Study of neutron-deficient isotopes of Fl in the $^{239}$Pu, $^{240}$Pu + $^{48}$Ca reactions

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Abstract. The results of the experiments aimed at the synthesis of Fl isotopes in the $^{239}$Pu + $^{48}$Ca and $^{240}$Pu + $^{48}$Ca reactions are presented. The experiment was performed using the Dubna gas-filled recoil separator at the U400 cyclotron. In the $^{239}$Pu + $^{48}$Ca experiment one decay of spontaneously fissioning $^{284}$Fl was detected at 245-MeV beam energy. In the $^{240}$Pu + $^{48}$Ca experiment three decay chains of $^{285}$Fl were detected at 245 MeV and four decays were assigned to $^{284}$Fl at the higher $^{48}$Ca beam energy of 250 MeV. The $\alpha$-decay energy of $^{285}$Fl was measured for the first time and decay properties of its descendants $^{281}$Cn, $^{277}$Ds, $^{273}$Hs, $^{269}$Sg, and $^{265}$Rf were determined more precisely. The cross section of the $^{239}$Pu($^{48}$Ca,3n)$^{284}$Fl reaction was observed to be about 20 times lower than those predicted by theoretical models and 50 times less than the value measured in the $^{244}$Pu+48Ca reaction. The cross sections of the $^{240}$Pu($^{48}$Ca,4-3n)$^{284,285}$Fl at both $^{48}$Ca energies are similar and exceed that observed in the reaction with lighter isotope $^{239}$Pu by a factor of 10. The decay properties of the synthesized nuclei and their production cross sections indicate rapid decrease of stability of superheavy nuclei with departing from the neutron number $N=184$ predicted to be the next magic number.

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1. Introduction

One of the outcomes of macroscopic-microscopic nuclear theory [1] predicts the existence of the island of stability of the super-heavy nuclei around \( Z=114 \) and \( N=184 \). The significant success in the synthesis and decay properties study of the superheavy nuclei was achieved in the FLNR JINR (Dubna, Russia) during last 15 years [2]. Six new super-heavy elements with \( Z=113-118 \) and more than 50 new isotopes with \( Z=104-118 \) were observed for the first time at the Dubna Gas-Filled Recoil Separator (DGFRS) in the irradiations of \(^{238}\text{U}, ^{237}\text{Np}, ^{242,244}\text{Pu}, ^{243}\text{Am}, ^{245,246}\text{Cm}, ^{247}\text{Bk} \) and \(^{249}\text{Cf} \) targets with accelerated \(^{48}\text{Ca} \) ion beam delivered by U-400 cyclotron. The first super-heavy nucleus with \( Z=114 \) was discovered in June, 1999 in the complete-fusion reaction \(^{244}\text{Pu} + ^{48}\text{Ca} \) performed by Dubna-Livermore collaboration. The detected parent nucleus and its decay chain were assigned to \(^{289}\text{Fl} \) formation (see [2] and Refs. therein). Later, the properties of lighter isotopes \(^{286-288}\text{Fl} \) were determined [3-5] both as in direct cross bombardment reactions of \(^{242}\text{Pu} \) and \(^{244}\text{Pu} \) targets with \(^{48}\text{Ca} \) ion beam at different beam energies corresponding to the maximum reaction yield with evaporation of 2-5 neutrons, and as descendant daughter nuclei in the decays of synthesized \(^{290-291}\text{Lv} \) and granddaughter of \(^{292}\text{118} \) in irradiations of \(^{245,248}\text{Cm} \) and \(^{249}\text{Cf} \) targets [5-7]. Experiments on the synthesis of \( \text{Fl} \) and \( \text{Lv} \) isotopes were repeated in other laboratories with IVO experimental setup [8-11], SHIP [12, 13], BGS [14, 15] and TASCA [16, 17]. Thus, the production cross-sections and decay properties of the isotopes of \(^{286-289}\text{114} \) and \(^{291-293}\text{116} \) were recently confirmed in the independent experiments.

Few attempts have been undertaken to expand the region of super-heavy nuclei with synthesis of elements 119 and 120 by using target nuclei ranging from \(^{238}\text{U} \) to \(^{240}\text{Cf} \) with projectiles heavier than \(^{48}\text{Ca} \), from \(^{50}\text{Ti} \) to \(^{64}\text{Ni} \). However, no decay chains of SHN were observed in these experiments. The upper cross section limits were set at \( 0.07 - 1.1 \) pb depending on the reaction studied [18-21]. The next step in the exploring of island of stability of the SHN could be performing the nuclear reactions leading to new isotopes at the edge of the new SHE region. For example, in irradiation of \(^{239}\text{Pu}, ^{240}\text{Pu} \) and \(^{249,251}\text{Cf} \) targets with \(^{48}\text{Ca} \) beam one could observe the formation and decay of both very light \(^{283-285}\text{Fl} \) isotopes and two the heaviest isotopes of 118 element: \(^{293}\text{118} \) and \(^{296}\text{118} \) [19].

In this work, we present the results of experiment on the synthesis of neutron-deficient Fl isotopes in the \(^{239}\text{Pu} + ^{48}\text{Ca} \) and \(^{240}\text{Pu} + ^{48}\text{Ca} \) fusion-evaporation reactions. These experiments have provided evidence for the SF decay of a new isotope, \(^{284}\text{Fl} \), and new properties of the \(^{286}\text{Fl} \) decay chain.

2. Experiment

The \(^{48}\text{Ca} \) ion beam was delivered to the target with a maximum intensity of 1.3 particle \( \mu \text{A} \). The beam energy was determined with a systematic uncertainty of 1 MeV by a time-of-flight system placed in front of the DGFRS.

The target materials were provided by Oak Ridge National Laboratory (ORNL) (\(^{240}\text{Pu} \), enrichment of 98.97%) and JINR (\(^{239}\text{Pu} \) and \(^{240}\text{Pu} \), enrichment of 98.2 and 92%, respectively). The impurities in the \(^{239}\text{Pu} \) and \(^{240}\text{Pu} \) targets mainly consisted of \(^{240}\text{Pu} \) and \(^{239}\text{Pu} \), respectively. The average thicknesses of the targets for the main isotopes were \( 0.50 \pm 0.05 \) mg/cm\(^2\) for \(^{239}\text{Pu} \) and \( 0.39 \pm 0.04 \) mg/cm\(^2\) for \(^{240}\text{Pu} \) (given uncertainties correspond to standard deviations of thicknesses measured for six sectors of each target). The targets were manufactured by depositing \( \text{PuO}_2 \) oxide onto \( 0.71-0.72 \) mg/cm\(^2\) Ti foils. Each target had an area of 5.4 cm\(^2\) in the shape of an arc segment with an angular extension of 60 deg and an average radius of 60 mm. The segments were mounted on a disk that was rotated at 1700 rpm such that the target was perpendicular to the direction of the incoming beam. In the course of the bombardment with the \(^{48}\text{Ca} \) beam, the target layers were systematically monitored by counting \( \alpha \)-particles from the decay of the target isotopes.

The lab-frame beam energies in the middle of the target layers, excitation energy ranges (with use of mass tables [20,21]) and beam doses for the experiments studied are summarized in Table I. For calculation of excitation-energy ranges of the resulting compound nuclei \(^{287,288}\text{Fl} \), we took into account the beam energy resolution, the small variation of the beam energy during irradiation, and the energy loss in the target.


Table I. Target, reaction-specific lab-frame beam energies in the middle of the target layers, corresponding excitation energy intervals, and total beam doses for the given reactions.

| Target | $E_{lab}$ (MeV) | $E^*$ (MeV) | Beam dose |
|--------|-----------------|-------------|-----------|
| $^{240}\text{Pu}$ | 245 | 36.5-41.1 | $4.0 \times 10^{18}$ |
| $^{240}\text{Pu}$ | 250 | 40.9-45.4 | $4.7 \times 10^{18}$ |
| $^{239}\text{Pu}$ | 245 | 35.4-40.0 | $1.4 \times 10^{19}$ |

Evaporation residues (ER) recoiling from the target were separated in flight from $^{48}\text{Ca}$ beam ions, scattered particles and transfer-reaction products by the DGFRS. The transmission efficiency of the separator for $Z=114$ nuclei was estimated to be about 35%. Recolls passed through a time-of-flight system (TOF) and were implanted in the detectors. The TOF system consists of two multi-wire proportional counters (MWPC) placed at a distance of 65 mm between them. The detectors and MWPC are placed in Pentane at a pressure of about 1.5 Torr. A 0.2-μm/cm² Mylar foil separates the detection system from the DGFRS volume, which is filled with hydrogen at a pressure of 1 Torr.

The array of Silicon detectors at the DGFRS final focus has been modified to increase the position resolution of recorded signals and subsequently reduce the probability of observing sequences of random events that mimic decay chains of implanted nuclei. The new detection system includes a 0.3-mm thick Double-sided Silicon Strip Detector (DSSD) manufactured by Micron Semiconductor Ltd (model BB-17). This large DSSD has 1-mm wide strips, 48 at the front side and 128 at the back side, creating over 6000 1-mm² pixels in one Silicon wafer. Such high pixilation helps to achieve superior position resolution for recoil-correlated decay sequences reducing potential random events. The recoil implantation counter was surrounded by six single Si-detectors, MICRON model MSX-7200, each 500 microns thick and having an active area of 65 mm by 120 mm. Two pairs of these Si-box detectors were mounted at the DSSD long side, and two detectors were used to close the Si-box geometry, one at each short side. All Si counters had a minimum amount of supporting frame material. The active detection length for the DSSD escape events was extended about 120 mm, from the DSSD surface towards the separator. The DSSD was backed by the single Si-veto detector (MICRON MSX-62), of 0.5 mm thickness and 48 mm by 128 mm active size matching the respective BB-17 area. This veto counter was mounted in the frame identical to the BB-17 support about 3 mm from the back surface of the DSSD. The signals from all detectors were processed using MESYTEC preamplifiers. This new Si-detector array was designed, assembled, commissioned off-line and provided by Oak Ridge National Laboratory.

The output signals from the MESYTEC linear-logarithmic preamplifiers serving BB-17 DSSD as well as linear preamplifiers serving MSX-7200 and MSX-62 detectors were split into two branches. One of these branches was processed with analog electronics similar to those used in previous DGFRS experiments, see [22]. The analog electronics system was used to create a dedicated low-background detection scheme for the nuclei to be investigated. This detection scheme allows the beam to be switched off after a predefined event sequence. The beam interruption occurs after the detection of a recoil signal with the expected implantation energy for $Z=114$ evaporation residues followed by an α-like signal in the implantation detector with an energy of 9.8-11.5 MeV, in the same front and back strips, i.e., in the same 1-mm² DSSD pixel.

The second branch of split preamplifier signals was processed using a digital electronics system based on XIAX Pixie-16 modules provided by ORNL. Such a digital pulse processing system was developed and applied successfully in earlier experiments on short-lived charged particle emitters at the Recoil Mass Separator (RMS) of ORNL’s Holifield Radioactive Ion Beam Facility [23,24].

The FWHM energy resolution of the implantation detector was 34 to 78 keV, while the summed signals recorded by the side and implantation detectors had an energy resolution of 147 to 263 keV.
Other experimental conditions, including the method of calibration of the detectors, were the same as in previous DGFRS experiments (see [2] and references therein).

3. Results

According to excitation functions measured in the reactions of $^{48}$Ca with target nuclei $^{238}$U-$^{249}$Cf (see [2] and references therein), the maximum of the cross section of the fusion-evaporation reaction $^{240}$Pu+$^{48}$Ca is expected at an excitation energy $E^*$ of the compound nucleus $^{285}$Fl of about 40 MeV. In the first experiment, performed at $^{48}$Ca energy $E_{\text{lab}}=245$ MeV (Table I), we observed three decay chains of $^{285}$Fl, the product of the $3n$-reaction channel (Fig. 1). This isotope was first registered in the $^{242}$Pu($^{48}$Ca,5$n$)$^{285}$Fl reaction at LBNL [25]. In their single decay chain, the $\alpha$ particle of the parent nucleus escaped from the front of the five-sided detector box leaving the energy of 1.64 MeV in the implantation detector. Four further $\alpha$ decays of nuclei from $^{281}$Cn to $^{269}$Sg were detected with full energy by the front detector only or simultaneously with side one.

![Figure 1. Decay properties of $^{285}$Fl and descendant nuclei observed in the $^{240}$Pu+$^{48}$Ca reaction at projectile energy of 245 MeV. The upper right rows for each chain show ER (in pink) energies and strip numbers (front/back). The left rows provide energies, time intervals between events and their strip numbers for $\alpha$ decay (in yellow) and SF (in green). Energies of summed signals are given in parentheses. Events marked with a shadow were registered during the beam-off periods. The $\alpha$-particle energy errors are shown by smaller italic numbers. The time interval for an SF event following a “missing $\alpha$” was measured from the preceding registered event and is shown in italics.](image)

In all three decay chains observed in the present $^{240}$Pu+$^{48}$Ca reaction, the $\alpha$ particle of $^{285}$Fl was registered by the focal-plane detector with full energy which resulted in the first measurement of its energy $E_\alpha$. The decay times of nuclei $^{285}$Fl-$^{265}$Rf as well as $E_\alpha$ values of $^{281}$Cn-$^{269}$Sg in three chains are in good agreement with those measured in the $^{242}$Pu($^{48}$Ca,5$n$)$^{285}$Fl reaction [25]. Only the energies of $\alpha$ particles of $^{277}$Ds in the third chain and $^{273}$Hs in the second and third chains are somewhat lower than values measured in [25] (by 73 and about 25 keV, respectively, taking into account energy resolutions of detectors). However, these deviations do not seem significant.

During the 527-hour $^{240}$Pu + 245-MeV $^{48}$Ca experiment when the beam was on the target, the total number of sequences consisting of ER-like events with $E_{\text{ER}}=6$-16 MeV and $\alpha$-like events with $E_\alpha=10.1$-10.7 MeV detected within 1.5 s in the same front and back strips of the focal-plane detector was only 57. In the first two decay chains of $^{285}$Fl shown in Fig. 1, the $\alpha$ decay of the parent nucleus...
switched the beam off. Three further $\alpha$ particles of $^{281}$Cn,$^{235}$Hs in the first case and four $\alpha$ particles of $^{281}$Cn,$^{269}$Sg and SF of $^{265}$Rf in the second chain were registered in the absence of beam-associated background. The total duration of beam-off intervals in this run was about 4.3 h. In the first chain, the beam-off pause was not manually prolonged and decays of $^{269}$Sg and $^{265}$Rf were registered when the beam was switched on. For beam-off $\alpha$ decays with $E_\alpha=8$-11 MeV, the probability of their detection as random events in any crossing of strips within period $\Delta t=3$ min was about $1.3\times10^{-4}$ [26]. For beam-on $\alpha$- and SF-like events with energies $E_\alpha=8$-11 MeV and $E_{SF}>130$ MeV, respectively, and $\Delta t=10$ min these values were $1.2\times10^{-2}$ and $1.3\times10^{-3}$, respectively. The decay of $^{281}$Cn in the second chain was registered by the focal-plane detector only with energy of 1.7 MeV while the beam was stopped by ER-$\alpha_1$ sequence. The probability of the random origin of an event with any energy in strips 19/37 within $\Delta t=1$ s was less than $7\times10^{-3}$. In the third chain, the first $\alpha$ particle did not stop the beam, for an unknown reason. Two subsequent $\alpha$ decays were detected by both the focal-plane and side detectors and could not switch the beam off. Thus, all these events as well as the fourth $\alpha$ particle and the SF event were observed during the beam-on time interval. However, the probability of detection of random beam-on events with $E_\alpha=8$-11 MeV within $\Delta t=5$ s was $1.0\times10^{-4}$.

In the third chain, no $\alpha$ particles with $E_\alpha=8$-11 MeV were found in strips 2/31 between decays attributed to $^{277}$Hs and $^{265}$Rf. This missing $\alpha$ event could be detected by the focal detector only with low energy-release, but the probability of detection of similar random beam-on events within the time interval of 491 s is rather large preventing the definite assignment of such an event to $^{269}$Sg. Missing this $\alpha$ particle and incomplete detection of the energy of $^{281}$Cn in the second chain are in accordance with the 85-% registration efficiency of the detectors. From the remaining 13 $\alpha$ particles, five decays were registered by the both implantation and side detectors which also corresponds to their detection efficiencies. In one of these events, the $\alpha$-particle energy of $^{277}$Ds absorbed by the focal detector was below the energy threshold, and the number of the front strip was not determined. However, the energy and corresponding strip number were registered in the back side of the detector.

In the second experiment, the energy of $^{48}$Ca was increased to 250 MeV for measurement of the excitation function of the $^{240}$Pu+$^{48}$Ca reaction and possible observation of the product of the 4$\pi$-evaporation channel, the new lightest isotope $^{284}$Fl. In this experiment, decay chains of $^{285}$Fl were not observed which indicates a decrease of the cross section of the 3n channel at this $^{48}$Ca energy ($\sigma_{3n}\leq1.3$ pb).

However, in this run, four ER-SF chains were found which are shown in Fig. 2. In addition, 27 recoil-fission (focal-plane only events) were found with $E_{R}=6$-16 MeV, $E_{F}>130$ MeV, and $\Delta t=100$ s, with a random distribution of R-F time intervals varying from 3.3 to 96.6 s, therefore, the total number of random ER-SF chains for $\Delta t=10$ ms is $3\times10^{-3}$.

![Figure 2](image-url)

**Figure 2.** Decay properties of nuclei assigned to $^{284}$Fl observed in the $^{240}$Pu+$^{48}$Ca (top) and $^{239}$Pu+$^{48}$Ca (bottom) reactions. The right rows for each chain show ER (in pink) energies and strip numbers (front/back). The left rows provide SF fragment (in green) energies, time intervals between events and their strip numbers. The decay chain shown in the right top corner was registered in two back strips.
The same nucleus $^{284}$Fl could be produced in the $3n$-evaporation channel of the reaction with the lighter Pu isotope, $^{239}$Pu+$^{48}$Ca, and its observation could make a cross-bombardment consistency check on the synthesis of the new Fl isotope. This experiment was performed at one $^{48}$Ca energy $E_{\text{lab}}=245$ MeV which corresponds to expected maximum of the cross section of the $3n$-evaporation channel [27].

In the $^{239}$Pu+$^{48}$Ca reaction short ER-SF chain with decay time of 0.53 ms was detected (see Fig. 2) as well as 30 another chains with $E_k=6-16$ MeV, $E_f>130$ MeV, and $\Delta t=100$ s (again all these fission-like events were detected by the focal-plane detector only). Like in the previous case, the number of random ER-SF chains for $\Delta t=10$ ms was estimated to be $3 \times 10^{-3}$.

The energy of the $^{48}$Ca in the reaction with $^{239}$Pu was the same as in the $^{240}$Pu experiment where the $3n$-reaction channel was observed. This also corresponds to the predicted cross-section maximum of the $^{239}$Pu($^{48}$Ca,3$n$)$^{284}$Fl reaction [27]. Based on these facts, one may assign this ER-SF chain to $^{284}$Fl. In that case, the average half-life of this isotope calculated for five decays would be equal to $T_{1/2}=2.5\pm0.8$ ms. The production cross section was found to be $\sigma_{3n}=0.23^{+0.09}_{-0.20}$ pb, which is lower by factor of 10 than that for the reaction with $^{240}$Pu. An upper cross-section limit of 0.47 pb can be set if this decay does not originate from $^{284}$Fl.

![Figure 3. Maximum of the production cross sections of Fl isotopes in the $^{48}$Ca-induced reactions with targets shown in figure vs. mass number of the compound nucleus. Data from DGFRS [4,5] are shown by squares as well as from BGS [25] and TASCA [16] by circles. Vertical error bars correspond to total uncertainties. The dashed line is drawn to guide the eye.](image)

4. Conclusion
In the $^{240}$Pu+$^{48}$Ca reaction, three decay chains of $^{285}$Fl were observed. The $\alpha$-particle energy of $^{285}$Fl ($E_{\alpha}=10.41\pm0.05$ MeV) was measured for the first time. The decay properties of isotopes $^{285}$Fl, $^{281}$Cn, $^{277}$Ds, $^{273}$Hs, $^{269}$Sg and $^{265}$Rf were determined more precisely. The cross section of the $^{240}$Pu($^{48}$Ca,3$n$)$^{285}$Fl reaction for 245-MeV projectiles was measured to be $2.5^{+2.9}_{-1.4}$ pb which exceeds that for $^{239}$Pu by a factor of 10 but is 2-4 times lower than that for $^{244}$Pu.

Two ER-SF decay chains observed in the $^{240}$Pu + 250-MeV $^{48}$Ca reaction with high energy-release of fission fragments ($E_{\text{SF}}=190$ and 234 MeV) are assigned to the new spontaneously fissioning isotope $^{284}$Fl. Two more ER-SF events registered with lower energy values ($E_{\text{SF}}=140$ and 168 MeV) could also originate from $^{284}$Fl (assignment to $^{240}$Pu+$^{244}$Am fission isomers is not rejected also). The cross section of the $^{240}$Pu($^{48}$Ca,4$n$)$^{284}$Fl reaction ($2.6^{+3.3}_{-1.2}$ pb) is similar to that for the $3n$-reaction channel.

In the $^{239}$Pu+$^{48}$Ca reaction, one ER-SF chain was observed with SF decay properties comparable with those registered for $^{284}$Fl in the reaction with $^{240}$Pu. The production cross section for this event, presumably the product of the $^{239}$Pu($^{48}$Ca,3$n$)$^{284}$Fl reaction, of 0.2 pb is 20 times lower than the
theoretically predicted value and 50 times lower than maximum fusion-evaporation cross section measured in the reaction with 244Pu.

The considerable drop of the evaporation cross sections of the 239Pu+48Ca and 240Pu+48Ca reactions as well as the decline of the half-life and dominance of spontaneous fission over α decay for 284Fl and increased growth of the α-decay energy of 288Fl compared to the heavier Fl isotopes indicate one is approaching the neutron-deficient border of stability of SHN.

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