66%
China	HFETR	125	7%
Belgium	BR-2	100	18%
India	DHRUVA	100	No data
Kazakhstan	EWG 1	60	No data
Japan	JMTR	50	49%
Netherlands	HFR	45	77%
Indonesia	GA S MPR	30	20%
Korea	HANARO	30	30%
Poland	MARIA	30	27%
Germany	FRJ-2 (DIDO)	23	54%
Egypt	ETRR-2	22	80%
Australia	OPAL	20	90%
Germany	FRM II	20	71%
Japan	JRR-3M	20	45%
Norway	HBWR	20	55%
South Africa	SAFARI-1	20	79%
Taiwan	TRR-II	20	No data
Algeria,	ES-SALAM	15	2%
Romania	TRIGA II	14	13%
Czech Republic	LVR-15 REZ	10	51%
Hungary	BUDAPEST R.R	10	40%
Libya	IRT-1	10	2%
Uzbekistan	WWR-CM	10	68%

Annex 4. TARGET ACTIVATION SOLUTIONS VIA SOFTWARE

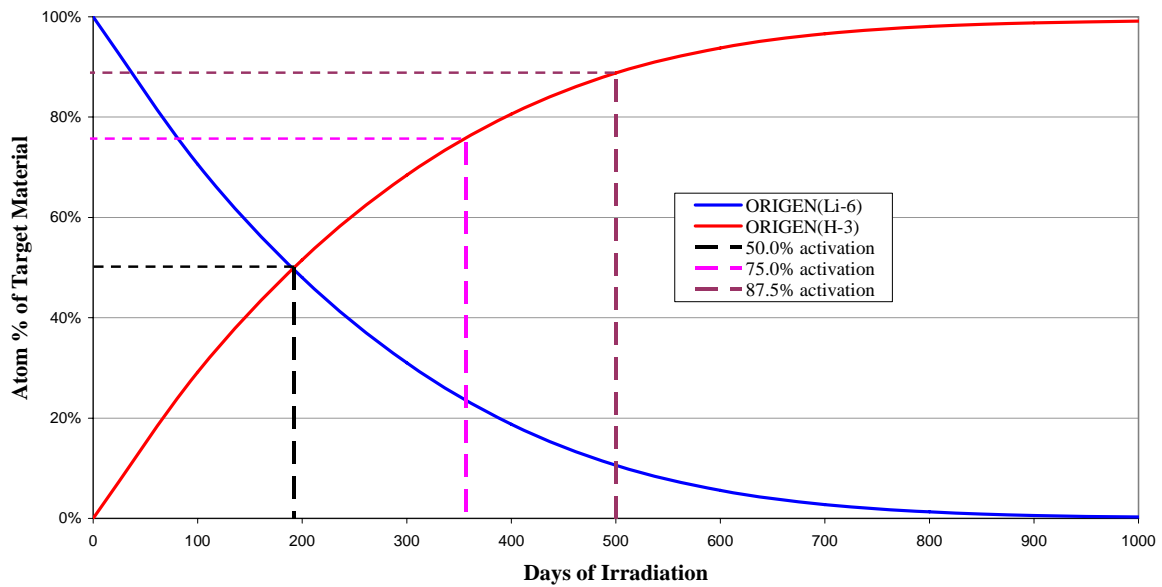
One of the most commonly used computer codes for the calculation of isotope generation, depletion and radioactive decay is known as ORIGEN (Oak Ridge Isotope Depletion and Generation Code) which is distributed by the Radiation Safety Information Computational Center (RSICC) at Oak Ridge National Laboratory, Oak Ridge, TN, USA. ORIGEN-type codes are designed for use in nuclear modelling applications to compute fuel depletion, actinide transmutation and decay, fission product build-up and decay, and radiation source terms for use in nuclear modelling applications.

In preparing this report the author made use of the target irradiation module of two different versions of ORIGEN¹⁹ for modelling the rate of production of Po-210 (via the reaction detailed in Equation 21). The two versions 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 using the Bateman equation methodology noted above.

After performing the relevant Bateman calculations (using Equation 19 above) and comparing the results to the ORIGEN output it became clear that the two versions of the program were using different values for the reaction cross section required by Equation 21. A literature search on this issue led to an interesting discussion of discrepancies in the tabulated values of the Bi-209 activation cross section by Letourneau et al²⁰. Letourneau notes that the ENDF²¹ figure for the reaction cross section of ≈ 24 mb is approximately 50% higher than the value of earlier determinations (the Letourneau paper argues that the real value for the relevant cross section is 17.9 mb).

While the difference between the two cross section values was sufficient to explain the discrepancy between the two versions of the ORIGEN program it provided no means of determining whether either value was valid for the irradiation case examined. Attempts to produce a valid comparison between values produced by ORIGEN and values produced using the Bateman methodology for irradiation of Bi-209 were inconclusive and so it was decided to perform equivalent calculations for a different irradiation case – the irradiation of Li-6 to produce tritium. The results of these calculations are plotted as Figure 16 below.

Figure 16 - ORIGEN generated data for irradiation of Li-6 in a thermal neutron flux of $1.25 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$



Even the most cursory examination of Figure 16 shows that it is virtually identical to Figure 8 which is a curve shape that should only arise if the product of decay or irradiation were stable. In the 1000 day period of irradiation modelled by ORIGEN there has been 0% decay of tritium as opposed to the ~15% that would have been expected.

Figure 17 - Decay corrected version of Figure 16

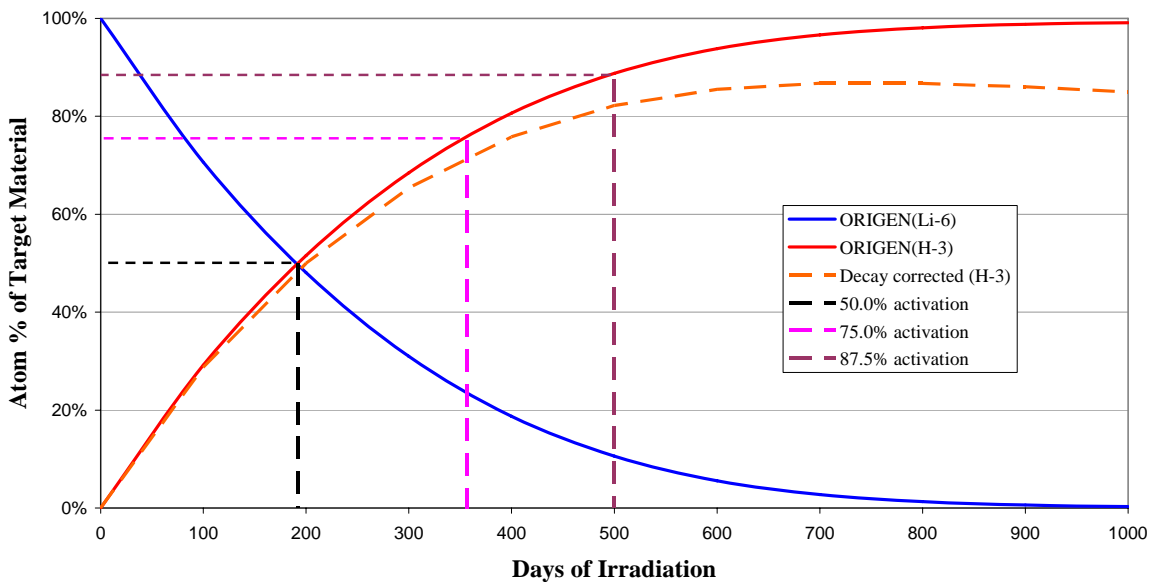


Figure 17 shows the expected amount of H-3 decay during the period of irradiation.

Another significant problem revealed by the ORIGEN plot is that the neutron flux has varied across the period of irradiation. In Figure 16, the period to reach 50% activation is 192 days, the period to reach 75% activation is only another 165 days, the period to reach 87.5% activation is only another 143 days and the period to reach 93.5% activation is shorter still at another 100 days. As shown in Figure 11, these intervals should be proportional to $\sigma_{\text{Li}} \phi$ and

each of the intervals should be equal to each other if the flux had been held constant across the period of irradiation. The flux during the first period was approximately 50% of the flux in the fourth period. In part, the problem arises because the versions of ORIGEN available to the author are intended for modelling power reactors and have not been validated for high-flux research reactors.

It is valid for ORIGEN to adjust the flux levels in the core across the period of irradiation.

For power reactors, the level of reactivity and the fissile material loading in the core changes continuously during operation. While it is common for research reactor operators to aim for stable free neutron flux levels, power reactor operators aim for stable specific reactor power levels.

The ORIGEN family of codes is well-documented and has been validated for different fuel types and reactor models by post irradiation examination. There are two likely explanations for the problematic results produced using ORIGEN in these irradiation cases, either:

- the author entered the inputs to ORIGEN code incorrectly; or
- the irradiation cases examined for this report were outside the area of validity of the ORIGEN versions and libraries used.

ORIGEN is a very useful tool-set for the irradiation cases it was designed to deal with, but care should be taken when applying any modelling tool to a novel problem. ORIGEN-ARP with standard libraries loaded is not the correct tool to use for the irradiation cases examined in this report.

Due to these difficulties in using ORIGEN for the irradiation cases of interest the calculations used in the main body of this paper were all done from first principles using the methodology in Annex 1

Annex 5. VARIATION IN CROSS SECTION VALUES FROM DIFFERING SOURCES

A variety of information sources were used for cross section data when preparing this report. The initial source was CRC Handbook of Chemistry and Physics 85th edition²² with cross-validation against the JEFF-3.1 Nuclear Data Library²³ and ENDF²⁴ via the IAEA's Nuclear Data Service. Figure 1 shows that there are two isomeric forms of Bi-210, a short lived ground-state (Bi-210) and a relatively long-lived metastable isomer (Bi-210m). Only a limited subset of the available data-sources indicated the branching ratio between Bi-210 and Bi-210m. In the case of the resonance integral, sources indicated a branching ratio of 50/50 while for thermal neutrons⁴, the branching ratio was given as approx. 70/30.

In the case of the production of the short-lived ground state of Bi-210 described in Figure 1, the quoted resonance integrals varied, between sources, by up to 10%. Resonance integrals values varied between 85-103 mb (out of a total resonance integral of 191-207 mb).

In the case of the thermal neutron cross section values the uncertainties were at rounding levels, values varied between 23-24 mb (out of a total activation cross section of 34 mb), but when considered in conjunction with the Latourneau²⁰ discussion it is clear that there are still uncertainties relating to the appropriate values to use for these cross sections.

In making calculations drawing upon these cross sections the underlying uncertainties in the cross section values needs to be factored into the overall uncertainty in the final result.

NOTES

Equations used in this report were produced using DS MathType 6.0

Figure 5 was taken from “*NTD Silicon Irradiation*”

http://www.ansto.gov.au/access/services/ntd_silicon_irradiation.html⁷

Figure 13 was obtained using the plot function from ENDF²¹ at

<http://www-nds.iaea.org/exfor/endl00.htm>

Figure 15 was taken from an article on “Spallation Neutron sources”¹⁸

All other figures, charts and tables used in this report were produced in MS Excel 2003 using data that was either:

- calculated by the author (Table 2, Table 1, Figure 8, Figure 9, Figure 10, Figure 11, Figure 12, Figure 2, Figure 3, Figure 4, Figure 6 and Figure 7)
- drawn from public databases (Table 3, Table 4, Figure 13, Figure 14, Figure 15 and Figure 1). or
- generated by use of the ORIGEN code (Figure 16 and Figure 17)

ENDNOTES – INCLUDING BIBLIOGRAPHY

1. 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).
2. Rhodes, R., “*The Making of the Atomic Bomb*”, Penguin Books, New York, 1986
3. See Table 3
4. See Table 2
5. Alfassi, Z.B., “*Activation Analysis*” CRC Press, Boca Raton, FL, 2005
6. Amos, P.E., and Kimnew, S., “*Silicon Irradiation Rig Design for Opal Reactor*”, International Group on Research Reactors (IGORR), IGORR 207, Lyon France
7. ANSTO, “*NTD Silicon Irradiation*” http://www.ansto.gov.au/access/services/ntd_silicon_irradiation.html, accessed 23 February 2008
8. Equation 4 and Equation 5 are the same relationship expressed in different terms.
9. Bateman, H., “*Solution of a System of Differential Equations Occurring in the Theory of Radio-active Transformations*,” Proc. Cambridge Phil. Soc. 15, 428, (1910).
10. Podgorsak, E.B., “*Radiation Physics Handbook for Medical Physicists*”, Springer, Germany, 2005
11. Lilley, J., “*Nuclear Physics*”, Wiley 2001
12. Two factors that are referred to as the “infinite multiplication factor” which is referred to as k_{∞} and the “effective multiplication factor” which is referred to k_{eff} and abbreviated to k_{eff} . Both k_{∞} and k_{eff} describe the level of criticality of the reactor core, k_{∞} is the ratio of the neutrons produced by fission in one generation to the number of neutrons lost through absorption in the preceding generation for an infinite reactor, while k_{eff} is a similar ratio for a finite reactor.
13. This plot has been obtained using the plot function from ENDF²¹ at <http://www-nds.iaea.org/exfor/endl00.htm>
14. “*DOE Fundamentals Handbook: Nuclear Physics and Reactor Theory*”, DOE-HDBK-1019/1-93, Washington, January 1993.
15. The character λ is commonly used (in different contexts) to refer to both the de Broglie wavelength and the decay constant. Every effort has been made to keep the two concepts separate within the report.
16. ANSTO 2008, Private Communication
17. Figures taken from IAEA database “Nuclear Research Reactors in the World” access date 10 February 2008 at <http://www.iaea.org/worldatom/rrdb/>
18. Figure from Cho, Y., “*Spallation Neutron Sources*”, www.akpa.org/article/ycho.pdf accessed 20 Feb 2008.
19. Calculations made use of ORIGEN-ARP included as part of the RSICC SCALE5 package and compared to the much older standalone version ORIGEN 2.0.
20. Letourneau et al “*Measurement of the ²¹⁰Po production induced by thermal neutron capture on ²⁰⁹Bi*” Annals of Nuclear Energy, 9 November 2005.

-
21. ENDF VII - Evaluated Nuclear Data Files, National Nuclear Data Centre, Brookhaven National Laboratory.
 22. Lide, D.R. ed., “*CRC Handbook of Chemistry and Physics*”, CD-ROM version 2005, CRC Press, Boca Raton, FL, 2005
 23. OECD/NEA, “*JEFF-3.1 Nuclear Data Library – JEFF report 21*”, Joint Evaluated Fission and Fusion (JEFF) Project, OECD/NEA No. 6190, Paris, 2006
 24. “*Evaluated Nuclear Data File (ENDF) Database Version of October 30, 2007*”, accessed via IAEA Nuclear Data Service, <http://www-nds.iaea.org/exfor/endl00.htm>,