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DICE: Apparatus for Detection of Internal Conversion Electrons

Lauren E. Reed[1*], *Dennis* Renisch[1,2], and *Christoph E.* Düllmann[1,2,3]

[1]Department Chemie - Standort TRIGA, Johannes Gutenberg-Universität Mainz, 55099 Mainz, Germany
[2]Helmholtz-Institut Mainz, 55099 Mainz, Germany
[3]GSI Helmholtzzentrum für Schwerionenforschung GmbH, 64291 Darmstadt, Germany

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 ^{239}Pu, which emit ^{235m}U 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 ^{207}Bi and ^{235m}U electron sources, demonstrating that DICE allows the detection and counting of IC electrons from ^{235m}U deexcitation. Our preliminary ^{235m}U 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 ^{235m}U.

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, ^{229m}Th and its potential to develop nuclear clocks that possibly outperform current atomic clocks by orders of magnitude by employing nuclear transitions rather than transitions in the electron shell [1-2]. One such ultra-low-lying isomer is ^{235m}U, the second-lowest known isomeric state, located at 76.8(5) eV excitation energy [2-4]. ^{235m}U ($I^\pi = 1/2^+$) is the first excited state of ^{235}U ($I^\pi = 7/2^-$) and deexcites by a highly converted (conversion coefficient $\alpha \approx 10^{21}$) E3 transition to ^{235}U [5]. The internal conversion (IC) process usually involves inner subshell electrons, but if the nuclear transition energy is very small, the inner subshell electrons have binding energies that exceed the isomer excitation energy. Therefore, IC must proceed in the outer electron subshells. The half-life of ^{235m}U has been reported to range from 24.7(3) min to 27.0(2) min, depending on the chemical environment, and is usually given as ≈ 26 min [6-12].

The low excitation energy of ^{235m}U implies the conversion electrons (CEs) to be emitted with very low kinetic energy. They can only travel a few atomic layers in solids [13]. One way to obtain ^{235m}U is as a recoil daughter from ^{239}Pu thin layers. These emit ^{235m}U, with the isomer being populated in the ^{239}Pu α-decay with a summed branching of 99.8% [14]. The quantification of ^{235m}U emitted from the sample allows for obtaining information on the recoil ion rate of the ^{239}Pu thin layers. The rate at which ^{235m}U becomes available for study is a key performance indicator of the recoil ion source. To determine source recoil efficiency, it is necessary to quantify the amount of $^{235(m)}$U recoil ions emitted from the source per time quanta and compare this with the theoretical amount of $^{235(m)}$U recoil expected in the same time quanta. While the ^{235}U ground state has a too large half-life ($7 \cdot 10^8$ a) to be observed via α-spectrometry, measurements of the isomer decay proceeding with ≈ 26 min half-life 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 ^{239}Pu thin layer recoil ion source was prepared by the molecular plating (MP) technique [15]. Recoiling $^{235(m)}$U nuclei can be collected in a suitable catcher, from which CEs can emerge. From the rate of emitted CEs, the number of implanted ^{235m}U nuclei can be determined, thus allowing the calculation of the ^{239}Pu thin layer recoil efficiency. The present work provides a technical overview of DICE and the first commissioning results.

2 DICE – Detection of Internal Conversion Elections

2.1 General setup

The DICE setup (Fig. 1) comprises a vacuum chamber evacuated by a turbomolecular pump (Agilent Turbo-V 511 Navigator) to ≈ $1 \cdot 10^{-6}$ mbar within 5 min. A Pirani gauge monitors the pressure. The chamber houses a two-stage multi-channel plate (MCP) stack and an insulated ^{235m}U 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

* Corresponding author: lreed@uni-mainz.de

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.

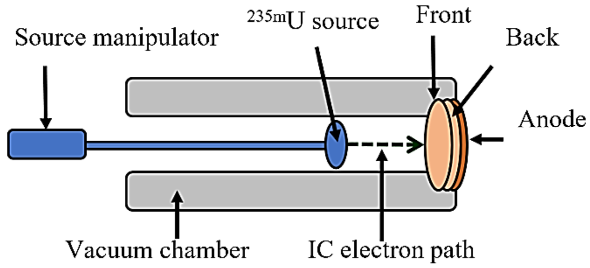

Fig. 1. Schematic of the Detection of Internal Conversion Electrons (DICE) apparatus.

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 $< 2\cdot10^{-6}$ 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 ^{239}Pu 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.

^{239}Pu stock solution in dilute HCl contained about 0.2 % ^{241}Am. It was prepared for ^{239}Pu plating by evaporation and redissolution with four aliquots of conc. HNO_3 and two aliquots of 0.1 M HNO_3.

3.2 ^{239}Pu source preparation and analytics

135 kBq of ^{239}Pu, dissolved in 10 µL 0.1 M HNO_3 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/cm^2. Voltages did not exceed 400 V. One minute before the end of plating, 1 mL of conc. NH_4OH 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 ^{239}Pu 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 ^{235m}U

A 16 µm Al foil was placed 1 mm above the ^{239}Pu recoil ion source under a vacuum of $\approx 1\cdot10^{-2}$ mbar until saturation (> 4 h). The ^{239}Pu layer was sufficiently thin to allow recoiling ^{235m}U (88 keV) to escape the layer and implant in the Al foil to a depth of ≈ 34 nm [19]. Some ^{239}Pu 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 ^{235m}U decay losses. The geometrical collection efficiency of ^{235m}U 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 ^{207}Bi

For first commissioning, a 29.5 kBq ^{207}Bi CE source ($t_{1/2}$ = 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. ^{207}Bi 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.

Test no.	Interval (s)	Time per run (s)	No. of runs	Discrim. thresh. (+ mV)	Source bias (- V)
1	10	60	13	0 – 140	0
2	10	60	15	60	0 -1400
3	10	60	13	0 – 140	900
4	10	300	2	60	900

3.5 Measurement of the ^{235m}U half-life

The ^{207}Bi results verified the correct operation of DICE. However, they cannot be directly applied to ^{235m}U as the CE energies between the two nuclides differ drastically; biasing affects ^{207}Bi CEs in the keV range much less than the ^{235m}U CEs in the eV range. Once DICE was proven to detect electrons with good repeatability and low noise, initial ^{235m}U CE measurements began. A foil with freshly collected ^{235m}U 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 (A-F) 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. ^{235m}U tests at (1, runs 1-4) lower MCP bias, and (2, runs A-F) higher MCP bias.

Test no.	Interval (s)	Total time (s)	No. of runs	Discrim. thresh. (+ mV)	Source bias (- V)
1	30	>9000	4	60	900
2	30	360	6	0, 10, 20, 30, 60, 90	900

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 ^{235m}U half-life via $N_t = N_0 e^{-(\ln(2)/t_{1/2})t}$ where N_t is the number of ^{235}U atoms at time t, N_0 is the number of ^{235m}U atoms at $t = 0$, and $t_{1/2}$ is the half-life of ^{235m}U.

4 Results and discussion

4.1 Source preparation and characterization

Radiographic imaging of the ^{239}Pu 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 ≈ 15.5 μg/cm^2, and an MP yield of 96(2) %, confirming efficient MP of the ^{239}Pu recoil ion source, was deduced from α-spectrometry.

4.2 DICE commissioning with ^{207}Bi

For the ^{207}Bi 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 ^{207}Bi source counts, rendering background increase insignificant. The number of electrons detected from the ^{207}Bi 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 ^{207}Bi CEs.

4.3 Measurement of the ^{235m}U half-life

Between the two MCP bias data set results (Tables 3 and 4), the preliminary average half-life of ^{235m}U deexcitation is 26.2(4) min, agreeing with the accepted value of ≈ 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 ^{239}Pu sputtering occurred during collection; this did not correlate with collection time. Fig. 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 ^{235m}U half-life.

Tables 3 and 4. Preliminary ^{235m}U 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.

Run no.	Half-life (min)
1	27.6 ± 0.5
2	27.8 ± 0.4
3	26.6 ± 1.0
4	28.9 ± 0.5
Mean	27.7 ± 0.9

Run no.	Half-life (min)
A	25.1 ± 0.6
B	23.8 ± 0.6
C	24.2 ± 0.5
D	21.0 ± 0.5
E	24.9 ± 0.6
F	24.9 ± 0.6
Mean	24.6 ± 0.6

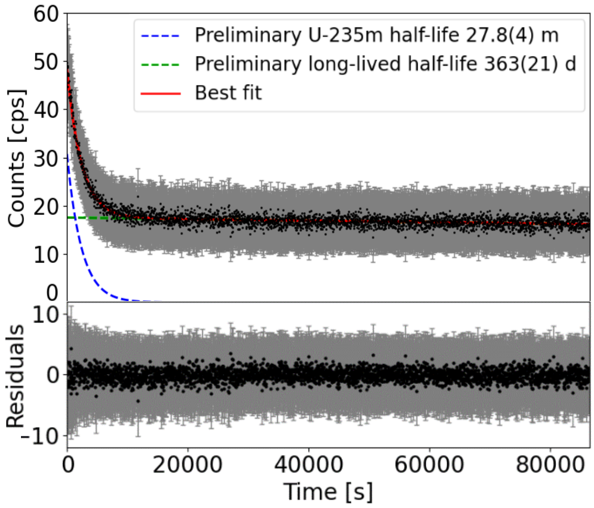

Fig. 2. Example ^{235m}U half-life determination from run 2 at the lower MCP bias. A preliminary ^{235m}U half-life of 27.8(4) m and longer-lived component of 363(21) d were obtained.

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 $^{235m}UO_3$ [7-8]. Assuming ^{235m}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 ^{235m}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 ^{235m}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 ^{235m}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 ^{235m}U emitted by ^{239}Pu-containing thin layers prepared to provide the exotic ^{235m}U for fundamental studies of its still scarcely studied properties. The short ^{235m}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 ^{235m}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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