Search for particle-stable tetraneutrons in thermal fission of ²³⁵U

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Background: The existence of a tetraneutron comprising four neutrons has long been debated.

Purpose: Motivated by a recent observation of particle-stable tetraneutrons, we investigated potential particle-stable tetraneutron emission in thermal neutron-induced ²³⁵U fission using a nuclear research reactor.

Methods: We performed γ -ray spectroscopy for a ⁸⁸SrCO₃ sample irradiated in a reactor core. Stable ⁸⁸Sr was expected to produce ⁹¹Sr by a tetraneutron-induced (⁴n, n) reaction; hence, observation of γ rays followed by β decay of ⁹¹Sr would indicate particle-stable tetraneutron emission.

Results: The γ -ray spectrum of an irradiated ⁸⁸SrCO₃ sample did not show any photopeak for ⁹¹Sr.

Conclusion: The emission rate of particle-stable tetraneutrons, if they exist, is estimated to be lower than 8×10^{-7} per fission at the 95% confidence level, assuming the cross sections of reactions induced by hypothetical particle-stable tetraneutrons.

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I. INTRODUCTION

Atomic nuclei composed of Z protons and N neutrons can be particle-stable systems, depending on Z and N. For each element with a fixed Z, a stable isotope satisfies $N \sim Z$; however, particle stability against neutron emission has a limit, which is known as the neutron drip line. A neutron drip line up to neon (N=10) has been established [1]. A highly debated question is whether a charge-neutral (Z=0) multineutron system can exist [2]; extensive searches for tetraneutrons (4 n) have been conducted using various methods.

In 2002, evidence of a bound tetraneutron formed in the breakup of ^{14}Be was reported [3]; a neutral particle was detected in a liquid scintillator with a signal significantly larger than that expected in the detection of a neutron. A ^{10}Be fragment was simultaneously detected leading the authors to conclude a bound tetraneutron was formed in the breakup of $^{14}\text{Be} \rightarrow ^{10}\text{Be} + ^4\text{n}$. However, an alternative interpretation that two or more neutrons from a resonant tetraneutron might rescatter in the liquid scintillator and cause such a large signal has also been suggested [4].

Later, a double-charge exchange reaction, $^4\text{He}(^8\text{He}, ^8\text{Be})$, was used to populate tetraneutron states. A peak at $0.83 \pm 0.65 \pm 1.25\,\text{MeV}$ above the 4n emission threshold was observed in the missing-mass spectrum, indicating the existence of a resonant state [5], although the statistical and systematic uncertainties made the result also consistent with a bound tetraneutron.

Recently, two experimental groups independently reported the presence of tetraneutrons in the form of bound [6] and resonant [7] states. Faestermann et~al. observed a peak structure in the $^{10}\mathrm{C}$ energy spectrum for the $^{7}\mathrm{Li}(^{7}\mathrm{Li}, ^{10}\mathrm{C})4n$ reaction. This was attributed to the formation of a bound tetraneutron state with an energy of $-0.42\pm0.16\,\mathrm{MeV}$, relative to the four-neutron threshold, along with the first excited state of $^{10}\mathrm{C}$ [6]. The negative energy suggests that the tetraneutron is particle-stable. Duer et~al. performed missing-mass spectroscopy on the $^{8}\mathrm{He}(p,p^{4}\mathrm{He})$ reaction and observed a resonance-like structure in the missing mass spectrum [7]. They determined the energy and width of the resonant state as $2.37\pm0.38\pm0.44\,\mathrm{MeV}$ and $1.75\pm0.22\pm0.30\,\mathrm{MeV}$, respectively.

From a theoretical perspective, a general consensus exists that a tetraneutron will not form a bound state if realistic two-body and three-body nuclear forces are used; i.e., the unexpected existence of a bound tetraneutron state has a profound impact on our understanding of the many-body neutron force [8–10]. The possible existence of an observable resonant state with a sufficiently narrow width is also under debate [2]. For example, the low-energy structures reported in Ref. [7] can be reproduced by considering the dineutron–dineutron correlation in the final state without assuming a tetraneutron resonant state [11].

In this work, we explore the possible emission of a hypothetically bound tetraneutron resulting from thermal neutron-induced fission of ²³⁵U in a nuclear research reactor. The dominant thermal fission process is binary fission, which leads to the emission of two heavy nuclear fragments together with 2.4 neutrons, on average; however, ternary fission involving a light nuclear fragment also occurs with a probability of 0.2% per fission [12,13]. ⁴He is the most frequently emitted nuclide, accounting for ~90% of ternary fission. Furthermore, hydrogen isotopes (tritons, protons, and deuterons, in

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descending order of emission rate), unstable helium isotopes (6 He, 8 He), and nuclides with $Z \geqslant 3$ are emitted in ternary fission. The emission rates of the two-neutron halo nuclei 11 Li and 14 Be, relative to those of 4 He, were measured as $(9.0 \pm 2.2) \times 10^{-8}$ and $(3.4 \pm 2.0) \times 10^{-8}$ [13], that is, on the order of 10^{-11} per fission. Under the assumption that the hypothetically bound tetraneutron is a ternary particle in uranium fission, the objective of this study was to evaluate the emission rate per fission.

An emitted tetraneutron is assumed to induce a secondary reaction with the nucleus in a sample irradiated in the reactor core, assuming that it has a sufficiently long half-life to travel to the sample from the fuel rod inside which it was produced. For example, a $(^4\mathbf{n}, n)$ reaction converts the stable isotope AZ into the isotope ^{A+3}Z . The $(^4\mathbf{n}, n)$ reaction may involve three-neutron transfer and/or one-neutron emission preceded by the formation of a compound nucleus $^{A+4}Z^*$. If ^{A+3}Z is unstable against β decay, a γ ray with a specific energy is emitted after β decay. The observation of γ rays from ^{A+3}Z indicates the existence of a bound tetraneutron unless a competing process forms ^{A+3}Z from a nuclide other than the AZ in the sample, such as an impurity.

The experimental procedure was the same as that used in the well-established instrumental neutron activation analysis (INAA) in radiochemistry [14]. In INAA, a trace element in a sample under irradiation is activated by neutron capture, i.e., the (n, γ) reaction, predominantly with thermal neutrons. The concentration of the trace element of interest can be determined using γ -ray spectroscopy, as the thermal neutron flux at the sample position and the neutron-capture cross section are known. In contrast to INAA, we determined the "tetraneutron flux" at the sample position using the concentration of ^{A}Z in the sample and the assumption of a $(^{4}n, n)$ reaction cross section.

To the best of our knowledge, only one study on tetraneutrons has been conducted in a research reactor, which was published in 1963 [15]. Two types of reactions, $^{14}\mathrm{N}(^4\mathrm{n},n)^{17}\mathrm{N}$ and $^{27}\mathrm{Al}(^4\mathrm{n},p2n)^{28}\mathrm{Mg}$, were investigated using samples of 3-amino-1,2,4-triazole (C₂H₄N₄) and pure aluminum. Neither delayed neutrons from $^{17}\mathrm{N}$ nor γ rays resulting from the $^{28}\mathrm{Mg}$ β decay were detected above the background level. The upper limits of tetraneutron emission per fission were estimated as 2×10^{-8} and 5×10^{-9} , from the neutron and γ -ray analyses, respectively.

Several attempts have been made to generate bound tetraneutrons or heavier multineutron systems via the spallation of heavy nuclei, such as uranium, using a high-energy beam (for details, see Section 2.2 in Ref. [2]). Detraz provided evidence for multineutron-bound states based on the observation of ⁷²Zn γ rays in a zinc sample after the spallation of tungsten by a 24 GeV proton beam [16]. However, De Boer *et al.* argued that high-energy tritons produced during spallation can penetrate a shield and induce the ⁷⁰Zn(t, p) ⁷²Zn reaction in the zinc sample [17]. Evidence of a bound multineutron system with $N \ge 6$ in α -induced ²³⁸U fission has also been reported [18], based on the observation of γ rays from ⁹²Sr, which were expected to be produced in a ⁸⁸Sr(N n, (N-4)n) ⁹²Sr reaction. Further, the cross section of photon-induced ²⁰⁹Bi(γ , 4n) ²⁰⁵Bi, slightly below and above

the threshold, has been measured by observing γ rays from 205 Bi [19]; nonobservation of the signal below the threshold and an estimated cross section above the threshold, much higher than predicted one, indicates the production of a resonant tetraneutron state rather than a bound state.

Motivated by the recent indication of bound tetraneutrons [6], we considered revisiting the tetraneutron search in a research reactor worthwhile. Contrary to spallation experiments with accelerators, the typical kinetic energy in thermal neutron-induced fission and possible secondary reactions is, at most, on the order of MeV per nucleon. Furthermore, a triton cannot produce ^{A+3}Z from AZ . Therefore, the aforementioned procedure is free of the drawbacks of the spallation method.

Here, we limit our discussion to tetraneutrons. Strictly speaking, the isotope ^{A+3}Z is converted from the stable isotope ^AZ by not only bound tetraneutrons but also bound multineutron systems with N > 4, including hexaneutrons ⁶n and octaneutrons ⁸n, which have been investigated far less than tetraneutrons, both experimentally and theoretically [2]. In particular, ab initio calculations of such systems are lacking. Whether the addition of neutrons to tetraneutrons stabilizes the system is an interesting question. Whereas a dedicated experiment in search of such an exotic system requires enormous effort, our method can probe any possible multineutron bound systems using equipment for INAA. The number of neutrons N cannot be determined directly even if a positive ^{A+3}Z signal is observed; however, this problem may be remedied through the search for more neutron-rich isotopes, such as ^{A+4}Z , similar to Ref. [18].

II. EXPERIMENT

The irradiation experiment was conducted using a hydraulic conveyer at the Kyoto University Research Reactor (KUR) in the Institute for Integrated Radiation and Nuclear Science, Kyoto University, with a thermal power of 5 MW during irradiation. We selected ⁸⁸Sr as the target ^AZ for conversion to ^{A+3}Z (⁸⁸Sr \rightarrow ⁹¹Sr) via the (⁴n, n) reaction, with a Q value of 20 MeV minus the binding energy of the tetraneutron. The main reasons for this selection are as follows:

- (1) For a given Z, the number of neutrons of the radioisotope ${}^{A+3}Z$, i.e., N+3=A-Z+3, should be as large as possible to eliminate the possibility of producing ${}^{A+3}Z$ from stable isotopes by means other than the $({}^4n, n)$ reaction. 88 Sr has the largest number of neutrons (N=50) among the stable isotopes of strontium.
- (2) The cross section for neutron capture ${}^{A}Z(n, \gamma)^{A+1}Z$ should be sufficiently small. For each radioisotope, including ${}^{A+1}Z$, the maximum allowed quantity to be used per day is stipulated by the institute, and the irradiation and cooling times should be optimized accordingly.
- (3) ⁸⁹Sr is an almost pure β emitter, implying that γ rays are hardly emitted after ⁸⁹Sr β decay. Indeed, only 908.96(3) keV γ rays are emitted with an intensity of $9.56(5) \times 10^{-5}$ [20]. This feature is beneficial for reducing the dead time of the germanium detector.

TABLE I. Neutron capture cross sections for thermal neutrons (σ_{th}) and resonance integrals (I_0) for ⁸⁸Sr, ⁸⁹Sr, and ⁹⁰Sr.

Nuclide	σ _{th} (mb)	<i>I</i> ₀ (mb)
⁸⁸ Sr	5.5 ± 0.4 [21]	24 [21]
⁸⁹ Sr	420 ± 40 [21]	749.8 [22]
⁹⁰ Sr	10.4 ± 1.4 [21]	104 ± 16 [21]

(4) The half-life of ^{A+3}Z should be at least comparable to the cooling time of the sample after irradiation.

As an irradiation sample, 570 mg of isotopically enriched 88 SrCO₃ (99.90% enrichment) was enclosed in a silica tube. The abundances of the strontium isotopes in the 88 SrCO₃ reagent were less than 0.01% for 84 Sr, 0.02% for 86 Sr, and 0.08% for 87 Sr. The sample was covered with a 0.5 mm-thick cadmium sheet to absorb thermal neutrons and prevent the undesirable activation of the sample owing to a high thermal-neutron capture cross section of 2×10^4 b for 113 Cd. In contrast, the cadmium sheet would have a negligible impact on a tetraneutron whose kinetic energy is significantly higher than that of thermal neutrons (\sim 0.025 eV). It was then placed in a capsule made of an aluminum alloy for the hydraulic conveyer.

Prior to the experiment, we evaluated two possible contributions to the production of 91 Sr without tetraneutrons: neutron capture of long-lived 90 Sr and triple neutron capture under the aforementioned conditions. For a conservative evaluation, the activation probability was estimated using neutron capture in an irradiation without a cadmium absorber, and we ignored the reduction of activated isotopes by β decay.

In general, the activation rate R of neutron capture [14] is expressed as

$$R = \Phi_{\rm th} \sigma_{\rm th} + \Phi_{\rm epi} I_0, \tag{1}$$

where Φ_{th} (Φ_{epi}) denotes the thermal (epithermal) neutron flux and σ_{th} represents the neutron-capture cross section for thermal neutrons. The resonance integral I_0 is defined as follows:

$$I_0 = \int_{E_{\min}}^{E_{\max}} \frac{\sigma(E)}{E} dE, \qquad (2)$$

where $\sigma(E)$ represents the neutron capture cross section as a function of the neutron energy E, and the integration interval $[E_{\rm min}, E_{\rm max}]$ refers to the energy region of epithermal neutrons. The nominal neutron flux at the hydraulic conveyer was $\Phi_{\rm th}=8.15\times 10^{13}\,(n/{\rm cm}^2/{\rm sec})$ and $\Phi_{\rm epi}=5.95\times 10^{12}\,(n/{\rm cm}^2/{\rm sec})$. The activation rates during a 2-h irradiation were calculated to be 4×10^{-9} for $^{88}{\rm Sr}\to ^{89}{\rm Sr}, 3\times 10^{-7}$ for $^{89}{\rm Sr}\to ^{90}{\rm Sr}$ and 1×10^{-8} for $^{90}{\rm Sr}\to ^{91}{\rm Sr}$ using the capture cross sections and resonance integrals for strontium isotopes, as summarized in Table I.

A portion of the 88 SrCO₃ reagent was analyzed via energy-filtered thermal ionization mass spectrometry (TIMS) [23]. Consequently, the 90 Sr signal was not observed, and we concluded that the 90 Sr/ 88 Sr ratio was below the detection limit of 2.7×10^{-12} [23,24]. As the number of 88 Sr atoms in the

sample was 2.32×10^{21} , the 91 Sr yield after the 2-h irradiation was estimated to be at most 7×10^{1} .

Similarly, the probability of triple-neutron capture, i.e.,

$$^{88}\text{Sr} \xrightarrow{(n,\gamma)} ^{89}\text{Sr} \xrightarrow{(n,\gamma)} ^{90}\text{Sr} \xrightarrow{(n,\gamma)} ^{91}\text{Sr}, \tag{3}$$

during a 2-h irradiation was estimated to be 2×10^{-24} , which corresponds to a $^{91}\mathrm{Sr}$ yield of 4×10^{-3} . Notably, the tripleneutron capture by $^{88}\mathrm{Sr}$ was negligible. For example, a similar calculation for the neighboring nuclide $^{89}\mathrm{Y}$ resulted in a probability 2×10^5 times higher.

These estimations confirm that the 88 Sr sample is a promising candidate for investigating bound tetraneutrons via the $(^4n, n)$ reaction.

The sample was irradiated in a hydraulic conveyer for 2 h, followed by approximately 11 h of cooling to reduce its radioactivity. We expected that the dominant radioisotopes after cooling would be $^{87m} \rm Sr$ (half-life: 2.815(12) h [25]) and $^{89} \rm Sr$ (half-life: 50.563(25) d [20]) with activities of 0.28 MBq and 0.49 MBq. After opening the capsule and removing the highly activated cadmium absorber inside a hot cell, we measured γ rays from the activated $^{88} \rm SrCO_3$ sample enclosed in the silica tube using a high-purity germanium detector in a hot laboratory. The distance between the sample and detector surface was 15 cm. We repeated the 30-min measurements 48 times, considering that the half-life of $^{91} \rm Sr$ is 9.65(6) h [26].

III. RESULTS AND DISCUSSION

Figure 1 shows the γ -ray spectrum for all the measurements, which was obtained by summing the dead-time-corrected spectrum for each measurement. In addition to the 389 keV photopeak of 87m Sr, we observed several photopeaks originating from radioisotopes, such as 82 Br, 115 Cd, and 23 Na because of the neutron capture by an impurity that was present in the silica or adhered to the surface of the silica tube. A calibration source containing 60 Co and 137 Cs was used for energy calibration and determination of the absolute photopeak efficiency. Furthermore, eight photopeaks of 82 Br ranging from 554.352(10) to 1474.895(10) keV [27] observed in each measurement were used for the *in situ* calibration.

We did not observe photopeaks at 749.8(1) keV or at 1024.3(1) keV originating from the 91 Sr β decay; their intensities per 91 Sr β decay are known to be $I_{\gamma}=23.7(8)\%$ and 33.5(11)%, respectively [26]. However, the sum peak at 1022 keV due to the annihilation observed in the γ -ray spectrum hindered the statistical estimation of the strength of the neighboring 1024.3(1) keV photopeak. Therefore, the upper limit of the 91 Sr yield was determined using the spectrum near 749.8(1) keV.

As shown in Fig. 2, a small 97 Zr photopeak at 743.36(3) keV [28] was identified near the expected 91 Sr photopeak. Hence, the spectrum was fitted with respect to the two peak functions plus a linear background aE + b. We found that the shape of the *in situ* 82 Br photopeaks changed monotonically during each measurement. We assumed that the parameters of the shift ΔE_j and width σ_j for the 82 Br photopeak at 776.511(10) keV in the *j*th measurement represented the behavior of the expected 91 Sr photopeak. Moreover,

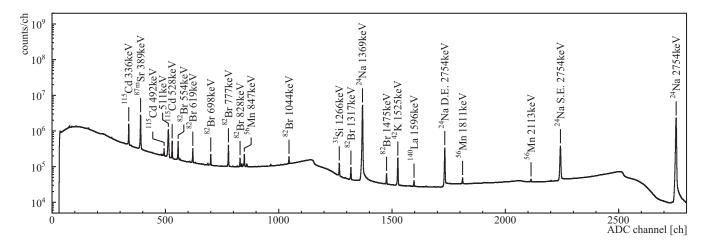


FIG. 1. Measured γ -ray spectrum. The dead-time-corrected spectra of all the measurements over 24 h were summed. S.E. and D.E. refer to the single and double escape peaks, respectively.

the change in photopeak efficiency during the measurements was corrected for when summing the spectrum from each measurement.

We fitted the summed spectrum from the first measurement until the N_{max} -th measurement ($1 \le N_{\text{max}} \le 48$) with the following function of the energy E with five free parameters $N_{^{91}\text{Sr}}$, $N_{^{97}\text{Zr}}$, $\sigma_{^{97}\text{Zr}}$, a, and b:

$$f(E; N_{^{91}Sr}, N_{^{97}Zr}, \sigma_{^{97}Zr}, a, b)$$

$$= \sum_{j=1}^{N_{\text{max}}} \frac{2^{-(j-1)\Delta t/T_{1/2}}N_{^{91}Sr}}{\sqrt{2\pi}\sigma_j} \exp\left[-\frac{(E - E_{^{91}Sr} - \Delta E_j)^2}{2\sigma_j^2}\right]$$

$$+ \frac{N_{^{97}Zr}}{\sqrt{2\pi}\sigma_{^{97}Zr}} \exp\left[-\frac{(E - E_{^{97}Zr})^2}{2\sigma_{^{97}Zr}^2}\right] + aE + b, \quad (4)$$

where N_{91} represents the area of the 91 Sr photopeak during the first measurement and N_{97}_{Zr} (σ_{97}_{Zr}) represents the area (width) of the ⁹⁷Zr photopeak in the summed spectrum. The literature values of the γ -ray energies for 91 Sr and 97 Zr were input as $E_{^{91}Sr}$ and $E_{^{97}Zr}$, respectively. The attenuation of ^{91}Sr radioactivity was incorporated into the factor $2^{-(j-1)\Delta t/T_{1/2}}$, where Δt represents the measurement time of 0.5 h and $T_{1/2}$ represents the half-life of 91Sr. Subsequently, the upper limit of N_{91}_{Sr} at the 95% confidence level (C.L.) was evaluated by integrating the probability density function, which was assumed to be Gaussian in the nonnegative physical region. By repeating the fitting for $N_{\text{max}} = 1, 2, \dots, 48$, the upper limit was minimized to $38.3_{-0.7}^{+0.8}$ for $N_{\text{max}} = 36$, where the systematic error stems from the uncertainty of the photopeak energy, i.e., ± 0.1 keV. The best fit to the spectrum with $N_{\text{max}} = 36$ and the fit function with $N_{\rm ^{91}Sr}$ being the upper limit at the 95% C.L. are displayed in Fig. 2.

Finally, the upper limit of the 91 Sr yield in the sample immediately after the 2-h irradiation was determined to be 1.0×10^7 using the intensity of $I_{\gamma} = 23.7(8)\%$ and a photopeak efficiency of 0.116(5)%.

To enable comparison with the previous experiment [15] and future experiments at KUR or other research reactors

with different configurations, we estimated the emission rate of bound tetraneutrons per fission (R_{1n}) which serves as an indicator of the experimental sensitivity. To relate the 91 Sr yield after irradiation (Y_{91} Sr) to the emission rate, we made the same assumptions as in Ref. [15]:

(1) The mean free path of bound tetraneutrons in the lightwater moderator was assumed to be 25 cm. Then, the probability that a tetraneutron produced uniformly in

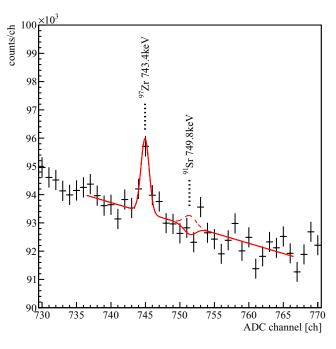


FIG. 2. Enlarged γ -ray spectrum in the vicinity of 749.8 keV. The first 36 measurements were used for the analysis. The solid line shows the best fit to the spectrum within a range of ± 15 ch from 751.4 ch (i.e., 749.8 keV). The negative peak at 751.4 ch reflects a negative $N_{\rm Pl}$ in the fitting function [Eq. (4)]. The dashed line indicates the 91 Sr photopeak with the upper limit of the strength at the 95% confidence level.

- the fuel rods strikes a unit area in the sample position was numerically calculated to be $1.0 \times 10^{-4}/\text{cm}^2$, using the configuration of the reactor core [29].
- (2) The cross section of the ${}^{88}\text{Sr}({}^{4}\text{n}, n){}^{91}\text{Sr}$ reaction was assumed to be 50 mb, ignoring the difference of the target nucleus. The assumption was made in Ref. [15] based on the (α, p) and (α, n) cross sections on light nuclei. Whereas this assumption is not grounded in that bound tetraneutrons would be much loosely bound than α particles, and that the Coulomb barrier between a tetraneutron and a target nucleus does not exist, we tentatively adopt the assumption for an order-of-magnitude estimation.

Using a fission rate of 1.6×10^{17} per second, which can be derived by dividing the thermal power of 5 MW by the released energy of 190 MeV during fission, the "tetraneutron flux" at the sample position was estimated to be $1.7 \times 10^{13} \times R_{4_n}/\text{cm}^2/\text{s}$. By multiplying it by the number of ⁸⁸Sr atoms in the sample and the (⁴n, n) cross section, the ⁹¹Sr yield can be expressed as

$$Y_{\rm {}^{91}Sr} = 1.3 \times 10^{13} \times R_{\rm {}^{4}n}. \tag{5}$$

Hence, we determined that the upper limit of R_{4n} was 8 × 10^{-7} per fission at the 95% C.L.

We consider γ -ray spectroscopy to have an advantage over counting delayed neutrons [15] because the γ -ray spectrum contains rich information, such as the contributions of impurities. Moreover, whereas β -delayed neutron emitters, e.g., ¹⁷N, have short lifetimes of <1 min, our method can be applied to moderately long-lived isotopes such as ⁹¹Sr, allowing the irradiation time to be significantly increased.

We demonstrated the feasibility of particle-stable tetraneutron search using an INAA-like approach in a nuclear reactor. In the first measurement with a 570 mg sample of $^{88}\mathrm{SrCO_3}$ irradiated for 2 h at KUR, γ rays from $^{91}\mathrm{Sr}$, which were expected to be produced by a tetraneutron-induced $^{88}\mathrm{Sr}(^4\mathrm{n},n)^{91}\mathrm{Sr}$ reaction, were not observed. Through fitting the γ -ray spectrum, the yield of $^{91}\mathrm{Sr}$ after irradiation was deduced to be less than 1.0×10^7 at the 95% C.L. This corresponds to an emission rate of particle-stable tetraneutrons, assuming they exist, of less than approximately 8×10^{-7} per fission at the 95% C.L., using the $(^4\mathrm{n},n)$ cross section and mean free path in the moderator conjectured in Ref. [15]. Modern calculations for reactions induced by tetraneutrons or heavier multineutron systems would help quantitative discussions on the emission rate.

IV. CONCLUSION

In the future, increasing the experimental sensitivity will be crucial. For example, extracting irradiated $^{88} SrCO_3$ powder from an activated silica tube will reduce the background in γ -ray spectroscopy. Additionally, the number of impurities can be reduced via chemical processing of the powder. Thus, a subtle signal owing to particle-stable multineutron systems may be observed in high-purity samples irradiated in a nuclear reactor.

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