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ENHANCEMENT OF GAMMA-RAY YIELD FROM FAST NEUTRONS INTERACTION WITH CADMIUM ISOTOPES

Purpose. This paper presents the results of investigation of γ-ray yield enhancement from ^{nat}Cd(n,xγ) reactions obtained in experiments with 14-MeV neutrons in comparison with theoretical spectrum calculated in Empire code.
Methods. The experiment was carried out applying TOF-technique for n-γ discrimination. The MCNP simulation of the experiment was performed and the influence of scattered neutrons background at gamma-ray spectrum shape was estimated.
Results. It was shown that despite TOF-technique thermal neutrons capture by ^{113}Cd isotope gives considerable contribution to gamma-spectrum induced in ^{nat}Cd sample by primary DT-neutrons. The thermal neutron flux was determined to be nearly constant at different locations of experimental hall.
Conclusions. For the correct measurements of the γ-production cross sections for cadmium samples the sufficient separation between primary neutrons and scattered ones must be provided.
Keywords: thermal neutrons, DT-neutrons, cadmium, gamma-spectrum, gamma-ray yield.

Introduction

The cross sections of gamma-rays production induced by neutron interactions are required for radiation damage and shielding calculations of the fission and fusion reactors. The 14-MeV neutrons are of utmost importance for these tasks because of the big number of open reaction channels at this incident neutrons energy. Almost all of interactions between neutrons and nuclei of interest are accompanied by the prompt gammas emission, and the (n,xγ) reactions cross sections must be precisely known for the modern and future reactor construction elements undergoing neutron exposure.

Despite large number of experiments performed with DT-neutrons they are still relevant for fusion reactors design. It is caused by disagreements between nuclear data from different authors [1] or between experimental results and theoretical calculations. In particular, the last ones were obtained in our experiments for Cd element [2], which is commonly used in the reactors industry. The measured gamma-ray spectrum appeared to be enhanced in comparison with theoretical calculations. In this work we present the investigation of this phenomenon.

Data analysis

Our experiments in gamma-ray spectroscopy were performed with 14-MeV neutrons for different samples, such as ^{nat}Fe, ^{nat}Bi, ^{nat}Cd, ^{nat}Ni, ^{nat}Sn and ^{nat}C [2–5]. All the measurements were carried out using the pulse neutron generator facility PNG-200 under the same geometry (samples had the torus shapes with outer and inner radii of 10÷16.5 cm and 1.5 cm, respectively) and with the same settings: for n-γ discrimination the time-of-flight (TOF) technique was applied with pulse repetition period

(PRP) of 140 ns and with γ-detection time window of 20 ns (the detailed description of experiments and DAQ can be found in [4]). A good agreement between experimental results and theoretical calculations performed in Empire code [6] was obtained for all elements except ^{nat}Cd sample, where notable discrepancy in the 5÷10 MeV energy range is observed (Fig. 1). Such enhancement of measured gamma-spectrum from ^{nat}Cd was also observed in works of other authors [7, 8].
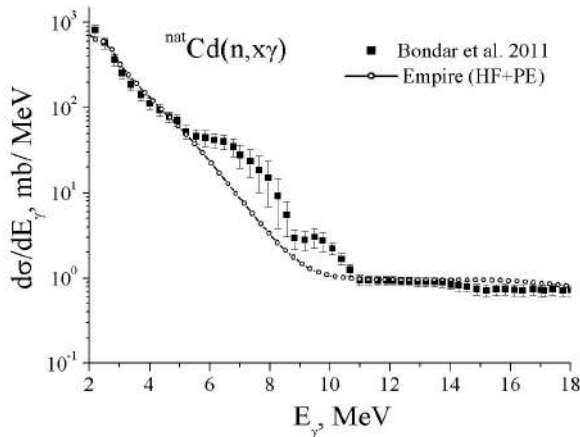

Figure 1: The experimental data and theoretical calculations of gamma-ray spectrum from ^{nat}Cd(n,xγ) reactions induced by 14-MeV neutrons.

The origin of this discrepancy can be caused by the experimental conditions such as presence of thermal neutrons background. The neutron scattering on construction elements and walls of the experimental hall leads to continuous neutron spectrum formation in the experimental hall, as well as the sample itself, and takes much longer than PRP time. Hence, despite TOF technique, the background gammas induced by low-energy neutrons interaction can also be detected in the time window. It is well-known that for ^{nat}Fe, ^{nat}Bi, ^{nat}Ni, ^{nat}Sn and ^{nat}C elements capture cross section even for thermal neutrons does not exceed several barns. But for ^{nat}Cd this value is about 2500 b, and (n,γ) reactions can cause an essential impact on gamma-ray spectrum.

An additional contribution to γ-spectrum is mainly expected to be from the ^{113}Cd(n,γ)^{114}Cd nuclear reaction. The ^{113}Cd isotope has 12.23% in natural abundance and average cross section of 20 000 b for thermal neutrons capture resulting in formation of compound nucleus ^{114}Cd with excitation energy 9.04 MeV. The de-excitation of this state occurs with high probability by the cascade E2 transitions through the discrete levels 0–558.45 keV–1283.7 keV–1991 keV–2670 keV, accompanied by gamma-rays with the energies 9.04 MeV, 8.48 MeV, 7.76 MeV, 7.05 MeV, and 6.37 MeV (Fig. 2).
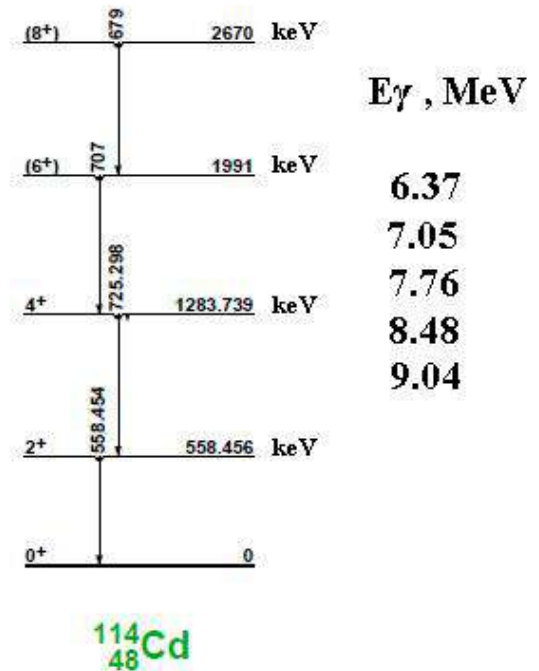

Figure 2: Possible γ-transitions from the ^{113}Cd(n,γ)^{114}Cd reactions at excitation energy of 9.04 MeV.

The energy resolution for these energies in experiment was near 1 MeV, so along with continuum transitions these gamma-rays due to neutron capture could provide smooth enhancement of gamma-spectrum within the energy interval 5÷10 MeV. The continuum transitions from ^{113}Cd(n,γ)^{114}Cd reactions give notable contribution below 7 MeV [9], therefore the observed double-humped spectrum can be formed by the sum of a group of peaks around 7 MeV and the continuous spectrum increasing at lower energies (left part), and discrete gamma-rays with energies from 7.05 to the 9.04 MeV in the right part of the spectrum. All these considerations indicate possible influence of low-energy neutrons. To figure this out, the scattered neutrons fluxes must be determined.

MCNP simulations

The 14-MeV and thermal neutrons fluxes were estimated by MCNP [10] simulations. The geometry of the experiment is presented in Fig. 3. The main components that can effectively moderate neutrons, such as concrete walls, floor, roof and oil filled bucket for high voltage supply were also built for simulations.
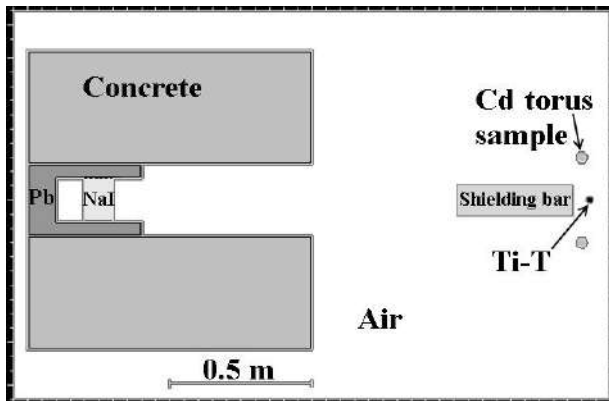

Figure 3: The MCNP geometry of the experiment.

The overall time of neutrons moderation to the thermal energies and subsequent diffusion before absorption is several hundreds of microseconds. The PNG-200 generates pulses of DT-neutrons with period of 0.14 μs, so it can be considered as constant neutrons source relative to the scattered neutrons lifetime in experimental hall. In such conditions the flux of thermal neutrons is statistically averaged and nearly constant during the measurements with strong dependence on intensity of DT-neutrons from the Ti-T target. Therefore the continuous and isotropic point-source of 14-MeV neutrons was specified in simulations.

The neutrons fluxes per one DT-neutron from the source were determined at different locations in the experimental hall: near the sample, behind the shielding bar and near the wall, which is at 3.5 m distance from Ti-T target. The results of simulated fluxes are shown in Table 1.

Table 1: Primary 14-MeV and scattered low-energy (0÷0.5 eV) neutron fluxes at different locations

Location	$F_{14}, 1/cm^2/DT-neutron$ (14 MeV)	$F_{th}, 1/cm^2/DT-neutron$ (0÷0.5 eV)
Near the sample	$2.92 \cdot 10^{-4}$	$9.66 \cdot 10^{-7}$
Behind shielding bar	$9.57 \cdot 10^{-6}$	$9.48 \cdot 10^{-7}$
Near the wall	$7.59 \cdot 10^{-7}$	$9.65 \cdot 10^{-7}$

The simulated neutron spectra near the sample within the 0.1÷15 MeV and 0÷0.5 eV energy ranges are shown in Fig. 4 The ratio between 14-MeV (F_{14}) and thermal (from 0 and up to 0.5 eV, F_{th}) neutron fluxes near the sample appeared to be $F_{14}/F_{th} = 302.3$.
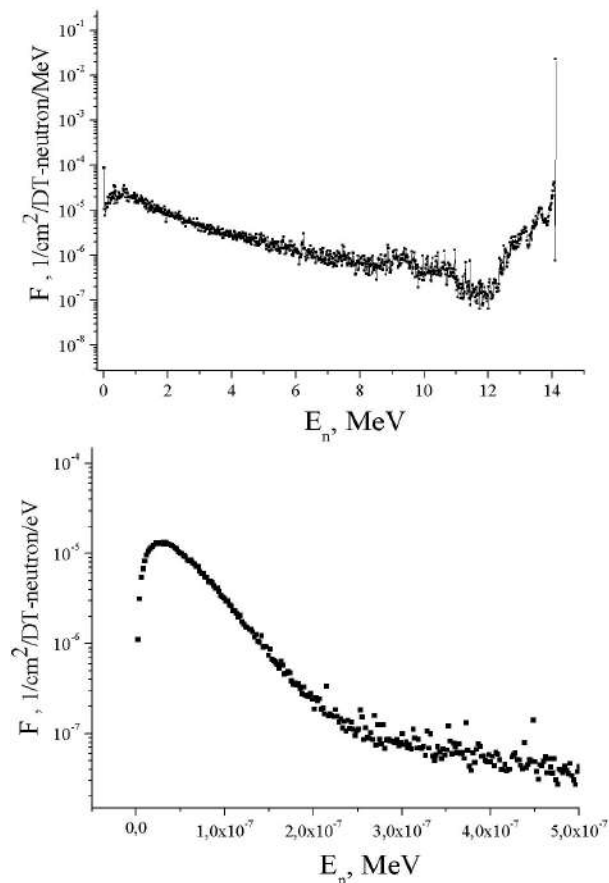

Figure 4: Simulated neutron spectra near the ^{nat}Cd sample.

The yield of gamma-rays from (n,xγ) reactions induced by 14-MeV neutrons in the

^{nat}Cd sample equals

$$Y_{14} = N_{14}F_{14}\alpha_n\sigma_{14}, \quad (1)$$

where N_{14} is the number of nuclei undergoing irradiation by 14 MeV neutrons; F_{14} is the 14-MeV neutron fluence; α_n – coefficient for 14-MeV neutrons beam attenuation in the sample; σ_{14} – cross section of gamma-production from (n,xγ) reactions induced by 14-MeV neutrons. In case of thermal neutrons, the high cross section of radiative capture by ^{113}Cd isotope limits the free path of thermal neutron in cadmium by the value of $\lambda_{th} = 1/\Sigma_{th} = 8.6 \cdot 10^{-3} cm$. As a result, all the thermal neutrons are absorbed within a thin ($x < 0.1cm$) layer of cadmium, and (n,γ) reactions rate equals to the total number of neutrons hitting the torus sample:

$$R_{14} = F_{th}(1 - exp(-x/\lambda_{th}))S = F_{th}S. \quad (2)$$

Here S is the area of the sample surface, and $(1 - exp(-x/\lambda_{th}))$ determines the neutrons attenuation at distance x inside the sample. The gamma-yield caused by thermal neutrons Y_{th} is the product of capture reactions rate R_{th} and multiplicity of capture γ-rays M_γ. During experiments the total yield $Y_{exp} = Y_{14} + Y_{th}$ is measured including both thermal and 14-MeV neutrons induced reactions, and the enhancement of gamma-yield caused by thermal neutrons contribution can be expressed as follows:

$$\frac{Y_{exp}}{Y_{14}} = 1 + \frac{Y_{th}}{Y_{14}} = 1 + \frac{F_{th}SM_\gamma}{N_{14}F_{14}\alpha_n\sigma_{14}}. \quad (3)$$

While the measured cross section of gamma-ray production σ_{exp} is proportional to the yield Y_{exp} (because Y_{th} was normalized to $N_{14}F_{14}$), and the response function of the spectrometer is the same for gamma-rays of different origins, the relation between different gamma-production cross sections correlates with corresponding yields. The cross section discrepancy between measured experimental data and theoretical calculations in the 5÷10 MeV energy range is

$$\frac{\int_5^{10} \frac{d\sigma_{exp}}{dE}\,dE}{\int_5^{10} \frac{d\sigma_{14}}{dE}\,dE} = 1.64 \pm 0.18, \quad (4)$$

while equation (3) gives close to this discrepancy value between experimental and theoreti-

cal yields

$$1 + \frac{F_{th}SM_\gamma N_{\gamma th}}{N_{14}F_{14}\alpha_n\sigma_{14}N_{\gamma 14}} = 1.47. \quad (5)$$

Here $M_\gamma = 4.1$ is average gamma-ray multiplicity of ^{113}Cd(n,γ)^{114}Cd reactions, $N_{\gamma th} = 10.2\%$ is part of gamma-spectrum within considered energy range induced by thermal neutrons [11] and $N_{\gamma 14} = 2.2\%$ is the same for 14-MeV neutrons obtained from Empire calculations.

The values in (4) and (5) confirm the idea of contribution from thermal neutrons to the measured spectrum. The excess of the measured gamma-production cross section within 5÷10 MeV is 60 mb, and one could assume that it is caused not by 14-MeV, but thermal neutrons interactions with ^{113}Cd nuclei. Then the cross section of gamma-ray production renormalized to the flux of thermal neutrons and number of ^{113}Cd nuclei equals 11 450 barn, which makes 12.7% of the total cross section of gamma-yield, namely 88 000 b [11].

The value obtained in (5) shows that the number of gamma-rays generated by thermal neutrons is about 47% of gamma-rays from 14-MeV neutrons and it is comparable with obtained discrepancy (4). Obviously, this contribution must be taken into account during gamma-spectra measurements from fast neutrons interactions. It can be seen from table 1 that the primary 14-MeV neutrons beam near the sample is bigger than at other locations by 30 (behind the shielding bar) and 385 (near the wall) times respectively, while the thermal neutrons flux is nearly constant regardless of the location of control volume in the experimental hall. It means that the changing of the sample position will not reduce the influence from scattered neutrons and the additional shielding is required. In particular, it can be realized with the help of neutronstop covering on walls, ceiling and floor. According to MCNP simulations, such type of shielding (with bricks of 10 cm thickness and 5% of ^{10}B addition by mass) leads to the reduction of thermal neutrons flux up to 10^2 times. But the clear gamma-spectra from 14-MeV neutrons can be obtained only taking into account the scattered ones.

Despite values in (4) and (5) are close, the experimental enhancement is still higher than

expected one from thermal neutrons. This bump was also observed in the experiments with high flux of thermal neutrons and enriched ^{113}Cd sample in [11], and it could not be reproduced by theoretical calculations. So this energy region is under the interest and requires additional investigations.

Conclusions

It was shown that the discrepancy in the gamma-spectrum from ^{nat}Cd sample is caused by the thermal neutrons interactions due to the big capture cross section with ^{113}Cd nuclei. The MCNP simulations confirmed this idea and helped to understand the irradiation conditions of the ex-

periment. According to obtained results, about 30% of all gamma-rays from (n,xγ) reactions induced in the ^{nat}Cd sample caused by thermal neutrons, and the flux of them is nearly constant at different points in the experimental hall. Therefore, the measurements of fast neutrons interactions with cadmium samples must be conducted very carefully taking into account possible contributions from thermal neutrons. The best way is to ensure the irradiation conditions with minimum scattered and maximum of 14-MeV neutrons fluxes and to provide sufficient separation between them. Our further investigation includes reexamination of gamma-production cross sections of ^{nat}Cd(n,xγ) reactions induced by 14-MeV neutrons.

REFERENCES

[1] Simakov S.P., Pavlik A., Vonach H., Hlavac S. Status of experimental and evaluated discrete γ-ray production at En=14.5 MeV / Final Report of Research Contract 7809/RB, performed under the CRP on Measurement, Calculation and Evaluation of Photon Production Data — 1998. — p.24.

[2] Bondar B.M., Bondar V.M., Gorbachenko O.M., Kadenko I.M. , Leshchenko B.Yu., Onishchuk Yu.M., Plujko V.A. Cross sections of prompt γ-ray production induced by interactions of fast neutrons with cadmium // Nuclear Physics and Atomic Energy — 2011. — Vol.12(2). — p.129-136.

[3] Bondar V.M., Gorbachenko O.M., Kadenko I.M., Leshchenko B.Yu., Onishchuk Yu.M., Plujko V.A. Spectra of gamma-rays in (n,xγ) reactions on ferrum and bismuth nuclei //Nuclear Physics and Atomic Energy — 2010. — Vol.11(3). — p.246-250.

[4] Kadenko I.M., Plujko V.A., Bondar B.M., Gorbachenko O.M., Leshchenko B.Yu., Solodovnyk K. M. Prompt gamma-rays from fast neutron capture in ^{nat}Ni// Nuclear physics and atomic energy — 2016. — Vol.17(2). — p.122-129.

[5] Kadenko I.M., Plujko V.A., Bondar B.M., Gorbachenko O.M., Leshchenko B.Yu., Solodovnyk K. M. Gamma-rays from ^{nat}Sn and ^{nat}C induced by fast neutrons // Nuclear physics and atomic energy — 2016. — Vol.17(4). — p.349-353.

[6] Herman M., Capote R., Carlson B.V., Oblozinsky P., et al. EMPIRE: Nuclear Reaction Model Code System for Data Evaluation // Nuclear Data Sheets — 2007. — V.108(12). — p.2655-2715.

[7] Bezotosnyy V.M., Gorbachev V.M., Surov L.M., Shvetsov M.S. Differential and total cross sections for gamma-production by inelastic interaction of 14 MeV neutrons with different nuclei // Vop. At. Nauki i Tekhn., Ser. Yadernye Konstanty — 1978. — Vol.3(30) — p.21.

[8] Nefedov Yu.Ya., Nagornyj V.I., Semenov V.I. The experimental results of the gamma-ray production cross-section and spectra at the nonelastic interaction 14 MeV neutrons with nuclei

of Na, S, Cl, Ti, V, Cr, Ni, Zn, Ge, Nb, Cd, In, Sn, Bi, ^{235}U and ^{238}U // Vop. At. Nauki i Tekhn., Ser. Yadernye Konstanty — 2000. — Vol.1. — p.7.

[9] Rusev G., Jandel M., Krtiˇcka M., Arnold C. W., Bredeweg T.A., Couture A., Moody W.A., Mosby S.M., and Ullmann J.L. Cascade γ rays following capture of thermal neutrons on ^{113}Cd // Phys.Rev.C 88 — 2013. — Vol.88(5) 057602.

[10] RSICC Computer code collection. MCNP4C. Monte Carlo N particle transport code system / Oak Ridge National Laboratory, USA — 2000. — p.898.

[11] Belgya T., Szentmiklósi L., Massarczyk R., Schwengner R., Junghans A.R., and Grosse E. High-resolution study of the ^{113}Cd(n,γ) spectrum by statistical decay model with discrete levels and transitions // EPJ Web Conf., — 2017. — Vol.146(0500) — p.4.

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УСИЛЕНИЕ ВЫХОДА ГАММА-КВАНТОВ ПРИ ВЗАИМОДЕЙСТВИИ БЫСТРЫХ НЕЙТРОНОВ С ИЗОТОПАМИ КАДМИЯ

В статье исследуется усиление гамма-выхода с ядерных реакций ^{nat}Cd(n,xγ) наблюдаемое в эксперименте с нейтронами с энергией 14 МэВ. Данное усиление отсутствует в теоретическом гамма-спектре, рассчитанном с помощью кода Empire. Эксперимент проводился с применением времяпролетной методики для n-γ дискриминации. Было проведено MCNP симуляцию эксперимента и оценено влияние фона от рассеянных нейтронов на форму гамма-спектра. Было показано, что несмотря на временную сепарацию событий, поглощение тепловых нейтронов изотопом ^{113}Cd дает существенный вклад в гамма-спектр с реакций ^{nat}Cd(n,xγ), вызванных прямыми DT-нейтронами. С помощью MCNP моделирования был определен флюенс тепловых нейтронов, который оказался постоянным в разных точках экспериментального зала. Сделано вывод о необходимости дополнительного снижении фона и сепарации между прямыми и рассеянными нейтронами для корректного измерения сечений выхода гамма-квантов для образцов кадмия.
Ключевые слова: тепловые нейтроны, DT-нейтроны, кадмий, гамма-спектр, гамма-выход.

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ПІДСИЛЕННЯ ВИХОДУ ГАММА-КВАНТІВ ПРИ ВЗАЄМОДІЇ ШВИДКИХ НЕЙТРОНІВ З ІЗОТОПАМИ КАДМІЮ

В статті досліджується підсилення гамма-виходу з ядерних реакцій ^{nat}Cd(n,xγ), що спостерігалося в експерименті з нейтронами з енергією 14-MeB. Дане підсилення відсутнє в гамма-спектрі, розрахованому в коді Empire. Експеримент проводився із застосуванням чаоспрольотної методики вимірювань для n-γ дискримінації. Було проведено MCNP симуляцію експерименту і оцінено вплив фону від розсіяних нейтронів на форму гамма-спектру. Було показано, що незважаючи на часову сепарацію подій, захват теплових нейтронів ізотопом ^{113}Cd дає суттєвий внесок в гамма-спектр з реакцій ^{nat}Cd(n,xγ), зумовлених прямими DT-нейтронами. За допомогою MCNP симуляції було визначено флюенс теплових нейтронів, який виявився сталим в різних точках експериментальної зали. Зроблено висновок, що для коректного вимірювання перерізів виходу гамма-квантів для зразків кадмію необхідно забезпечувати додаткову сепарацію між прямими та розсіяними нейтронами.

Ключові слова: теплові нейтрони, DT-нейтрони, кадмій, гамма-спектр, гамма-вихід.

СПИСОК ВИКОРИСТАНОЇ ЛІТЕРАТУРИ

[1] Simakov S.P., Pavlik A., Vonach H., Hlavac S. Status of experimental and evaluated discrete γ-ray production at En=14.5 MeV / Final Report of Research Contract 7809/RB, performed under the CRP on Measurement, Calculation and Evaluation of Photon Production Data — 1998. — p.24.

[2] Бондар Б.М., Бондар В.М., Горбаченко О.М., Каденко І.М., Лещенко Б.Ю., Оніщук Ю.М., Плюйко В.А. Диференціальні перерізи вильоту гамма-квантів при взаємодії нейтронів з ядрами кадмію // Ядерна фізика та енергетика — 2011. — №12(2). — с.129-136.

[3] Бондар В.М., Горбаченко О.М., Каденко І.М., Лещенко Б.Ю., Оніщук Ю.М., Плюйко В.А. Спектри гамма-квантів у (n,xγ) реакціях на ядрах заліза та вісмуту // Ядерна фізика та енергетика — 2010. — №11(3). — с.246-250.

[4] Каденко І.М., Плюйко В.А., Бондар Б.М., Горбаченко О.М., Лещенко Б.Ю., Солодовник К.М. Миттєві гамма-кванти при поглинанні швидких нейтронів ядрами ^{nat}Ni // Ядерна фізика та енергетика — 2016. — №17(2). — с.122-129.

[5] Каденко І.М., Плюйко В.А., Бондар Б.М., Горбаченко О.М., Лещенко Б.Ю., Солодовник К.М. Гамма-випромінювання з ^{nat}Sn та ^{nat}C під дією нейтронів // Ядерна фізика та енергетика — 2016. — №17(4). — с.349-353.

[6] Herman M., Capote R., Carlson B.V., Oblozinsky P., et al. EMPIRE: Nuclear Reaction Model Code System for Data Evaluation // Nuclear Data Sheets — 2007. — V.108(12). — p.2655-2715.

[7] Безотосный В.М., Горбачев В.М., Суров Л.М., Швецов М.С. Полные сечения образования γ-квантов при неупругом взаимодействии 14-МеВ нейтронов с различными ядрами // Вопросы атомной науки и техники, серия ядерные константы — 1978. — №3(30) — с.21.

[8] Нефедов Ю.Я., Нагорный В.И., Семенов В.И., Житник А.К., Орлов Р.А., Шмаров А.Е. Результаты измерений сечений и спектров образования гамма-квантов при неупругом взаимодействии 14 МэВ нейтронов с ядрами Na, S, Cl, Ti, V, Cr, Ni, Zn, Ge, Nb, Cd, In, Sn, Bi, ^{235}U and ^{238}U // Вопросы атомной науки и техники, серия ядерные константы — 2000. — №1. — с.7.

[9] Rusev G., Jandel M., Krtička M., Arnold C. W., Bredeweg T.A., Couture A., Moody W.A., Mosby S.M., and Ullmann J.L. Cascade γ rays following capture of thermal neutrons on ^{113}Cd // Phys.Rev.C 88 — 2013. — Vol.88(5) 057602.

[10] RSICC Computer code collection. MCNP4C. Monte Carlo N particle transport code system / Oak Ridge National Laboratory, USA — 2000. — p.898.

[11] Belgya T., Szentmiklósi L., Massarczyk R., Schwengner R., Junghans A.R., and Grosse E. High-resolution study of the ^{113}Cd(n,γ) spectrum by statistical decay model with discrete levels and transitions // EPJ Web Conf., — 2017. — Vol.146(0500) — p.4.