Activation of $^{197}$Au and $^{209}$Bi in a fast spectrum sub-critical assembly composed of 500 kg natural uranium irradiated with 1 and 4 GeV deuterons

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Article history:
Received 21 May 2013
Received in revised form 10 September 2013
Accepted 11 September 2013
Available online 10 October 2013

Keywords:
Accelerator Driven System (ADS)
Uranium target-blanket
Gold activation
Bismuth activation
MCNPX
Gamma spectrometry

1. Introduction

An Accelerator Driven System (ADS) involves coupling an intermediate-energy (>150 MeV/nucleon) ion beam with a sub-critical nuclear assembly. High numbers of spallation neutrons produced by the interaction of the ion beam with a heavy metal target are used to sustain fission reactions within the assembly. Accelerator Driven Systems are proposed as a means for both power production and nuclear waste transmutation (Bowman, 1992; Carminati et al., 1993).

The upper energy limit of neutrons in an ADS is above the range that evaluated cross section data sets extend to. Monte Carlo physics models are thus relied upon to simulate the high energy region of the neutron spectrum. It is important to experimentally validate these models as high energy particles, especially neutrons all contribute to reactions and neutron balance within the sub-critical assembly.

In this work, $(n,xn)$ threshold reactions in gold and bismuth samples were used to experimentally probe the high energy region of the neutron spectrum in the Quinta assembly (see Section 2.1 for details), while the $^{197}$Au$(n,\gamma)$ reaction was used for the detection of slow neutrons. Simulations using MCNPX with the INCL4-INCL4 cascade and ABLA fission/evaporation models were compared with the experimentally measured data. This was to assess the ability of the code to correctly model neutron production and transport of spallation neutrons within a fast Accelerator Driven System. Liquid lead–bismuth eutectic (Fazio et al., 2008; Gromov et al., 1997) and lead-gold eutectic (Medarde et al., 2011) have also been proposed as possible target materials for future ADS designs and so these activation studies may prove useful.

2. Experimental

2.1. The Quinta assembly

The Quinta assembly, located at the Joint Institute for Nuclear Research (JINR), Dubna, Russia, consists of a total of 512 kg of natural uranium. It is composed of five sections, each being 114 mm long and separated by a 17 mm air gap which allows the placement of samples mounted onto special plates (refer to Fig. 1(a) and (b)). The uranium exists as many cylindrical rods, where each rod is 36 mm in diameter, 104 mm in length and 1.72 kg in mass. These
dimensions are inclusive of a 1 mm thick impervious outer aluminium casing which is present for safety reasons. Excluding the first section, 61 rods are arranged in a hexagonal lattice with a pitch size of 36 mm and enclosed in a hexagonal aluminium container with a wall thickness of 5 mm. The first section contains only 54 rods and the removal of the central 7 rods is to create a beam window. This beam window is 80 mm in diameter and serves to reduce the loss of backward emitted/scattered neutrons. The front and back of each section are then bounded by additional aluminium plates 350 × 350 × 5 mm.

The entire uranium target-blanket reaches 638 mm in length and a mass of 538 kg including all construction materials. The five sections are mounted onto a single slab of aluminium with thickness 25 mm and surrounded by lead bricks 100 mm thick on all six sides. This serves as a neutron reflector and to some extent as a biological shielding for γ-rays. The top section of the lead is further supported by 16 mm of aluminium. The front of the lead castle has a square window 150 × 150 mm. Sample plates can be inserted and removed quickly into the air gaps between the sections, as well as on the front and back of the target by the presence of slots and lids located on the roof of the lead castle. The sample plates are labelled 0–5, starting from the direction of the incident beam (as shown in Fig. 1(b)) and will be referred to accordingly throughout this text.

2.2. Irradiation details

The Quinta target was irradiated with a pulsed deuteron beam of energies 1 and 4 GeV extracted from the Nuclotron accelerator, also located at the JINR. The two irradiation periods lasted approximately 21 and 17.5 h for the 1 and 4 GeV irradiations, respectively. Further details of the irradiation may be seen in Table 1. The distribution of deuterons as extracted from the Nuclotron accelerator is shown in Fig. 2. The beam position and beam shape was determined using two separate techniques. The first involved measuring the yield of reaction products produced by beam induced 27Al(d,f)24Na reactions. The complete experimental methodology and results detailing the determination of the deuteron beam fluence, beam position and shape has been published previously (Furman et al., 2012). Nevertheless, for completeness, a brief summary from Furman et al. (2012) is provided in the following paragraph.

Three independent aluminium foil monitors were placed between the beam output and the front of the Quinta target. The three monitors were placed between 2 and 3 m away from the Quinta target to avoid backscattered neutrons contaminating the measurement. Only three experimental values for the 27Al(d,x)24Na cross section in the GeV range (at 2.33 (Banaigs et al., 1971), 6.0 and 7.3 GeV (Kozma and Yanovsky, 1990)) are known. Therefore, the cross section at 1 and 4 GeV energies were estimated from interpolating the curve fitted to experimental data in the energy range of 0.1–7.3 GeV. The 27Al(d,x)24Na cross section was found to be 16.4 ± 1.6 mb and 14.5 ± 1.5 mb for 1 and 4 GeV deuterons, respectively. The three foils were each analysed independently by different groups and good agreement between all three measurements was reached. The weighted mean value of the total number of deuterons on the Quinta target was found to be (1.50 ± 0.16) × 1013 and (1.94 ± 0.20) × 1013 for the 1 and 4 GeV irradiations, respectively.

The beam position and beam shape was determined using two separate techniques. The first involved measuring the yield of reaction products produced by beam induced reactions on natural copper foils (method described in Svoboda (2011)). The other method utilised an array of fission track detectors measuring beam induced natPb(d,f) reactions (Zhuk et al., 2008). Both the copper activation foils and fission track detectors were placed directly in front of the Quinta target on the surface of the lead castle. The beam shape was fitted to a Gaussian and the resulting FWHM in the vertical and horizontal directions, as well as the positioning of the beam centre is presented in Table 1.

2.3. Activation analysis of samples

A total of five bismuth and three gold foils (purity > 99.99%) were placed inside the Quinta target for each of 1 GeV and 4 GeV irradiations. Each of the foils were sandwiched between two pieces of ~100 µm thick muscovite mica which were used as SSNTD’s for fission rate measurement (these results are not discussed in the present paper).

Table 1
<table>
<thead>
<tr>
<th>Incident ion</th>
<th>Deuteron</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ion energy</td>
<td>1 GeV</td>
</tr>
<tr>
<td>Irradiation duration</td>
<td>21 h 7 min</td>
</tr>
<tr>
<td>Deuterons on target</td>
<td>2.6 ± 0.3</td>
</tr>
<tr>
<td>FWHM &lt;i&gt;x&lt;/i&gt; (cm)</td>
<td>3.5 ± 0.3</td>
</tr>
<tr>
<td>&lt;i&gt;x&lt;/i&gt; coordinate of the beam centre on the &lt;i&gt;x&lt;/i&gt;-&lt;i&gt;y&lt;/i&gt; plane.</td>
<td></td>
</tr>
</tbody>
</table>
The samples were mounted onto the back of 2 mm thick aluminium plates and inserted into gaps labelled 1, 2, 3, 4 and 5 (Fig. 1(b)). The gold samples were only placed in gaps 1, 2 and 3. For the 1 GeV irradiation, the samples were positioned –4 cm from the target axis. However, for the 4 GeV irradiation, to prevent over-exposing the mica (in contact with sample foils) with fission tracks (this is independent of the activation measurements), the samples were lowered to –6 cm. The positioning of the samples on the plates is presented in Table 2.

After irradiation, the samples were removed and transported away to be analysed with gamma spectrometry. All 16 samples were measured using a HPGe n-type coaxial (model: GR-1819) detector, manufactured by CANBERRA Industries with relative efficiency 18% (at 1.33 MeV), as determined by the manufacturer. Measurements began 1.5 h after irradiation had stopped, continuing for up to 6 days afterwards. The spectra collection times ranged from 15 min to just over 3 h. All spectra were analysed using the HYPERMET-PC software (Fazekas et al., 1997).

### 2.3.1. Reaction rate determination

The isotopes detected in the bismuth and gold samples are listed in Table 3. All of these isotopes were detected in at least one of the measured samples with the exception of 195Au. The half life of 195Au is simply too long (186 days) for there to be sufficient build up of activity in the relatively short irradiation time of this experiment.

The production rate, $R_{expt}$ of each of the selected isotopes (per target atom per incident deuteron) was determined using the following equation:

$$ R_{expt} = \frac{N_C}{L_{expt}} \frac{B}{SD} \frac{M_{m}}{\sigma_m N_A} t_{irr} \Phi $$

where $N_C$ is the count rate of peak of interest; $C$ the correction for decay during counting time (see Eq. (2)); $I$, the emission probability of measured $\gamma$-ray; $\sigma_p$ the detector full-energy peak efficiency; $T$ the correction due to True Coincidence Summing (TCS); $B$ the correction due to de-activated nuclei; $m$ mass of activation foil; $\Phi$ the isotopic fraction of activation foil; $M_m$ the molar mass of activation foil; $\tau$ the deuteron irradiation time; $\Phi$ the total number of incident deuterons.

### Table 2

<table>
<thead>
<tr>
<th>Sample</th>
<th>1 GeV</th>
<th>4 GeV</th>
</tr>
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<tr>
<td></td>
<td>x</td>
<td>y</td>
</tr>
<tr>
<td>1</td>
<td>0</td>
<td>–4</td>
</tr>
<tr>
<td>2</td>
<td>0</td>
<td>–4</td>
</tr>
<tr>
<td>3</td>
<td>0</td>
<td>–4</td>
</tr>
<tr>
<td>4</td>
<td>0</td>
<td>–4</td>
</tr>
<tr>
<td>5</td>
<td>0</td>
<td>–4</td>
</tr>
</tbody>
</table>

*a See Section 4.3 for explanation.*

### Table 3

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Product</th>
<th>$E_n$</th>
<th>$E(\sigma_{max})$</th>
<th>Half-life</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{209}$Bi(n,4n)</td>
<td>$^{205}$Bi</td>
<td>22.6</td>
<td>34</td>
<td>6.24 d</td>
</tr>
<tr>
<td>$^{209}$Bi(n,5n)</td>
<td>$^{207}$Bi</td>
<td>29.6</td>
<td>44</td>
<td>15.3 d</td>
</tr>
<tr>
<td>$^{209}$Bi(n,6n)</td>
<td>$^{206}$Bi</td>
<td>38.1</td>
<td>55</td>
<td>11.2 h</td>
</tr>
<tr>
<td>$^{209}$Bi(n,7n)</td>
<td>$^{207}$Bi</td>
<td>45.4</td>
<td>66</td>
<td>11.8 h</td>
</tr>
<tr>
<td>$^{197}$Au(n,γ)</td>
<td>$^{198}$Au</td>
<td>0</td>
<td>2</td>
<td>2.69 d</td>
</tr>
<tr>
<td>$^{197}$Au(n,2n)</td>
<td>$^{195}$Au</td>
<td>8.11</td>
<td>15</td>
<td>6.17 d</td>
</tr>
<tr>
<td>$^{197}$Au(n,3n)</td>
<td>$^{194}$Au</td>
<td>14.8</td>
<td>24</td>
<td>186 d</td>
</tr>
<tr>
<td>$^{197}$Au(n,4n)</td>
<td>$^{192}$Au</td>
<td>23.2</td>
<td>34</td>
<td>38.0 h</td>
</tr>
<tr>
<td>$^{197}$Au(n,5n)</td>
<td>$^{191}$Au</td>
<td>30.2</td>
<td>44</td>
<td>17.7 h</td>
</tr>
<tr>
<td>$^{197}$Au(n,6n)</td>
<td>$^{190}$Au</td>
<td>38.9</td>
<td>54</td>
<td>4.94 h</td>
</tr>
<tr>
<td>$^{197}$Au(n,7n)</td>
<td>$^{189}$Au</td>
<td>46.0</td>
<td>65</td>
<td>3.18 h</td>
</tr>
</tbody>
</table>

Due to pulsed beam (see Eq. (3)); $S$ the correction due to activity saturation (see Eq. (4)); $D$ the correction for decay between end of irradiation and beginning of counting (see Eq. (5)); $M_m$ the molar mass of activation foil; $\Phi$ the isotopic fraction of activation foil; $m$ the mass of activation foil (in grams); $N_A$ Avogadro’s number; $t_{irr}$ the deuteron irradiation time; $\Phi$ the total number of incident deuterons.

$$ C = \frac{t_{irr}}{1 - e^{-t_{irr}/\lambda}} $$

$$ B = \frac{t_{irr}(\sum W e^{-t_{irr}/\lambda})}{1 - e^{-t_{irr}/\lambda}} $$

$$ S = 1 - e^{-t_{irr}/\lambda} $$

$$ D = e^{-t_{irr}/\lambda} $$

where $\lambda$ is the decay constant of produced nuclide; $t_r$ the real-time of measurement interval; $W$ the ratio of number of deuterons in pulse to total number of deuterons; $t_0$ the time from given deuteron pulse to end of irradiation period; $t_i$ the time between end of irradiation and beginning of counting.

The $^{197}$Au(n,2n) reaction produces two isomers of $^{196}$Au. If spectrometry measurements take place prior to the complete decay of $^{196}$Au (half life 9.6 h), then this must be corrected for (see Soete et al. (1972) for details).

The method used in this work does not take into consideration the depletion of the target material, nor possible activation of the produced parent nuclide. The depletion-activation model introduced by Abdel-Rahman and Podgorsak (2005) can be used to correctly account for this. However, due to the low specific activities produced and the activation factor, $m = \sigma_0 \lambda \ll 10^{-3}$ the saturation model used here is expected to be valid.
2.3.2. Determination of detector efficiency

The total and full-energy peak efficiency of the HPGe detector was determined through both Monte Carlo and experimental means. Eleven sources of verified activity, including \( ^{241}\text{Am}, \ ^{133}\text{Ba}, \ ^{139}\text{Ce}, \ ^{57}\text{Co}, \ ^{137}\text{Cs}, \ ^{54}\text{Mn}, \ ^{113}\text{Sn}, \ ^{228}\text{Th} \) and \( ^{88}\text{Y} \) were used. Four of these sources \( ^{241}\text{Am}, \ ^{139}\text{Ce}, \ ^{137}\text{Cs} \) and \( ^{54}\text{Mn} \) are single line photon emitters and hence appropriate for total efficiency calibration. It is however necessary to first subtract the low-energy components of the spectrum caused by the emission of X-rays originating from electron capture or internal conversion (Debertin and Helmer, 1988). \(^{60}\text{Co} \) emits two photons close in energy \( =1332 \text{ keV} \) and so total efficiency may be determined for the mean of these energies. In the case of \(^{88}\text{Y} \), two photons are emitted that are far apart in energy \( (E_1 = 898, \ E_2 = 1836 \text{ keV}) \). The total efficiency at 898 keV was found by interpolating the already measured data from the sources mentioned above which could then be used to calculate the total efficiency at 1836 keV (Debertin and Helmer, 1988).

To verify and interpolate the experimentally measured full-energy peak and total efficiency, Monte Carlo simulations of the detector were performed (Ewa et al., 2001; Hardy et al., 2002; Helmer et al., 2003, 2004). Quality assurance data sheets and technical drawings of the detector dimensions provided by the manufacturer were used as a basis to model the detector geometry in the simulation. However, slight modifications of the manufacturers specifications were required to achieve better agreement with the experimentally measured data (Hardy et al., 2002; Helmer et al., 2003). In the simulations, it is important to include all materials surrounding the detector, such as the presence of lead shielding.

The effects of random summing (pile-up) may ordinarily be corrected for using pile-up rejection electronics. However, as none were utilised here, it was necessary to correct for it mathematically (Gilmore, 2008). True Coincident Summing (TCS) corrections were calculated using the “TrueCoinc” (Sudár, 2008) program which utilises decay scheme data of the ENSDF format (NNDC, 2012).

The experimental and calculated total and full-energy peak efficiency of the GR-1819 CANBERRA detector are shown in Fig. 3. Measurements were performed at three distances from the detector cap (1.0, 3.5 and 13.2 cm). All samples irradiated in the Quinta setup had relatively low activities, therefore to ensure adequate counting statistics, they were all measured at the 1.0 cm position. Reasonable agreement between the experimentally measured and simulated efficiencies was reached. However, a minor systematic difference between the experimentally measured and simulated values at the 1.0 cm position is noted, and this was incorporated into the uncertainty of the Quinta sample measurements (Section 4.4).

The calibration sources used for the efficiency measurements were all effective point sources. This does not reflect the geometry of the samples measured which were square foils (1 cm²) with thicknesses 50 μm and 250 μm for the gold and bismuth foils, respectively. Gold and bismuth are both dense, high Z material which cause significant scattering of photons within the sample itself. To correct for all of these, further Monte Carlo simulations with the specific geometry of the gold and bismuth foils were performed.

3. Monte Carlo simulations

The irradiation of the Quinta setup was simulated using MCNPX 2.7 (Pelowitz, 2011). The entire geometry of the Quinta assembly (Fig. 1) was programmed into the input file. The model was “irradiated” with 1 GeV and 4 GeV deuterons, with a 2° angle of deviation from the target axis on the xz-plane. The experimentally measured beam position and Gaussian shape (Table 1) was emulated in the simulation. Deuterons, neutrons, protons, pions (charged and neutral), muons and photons were all transported. Electrons were not transported as they have negligible effect on the overall results and only serve to slow the simulation down significantly (Hashemi-Nezhad et al., 2008). MCNPX models particle reactions and transport using a combination of cross section data sets and model physics. All transport cross section sets used here were from the ENDF/B-VII.1 libraries (Chadwick et al., 2011) and the INCL4-ABLA physics model utilised. The INCL4 intra-nuclear cascade model (Boudard et al., 2002) and the ABLA fission-evaporation model (Junghans et al., 1998) were chosen here to provide consistency with previous calculations performed on the Energy plus Transmutation setup which consists of lead target-uranium blanket (see e.g. Adam et al., 2011; Borger et al., 2011, 2013; Krása et al., 2010). As an example, the particle spectra for neutron, proton, pion (π”), deuteron and photon, in position of sample 2 is shown in Fig. 4.

3.1. Cross sections

To calculate the production rate of the selected products (Table 3) it is necessary to include reactions induced by neutrons, protons and deuterons. Cross section data from TENDL-2009 (Koning and Roehman, 2009) evaluation was used for \(^{209}\text{Bi}(n,4n)\) Bi reaction. As no evaluated data was available for \(^{209}\text{Bi}(n,xn)\), \( x = 5, 6, 7 \) reactions, these were calculated using the TALYS 1.4 code (Koning et al., 2008) and compared to experimental data obtained from Kim et al. (1998), Svoboda et al. (2011) and Vrzalová et al. (2013). \(^{209}\text{Bi}(p,x)\) data was obtained from experimental measurements made by Bell and Skarsgard (1956) and Michel et al. (2002). \(^{209}\text{Bi}(d,x)\) reaction data was obtained from Gonchar et al. (1994) and Somolová et al. (2010). TALYS was not used to calculate proton and deuteron induced cross sections because in our calculations, there were large unexplained discrepancies with the experimentally measured cross sections.

The cross section for \(^{197}\text{Au}(n,\gamma)\) Au was from the ENDF/B-VII library, whilst cross sections for \(^{197}\text{Au}(n,2n)\) Au, \(^{197}\text{Au}(p,d)\) Au, \(^{197}\text{Au}(p,\alpha)\) Au and \(^{197}\text{Au}(n,4n)\) Au were from TENDL-2009. \(^{197}\text{Au}(n,xn)\), \( x = 5, 6, 7 \) cross sections were all calculated using the TALYS 1.4 code. Proton cross section data was available for \(^{197}\text{Au}(p,x)\) reactions only (Kavanagh and Bell, 1961; Michel et al., 1997; Szlecesenyi et al., 1997). No deuteron-induced reaction cross section data for reactions of interest was found in the EXFOR database or open literature.
The ENDF/B-VII \((n,\gamma)\) cross section extends up to only 30 MeV, while neutron energies in Quinta can extend all the way up to about half that of the incident deuteron energy (Cugnon et al., 1997). The \(^{197}\text{Au}(n,\gamma)\) reaction cross section is dominated in the resonance region for neutrons with energies less than 0.01 MeV. The \((n,\gamma)\) cross section extends up to only 30 MeV, while neutron energies in Quinta can extend all the way up to 200 and 250 MeV, respectively. Above this range, the cross sections were assumed to remain constant.

### 3.2. Calculating reaction rates

The theoretical reaction rates, \(R_{\text{calc}}\), were calculated by folding the particle spectrum with the corresponding reaction cross section in the following way:

\[
R_{\text{calc}} = \int_{E_\text{th}}^{E_\text{max}} \sigma(E) \phi(E) \, dE
\]  

Here, \(\phi(E)\) is the flux (in units of neutrons cm\(^{-2}\) MeV\(^{-1}\) incident deuteron \(^{-1}\)), \(\sigma(E)\) is the energy dependent cross section and \(E_{\text{th}}\) is the threshold energy of the reaction. For reactions that have evaluated cross section data, this equation can be solved automatically in MCNPX by combining an F4 tally with an FM tally multiplier. For cross sections derived from TALYS based calculation or measured experimental data from the literature, Eq. (6) was determined by inputting the cross section data into MCNPX as a user supplied dose function with the DE and DF cards.

Due to the lack of cross section data for \(^{209}\text{Bi}\) and \(^{197}\text{Au}\), the \((n,4n)\) data is from the TENDL-2009 evaluation, while the others are calculated using the TALYS code.

### Table 4

Proportion of total \(^{206}\text{Bi}\), \(^{207}\text{Bi}\) and \(^{209}\text{Bi}\) produced due to neutron, proton and deuteron induced reactions in the 1 and 4 GeV irradiations as calculated with MCNPX.

In the case of the 4 GeV irradiation, the contribution of deuteron induced reactions is negligible. Refer to Table 2 for sample coordinates.

<table>
<thead>
<tr>
<th>Product</th>
<th>Sample</th>
<th>(1) GeV (% reactions)</th>
<th>(4) GeV (% reactions)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Neutrons</td>
<td>Protons</td>
<td>Deuterons</td>
</tr>
<tr>
<td>(^{206}\text{Bi})</td>
<td>1</td>
<td>81</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>92</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>90</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>90</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>91</td>
<td>4</td>
</tr>
<tr>
<td>(^{207}\text{Bi})</td>
<td>1</td>
<td>67</td>
<td>11</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>83</td>
<td>11</td>
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</tr>
<tr>
<td></td>
<td>5</td>
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<td>9</td>
</tr>
<tr>
<td>(^{209}\text{Bi})</td>
<td>1</td>
<td>62</td>
<td>16</td>
</tr>
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<td>2</td>
<td>78</td>
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<tr>
<td>(^{207}\text{Bi})</td>
<td>1</td>
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<tr>
<td></td>
<td>5</td>
<td>80</td>
<td>14</td>
</tr>
</tbody>
</table>

### Fig. 4

Particle spectra including neutron, proton, pion, photon and deuteron in Quinta assembly in location of sample 2 (see Table 2 for precise sample positioning) as calculated with INCL4-ABLA model. Equal logarithmic energy binning with 20 intervals per decade is used.

### Fig. 5

\((n,xn)\) reaction cross sections for \(^{206}\text{Bi}\) and \(^{197}\text{Au}\). The \((n,4n)\) data is from the TENDL-2009 evaluation, while the others are calculated using the TALYS code.

\[
R_{\text{calc}} = \int_{E_\text{th}}^{E_\text{max}} \sigma(E) \phi(E) \, dE
\]
4. Results and discussion

4.1. $^{209}$Bi activation

The experimental and calculated data for the reaction rates of the $^{209}$Bi samples for both 1 and 4 GeV deuteron irradiations of the Quinta assembly are presented in Fig. 6. The breakdown of neutron, proton and deuteron-induced reactions is presented in Table 4.

In general, good agreement between experimental and calculated values is achieved within the limits of measurement uncertainties. The exception is sample 1 of the 1 GeV irradiation, where the experimental value is consistently higher than the calculated value. The $R_{exp}/R_{calc}$ values are 1.6, 2.0 and 3.2 for $^{209}$Bi, $^{208}$Bi and $^{207}$Bi production, respectively. The 1 GeV samples were placed much closer to the centre of the incident deuteron beam than the 4 GeV samples. The FWHM of the 1 GeV beam is also more than twice the size of the FWHM of the 4 GeV beam (Table 1). As such, the neutron spectrum in 1 GeV sample location is much harder than for the 4 GeV (Fig. 4). The threshold energies for all four bismuth reactions are above 20 MeV (Table 3). These high energy neutrons arise from the intra-nuclear cascade stage of the nuclear disintegration of the incident beam deuterons (Oppenheimer, 1935) and are emitted with a strong forward direction. From Table 4 it is clear that the percentage of deuteron-induced reactions in the first sample is much larger than for any of the other samples. However, inclusion of this contribution is still not enough to make a good agreement. Therefore, it is likely the discrepancy in the first sample is due to an error in the beam centre location on the target and/or positioning of samples on plate 1 for the 1 GeV irradiation. The validity of this claim was investigated further and is discussed in Section 4.3.

4.2. $^{197}$Au activation

The gold activation results (Fig. 7) produced a more mixed agreement between experimental and calculated reaction rates. As with the bismuth results, sample 1 of the 1 GeV irradiation reveals an experimental value higher than the calculated value, but only for the high energy threshold reactions (>20 MeV). The $R_{exp}/R_{calc}$ values are 1.9, 2.2, 2.0 and 1.5 for $^{194}$Au, $^{193}$Au, $^{192}$Au and $^{191}$Au production, respectively. This is discussed further in Section 4.3. The lower energy threshold reactions, (n,γ) and (n,2n) do not show this discrepancy. Neutrons with energies <20 MeV originate from low energy pre-equilibrium emission, evaporation, (n,xn) and fission reactions, majority of which are emitted isotropically (Filges and Goldenbaum, 2010). These lower energy reactions would therefore be less affected by small variations in positioning of the samples and/or incident beam.

The calculated value of the (n,γ) reaction rate is systematically about 20% lower than the experimentally determined value and in all samples outside of the measurement uncertainties. Other reaction channels that lead to the production of $^{198}$Au include $^{197}$Au(d,p)$^{198}$Au. This reaction has relatively low cross section (Koning and Rochman, 2009) and is directly dependent on the deuteron flux in the sample positions. The $^{197}$Au(d,p)$^{198}$Au reaction channel contribution was therefore deemed negligible for both 1 and 4 GeV irradiations.

Calculations performed by Hashemi-Nezhad et al. (2011) of a 1 GeV proton beam on a uranium target has shown that the CEM03 model (Mashnik et al., 2006) has a total neutron yield 10% higher than calculations using the INCL4-ABLA models. Calculations performed in this work for the 1 GeV deuteron beam incident on the Quinta target-assembly have revealed the total neutron creation is 1.2% higher for the CEM03 model compared

Fig. 6. The experimental and Monte Carlo calculated axial distribution of $^{209}$Bi activation in the Quinta assembly under 1 and 4 GeV deuteron irradiation. Production rate of $^{209}$Bi, $^{208}$Bi, $^{207}$Bi and $^{206}$Bi due to neutron, proton and deuteron-induced reactions is shown.
to the INCL4-ABLA models. A comparison of the neutron spectra as calculated with CEM03 and INCL4-ABLA in the position of sample 2 for the 1 GeV deuteron irradiation is shown in Fig. 8. It is immediately noticeable that INCL4-ABLA produces 25% more neutrons above 20 MeV, despite the total number of neutrons produced being 1.2% less than CEM03. Therefore, CEM03 must produce more neutrons than INCL4-ABLA for neutron energies less than 20 MeV. Fig. 8 also contains an overlay of the relevant part of the $^{197}$Au($n,\alpha$) cross sections showing that the reaction is most sensitive to neutrons less than 20 MeV. Calculations using the CEM03 model have shown it leads to an increased $^{197}$Au($n,\alpha$) reaction rate of 5–10%. This is enough to achieve better agreement with the experimental results. The variations between each of the physics models including total neutron yield and neutron spectra serves to highlight the importance of experimentally validating each of the physics models.

Finally, there is a peculiarly large disagreement between the experimental and calculated result for the $^{192}$Au production rate. The experimental result is roughly two times higher than the calculation. This discrepancy is unlikely to be due to an error in the cross section calculation because for there to be agreement, the $^{197}$Au$(n,6n)$ cross section would need to be larger than the $(n,4n)$ cross section and also deviate significantly from the $^{209}$Bi$(n,6n)$ cross section (Fig. 5). This is considered illogical. Another considered possibility was the presence of $^{192}$Ir which emits the same signature $\gamma$-rays upon $\beta^-$ decaying. However, the relatively long half life of $^{192}$Ir (74 days) would not allow significant activity build up in the short irradiation times experienced here. Additionally, multiple spectra of each sample were taken over a period of time, and peaks attributed to $^{192}$Au were absent from all but the first spectra of each sample, which implies it must be short lived. The possibility of any “interfering nuclear reactions” (Soete et al., 1972) is
being considered and the cause of this large discrepancy is still not known and is currently under investigation.

4.3. Effect of sample/beam positioning on reaction rates

In order to investigate the effect of beam position and/or sample placement on the reaction rates of sample 1 in the 1 GeV irradiation, further Monte Carlo simulations were carried out. Two methods were tested, firstly, the beam was shifted down by up to 1.0 cm and secondly, the sample was shifted up by up to 1.0 cm. If the elevated experimental reaction rate in sample 1 of the 1 GeV irradiation is caused by an error in the beam position and/or sample placement it is unlikely to be more than 1 cm.

Analysis of the results suggested that the discrepancy is most likely due to a ~0.5 cm vertical displacement of the sample. This probably occurred when mounting the sample onto the plate and/or when lowering the plate into the slot between the Quinta sections. Error in the positioning of the beam was deemed not to be the cause as this led to further inconsistencies of the reaction rates of the other four samples. Samples placed closer to the incident beam would experience a lot more direct deuteron-induced reactions and an increased high energy neutron flux.

4.4. Uncertainties and possible sources of error

Sources of uncertainty in the experimental data arise from the 11% uncertainty in the measured number of incident deuterons. Statistical uncertainties from counting statistics ranged between 1% and 10% and in rare cases was as high as 20%, depending on the reaction and sample involved. An additional 6% is attributed to the full-energy peak efficiency, TCS and the emission probability of detected γ-ray.

Two sources of uncertainty were considered for the Monte Carlo results. The statistical uncertainties of the simulations were kept below 5% in all cases. Uncertainties in the cross sections were estimated from the experimentally measured data where possible. The precision of the calculated reaction rates is therefore limited by the uncertainties of the experimentally measured cross sections, which unfortunately in some cases were quite large.

5. Conclusion

The activation of gold and bismuth was studied in the 512 kg natural uranium target-blanket subcritical Quinta assembly irradiated with 1 and 4 GeV deuterons. Cross sections for neutron, proton and deuteron induced reactions were retrieved from the literature where available, and/or in the case of neutron-induced reactions were calculated with the TALYS code. Simulations with MCNPX 2.7 utilising the INCL4-ABLA physics model option revealed good agreement with the experimentally measured reaction rates of the bismuth samples. However, comparisons between experimental and Monte Carlo for the gold sample revealed more mixed results. The 197Au(n,γ) reaction used for detection of slow neutrons showed better agreement with the CEM03 model than INCL4-ABLA. It was also found that small variations in sample positioning relative to the incident deuteron beam can have dramatic effect on reactions with high threshold energies (>20 MeV). Reactions occurring with lower energy thresholds (<20 MeV) are not affected as much.

The observed agreement between the experimental and simulated results in this study has indicated potential of the ability for the INCL4-ABLA models to successfully model the production and transport of neutrons up to about 100 MeV in fast Accelerator Driven Systems irradiated with 1 and 4 GeV deuterons. Additionally, verification of the 197Au and 209Bi activation cross sections used here is also provided. However, the lack of experimental samples, especially in the radial direction, limits this conclusion to only the axial direction at this stage. Further experimental studies will be required to provide more conclusive evidence of the ability of INCL4-ABLA to model the neutronics of a fast ADS in general.

Acknowledgments

The author’s would like to thank the Joint Institute of Nuclear Research, particularly the Veksler and Baldin Laboratory and the staff of the Nuclotron accelerator for providing access to the research facilities used in these experiments. Additionally, we also would like to thank Graeme McDonnell from Nu Scientific for providing the technical drawings of the Canberra HPGe detector and Professor Igor Zhuk of the Joint Institute for Power and Nuclear Research, Belarus for providing additional information on the beam parameters.

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