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Reexamination of the Claim of Marinov *et al.* on Discovery of Element 112

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Abstract

Marinov *et al.* have detected spontaneous fission events in sources separated from tungsten targets irradiated with 24 GeV protons. These fission events could not be attributed to actinides or to any other known isotope. Marinov *et al.* propose that fission events are due to production of element 112 (Eka-Hg) in the tungsten target. We have addressed Marinov's claim with a new analysis of their data and modern theoretical model calculations of possible interactions. Using data available in the literature the spontaneous fission half-life of the Eka-Hg was estimated to be ~74 days. This is dramatically longer than the half-life obtained for $^{283}_{112}\mathrm{Cn}$, produced in the fusion of energetic $^{48}\mathrm{Ca}$ ions with $^{238}\mathrm{U}$. Monte Carlo calculations show that enough Sr isotopes are produced in the tungsten target to make the production of element 112 via fusion of Sr and W feasible; however, if such fusion was possible it had to be deep sub-barrier fusion.

Keywords

Element 112, Tungsten Target, Spallation Products, Heavy Ion Fusion

1. Introduction

Forty seven years ago Marinov *et al.* [1] [2] [3] reported evidence for possible synthesis of a super heavy (SH) element (Z = 112) in the irradiation of tungsten target with 24 GeV protons.

They irradiated three W-targets, each with mass of 33 g and thickness 120 g·cm^{-2} using the CERN PS accelerator. These targets are referred to as W1, W2 and W3. Table 1 gives some information on irradiation of the targets [1]. The W1 target was available for analysis 3 - 4 month after the end of the irradiation.

Table 1. Irradiation data for three tungsten targets from [1].

Target	Irradiation period	Number of protons on target	Time between end of irradiation and start of analysis
W1	About a year	2×10^{18}	3 to 4 months
W2	About 4 months	7×10^{17}	A few days
W3	NA	NA	NA

For reasons given in [1] most of the reported experimental results and findings are for W2-target and we will focus on this target as well.

The synthesis of isotope ${}^{283}_{112}\mathrm{Cn}$ of element 112 via ${}^{48}_{20}\mathrm{Ca} + {}^{238}_{92}\mathrm{U} \rightarrow {}^{286}_{112}\mathrm{Cn} \rightarrow {}^{283}_{112}\mathrm{Cn} + 3\mathrm{n}$ reaction was reported in 1999 [4] [5]. Prior to that in 1996 [6], production of ^{277}Cn in ^{208}Pb (^{70}Zn, n) reaction has been reported. The half-life of ^{283}Cn is 4 s and ^{277}Cn has a half-life of 0.69 ms.

Discovery of the short-lived isotopes of element 112 does not rule out the existence of long-lived isotopes of this element.

Marinov *et al.* [1] [2] [3] suggest that neutron deficient long lived isotopes of element 112 can exist and may be produced with higher cross-sections.

Their claim is based on the following assumptions and observations:

1) Element 112 would be a chemical homologue of mercury (Eka-Hg).

2) Detection of spontaneous fission events in Eka-Hg sources separated from proton irradiated W-targets, which could not be attributed to actinides or to any other known isotope.

Heavy ion fusion experiments such as [4] [6] obviously will not be able to detect the long-lived SH isotopes because very few atoms of these are produced. On the other side, experiments like that of the Marinov *et al.* will not be able to detect the short-lived SH isotopes because of the required long chemical separation times.

Several attempts to reproduce the experimental findings of Marinov *et al.* were inconclusive, see e.g. [7] [8] [9] [10]. The rejection by Barber *et al.* [11] of the claim of Marinov *et al.* was recently addressed by Brandt *et al.* [12] who point out that presently unexplained findings may indicate novel reaction paths leading to unexpected results.

In this paper we use the experimental results given in the publications of Marinov *et al.* to investigate the feasibility of reproducing their findings in possible future experiments.

2. Calculation of Spontaneous Fission Half-Life of Eka-Hg

Table 2 gives a timeline of the experiments and measurements on W2 target after the end of the proton irradiation, taken mainly from [1]. For those periods where no information was available in the literature, reasonable times have been assigned by us. The timeline given in the last column of **Table 2** is used in this report.

The most unambiguous experimental results reported [1] are the fission

Table 2. Timeline of experiments and measurements after the end of the irradiation.

Chemical separation and alpha activity measurements			
Row number	**Description**	**Experimental times from Ref. 1**	**Times used in the calculations**
1	Time from end of irradiation to start of chemical separation	A few days	4 d
2	Chemical separation period	NA	2 d
3	Alpha counting of Eka-Hg source from W2	406 h (16.92 d)	16.92 d
4		236 h (9.83 d)	9.83 d
5	Time interval between two alpha counting (3 and 4 above)	24 d	24 d
6	Alpha counting of Eka-Hg source from W1 sample	280 h (11.7 d)	11.7 d
7	Time gap between alpha counting of the W2 and W1 sources	NA	0.5 d
8	Background alpha counting	10 d	10 d
9	Total time from end of irradiation to end of alpha counting	NA	79 d
	Fission event detection		
10	Time gap between end of alpha counting and start of fission track recording	NA	0 d
11	First Makrofol foil exposure	7 d	7 d
12	Second Makrofol foil exposure	14 d	14 d
13	Time gap between two exposures	4 d	4 d
14	Time spent for thickness reduction of Eka-Hg source from W2	NA	1 d
15	First Makrofol foil exposure (reduced sample thickness)	8 d	8 d
16	Second Makrofol foil exposure (reduced sample thickness)	8 d	8 d
17	Time gap between two Makrofol exposures	13 d	13 d

events recorded after attempts to reduce the thickness of the Eka-Hg source prepared from W2 target, (Table 2, rows 15 - 17). The spontaneous fission half-life of Eka-Hg can be calculated using these data.

The fission events in the source were recorded as tracks in Makrofol KG polycarbonate foils (hereafter referred to as Makrofol) that were placed in close contact with the surface of the source.

First of all one need to convert the observed fission tracks in the Makrofol foils to the number of fission events in the source.

Fission track density ρ (tracks cm^{-2}) in a foil in contact with a fission source is related to the number of fission events per unit volume of the source, N_v by the following relationship [13] [14]

$$\rho = n\varepsilon\mu dN_v \tag{1}$$

where:

n is number of fragments per fission event (we use $n = 2$);

ε is fission track detection efficiency of Makrofol foil which is ~1;

μ is a parameter that depends on fission source thickness d and range of the fission fragments in the source material, R.

For a thin source ($d < R$) equation 1 can be written as

$$N_c = \eta N_f \tag{2}$$

where N_c is total number of fission tracks detected in the Makrofol foil, N_f is number of fission events in the source and η *is*

$$\eta = n\varepsilon\mu \tag{3}$$

For a thin source, μ is given by [13] [14]

$$\mu = \frac{1}{2}\left(1 - \frac{d}{2R}\right) \tag{4}$$

Determination of μ requires knowledge of source thickness, and range of fission fragments. The authors of [1] mention that the thickness of the Eka-Hg source was 2 mg·cm^{-2}, but the density of the source material is not given. If we assume that source material has a density equivalent to the density of HgO (11.14 g·cm^{-3}), thickness of the source and range of fission fragments in the source material may be calculated as 1.8 μm and 8 μm, respectively.

Using the Equation (4) and values of n and ε we obtain $\eta = 0.89$.

The first Makrofol foil was in contact with W2 source for 8 days and showed 28 tracks (row 15 of **Table 2**). Thus

$$\frac{28}{\eta} = N_1\left(1 - \mathrm{e}^{-8\lambda}\right) \tag{5}$$

where N_1 is the number of Eka-Hg nuclei in the source at the start of the 1st Makrofol exposure (row 15 of **Table 2**) and λ is the spontaneousfission decay constant.

The second Makrofol exposure (row 16 of **Table 2**) started 13 days after the end of the first one and the number of tracks in this foil after 8 days of exposure was 23. The number of Eka-Hg nuclei at the start of the second exposure is

$$N_1' = N_1\mathrm{e}^{-(8+13)\lambda}$$

Thus for the second Makrofol exposure we get

$$\frac{23}{\eta} = N_1\mathrm{e}^{-21\lambda}\left(1 - \mathrm{e}^{-8\lambda}\right) \tag{6}$$

From Equations (5) and (6) we obtain: $\lambda = 9.37 \times 10^{-3}$ d^{-1} and $t_{1/2} \approx 74$ d.

The number N_1 of Eka-Hg at the start of the fission track measurements can be calculated using Equation (5) or Equation (6);

$$N_1 = 436$$

Taking into account that fission track recording (row 15 of **Table 2**) started 105 days after the end of the irradiation, the number of Eka-Hg at the end of the irradiation was:

$$N = 1165$$

This number must be corrected for the number of the produced Eka-Hg nuclei that have decayed in the course of the 4 month irradiation period.

The total number of 24 GeV protons that was delivered to W2 target from the PS accelerator of CERN in late 1960s was 7×10^{17}. With a pulse repetition time of 2.4 s and number of protons per pulse of 10^{12} [15] [16] total actual target exposure time has been ~1.68×10^6 s (~19 days). This implies that ~84% of the irradiation time (~4 month) the target did not receive any proton while the produced Eka-Hg nuclei were decaying. Therefore a large number of the nuclei of interest have been lost prior to the start of chemical separation. An accurate calculation of this lost activity requires knowledge of the exact timeline of the irradiation and pulse intensity distribution during the irradiation period. In absence of such data and taking into account the calculated half-life, we estimate that at least 60% of the activity survived to the end of the irradiation. Thus the corrected number of the produced Eka-Hg is

$$N_0 \approx 1941$$

3. Production of Spallation Residues in Interaction of 24 GeV Protons with Tungsten Target

It is assumed that the production of element 112 in a W-target irradiated with 24 GeV protons is possible via fusion of Sr isotopes (spallation products) with W nuclei of the target. Marinov *et al.* [3] and Kolb *et al.* [17] suggest
$^{88}_{38}\mathrm{Sr} + {}^{184}_{74}\mathrm{W} \rightarrow {}^{272}_{112}\mathrm{Cn}$ and $^{86}_{38}\mathrm{Sr} + {}^{186}_{74}\mathrm{W} \rightarrow {}^{272}_{112}\mathrm{Cn}$ reactions.

We used HTAPE card in the MCNPX 2.7 code [18] to calculate the types and numbers of residual nuclei produced in the interaction of 24 GeV protons with a ^{nat}W-target. The target specifications were same as given by [1], *i.e.* cylindrical ^{nat}W targets of thickness (length) 120 g·cm^{-2} and mass of 33 g, which translates to a cylinder of diameter 0.6 cm and length 6.2 cm. In the calculations the direction of the proton beam coincided with the target axis.

Figure 1(a) shows the charge distribution of all isotopes (spallation products) and **Figure 1(b)** illustrates the yield of Sr isotopes (Z = 38) produced in the interactions. All together sixteen isotopes of Sr with (A = 77 to 92) with total yield of 8.32×10^{-3} per proton are produced. The spallation residue yield is strongly dependent on the target length (along the beam line); for a shorter target length it will be less than the above given figure.

From **Figure 1(b)** it is evident that most abundantly produced isotopes of Sr (62%) are those with mass numbers of 82, 83 and 84. Fusion of these isotopes with ^{184}W and ^{186}W will result in

$$^{82}_{38}\mathrm{Sr} + {}^{184}_{74}\mathrm{W} \rightarrow {}^{266}_{112}\mathrm{Cn} \quad \text{and} \quad {}^{82}_{38}\mathrm{Sr} + {}^{186}_{74}\mathrm{W} \rightarrow {}^{268}_{112}\mathrm{Cn}$$

$$^{83}_{38}\mathrm{Sr} + {}^{184}_{74}\mathrm{W} \rightarrow {}^{267}_{112}\mathrm{Cn} \quad \text{and} \quad {}^{83}_{38}\mathrm{Sr} + {}^{186}_{74}\mathrm{W} \rightarrow {}^{269}_{112}\mathrm{Cn}$$

$$^{84}_{38}\mathrm{Sr} + {}^{184}_{74}\mathrm{W} \rightarrow {}^{268}_{112}\mathrm{Cn} \quad \text{and} \quad {}^{84}_{38}\mathrm{Sr} + {}^{186}_{74}\mathrm{W} \rightarrow {}^{270}_{112}\mathrm{Cn}$$

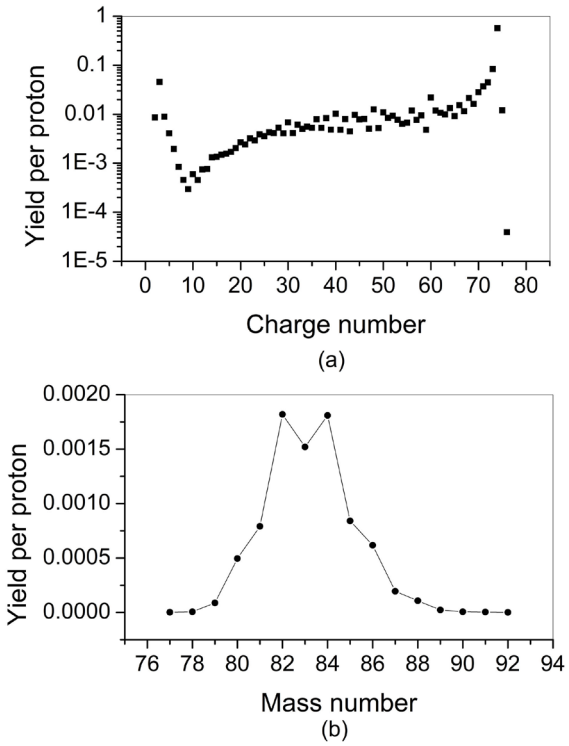

Figure 1. Spallation residues produced in 24 GeV p + ^{nat}W interactions. (a) Yield of all spallation products (b) Yield of Sr isotopes. Lines connecting data points are to guide the eye.

The mass numbers of Eka-Hg in the above given interactions are less than 272 given by Marinov *et al.* [3] and the products are neutron deficient isotopes.

Combined production rate of these three Sr isotopes ($^{82\text{-}84}$Sr) is 5.15×10^{-3} per proton which is 7 times higher than the combined production rates of ^{88}Sr and ^{86}Sr (7.25×10^{-4}), considered by Marinov *et al.* to be the principal participants in the fusion reaction with W nuclei [3].

4. Estimation of the Eka-Hg Production Cross-Section

An accurate estimation of the production cross-section of Eka-Hg with the available data is not possible for the following reasons:

1) The exact number of protons that hit the target is not known. From the publications it is not clear how the targets were irradiated? What was the proton direction with respect to the target axis? How was the beam fluence measured? Do the given proton numbers refer to those that hit the target or are they the numbers of the protons that were extracted from the accelerator?

Lack of this information in the publications implies that measurement of the production cross-section of Eka-Hg was not part of the experimental plan of Marinov *et al.*

2) As already mentioned, 16 isotopes of Sr are produced with different production rates. Out of these ^{88}Sr and ^{86}Sr have been nominated to produce ^{272}Cn via fusion with W nuclei. Each Sr isotope has its own fusion reaction cross-section with

a given W-isotope of ^{nat}W.

However a rough estimation of the production cross-section of ^{272}Cn is possible with the following assumptions:

1) The given proton numbers are actually those that hit the target.

2) The beam direction was along the target axis.

3) Cross-sections for $^{88}_{38}Sr + ^{184}_{74}W \rightarrow ^{272}_{112}Cn$ and $^{86}_{38}Sr + ^{186}_{74}W \rightarrow ^{272}_{112}Cn$ reactions are similar to each other.

Then, using the calculated yields of the Sr isotopes of the interest, isotope fractions of the W-isotopes in ^{nat}W and a beam dose of 7×10^{17} protons, one obtains a production cross-section of 1.6 nb.

5. Case of Uranium Target

Production of element 112 is possible via fusion of ^{238}U with Ca. Similar to the case of the W-target, we calculated the spallation residue yields in the interaction of 24 GeV protons with a U-target, of the same shape and dimensions as the W-target. The mass of U-target will be slightly less (~1.5%) than that of the W because of the difference in densities.

Figure 2(a) shows the charge distribution of all spallation residues produced in the interactions. Obviously the hump in the middle of the distribution is populated mainly with the ^{238}U-fission products.

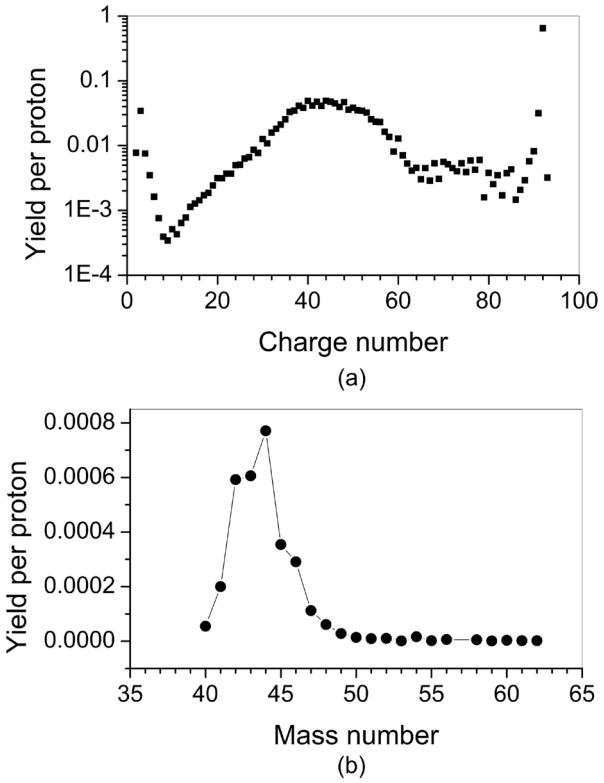

Figure 2. Spallation residues produced in 24 GeV p + ^{238}U interactions. (a) Yield of all spallation products; (b) Yield of Ca isotopes. Lines connecting data points are to guide the eye.

All together 22 isotopes of Ca with A = 40 - 62 are produced, with a total yield of 3.14×10^{-3} per proton. This is about 2.7 times less than the yield of Sr-isotopes produced in the W-target. Isotopes ^{42,43,44}Ca make about 63% of the total Ca yield. The yield of ^{48}Ca relevant to the experiments of Oganessian *et al.* [4] is less than 2% of the total Ca yield.

If isotopes ^{286}Cn and ^{283}Cn were produced in the proton irradiated U-target, they would not be detectable by chemical separation method because of their very short half-lives. If fusion of Ca isotopes (from the spallation residues) with ^{238}U nuclei in the target was possible then in order to produce the isotope ^{272}Cn, the post fusion compound nuclei must evaporate between 6 and 28 neutrons which is highly improbable for energetic reasons.

Boos *et al.* [19] irradiated a uranium target of mass 65 g with $(4 \pm 1) \times 10^{17}$ protons of 24 GeV using the CERN PS accelerator. The irradiation period was 20 days and chemical separation process of the target started three weeks after the end of the irradiation. They prepared two thin sources of HgS as carrier for Eka-Hg. For detection of the possible spontaneous fission activity in the HgS sources mica foils were placed in close contact with source 1 and 2 for periods of 2.6 y and 0.6 y, respectively. The mica foil placements on source 1 and 2 started 2.4 month and 1.4 month after the end of the irradiation, respectively. After exposure the mica foils were etched and scanned under an optical microscope; not a single fission track was detected.

The theoretical calculations given in this section confirm the experimental observations of Boos *et al.* [19].

6. Energy Distribution of the Spallation Products

Figure 3 shows the energy distribution of the all residual nuclei in the target volume, produced in interaction of the 24 GeV protons with ^{nat}W-target. As can be seen, almost all of the residual nuclei, regardless of their charge and mass have energies less than 100 MeV, well below (about one third of) the coulomb barrier

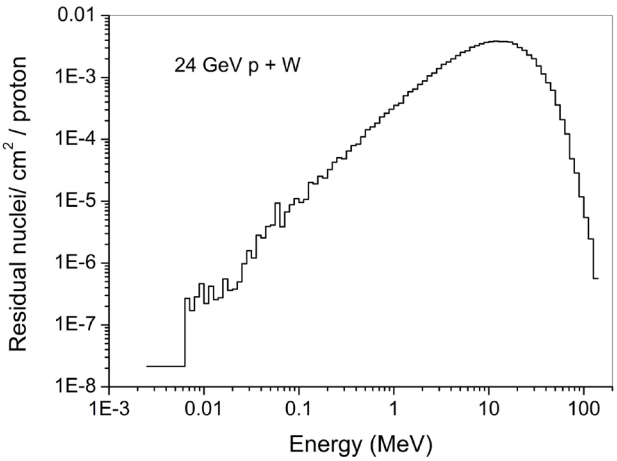

Figure 3. Energy distribution of the spallation residues produced in the interaction of 24 GeV protons with tungsten target of thickness 120 g·cm^{-2} and total mass of 33 g.

height of the (Sr + W) system. Therefore, if fusion of these nuclei in proton irradiated W-target were possible, then it had to be deeply sub-barrier fusion.

7. Effects of Proton Beam Energy on the Energy Distribution of Spallation Residues

Figure 4 shows the spectra of the neutrons and protons in interaction of 24 GeV protons with a tungsten target of thickness 120 g·cm^{-2} and mass 33 g. On average for every incident proton, there are 0.71 protons with energy greater than 23.7 GeV in the exit channel. Neutrons carry 5.2% and protons 80.6% of the beam energy. With energy and target size [1] used in the calculations majority of the beam protons leave the target without being engaged in an inelastic nuclear interactions and only lose small fraction of their energy via electronic interactions.

To increase the production rate of Sr nuclei in the W-target one need to:

1) Increase the target length to reduce number of the non-interacting beam protons.

2) Increase the beam dose at optimum proton energy.

Figure 5 shows the energy distribution of the heavy spallation residue (Z > 2) at different incident proton energies in interaction with a tungsten target of length 20 cm and diameter 0.6 cm. The energies of the spallation residue correspond to those at the moment of their production (*i.e.* maximum possible energy) and the energy degradation in the target environment is not taken into account. We chose a target with small diameter to reduce the amount of the unnecessary and disruptive radioactivity production. Obviously for a target of diameter 0.6 cm one prefers to have a proton beam of diameter less than 0.6 cm.

Spectra of the heavy ion residue at beam energies in the range of 3 GeV < $E_p \leq$ 24 GeV do not change significantly. For clarity of the figure in Figure 5 only the spectra at E_p > 5 GeV are shown. Most important, the total number of the produced ions per incident proton at different beam energies remains almost constant (Table 3). This is expected because; the heavy spallation residue mainly result from the proton induced fission of the target tungsten nuclei, a process that takes place after the cascade, pre-equilibrium and evaporation stages of the spallation reaction.

Table 3. Total number of heavy spallation residues produced on irradiation of a 20 cm long W target with protons of different energies.

Proton energy (GeV)	Total number of heavy ion residue per proton	Statistical uncertainty
0.5	2.5500E−02	1.10E−04
1	2.4400E−02	1.07E−04
5	2.5400E−02	1.14E−04
10	2.2200E−02	9.56E−05
15	2.2500E−02	9.69E−05
20	2.2900E−02	1.01E−04
24	2.3100E−02	1.02E−04

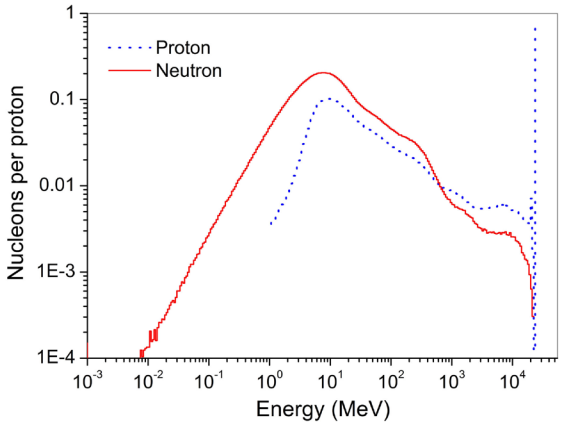

Figure 4. Spectra of the neutrons and protons escape the W-target (diameter 0.6 and length 6.2 cm) on its irradiation with 24 GeV protons.

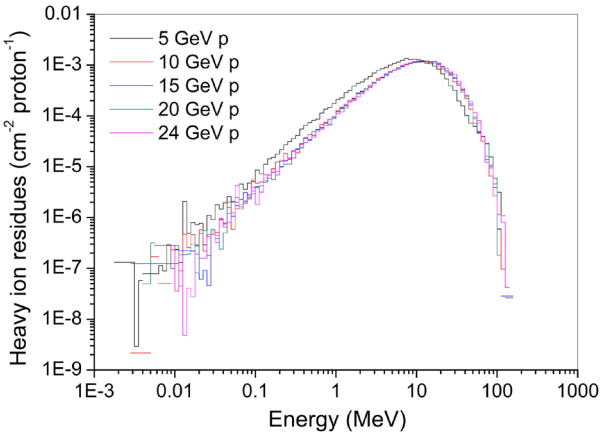

Figure 5. Energy distribution of the heavy nuclei (Z > 2) in irradiation of a W-target of diameter 0.6 cm and length 20 cm with protons of different energies.

Thus we conclude that if one intends to search for the interaction of any heavy spallation residue with the target nuclei, very high energy proton beam is not required.

8. Discussion

From the available published data [1] the half-life of the spontaneous fission events detected in the source from W2 target was estimated to be ~74 days.

Monte Carlo calculations show that, in the irradiation of a tungsten target with 24 GeV protons 16 isotopes of Sr are produced. The yields of these isotopes are sufficiently high to justify the assumption of the fusion reaction of Sr + W in the target. However, because of low energies of the spallation residues, if fusion of Sr and W nuclei in proton irradiated W-target were possible, then it had to be deeply sub-barrier fusion.

If sub-barrier fusion of (U + Ca) system is possible, auranium target irradiated with 24 GeV protons is not expected to showany isotopes of element 112. Most abundantly produced Ca isotopes are ^{42,43,44,45}Ca where the product mass range

goes from ^{40}Ca to ^{62}Ca. The post fusion compound nuclei need to evaporate 6 to 28 neutrons to reach mass 272 which is very unlikely. Short lived isotopes of element 112 produced via irradiation of ^{238}U with Ca ions will not be detectable because of the required long chemical separation and sample preparation times.

The proton dose of the W1 target was 2×10^{18} over an irradiation period of about one year. Similar to the case of the W2 target, it is estimated that the actual irradiation period during which the W1 target was receiving protons, was about 58 days. The estimated half-life and duration of the proton irradiation of the W1 target and considering that fission track recording started ~200 days after the end of the irradiation, failure to detect any spontaneous fission activity in source prepared from W1-target may therefore be justified.

Monte Carlo calculations using the MCNPX code show that energy distribution of the heavy ion spallation residue do not change for a very wide range of the incident proton energies. Therefore, if Sr + W fusion in proton irradiated W-target is possible, then one does not need to have very high energy proton beam. A relatively long target at a proton energy of ≥3 GeV will be sufficient to produce the desired number of Sr nuclei in the tungsten target.

9. Conclusions

Findings of Marinov *et al.* imply the following possibilities:

1) Production of super-heavy element in proton irradiated tungsten target.

2) Production of very long-lived isotope of SHE which are not known so far.

3) Production of SHE in deep sub-barrier fusion reactions.

4) Production of SHE with much higher cross-sections than those known so far.

We believe that new experiments with today's knowledge, experimental facilities and techniques are required to examine and test the reproducibility or otherwise disprove the Marinov *et al.* claims.

As strontium isotopes in the proton irradiated W-target will not have energies greater than 100 MeV (fission fragment energy), the most straightforward method for examining the Z = 112 production in a W-target is to irradiate a thin foil of W with a very heavy dose of 100 MeV Sr ions and look for fission events.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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