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# Spontaneous fission of light californium isotopes produced in $^{206,207,208}$Pb + $^{34,36}$S reactions; new nuclide $^{238}$Cf

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**Abstract**

In bombardments of $^{206,207,208}$Pb with $^{34}$S and $^{206}$Pb with $^{36}$S, we identified a new spontaneously fissioning isotope $^{238}$Cf with $T_{sf} \approx T_{1/2} = 21 \pm 2$ ms and obtained evidence of the production of a new isotope $^{237}$Cf with $T_{1/2} = 2.1 \pm 0.3$ s. The spontaneous-fission (SF) decay mode was established for $^{240}$Cf; its SF branch was estimated to be $b_{sf} \sim 2 \times 10^{-2}$. We measured also $b_{sf} \leqslant 1.4 \times 10^{-4}$ for $^{242}$Cf and estimated $b_{sf} \sim 10^{-1}$ for $^{237}$Cf. The production cross sections of $^{238}$Cf in the $^{206,207,208}$Pb + $^{34}$S reactions were measured to be in the range of 0.3 to 1.1 nb. Finally, we probed the influence of the neutron excess in the $N = 20$ projectile $^{36}$S on cross sections of fusion-evaporation reactions occurring on lead targets.

*Keywords:* RADIOACTIVITY $^{237,238,240}$Cf (SF); Measured $T_{1/2}$, deduced spontaneous fission branches. $^{242}$Cf (SF); Measured upper limit for spontaneous-fission branch. $^{246}$Fm; Measured $T_{1/2}$. NUCLEAR REACTIONS $^{206,207,208}$Pb($^{34}$S, X), $^{206}$Pb($^{36}$S, X), $E = 215$ MeV; $^{208}$Pb($^{40}$Ar, X), $E = 225$ MeV; Measured production $\sigma$ for spontaneously fissioning $^{237,238,240}$Cf nuclei. Isotopically enriched targets.

## 1. Introduction

Until recently, the 35.7 h $\alpha$-decaying isotope $^{246}$Cf [1], having a tiny spontaneous fission (SF) branch, $b_{sf} = 2 \times 10^{-6}$, remained the lightest Cf isotope whose SF decay mode was established and characterized. At the same time, seven lighter Cf isotopes down to $^{239}$Cf had already been produced and identified by their $\alpha$ decay [1,2].

We report here on our experiments designed to probe the SF decay of the lighter

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known even-even isotopes of Cf and to produce new, even more neutron-deficient Cf species. For the production of light Cf isotopes we used fusion-evaporation reactions induced by $^{34}$S and $^{36}$S projectiles on $^{206,207,208}$Pb target nuclei. In general, the available reaction systems $^{204,206,207,208}\mathrm{Pb} + {}^{32,33,34,36}\mathrm{S}$ provide not only the possibility of producing the lightest new Cf nuclei but also an opportunity to study the influence of nuclear structure and nucleon composition of both reaction partners on cross sections of fusion-evaporation reactions leading to very heavy, highly fissionable evaporation residues. At present, experimental information of this kind is very scarce or virtually absent. Yet this knowledge is of far-reaching importance for both understanding the mechanism of fusion of two complex nuclei and finding out the most prolific ways for the synthesis of new heavy and superheavy nuclides. An especially interesting case is associated with the neutron-rich $^{36}$S projectile which involves the magic number of neutrons, $N = 20$, and can be considered to be similar, in a sense, to the famous projectile $^{48}$Ca. However, the fusion-evaporation reactions between Pb and S nuclei have never been studied before. The cross-section measurements performed in our work represent a first step in this direction.

## 2. Experimental technique

The irradiations were performed at the Dubna U400 cyclotron by using beams of $^{34,36}$S and $^{40}$Ar projectiles with incident energies of 215 and 225 MeV, respectively. For the present studies, it was important to obtain appropriately intense beams of $^{34}$S and $^{36}$S at a reasonably low consumption of isotopically enriched sulphur materials (the abundances of $^{34}$S and $^{36}$S in naturally occurring sulphur are 4.21% and 0.02%, respectively). A special technique was developed for the production and acceleration of these ions by using a solid (ZnS enriched in $^{34}$S to $\approx 40\%$ or in $^{36}$S to $\approx 24\%$) as a working material to feed the discharge chamber of a PIG ion source of sputtering type. This technique allowed us to achieve an average consumption of the working material of $\approx 10$ $\mathrm{mg \cdot h^{-1}}$ $\mathrm{p\mu A^{-1}}$ and to provide a possibility of recovering $\approx 60\%$ of the material. Average intensities of $^{34}$S and $^{36}$S beams applied to Pb targets were about $8 \times 10^{12}$ and $3 \times 10^{12}$ pps, respectively.

In our experiments we employed the wheel system described in Ref. [3]. A beam of accelerated particles tangentially struck the lateral surface of a cooled copper cylinder onto which 2 to 3 $\mathrm{mg \cdot cm^{-2}}$ of the metallic target material was deposited. This cylindrical target (serving simultaneously as a recoil catcher) rotated at a constant velocity. For each particular bombardment, the period of the wheel revolution, $T_{\mathrm{rev}}$, was chosen according to the expected half-life value of a SF activity under study. Mica fission-fragment detectors arranged all around the rotating target cylinder (except for the beam-input zone, see Fig. 5 in Ref. [3]) were used for the detection of SF events. The mica detectors covered, without any gaps, the time interval of 0.06 $T_{\mathrm{rev}}$ to 0.85 $T_{\mathrm{rev}}$. The metallic layers of isotopically enriched $^{208}$Pb (99.0%), $^{207}$Pb (93.2%), and $^{206}$Pb (94.9%) were deposited onto the target cylinder by evaporation in vacuum.

Earlier, this technique was extensively used in experiments aimed at synthesizing transfermium nuclides (see, e.g., Refs. [3,4]) where it permitted the detection of SF species produced with picobarn cross sections. More recently, it was employed in the experiments that led to the discovery of a new region of EC($\beta^+$)-delayed fission around $^{180}$Hg–$^{188}$Pb–$^{196}$Po [5], in the studies of the stability of the K-isomeric states of $^{250}$Fm and $^{254}$102 against SF [6], in searches for carbon radioactivity of $^{114}$Ba [7], as well as in other studies [8].

## 3. Results and discussion

To probe the SF stability of the 3.4 min isotope $^{242}$Cf [1], we employed the $^{208}$Pb($^{40}$Ar, 2n) reaction leading to the $\alpha$-decaying nucleus $^{246}$Fm ($b_\alpha = 92\%$) and thus to $^{242}$Cf. Since $^{246}$Fm possesses a SF branch, $b_{sf} = (8 \pm 3)\%$ [1], its SF detection could be used to calibrate the yield of $^{246}$Fm and hence that of $^{242}$Cf. Two $^{208}$Pb + $^{40}$Ar bombardments were performed at two different periods of the target wheel revolution, $T_{rev} = 10$ s and 17 min, which were chosen according to the $T_{1/2}$ values of $^{246}$Fm and $^{242}$Cf. In the first bombardment with the $^{40}$Ar beam dose of $3 \times 10^{17}$ we observed 840 SF events distributed in time with $T_{1/2} = 1.5 \pm 0.1$ s [1]), without indications for any longer-lived SF components. This half-life value agrees well with the most accurate previous determinations of $1.2 \pm 0.2$ s and $1.5 \pm 0.3$ s reported for $^{246}$Fm in Refs. [10,11], respectively. From our data, the $^{246}$Fm production cross section was determined to be $19 \pm 9$ nb. The second $^{208}$Pb + $^{40}$Ar bombardment with the beam dose of $1.4 \times 10^{18}$ was performed to search for SF of $^{242}$Cf. Only 5 SF events were detected and thus the SF stability of $^{242}$Cf was proven to be rather high. It is characterized by $b_{sf} \leqslant 1.4 \times 10^{-4}$ and $T_{sf} \geqslant 1.5 \times 10^6$ s.

The SF stability of $^{240}$Cf was probed by using the $^{208}$Pb + $^{34}$S reaction. In a bombardment performed with the $^{34}$S beam dose of $1.2 \times 10^{18}$ at $T_{rev} = 5$ min we revealed a fission activity (65 events) with $T_{1/2} = 0.9 \pm 0.2$ min, at a cross-section level of 20 pb (see Table 1 and Fig. 1). We assigned this activity to the SF branch of the $\alpha$-decaying nuclide $^{240}$Cf on the basis of the following observations and arguments. First, its $T_{1/2}$ value agrees with $T_{1/2} = 1.06 \pm 0.15$ min known for the $\alpha$ decay of $^{240}$Cf [1]. Second, the cold-fusion-type reactions $^{206,207,208}$Pb + $^{34}$S specified by the so-called minimum excitation energy $E^*_{min}$ of the composite systems (i.e., the excitation energy at the Bass fusion barrier [13]) of about 33.5 MeV are similar to the well-studied $^{206,207,208}$Pb + $^{40}$Ar reactions characterized by $E^*_{min} \approx 31.5$ MeV. Hence, 2n- to 4n-evaporation channels are expected to be the most probable ones in the $^{208}$Pb + $^{34}$S system, as it is the case in the reactions $^{208}$Pb($^{40}$Ar, $x$n) (for a summary of measured

[1] Please note that all half-life values reported in the present paper were determined by using the maximum-likelihood method [9], with confidence limits corresponding to a decrease in the likelihood function by $e$ times with respect to its maximum value; the indicated confidence limits reflect statistical uncertainties only.

Table 1
Summary of experimental results on the production of SF activities in bombardments of $^{206,207,208}$Pb target nuclei with $^{34}$S and $^{36}$S projectiles

| Reaction | $T_{rev}$ | Beam dose ($10^{17}$) | $N_{sf}$ [a] | $T_{1/2}$ [b] | Assignment | $x$n channel | $\sigma_{sf}$ [c] (nb) |
|---|---|---|---|---|---|---|---|
| $^{208}$Pb+$^{34}$S | 5 min | 12 | 65 | $0.9\pm0.2$ min | $^{240}$Cf | 2n | $0.02\pm0.01$ |
| $^{206}$Pb+$^{36}$S | 5 min | 1.5 | 38 | $0.8^{+0.3}_{-0.2}$ min | $^{240}$Cf | 2n | $0.10\pm0.05$ |
| $^{208}$Pb+$^{34}$S | 0.2 s | 3 | 387 | $20\pm2$ ms | $^{238}$Cf | 4n | $0.5\ \pm0.2$ |
| $^{207}$Pb+$^{34}$S | 0.2 s | 1.5 | 425 | $26\pm4$ ms | $^{238}$Cf | 3n | $1.1\ \pm0.5$ |
| $^{206}$Pb+$^{34}$S | 0.2 s | 3 | 244 | $25^{+9}_{-6}$ ms | $^{238}$Cf | 2n | $0.3\ \pm0.1$ |
| $^{206}$Pb+$^{36}$S | 0.2 s | 0.5 | 100 | $15^{+4}_{-3}$ ms | $^{238}$Cf | 4n | $0.7\ \pm0.3$ |
| $^{207}$Pb+$^{34}$S | 15 s | 4 | 63 | $2.4^{+0.8}_{-0.4}$ s | $^{237}$Cf | 4n | $0.05\pm0.02$ |
| $^{206}$Pb+$^{34}$S | 15 s | 10 | 121 | $1.9\pm0.3$ s | $^{237}$Cf | 3n | $0.05\pm0.02$ |

[a] Total number of detected SF events.
[b] Deduced by using the maximum-likelihood method [9]; the indicated confidence limits of $T_{1/2}$ reflect statistical uncertainties only. In calculating $T_{1/2}$ values for $^{238}$Cf, small contributions from longer-lived SF activities of $^{240}$CF and $^{237}$Cf were taken into account.
[c] Cross sections for SF branches. The $\sigma_{sf}$ values were estimated from thick-target yields assuming $\Delta E_{FWHM} = 9\pm2$, $10\pm2$, and $12\pm2$ MeV for the widths of the excitation functions of the 2n-, 3n-, and 4n-evaporation channels, respectively (see, e.g., Refs. [6,8,12] and references therein). In the case of $^{238}$Cf ($b_{sf} \approx 1$), the $\sigma_{sf}$ values give total production cross sections.

cross sections of the $^{206,207,208}$Pb + $^{40}$Ar fusion-evaporation reactions, see Ref. [12]). We note, however, that the $^{34}$S-induced reactions on targets of $^{206,207,208}$Pb lead to Cf nuclides which are more neutron-deficient ($N$ = 138–142) compared to Fm nuclides ($N$ = 142–146) produced by the $^{40}$Ar-induced reactions. This fact should be reflected by a corresponding decrease in cross sections for $x$n channels in the $^{34}$S case. Finally, the odd-$A$ nucleus $^{239}$Cf as well as the isotope $^{238}$Cf (see below) cannot be sources of the 0.9 min fission activity, whereas a possible contribution from EC-delayed fission with $T_{1/2} = 2.4 \pm 0.1$ min in the decay chain $^{238}\text{Bk} \xrightarrow{EC} {}^{238}\text{Cm}$ [8,14] is expected to be small. Assuming for the $^{208}$Pb($^{34}$S, 2n) reaction a cross section of $\sim 1$ nb , we obtain an order-of-magnitude estimate $b_{sf} \sim 2 \times 10^{-2}$ for $^{240}$Cf. Evidently, a more accurate $b_{sf}$ determination for $^{240}$Cf requires an absolute measurement of the $^{208}$Pb($^{34}$S, 2n) reaction cross section.

The observation of the SF decay mode of $^{240}$Cf was confirmed by the results of a bombardment of $^{206}$Pb with $^{36}$S that we performed at $T_{rev}$ = 5 min. As Table 1 shows, some 40 SF events distributed in time with $T_{1/2} = 0.8^{+0.3}_{-0.2}$ min were detected in this bombardment. This result provides also a possibility of making the interesting comparison between the 2n-evaporation cross sections of the complete fusion reactions $^{206}$Pb + $^{36}$S and $^{208}$Pb + $^{34}$S with $E^*_{min}$ = 29.8 and 33.5 MeV, respectively, leading to the same compound nucleus $^{242}$Cf. For the $^{36}$S-induced reaction, the measured 2n-evaporation cross section proved to be $\approx 5$ times larger compared to the $^{34}$S case. This may be due to the lowered $E^*_{min}$ value in the former reaction, which is somewhat more appropriate for the sub-barrier 2n-evaporation channel.

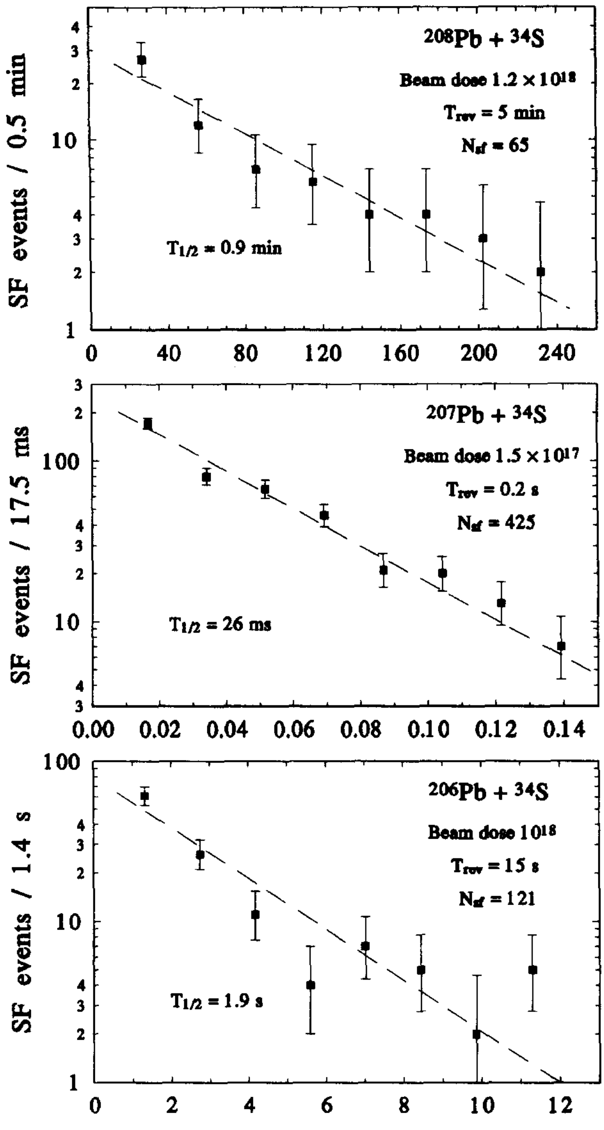


Fig. 1. Time distributions of SF events detected in some particular bombardments $^{208}Pb + ^{34}S$, $^{207}Pb + ^{34}S$. and $^{206}Pb + ^{34}S$. See also Table 1.

To explore the SF stability of still lighter, unknown isotopes of Cf, we carried out two $^{207}Pb + ^{34}S$ bombardments with the wheel revolution periods $T_{rev} = 0.2$ and 15 s. With $T_{rev} = 0.2$ s, a short-lived fission activity was discovered – we detected 425 fission events distributed in time according to $T_{1/2} = 26$ ms (see Fig. 1 and Table 1). The yield of these events corresponds to a cross section of about 1 nb. With $T_{rev} = 15$ s, we observed 63 fission events distributed in time with $T_{1/2} \approx 2$ s (Table 1); these events appeared with a cross section of $\approx 50$ pb.

Further, we made two $^{206}$Pb + $^{34}$S bombardments, again with $T_{rev} = 0.2$ and 15 s. As Table 1 and Fig. 1 show, the pattern of fission events observed was qualitatively similar to that of the $^{207}$Pb + $^{34}$S case.

Thus, we detected two new fission activities. It is quite clear that neither EC-delayed fission of neutron-deficient Bk or Am nuclides nor SF of light Cm or Pu nuclides can provide a source of the very short-lived fission activity with $T_{1/2} \approx 25$ ms. As can be inferred from systematics [15], the appearance of a new spontaneously fissioning isomer with such a half-life value is also absolutely improbable in the region of neutron-deficient actinide nuclei. From these considerations and from the data presented in Table 1 it follows that the short-lived SF activity should belong to the new Cf isotope, $^{238}$Cf, produced via the 3n- and 2n-evaporation channels on $^{207}$Pb and $^{206}$Pb, respectively. To verify this assignment once again, we performed a bombardment of $^{208}$Pb + $^{34}$S at $T_{rev} = 0.2$ s. Indeed, we again detected the 20 ms SF activity (387 events) with a cross section of $\approx 0.5$ nb.

Finally, we clearly identified the $\approx 20$ ms SF activity in a bombardment of $^{206}$Pb with $^{36}$S carried out at $T_{rev} = 0.2$ s. This allows us to compare straightforwardly the cross sections of the fusion-evaporation reactions $^{206}$Pb($^{36}$S, 4n) and $^{208}$Pb($^{34}$S, 4n) leading to $^{238}$Cf. As seen from Table 1, these cross sections proved to be virtually equal.

From predictions [16,17] and experimental systematics shown in Fig. 2, $^{238}$Cf is expected to have a partial $\alpha$-decay half-life of a few seconds. Therefore we conclude

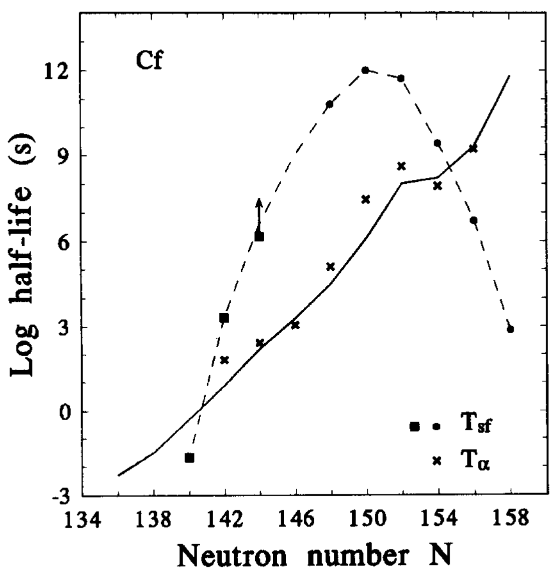


Fig. 2. Experimental partial half-lives for $\alpha$ and SF decay of the even–even Cf isotopes. The squares show the data of the present work. The solid line shows partial $\alpha$ half-lives calculated by Smolanczuk and Sobiczewski [17].

Table 2
Decay properties of the light Cf isotopes studied in the present work

| Isotope | Half-life | $b_{sf}$ | $T_{sf}$ |
|---|---|---|---|
| $^{242}$Cf | 3.4 ±0.2 min [a] | $\leqslant 1.4\times10^{-4}$ | $\geqslant 1.5\times10^{6}$ s |
| $^{240}$Cf | 1.06±0.15 min [a] | $\sim 2\times10^{-2}$ | $\sim 3\times10^{3}$ s |
| $^{238}$Cf | 21 ±2 ms | $\approx 1$ | ≈ 21 ms |
| $^{237}$Cf | 2.1 ±0.3 s | $\sim 10^{-1}$ | ~ 20 s |

[a] Data from Ref. [1].

that the isotope $^{238}$Cf gives a new example of a short-lived spontaneously fissioning nucleus. By analyzing the whole set of data from the four reactions employed to produce $^{238}$Cf (see Table 1), we determined its half-life to be 21 ± 2 ms.

Although some additional bombardments would be desirable to exclude completely few minor SF or EC-delayed fission sources (e.g., $^{236}$Bk), the most probable origin of the SF activity with $T_{1/2} = 2.1 \pm 0.3$ s seems to be due to a perceptible SF branch of the new, odd-*A* isotope $^{237}$Cf which is expected [16,17] to be predominantly an $\alpha$ emitter. In this case, the evaluation of the SF branch is essentially dependent on the assumptions regarding the absolute cross sections of the $^{207}$Pb($^{34}$S, 4n) or $^{206}$Pb($^{34}$S, 3n) reactions. Using a cross-section value of 0.5 nb, we obtain an order-of-magnitude estimate $b_{sf} \sim 10^{-1}$ and, accordingly, a SF hindrance factor of $\gtrsim 10^3$.

The decay properties of the light isotopes of Cf studied in the present work are summarized in Table 2.

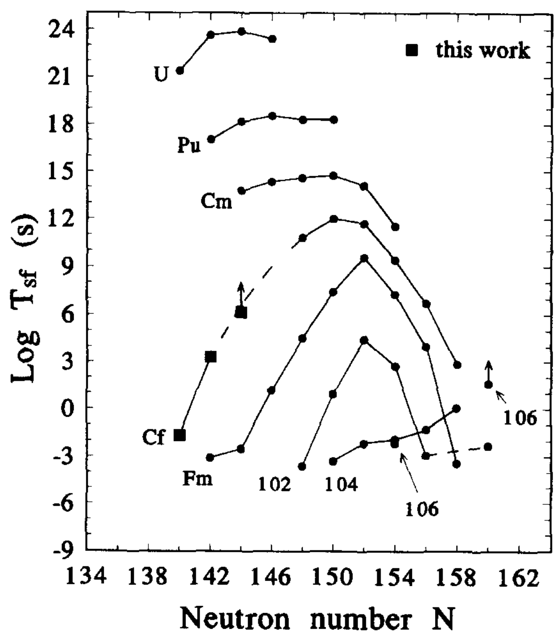


Fig. 3. Systematics of partial SF half-lives for even–even nuclei with $Z = 92$ through 106. For origins of the data points, see Refs. [1,6,18,19] and references therein.

## 4. Conclusion

We have explored the SF stability of the light Cf nuclei in a wide range of $N$. The dramatic effect of the deformed neutron shell $N = 152$ on the SF half-lives was demonstrated by revealing a $T_{sf}$ decrease from $6 \times 10^{10}$ s for $^{246}$Cf down to $2 \times 10^{-2}$ s for $^{238}$Cf (see Fig. 3). Our experiments resulted in the production of the new nuclide $^{238}$Cf and gave an indication of the production of the new isotope $^{237}$Cf. The identification of $^{238}$Cf was confirmed in recent experiments [20] performed by using the Dubna gas-filled recoil separator [21].

We obtained also the first experimental information about cross sections $\sigma_{xn}$ of $^{34}$S- and $^{36}$S-induced fusion-evaporation reactions occurring on $^{206,207,208}$Pb target nuclei. The measured $\sigma_{xn}$ values were found to be in the range of 0.3 to 1.1 nb for $x = 2$ to 4. A comparative study of the $^{206}$Pb($^{36}$S,$x$n) and $^{208}$Pb($^{34}$S,$x$n) reactions with $x = 2,4$ leading to the same compound nuclei and final products has shown that the $\sigma_{2n}$ value is some 5 times larger in the $^{36}$S case, while $\sigma_{4n}$ values are practically equal. An interesting extension of the present studies will be associated with the use of $^{32}$S projectile to attempt the further production of new, ultra-neutron-deficient Cf species.

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