DICE: Apparatus for Detection of Internal Conversion Electrons

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Abstract. An apparatus for the Detection of Internal Conversion Electrons (DICE) has been built and commissioned at Johannes Gutenberg University Mainz (JGU) for the study of recoil ion sources of 235mPu, which emit 235mU nuclei that deexcite to the ground state by emitting low-energy internal conversion (IC) electrons. We present an overview of DICE and its commissioning with 209Bi and 235mU electron sources, demonstrating that DICE allows the detection and counting of IC electrons from 235mU deexcitation. Our preliminary 235mU half-life of ≈ 26 min agrees with literature. DICE is thus an interesting tool to broaden analytical capabilities for recoil ion source characterization via studies of the peculiar decay of 235mU.

1 Introduction

Ultra-low-lying isomers have excitation energies of < 100 eV, rather than more typical isomers with energies of tens to hundreds of keV. These ultra-low-lying isomers, which bridge nuclear and atomic physics and are of interest in the particle physics community, have recently become a topic of great interest, mainly due to the lowest known isomer, 229mTh and its potential to develop nuclear clocks depending on the chemical environment, or of tens to hundreds of keV. While the 229mTh ground state has a too large half-life (7·1018 a) to be observed via α-spectrometry, measurements of the isomer decay proceeding with ≈ 26 half-lives are highly sensitive. To efficiently detect the corresponding low-energy CEs, the Detection of Internal Conversion Electrons (DICE) setup has been built and commissioned at JGU. For the present work, a 22 mm diameter 235Pu thin layer recoil ion source was prepared by the molecular plating (MP) technique [15]. Recoiling 235mU nuclei can be collected in a suitable catcher, from which CEs can emerge. From the rate of emitted CEs, the number of implanted 235mU nuclei can be determined, thus allowing the calculation of the 235Pu thin layer recoil efficiency. The present work provides a technical overview of DICE and the first commissioning results.

2 DICE – Detection of Internal Conversion Electrons

2.1 General setup

The DICE setup (Fig. 1) comprises a vacuum chamber evacuated by a turbomolecular pump (Agilent Turbo-V 511 Navigator) to ≈ 1·10⁻⁶ mbar within 5 min. A Pirani gauge monitors the pressure. The chamber houses a two-stage multi-channel plate (MCP) stack and an insulated 235mU source holder placed directly in front of the stack. The source holder is connected to a rod, which exits the chamber and allows manipulation of the source-to-MCP distance by up to 25 mm. The holder accommodates samples of up to 22 mm diameter. The source can be

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biased up to -2 kV using a high voltage (HV) power supply (NHQ 203M) to propel the low-energy CEs. Impulses detected from the MCP stack are decoupled, amplified and discriminated. The signal is then sent to a field-programmable gate array (FPGA) board, which computes the data and displays them in the chosen format on the computer.

![Source manipulator](image)

**2.2 Multi-channel plate (MCP) stack**

The MCP stack (DET40 MCP detector with timing anode, RoentDek Handels GmbH) is mounted inside the vacuum chamber flange. It has a Chevron-style configuration (two-stage stack) with a 12 µm pore diameter and can be used to count individual particles such as electrons, photons and ions with respect to time. Each plate features a 45 mm active diameter, a thickness of 1 mm, a bias angle of 20°, and an open area ratio of 70 %. The operating pressure is <2·10⁻⁶ mbar, and the maximum count rate is ~1 MHz. The front plate was always grounded, the back plate (B) was biased to +1900 V, and the anode (A) to +2100 V to ensure attraction and acceleration of the electrons. Later tests had B biased to +2200 V and A to +2500 V.

**2.3 Control and acquisition systems**

MCP signals coming from the anode were decoupled (RoentDek HFSD-BNC), pre-amplified (ORTEC VT120C +12 V fast pre-amp) and discriminated (ORTEC 584 constant fraction discriminator). Signals were counted by the FPGA (TUL PYNQ-Z2) board, which was python programmable in Jupyter notebook.

**3 Commissioning of DICE**

**3.1 Reagents and materials**

A 239Pu thin layer was deposited on a 525 µm Si wafer substrate by MP [15]. A Pt-coiled wire (Goodfellows, 99.9 %, 1 mm thickness) served as the anode [16]. All reagents were of high-purity chemical grade (Sigma-Aldrich). A MP cell, made from Kel-F to minimize Pu absorption, was set up with a 22 mm diameter open deposition area at the bottom of the cell [17]. Ethylene-propylene-diene-monomer (EPDM) gaskets, resistant against N,N-dimethylformamide (DMF), were used. All MP apparatus and substrates were washed with isopropanol, then acetone, and subsequently again with isopropanol before plating. After washing, the anode was additionally etched with 6 M HCl, washed with water and then isopropanol. Blank platings were performed three times before starting MP.

239Pu stock solution in dilute HCl contained about 0.2 % 241Am. It was prepared for 239Pu plating by evaporation and redissolution with four aliquots of conc. HNO3 and two aliquots of 0.1 M HNO3.

**3.2 239Pu source preparation and analytics**

135 kBq of 239Pu, dissolved in 10 µL 0.1 M HNO3 was pipetted into 10 mL DMF, mixed, and placed in the chimney of the cell. MP was carried out for 75 min at room temperature and a constant current density of 0.9 mA/cm². Voltages did not exceed 400 V. One minute before the end of plating, 1 mL of conc. NH4OH was added to the solution to prevent redissolution. The finished source was allowed to dry in the air [18]. It was characterized by radiographic imaging (FUJIFILM FLA 7000), scanning electron microscopy (SEM) at 20 kV (Philips XL30), and α-spectrometry (Canberra alpha analyst with Si p-i-n detector). After MP, the 239Pu content of an aliquot of the supernatant solution was measured and compared to that of an aliquot taken before MP had begun to give the indirectly determined yield. The direct yield was obtained from an α-measurement of the source.

**3.3 Collection of recoiling 235mU**

A 16 µm Al foil was placed 1 mm above the 239Pu recoil ion source under a vacuum of ≈1·10⁻² mbar until saturation (>4 h). The 239Pu layer was sufficiently thin to allow recoiling 235mU (88 keV) to escape the layer and implant in the Al foil to a depth of ≈34 nm [19]. Some 239Pu sputtering may also have occurred. Quick-release valves on the vacuum chamber allowed for fast (within 10 min) removal of the foil, which was then transferred to DICE for measurement, thus minimizing 235mU decay losses. The geometrical collection efficiency of 235mU recoils collected in the foil at saturation was calculated to be 43 % using the non-point source equation for geometrical efficiency.

**3.4 DICE commissioning with 207Bi**

For first commissioning, a 29.5 kBq 207Bi CE source (1/2 = 31.55 a) was mounted inside DICE. Four tests (Table 1) were performed at MCP stack bias voltages of; B: +1900 V, A: +2200 V (lower MCP bias) and B: +2200 V, A: +2500 V (higher MCP bias). Registered counts were summed over a defined interval, recorded, and then the counts reset and the process repeated for the total run time. Test no. 1 identified the optimum discriminator threshold at grounded source bias, and test no. 2 identified the optimum source bias at the optimum discriminator threshold found in test no. 1. Test no. 3 identified the optimum discriminator threshold at the optimum source bias found in test no. 2. Test no. 4 was longer runs at the optimum source bias and optimum discriminator threshold at that bias to assess system
stability and experiment reproducibility. All parameters other than the independent variable were kept constant for each test. The discriminator threshold was raised in +10 mV intervals and the source bias in -100 V intervals. The background was recorded at each MCP stack bias voltage.

Table 1. 207Bi source tests 1-4 at lower MCP bias and repeated at higher MCP bias. Optimum discrim. thresh. and source bias was found to be + 60 mV and ~ 900 V, respectively, for lower and higher MCP biases.

<table>
<thead>
<tr>
<th>Test no.</th>
<th>Interval (s)</th>
<th>Time per run (s)</th>
<th>No. of runs</th>
<th>Discrim. thresh. (+ mV)</th>
<th>Source bias (- V)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>10</td>
<td>60</td>
<td>15</td>
<td>0 – 140</td>
<td>0</td>
</tr>
<tr>
<td>2</td>
<td>10</td>
<td>60</td>
<td>15</td>
<td>0 – 1400</td>
<td>0 – 1400</td>
</tr>
<tr>
<td>3</td>
<td>10</td>
<td>60</td>
<td>13</td>
<td>0 – 140</td>
<td>900</td>
</tr>
<tr>
<td>4</td>
<td>10</td>
<td>300</td>
<td>2</td>
<td>60</td>
<td>900</td>
</tr>
</tbody>
</table>

3.5 Measurement of the 235mU half-life

The 207Bi results verified the correct operation of DICE. However, they cannot be directly applied to 235mU as the CE energies between the two nuclides differ drastically; biasing affects 207Bi CEs in the keV range much less than the 235mU CEs in the eV range. Once DICE was proven to detect electrons with good repeatability and low noise, initial 235mU CE measurements began. A foil with freshly collected 235mU was mounted inside DICE for each run. At the lower MCP stack bias, four runs (1-4) were performed (Table 2, test no. 1). Afterwards, an α-spectrometric measurement of the foil was performed to quantify Pu sputtering during collection. At the higher MCP stack bias, six runs were performed (Table 2, test no. 2). The discriminator threshold was varied (+ 0, 10, 20, 30, 60 and 90 mV) for testing purposes. All other parameters were unchanged.

Table 2. 235mU tests at (1, runs 1-4) lower MCP bias, and (2, runs A-F) higher MCP bias.

<table>
<thead>
<tr>
<th>Test no.</th>
<th>Interval (s)</th>
<th>Total time (s)</th>
<th>No. of runs</th>
<th>Discrim. thresh. (+ mV)</th>
<th>Source bias (- V)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>30</td>
<td>&gt;9000</td>
<td>4</td>
<td>60</td>
<td>900</td>
</tr>
<tr>
<td>2</td>
<td>30</td>
<td>360</td>
<td>6</td>
<td>0, 10, 20, 30, 60, 90</td>
<td>900</td>
</tr>
</tbody>
</table>

All results were corrected for decay loss between the end of collection and the start of counting. Uncertainty in counts was given as the square root of counts. Data were analyzed using a two-component exponential decay fit using a python-based least squares fitting model [20].

An average constant background determined independently was subtracted from the data sets. The point of intersection from the two best fits for each component was used to calculate the 235mU half-life via \( N_1 = N_0 e^{-\ln(2)/t_{1/2}} \) where \( N_1 \) is the number of 235mU atoms at time \( t \), \( N_0 \) is the number of 235mU atoms at \( t = 0 \), and \( t_{1/2} \) is the half-life of 235mU.

4 Results and discussion

4.1 Source preparation and characterization

Radiographic imaging of the 239Pu source showed a homogeneous activity distribution. In SEM images, the source surface looked smooth and crack-free. An activity of 135 kBq, corresponding to an areal density of \( \approx 15.5 \mu g/cm^2 \), and an MP yield of 96(2) %, confirming efficient MP of the 239Pu recoil ion source, was deduced from α-spectrometry.

4.2 DICE commissioning with 207Bi

For the 207Bi source tests (Table 1), background subtracted average counts were 39(2) cps and 2455(4) cps for the lower MCP stack bias and the higher MCP stack biasing, respectively. Errors were small, indicating excellent reproducibility. Background rates, due to, e.g., electronics noise, were low, i.e., in the 3-5 cps range. The background was slightly higher at the higher MCP stack bias than at the lower one. This may be due to the increased bias voltage attracting particles inside the chamber more efficiently. This also occurred for 207Bi source counts, rendering background increase insignificant. The number of electrons detected from the 207Bi source increased more strongly than the background rate for the higher MCP bias, suggesting that B: +2200 V, A: +2500 V biasing is more efficient for the detection of 207Bi CEs.

4.3 Measurement of the 235mU half-life

Between the two MCP bias data set results (Tables 3 and 4), the preliminary average half-life of 235mU deexcitation is 26.2(4) min, agreeing with the accepted value of \( \approx 26 \) min [6-12]. Run D was excluded as an anomaly due to the lower counts recorded and its much shorter half-life (21.0(5) min). Results from runs 1-4 showed small and varying amounts of 239Pu sputtering occurred during collection; this did not correlate with collection time. Figure 2 shows the exponentially decaying fit (\( R^2=0.998 \)) to the experimental data and the associated residuals for run 2. There was no correlation between discriminator threshold and half-life. The characterization of the background is ongoing and is needed to reach a data quality that is high enough to be sensitive to the variations of the 235mU half-life.

Table 3 and 4. Preliminary 235mU half-life and uncertainties (1 sigma confidence level) for runs at the lower MCP bias (left) and at the higher MCP bias (right). Mean uncertainty is the weighted standard deviation.

<table>
<thead>
<tr>
<th>Run no.</th>
<th>Half-life (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>27.6 ± 0.5</td>
</tr>
<tr>
<td>2</td>
<td>27.8 ± 0.4</td>
</tr>
<tr>
<td>3</td>
<td>26.6 ± 1.0</td>
</tr>
<tr>
<td>4</td>
<td>28.9 ± 0.5</td>
</tr>
<tr>
<td>Mean</td>
<td>27.7 ± 0.9</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Run no.</th>
<th>Half-life (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>25.1 ± 0.6</td>
</tr>
<tr>
<td>B</td>
<td>23.8 ± 0.6</td>
</tr>
<tr>
<td>C</td>
<td>24.2 ± 0.5</td>
</tr>
<tr>
<td>D</td>
<td>21.0 ± 0.5</td>
</tr>
<tr>
<td>E</td>
<td>24.9 ± 0.6</td>
</tr>
<tr>
<td>F</td>
<td>24.9 ± 0.6</td>
</tr>
<tr>
<td>Mean</td>
<td>24.6 ± 0.6</td>
</tr>
</tbody>
</table>
The half-life of 26.2(4) min between the two datasets agrees perfectly with 26.1(1) min obtained by Neve de Mevergnies for $^{235}$UO$_2$ [7-8]. Assuming $^{235}$U in the present experiment to be mainly in the +6 state, the results of [7-8] agree with our half-life. Neve de Mevergnies obtained a $^{235}$U half-life of 25.7(3) min with V catcher foil (average Pauling electronegativity of 1.63). Although der Mevergnies did not use Al substrate (average Pauling electronegativity of 1.61), results with V foil are very close to this experiment [11]. Izawa and Yamanaka found $^{235}$U half-life to be 25.7 ± 0.4 mins, which also agrees nicely with the present study’s half-life [12].

5 Conclusion and outlook

The DICE apparatus built and commissioned at JGU allows the detection and counting of CEs from $^{235}$U deexcitation. The preliminary half-life of 26.2(4) min agrees well with literature [6-12]. However, results among runs are not yet fully reproducible, with standard uncertainties in half-life of 3 % and 2 % for runs at lower and higher MCP stack bias, respectively. The higher MCP stack bias of B: +2200 V, A: +2500 V appear best for detecting both high- and low-energy electrons. DICE is thus well suited to quantify the rate of $^{235}$U emitted by $^{239}$Pu-containing thin layers prepared to provide the exotic $^{235}$U for fundamental studies of its still scarcely studied properties. The short $^{235}$U half-life and correspondingly high specific activity render $^{239}$Pu spiking of other α-decay recoil ion sources that emit long-lived (hence low-activity and thus hard to quantify) daughters attractive for performance studies of such sources. Upon final commissioning, DICE also promises to allow further study of the intriguing variation of $^{235}$U half-life depending on the chemical environment, which differs in literature by up to 9.5 %. Our preliminary studies suggest that after final optimization of background suppression, DICE should be sensitive to this half-life variation.

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References

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