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ELEMENTARY PARTICLES AND FIELDS
Experiment

Calculation of Neutron and Gamma Yields of (α, n) and $(\alpha, n\gamma)$ Reactions by Means of a New Version of the NeuCBOT Program for low background Experiments

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Abstract—(α, n) and $(\alpha, n\gamma)$ reactions induced by the alpha decays of uranium, thorium, and their daughter nuclides generate the intrinsic neutron and gamma backgrounds in modern ultralow background neutrino and dark matter detectors. In order to minimize the background, it is essential to select materials on the basis of a detailed analysis of relative concentrations of radionuclides and calculations of neutron and gamma-radiation yields. The NeuCBOT (Neutron Calculator Based On TALYS) program makes it possible to perform such calculations. The present article gives a review of a new version of the NeuCBOT program and a comparison of the results of calculations with different software tools.

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1. INTRODUCTION

(α, n) and $(\alpha, n\gamma)$ reactions occurring on various target nuclei attract the attention of researchers not only because of interest in them as such but also because of the need for considering these reactions and taking them into account in a number of fundamental and applied problems. The fundamental problems in question include those of studying the structure of nuclei, processes that accompany supernova explosions, the formation of neutron stars, and nucleosynthesis. In particular, the reaction $^{13}\mathrm{C}(\alpha, n)^{16}\mathrm{O}$ is assumed to be one of the main neutron sources in s-processes [1–4]. Because of enlargement of dimensions of neutrino detectors and facilities aimed at direct dark matter searches and because of improvement of their sensitivity, neutrons from (α, n) reactions became one of the dominant background components. The interaction of these neutrons with a working medium is virtually indistinguishable from inverse beta-decay reactions or from the scattering of a dark matter particle off a nucleus. Furthermore, (α, n) reactions are of importance in applied problems associated with the operation of nuclear power plants, nuclear waste disposal, and nonproliferation control. For example, inverse reactions, including the aforementioned process $^{13}\mathrm{C}(\alpha, n)^{16}\mathrm{O}$, are studied in order to obtain deeper insight into particle-energy-dependent neutron interactions with oxygen isotopes in the reactor core. Noninvasive analyses based on the detection of neutrons originating from alpha particle interaction with light fuel components can be performed in order to determine the level of fuel enrichment. In order to test the situation in underground repositories of waste nuclear fuel, it is necessary, among other things, to take into account the flux of neutrons produced in (α, n) reactions.

Such investigations mostly give calculated or measured cross sections, spectra, and yields of particles. Relevant experiments were predominantly performed between the 1950s and 1990s. For many nuclei, their results are still poorly consistent. On the other hand, the codes used for respective calculations may yield values differing by a few tens of percent or even in order of magnitude. This unsatisfactory situation has recently quickened interest in the subject, with the result that there appeared new articles containing fresh experimental data [4–6] and new software tools [7–10]. Also, international workshops on this topic were held [11–13]. The present article is devoted to developing the calculations of yields and spectra of neutrons and gammas from (α, n) and $(\alpha, n\gamma)$ reactions on the basis of NeuCBOT (Neutron Calculator Based On TALYS) utility and to

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reviewing the results based on it with an eye to further applications—first of all, for needs of low background experiments.

2. BACKGROUND FROM (α, n) AND $(\alpha, n\gamma)$ REACTIONS IN NEUTRINO EXPERIMENTS AND EXPERIMENTS AIMED AT DARK MATTER SEARCHES

Any detector consists of various materials. Each material contains some, even though extremely small, amount of radioactive impurities, including ^{232}Th, ^{235}U, ^{238}U, and their daughter nuclides. Upon undergoing decay, these nuclei may generate alpha particles, which, in turn, induce (α, n) and $(\alpha, n\gamma)$ reactions. As indicated in the Introduction, neutrons giving rise to background signals that resemble strongly signals from interactions of a neutrino and dark matter particles with a target material are hazardous for low background detectors. The effect of this background on measurements can be suppressed via selecting and/or producing such materials for detector components that would contain minimal amounts of uranium and thorium isotopes and which would generate minimum neutron yields from (α, n) reactions. The respective residual neutron background N_j for a specific material used in a particular detector unit[6)] carrying a subscript j can be calculated by the formula

$$N_j = (1 - \varepsilon_j) m T \sum_i a_i Y_i, \qquad (1)$$

where $(1 - \varepsilon_j)$ is the probability of not registering a neutron emitted in the detector unit j obtained as a result of Monte Carlo simulation; m is the material mass; T is the data-taking time; the index i indicates the decay chain under consideration or its part; a_i is the specific activity of the material; and Y_i is the neutron yield calculated on the basis of a dedicated code that takes into account the composition of the material. The specific activity is measured by three methods—namely, inductively coupled plasma mass spectrometry (ICP-MS), gamma spectrometry based on high purity Ge (HPGe) detectors, and polonium extraction [14]. These methods supplement one another since they are sensitive to different nuclides from decay chains.

The gamma background from $(\alpha, n\gamma)$ reactions is calculated in the same way with the only difference that, in expression (1), the gamma yields are used instead of the neutron yields.

[6)] A central detector or a water Cherenkov veto can be taken for an individual unit.

3. PROCEDURE FOR CALCULATING NEUTRON AND GAMMA-RAY YIELDS

In order to calculate the neutron yields and spectra, one can use one of the following programs: NeuCBOT [8, 10], SOURCES4A [15] or SOURCES4C [16], NEDIS-2.0 [17–20], USD [21], and SaG4n [9]. All of these utilities, with the exception of the last one, employ the same computational procedure.

For a single interaction in which the incident alpha particle has an energy E_α, while the emitted neutron has an energy E_n, the neutron yield Y is calculated according to the formula

$$Y(E_\alpha, E_n) = \int_0^{E_\alpha} \frac{\sigma(E'_\alpha, E_n)}{\xi(E'_\alpha)} dE'_\alpha, \qquad (2)$$

where $\sigma(E'_\alpha, E_n)$ is the reaction cross section and $\xi(E'_\alpha)$ is the material stopping power. In this expression, the integral is taken from zero since it is assumed that the probability for alpha particle capture is low, so that the particle moving in matter may lose its whole energy.

If it is necessary to calculate the neutron yield for a material of complex composition and for several alpha particles, then expression (2) takes a more complicated form; that is,

$$Y_i(E_n) \qquad (3)$$
$$= \sum_\alpha P_\alpha \sum_m \frac{N_\mathrm{A} C_m}{A_m} \int_0^{E_\alpha} \frac{\sigma_m(E'_\alpha, E_n)}{\xi(E'_\alpha)} dE'_\alpha,$$

where the first summation is performed over all alpha particles, while the second sum is taken over all nuclides according to the composition of the material and the indicated abundance of isotopes (natural or preset); P_α is the weight specified for the alpha particle of initial energy E_α or the branching ratio; N_A is Avogadro's number; C_m is the mass fraction of the nuclide; and A_m is its mass number.

In the case of employing the SaG4n program, the calculations are performed in a radically different way. The SaG4n program relies on the Geant4 simulation package [22–24], and this permits, within a virtual experiment, explicitly accomplishing the transport of incident alpha particles for facilities whose geometry may be complicated to any reasonable extent and simulating possible (α, n) reactions and parameters of product neutrons, as well as further motion of the particles, if necessary. Moreover, Geant4 also makes it possible to examine more complicated situations where there occurs associated production of neutrons with gammas or even with other particles (if this is possible). Thus, the SaG4n simulation

of (α, n) and $(\alpha, n\gamma)$ reactions is maximally close to reality. However, the SaG4n program has a number of disadvantages. In applying this utility, it is more difficult than in other ones to specify input data and to process output information. Moreover, SaG4n consumes much more computer time because of a simulation of each individual particle and reaction. Although a simulation with an ordinary PC takes, on average, several hours for simple cases involving one or a few materials for each decay chain or a set of alpha particles, the time of computations may increase to several days in the cases of improving the accuracy or employing a model of more complicated geometry. The situation may also become critical if it is necessary to determine particle yields for a list of materials that includes several tens or even several hundred items. Therefore, the application of other tools, such as NeuCBOT or SOURCES4, that may provide a similar accuracy of calculations within a shorter time is quite justified.

4. NeuCBOT

The NeuCBOT program was invented as a tool for evaluating the neutron background from (α, n) reactions in low background experiments, even though the development was conducted within the needs of the DarkSide-50 project aimed at dark matter searches. Shawn Westerdale was the author of the two first versions of this utility. Starting from the third version, the team of developers that includes three persons (S. Westerdale, M. Gromov, and I. Goncharenko) has been working on upgrading and improving it. The NeuCBOT program is written in the Python language and is adapted for running from a console using operating system from the Linux family. The program is distributed as free software under GNU General Public License v3.0. Different versions of the code and a user manual for its application are stored on the GitHub website of the project [10].

NeuCBOT relies, as follows from its spelt-out name—Neutron Calculator Based On TALYS—on the nuclear-reaction database (library) calculated by means of the TALYS code [25–27]. This database contains information about the cross sections for (α, n) and $(\alpha, n\gamma)$ reactions and about the spectra of emitted particles. The original version of NeuCBOT, denoted by v-1.0, employed the output data of TALYS-1.6 [26] or, in other words, information from the TENDL-2015 library [28]. A transition to TALYS-1.95 [26] and, accordingly, to TENDL-2019 [29] was accomplished in the second version (v-2.0). In the third version (v-3.0), support of the JENDL/AN-2005 database of evaluated cross sections [30, 31] was included as a supplement and an alternative to the TENDL-2019 database. For all versions of the NeuCBOT program, the stopping powers of various materials are taken from the database obtained by applying the set of the SRIM codes. If, for input information, the user indicates, instead of the list of alpha particles with preset energies and production branching ratios, nuclides that may undergo alpha decay or even trigger a decay chain, then information about the possible decay channels and the alpha particle energies is generated and is saved in NeuCBOT as a local database resulting from the search for respective data in ENSDF (Evaluated Nuclear Structure Data File) [32].

There are also other distinctions between the NeuCBOT versions. The first two versions permit calculating only the yields and spectra of neutrons, taking no account of the possibility of gamma-ray emission. In the third version, this drawback is taken into account and corrected, so that the yields and spectra of gammas can be obtained. Additionally, a transition from Python 2 to Python 3 was accomplished in the third version.

The third version of the program has a rather large number of advantages and disadvantages. The advantages include the following:

1. Easy to use: can be used by non-experts out-of-the-box after downloading.

2. Flexibility: the program is appropriate for experiments that employ various materials, which have various levels of contamination with alpha radioactive nuclei, and various assumptions on the presence of secular equilibrium.

3. Speed: if TENDL data have already been uploaded or preliminarily calculated, NeuCBOT yields there result within several minutes, but, if TALYS should be launched, the procedure lasts from several tens of minutes to several hours.

4. The code is written in the widespread and updated Python programming language. NeuCBOT is readily modifiable and adaptable to various needs.

5. Minimal set of dependences. Python 3 programming language and Bash command shell. Optionally, TALYS version 1.95 or higher.

6. Set of TALYS output files (set of libraries) for each chemical element. These files can be additionally uploaded from a remote store to the program directory by means of a script embedded in NeuCBOT. In this way, it is possible to dispense with launching TALYS and to

enhance the rate of computations of the yields and spectra by one to two orders of magnitude. Also, it is possible to upload data on reaction cross sections from JENDL, provided that they are available for the nuclides being considered.

7. If data obtained with the aid of TALYS are used, information about particle spectra is retrieved in addition to neutron and gamma yields.

8. alpha particle energies range between 0 and 10 MeV.

9. The code takes into account possible violation of secular equilibrium in the ^{238}U decay chain because of the presence of two long-lived nuclides—namely, ^{226}Ra and ^{210}Pb, whose half-lives are 1602 and 22.3 yr, respectively, and which, in contrast to other long-lived nuclei of this family, can be accumulated in materials. The former, ^{226}Ra, may be accumulated since it is an alkali-earth metal and, under the effect of one chemical process or another, go away from the primary material, where preceding members of the family, which belong to actinides remain. The accumulation of ^{210}Pb is due to the presence of ^{222}Rn in the gaseous phase under normal conditions. As a component of air, ^{222}Rn is readily transported, whereupon its daughter nuclei decay fast to the first long-lived nuclide, and this is precisely ^{210}Pb. If the calculations is performed under the assumption of broken secular equilibrium in the ^{238}U chain, equilibrium is assumed to be conserved in four parts of the series: from ^{238}U to ^{226}Ra, from ^{226}Ra (inclusive) to ^{210}Pb, and from ^{210}Pb (inclusive) to stable ^{206}Pb.

The following disadvantages are worthy of note:

1. The reactions are examined only in the bulk of materials without allowance for any edge effect (infinite medium). As a result, there is no option for the cases of a surface contaminations or thin films.

2. There is no possibility of specifying the configuration of objects, and this prevents analysis of complicated components of experimental facilities, including multilayered structures. Only a homogeneous distribution of contaminations over the materials is allowed.

3. No interaction of secondary particles (neutrons and gammas) after their production with surrounding media is taken into account.

4. Experimental data obtained over the past few years are not included in the libraries used.

5. Partial cross sections for interactions of excited states of daughter nuclei are not included in the calculations, and mechanisms for taking such interactions into account are not specified.

6. There is no option for calculating the realistic energy loss of alpha particles before the commencement of reactions.

5. COMPARISON OF THE RESULTS OF NEUTRON YIELD CALCULATIONS BASED ON VARIOUS VERSIONS OF NeuCBOT AND ALTERNATIVE PROGRAMS

The use of (α, n) cross sections obtained from the TALYS code exclusively was a serious drawback of the original version of the NeuCBOT program. Although the approaches used in TALYS to calculating nuclear reactions are tested and tuned on the basis of available experimental data, a good level of agreement cannot be attained for a number of cases. In particular, this is so for light nuclei. As was compellingly shown in [9], TALYS predicts smooth dependences of (α, n) cross sections on the energy of incident alpha particles in the energy range between 0 and 10 MeV, but, from experiments, we know that the dependences in question have either more features or a even a resonance structure. It is also worth noting that, for light nuclei, the cross sections calculated using TALYS are on average larger than the cross sections obtained from experiments. As a result, the NeuCBOT program systematically yielded overestimated neutron yields for organic compounds, for example, or for aluminum, silicon, and boron.

In [33] and [9], available experimental data were compared with the results of calculations performed on the basis of different codes by employing various libraries of (α, n) cross sections. It is shown that the effect of cross-section values on the ultimate result is significant, reaching several tens of percent or even one order of magnitude. Especially good agreement could be attained upon employing the JENDL/AN-2005 database of evaluated cross sections. Therefore, this database was included in NeuCBOT, with the result that agreement of the results of calculations with experimental values expectedly became better. By way of example, the respective comparison for the ^{232}Th chain is shown in Figs. 1 and 2.

If we examine attentively, the results presented in the figures, the following two conclusions suggest themselves. It is worth noting that these conclusions remain valid for other materials and other decay chains—that is, they are quite general. First, all

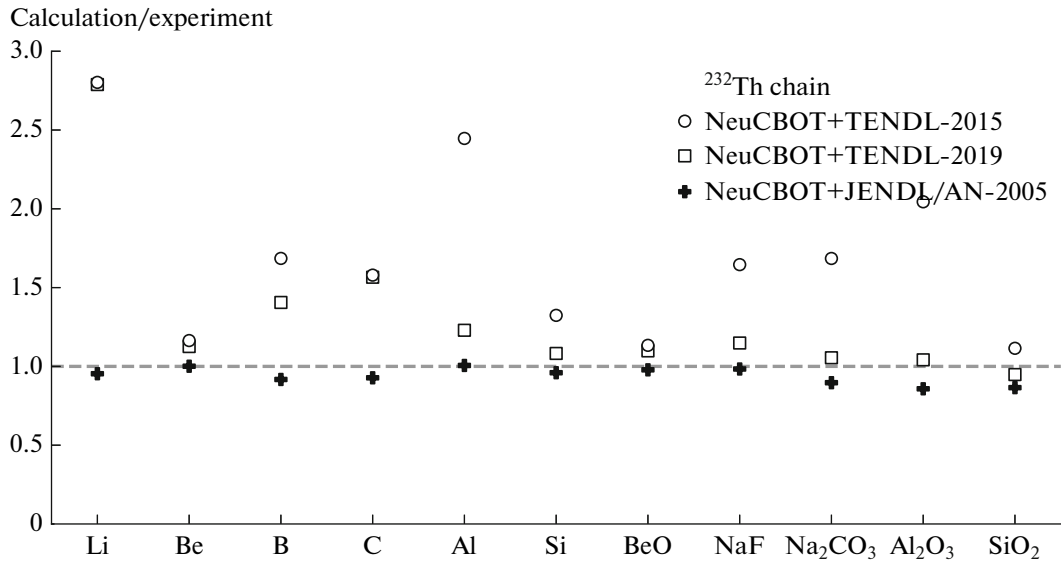

Fig. 1. Comparison of the neutron yields calculated for light nuclei by applying different versions of NeuCBOT. The numerical values are normalized to experimental data in order to demonstrate agreement between the calculations and measurements. The comparison in question is performed only for the thorium series.

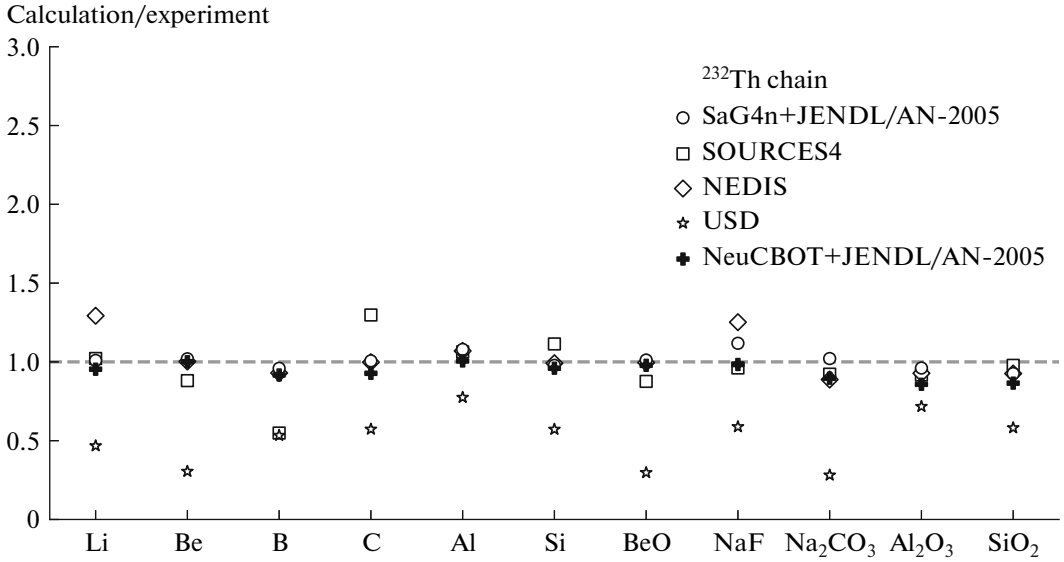

Fig. 2. Comparison of neutron yields calculated for light nuclei by applying different codes, including the upgraded version of NeuCBOT. The numerical values are normalized to experimental data in order to demonstrate agreement between the calculations and measurements. The comparison in question is performed only for the thorium series.

utilities, with the exception of USD, presently give close results, differing, in almost all of the cases, by not more than 20 to 30%. To some extent, this state of affairs is explained by common input experimental and evaluated data and by a computational procedure common to all of them, with the exception of the SaG4n package, as well as by timely support and upgrade of programs. The aforementioned level of the spread of the results is adequate to modern knowledge in these realms. Basically, the spread can be reduced upon the appearance of new precise experimental data—primarily for cross sections. Second, there is good agreement between the results obtained using SaG4n and NeuCBOT. This means that, for a number of not very complicated problems and for estimations, one can reliably apply NeuCBOT, thereby enhancing the rate of calculations without harboring

Table 1. Results of NeuCBOT calculations for neutron yields from (α, n) reactions for a number of metals and alloys that can be used as structural materials

Material	Neutron yield, 10^{-8} neutrons per decay of parent nucleus				
	^{232}Th	^{235}U	^{238}U upper	^{238}U middle	^{238}U lower
Cu	27	1.3	0	2.7	0
Cu20Ti80	510	180	0.15	200	0.7
Ti	620	220	0.18	240	0.9
VT1-00	625	230	0.4	240	1.1
VT1-0	630	230	0.6	250	1.45
Stainless steel 08X18H10T	190	39	0.13	51	0.18

Table 2. Results of NeuCBOT calculations for gamma yields from $(\alpha, n\gamma)$ reactions for a number of metals and alloys that can be used as structural materials

Material	Gamma yields, 10^{-10} gammas per decay of parent nucleus				
	^{232}Th	^{235}U	^{238}U upper	^{238}U middle	^{238}U lower
Cu	62	29.5	0.02	23	0.16
Cu20Ti80	72	29	0.06	29	0.18
Ti	74	29	0.07	30	0.19
VT1-00	74	29	0.11	30	0.23
VT1-0	74	30	0.16	30	0.27
Stainless steel 08X18H10T	32	23	0.4	16.4	1.3

doubts as to whether this would lead to serious errors or enhance substantially the systematic uncertainty.

6. NeuBOT CALCULATION OF THE GAMMA YIELD FROM $(\alpha, n\gamma)$ REACTIONS

The procedure for calculating gamma yields from $(\alpha, n\gamma)$ reactions does not differ from the approach used in calculating the neutron yields. Simultaneously, the TALYS code makes it possible to take into account a wide variety of reactions and to obtain, in output files, information about emitted particles in a unified form. Therefore, the inclusion of the calculation for $(\alpha, n\gamma)$ reactions in the NeuCBOT program is straightforward. From tests performed after this was done, it was found that, as might have been expected, the gamma yields are several orders of magnitude less than the neutron yields. For example, Tables 1 and 2 give respective values for various metals and alloys that can be used as structural materials in low background detectors. These values differ by one to three orders of magnitude. Such low gamma yields, which are less than 10^{-8} particles per decay of the parent nucleus (the first one in the chain) give grounds to disregard this background in the majority of cases.

In addition to information about gamma yields, the NeuCBOT program also outputs particle spectra in the same format as in the case of neutrons.

7. CONCLUSIONS

Owing to the inclusion of the evaluated cross sections from the JENDL/AN-2005 database in the new version of the NeuCBOT program, agreement of the neutron yields calculated for light nuclei with experimental data and with the results of calculations based on different software tools became substantially better. The deviation of the results of NeuCBOT calculations from available measured values does not exceed 20% in the majority of cases. The similarity of values calculated using the SaG4n package and the NeuCBOT program deserves particular attention, since SaG4n permits simulating tracks of alpha particles in matter and subsequent (α, n) reactions, as well as most closely reproducing physics processes. In view of the simplicity, speed, and flexibility of the NeuCBOT program, this tool is quite convenient and practicable for problems where there are no bodies complicated in shape and composition and where

there are no stringent requirements on the accuracy of calculations.

The updated version of the NeuCBOT program makes it possible to calculate gamma yields and spectra for $(\alpha, n\gamma)$ reactions. In addition, its code was fully recast from the Python 2 to the Python 3 language.

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