

This is a repository copy of Neutron production in (α, n) reactions in SOURCES4.

White Rose Research Online URL for this paper: <u>https://eprints.whiterose.ac.uk/205707/</u>

Version: Published Version

Proceedings Paper:

Kudryavtsev, V.A. orcid.org/0000-0002-7018-5827, Krawczun, P. and Bocheva, R. (2023) Neutron production in (α ,n) reactions in SOURCES4. In: SciPost Physics Proceedings. 14th International Conference on Identification of Dark Matter, 18-22 Jul 2022, Vienna, Austria. Stichting SciPost, 018.1-018.7.

https://doi.org/10.21468/scipostphysproc.12.018

Reuse

This article is distributed under the terms of the Creative Commons Attribution (CC BY) licence. This licence allows you to distribute, remix, tweak, and build upon the work, even commercially, as long as you credit the authors for the original work. More information and the full terms of the licence here: https://creativecommons.org/licenses/

Takedown

If you consider content in White Rose Research Online to be in breach of UK law, please notify us by emailing eprints@whiterose.ac.uk including the URL of the record and the reason for the withdrawal request.



eprints@whiterose.ac.uk https://eprints.whiterose.ac.uk/

Neutron production in (α, n) reactions in SOURCES4

Vitaly A. Kudryavtsev*, Piotr Krawczun and Rayna Bocheva

Department of Physics and Astronomy, University of Sheffield, Sheffield, S3 7RH, UK

* v.kudryavtsev@sheffield.ac.uk



14th International Conference on Identification of Dark Matter Vienna, Austria, 18-22 July 2022 doi:10.21468/SciPostPhysProc.12

Abstract

Neutrons produced in spontaneous fission and (α, n) reactions can induce background events in underground experiments looking for rare processes. A number of computer codes are available to calculate cross-sections of (α, n) reactions, branching ratios to various states and neutron yields. SOURCES4 code has been used in this work to calculate neutron yields and energy spectra with input cross-sections and branching ratios taken from experimental data and models from EMPIRE2.19/3.2.3 and TALYS1.9 codes. A comparison of SOURCES4 calculations with experimental data from alpha beams and radioactive decay chains is presented.

© Opyright V. A. Kudryavtsev *et al*. This work is licensed under the Creative Commons Attribution 4.0 International License. Published by the SciPost Foundation. Received 30-09-2022 Accepted 28-04-2023 Published 03-07-2023 doi:10.21468/SciPostPhysProc.12.018



Neutrons from radioactivity in environment and detector components affect the sensitivity of low-background experiments, constructed in underground laboratories and searching for rare events such as dark matter or neutrino interactions. These neutrons are produced in spontaneous fission and (α, n) reactions and can mimic, for instance, nuclear recoils from WIMP-nucleus interactions in direct dark matter searches.

Several computer codes are available to calculate neutron yields and energy spectra form these processes (see, for instance, Refs. [1–4]). Spontaneous fission (SF) is well described by the parameterisation [5] with parameters tuned to the measurements. The neutron yield from SF does not depend on the material where the neutron emission happens, but only on the concentration of ²³⁸U (neglecting other radioactive isotopes with very small neutron yields from this process). To characterise a background in an experiment, an accurate calculation of the neutron yield from (α , n) reactions is needed, where an alpha particle is originated from the decays of radioactive isotopes of uranium and thorium and their daughters. The codes usually use as inputs cross-sections of these reactions and transition probabilities to excited states together with energy losses of alphas as they travel through the material until they stop. A comparison of neutron yields and spectra as calculated in different codes and experimental data has been given in several papers, see for instance Refs. [2–4, 6, 7]. In this paper, we report the new calculations of neutron production with the SOURCES4 code that includes "optimised" cross-sections and branching ratios to excited states. This "optimisation" includes a combination of recent experimental data for the cross-sections and a model where the data are not available. We focus here on (α , n) reactions caused by alphas up to 10 MeV as typical for radioactive decay chains of ²³⁸U, ²³⁵U and ²³²Th.

2 Neutron production in the SOURCES4 code

The nuclear physics code SOURCES4 [4] has been used for a long time in a number of applications, including particle physics and particle astrophysics experiments located in deep underground laboratories. The code libraries contain alpha emission lines from radioactive isotopes, cross-sections of (α, n) reactions from calculations and experimental data, probabilities of transitions to different final states of the daughter nucleus and parameterisations for energy losses of alphas in different materials. The code calculates the neutron production rates (or yields) and energy spectra of emitted neutrons for several types of problems. We consider here the thick target approach where the size of the material sample is assumed to be much bigger than the range of alphas so edge effects can be neglected. The version SOURCES4A [8] of the code is used here for historical reasons. Previous tests confirmed that this version give the same result as the most recent version SOURCES4C [4] if the same cross-sections and transition probabilities to excited states are used in both versions. A big advantage of the code is its flexibility so that a user can choose what cross-section of (α, n) reaction is used for a particular isotope in a material sample. The user can also add more cross-sections to the library.

The original code calculates neutron production for alpha energies up to 6.5 MeV and is not fully suitable for calculation of neutrons from radioactive processes that involve alphas with energies up to about 9 MeV. The original code SOURCES4A was modified to remove the 6.5 MeV energy cut and an updated version now allows calculations of neutron production from alphas with energies up to 10 MeV [9, 10]. Libraries were initially updated to include cross-sections from the EMPIRE2.19 code [11] and to extend energy range for alphas up to 10 MeV [9, 10, 12–14]. A comparison of cross-sections from EMPIRE2.19 with some experimental data was published in Refs. [10, 14] and the results from the modified SOURCES4A code were used in a number of dark matter experiments (see, for example, Refs. [15–18]).

The user input to SOURCES4A includes either the energy of an alpha particle or the *Z* and *A* of the radioactive isotope (or several isotopes in the case of decay chains, for instance) with the number of atoms per unit volume. The user should also specify material composition (where the alpha sources are located) and isotopic composition for each element (only isotopes with cross-sections present in the code library can be included).

The output of SOURCES4A includes the neutron yield and spectra for the sum of the ground and all excited states, as well as neutron spectra for individual states. The output also includes neutron production spectra on each isotope in the material sample. In the case of decay chains, neutron production from individual radioactive isotopes on each isotope in the material sample is also returned by the code. The code does not generate gammas produced from de-excitation of a nucleus in the final state.

Recently, the libraries of SOURCES4A have been updated [19] to include some crosssections calculated with TALYS1.9 [20] and the newer version of EMPIRE3.2.3 [21]. Ref. [19] includes comparison of cross-sections from TALYS1.9, EMPIRE2.19, EMPIRE3.2.3 and experimental data, as well as the results for neutron yields calculations using SOURCES4A with different cross-sections. The most recent development includes an update of the libraries of cross-sections and branching ratios in SOURCES4A so the most reliable data are used when available and the model calculations from either TALYS1.9 or EMPIRE2.19/3.2.3 are used for isotopes and alpha energies where the data are scarce or unavailable. The same model is used for branching ratios since there are very few data on this in literature. We will refer to this approach and cross-sections as "optimised".

3 SOURCES4 results and comparison with other codes and experimental data

Here we present the results from SOURCES4A calculations using "optimised" cross-sections and branching ratios. Where possible, the measured cross-sections have been used as an input to neutron yield and spectra calculations. If different sets of data are inconsistent, or there are no data for a particular isotope, the model from either TALYS1.9 or EMPIRE2.19/3.2.3 is used. Branching ratios were calculated using TALYS1.9 or EMPIRE2.19/3.2.3. The results of these calculations for a number of elements are compared to different data sets that were obtained from α -particle beams interacting with thick targets composed of different elements/isotopes. This comparison provides an independent test of neutron yield calculations. First results have been presented at the LRT2022 workshop and published in [22].

As an example, we show in Figure 1 the neutron yield from fluorine as a function of α -particle energy. The measurements for natural fluorine have been reported in Refs. [23–25]. We have chosen fluorine here as it is a quite common material in underground experiments (used as a light reflector) and has a very high neutron yield due to (α , n) reactions. SOURCES4A calculations use the cross-sections for ¹⁹F from Ref. [26] up to 6.7 MeV. Above this energy and for branching ratios the model from TALYS1.9 [20] was implemented (see also Ref. [27] for the comparison of data and models for ¹⁹F and some other isotopes). The neutron yields from SOURCES4A are in good agreement with data.

Figure 2 shows the neutron energy spectrum from fluorine as calculated by SOURCES4A in comparison with the measurements from Ref. [25] for 5 MeV alpha beam. Again, a good agreement is seen. A similar agreement (within about 20%) was observed for most isotopes checked.

Figure 3 shows the neutron yield for several materials from 238 U decay chain as calculated by different codes in comparison with experimental data. The neutron yield is given as the number of neutrons per 10⁶ alphas to be directly compared with other publications [3,6]. The decay chain is assumed to be in equilibrium. There are 8 alphas in the 238 U chain so to convert this to the total neutron production rate per gram per second per ppb of uranium the value from the figure needs to be multiplied by $8 \times 1.245 \times 10^{-5}/10^{6}$. Here again we have used optimised cross-sections in SOURCES4A – a combination of experimentally measured crosssections and calculations either with TALYS1.9 or EMPIRE2.19/3.2.3. Data from the NEDIS-2.0 code were taken from [28]. USD data were obtained with the web-based toolkit developed at the University of South Dakota (USA) [1] and reported in Ref. [6]. NeuCBOT calculations have been reported in Ref. [2]. Yields from GEANT4 simulations using two libraries JENDL [29] and TENDL-2017 [20] were calculated in Ref. [3]. The "experimental data" were not the results of direct measurements of neutron yields from the whole decay chain but were evaluated from the measured neutron yields for different alpha energies as reported in Ref. [6]. A good agreement between SOURCES4A and evaluated data is seen for most materials.

Ref. [30] reported direct measurement of neutron yields from uranium and thorium decay chains and the comparison of SOURCES4A output with these data for thorium chain is shown in Figure 4. The calculations are again in very good agreement with measurements.



Figure 1: Neutron yield from fluorine as a function of α energy. SOURCES4A calculations with cross-sections from Ref. [26] up to 6.7 MeV and TALYS1.9 model above 6.7 MeV, are plotted together with the data from Refs. [23–25].



Figure 2: Neutron energy spectra from SOURCES4A and measurements from Ref. [25] for a 5 MeV energy alpha beam hitting a fluorine target.



Figure 3: Neutron yields from 238 U decay chain in several materials as calculated by different codes in comparison with measurements. Neutron yield is given as the number of neutrons per 10⁶ alphas. See text for details.

4 Conclusion

We have presented the "optimised" approach to the inputs to SOURCES4A code when the cross-sections are taken from existing experimental data where consistency between different data sets is observed, complemented with models from either TALYS or EMPIRE nuclear reaction codes. With this input, we have calculated neutron yields as functions of alpha energy



Figure 4: Neutron yields from 238 U decay chain in several materials as calculated by SOURCES4A in comparison with measurements. Neutron yield is given as the number of neutrons per 10^6 alphas. See text for details.

for a number of elements and materials and compared them to the measurements carried out with alpha particle beams. A good agreement is seen for most isotopes and materials tested. Calculated neutron energy spectra also show a reasonable agreement with the measurements. We have also presented a comparison of SOURCES4A calculations of neutron yields from radioactive decay chains with measurements and other codes.

Acknowledgements

We acknowledge support from the UKRI-STFC and the University of Sheffield.

References

- [1] D.-M. Mei, C. Zhang and A. Hime, *Evaluation of* (α, n) *induced neutrons as a background for dark matter experiments*, Nucl. Instrum. Methods Phys. Res. A: Accel. Spectrom. Detect. Assoc. Equip. **606**, 651 (2009), doi:10.1016/j.nima.2009.04.032.
- [2] S. Westerdale and P. D. Meyers, *Radiogenic neutron yield calculations for low-background experiments*, Nucl. Instrum. Methods Phys. Res. A: Accel. Spectrom. Detect. Assoc. Equip. 875, 57 (2017), doi:10.1016/j.nima.2017.09.007.
- [3] E. Mendoza et al., *Neutron production induced by α-decay with GEANT4*, Nucl. Instrum. Methods Phys. Res. A: Accel. Spectrom. Detect. Assoc. Equip. **960**, 163659 (2020), doi:10.1016/j.nima.2020.163659.
- [4] W. B. Wilson, R. T. Perry, W. Charlton, T. A. Parish and E. F. Shores, SOURCES: A code for calculating (α, n), spontaneous fission, and delayed neutron sources and spectra, Radiat. Prot. Dosim. 115, 117 (2005), doi:10.1093/rpd/nci260.
- [5] B. E. Watt, Energy spectrum of neutrons from thermal fission of ²³⁵U, Phys. Rev. 87, 1037 (1952), doi:10.1103/PhysRev.87.1037.
- [6] A. C. Fernandes, A. A. Kling and G. N. Vlaskin, *Comparison of thick-target* (α, n) yield calculation codes, EPJ Web Conf. **153**, 07021 (2017), doi:10.1051/epjconf/201715307021.

- [7] J. Cooley, J. K. Palladino, H. Qiu, M. Selvi, S. Scorza and C. Zhang, *Input comparison of radiogenic neutron estimates for ultra-low background experiments*, Nucl. Instrum. Methods Phys. Res. A: Accel. Spectrom. Detect. Assoc. Equip. 888, 110 (2018), doi:10.1016/j.nima.2017.11.028.
- [8] W. B. Wilson et al., SOURCES4A: A code for calculating (α, n), spontaneous fission, and delayed neutron sources and spectra, Technical Report LA-13639-MS (1999), doi:10.2172/15215.
- [9] M. J. Carson et al., Neutron background in large-scale xenon detectors for dark matter searches, Astropart. Phys. 21, 667 (2004), doi:10.1016/j.astropartphys.2004.05.001.
- [10] V. Tomasello, V. A. Kudryavtsev and M. Robinson, *Calculation of neutron background for underground experiments*, Nucl. Instrum. Methods Phys. Res. A: Accel. Spectrom. Detect. Assoc. Equip. 595, 431 (2008), doi:10.1016/j.nima.2008.07.071.
- M. Herman et al., *EMPIRE: Nuclear reaction model code system for data evaluation*, Nucl. Data Sheets 108, 2655 (2007), doi:10.1016/J.NDS.2007.11.003.
- [12] R. Lemrani, M. Robinson, V. A. Kudryavtsev, M. De Jesus, G. Gerbier and N. J. C. Spooner, *Low-energy neutron propagation in MCNPX and GEANT4*, Nucl. Instrum. Methods Phys. Res. A: Accel. Spectrom. Detect. Assoc. Equip. 560, 454 (2006), doi:10.1016/j.nima.2005.12.238.
- [13] V. Tomasello, M. Robinson and V. A. Kudryavtsev, *Radioactive background* in a cryogenic dark matter experiment, Astropart. Phys. 34, 70 (2010), doi:10.1016/j.astropartphys.2010.05.005.
- [14] V. Tomasello, *Background simulations for a large-scale cryogenic dark matter experiment*, PhD. Thesis, University of Sheffield (2009).
- [15] G. Angloher et al., EURECA conceptual design report, Phys. Dark Universe 3, 41 (2014), doi:10.1016/j.dark.2014.03.004.
- [16] E. Armengaud et al., Background studies for the EDELWEISS dark matter experiment, Astropart. Phys. 47, 1 (2013), doi:10.1016/j.astropartphys.2013.05.004.
- [17] D. S. Akerib et al., Projected WIMP sensitivity of the LUX-ZEPLIN dark matter experiment, Phys. Rev. D 101, 052002 (2020), doi:10.1103/PhysRevD.101.052002.
- [18] E. Aprile et al., XENON1T dark matter data analysis: Signal reconstruction, calibration, and event selection, Phys. Rev. D 100, 052014 (2019), doi:10.1103/PhysRevD.100.052014.
- [19] V. A. Kudryavtsev, P. Zakhary and B. Easeman, *Neutron production in (α, n) reactions*, Nucl. Instrum. Methods Phys. Res. A: Accel. Spectrom. Detect. Assoc. Equip. **972**, 164095 (2020), doi:10.1016/j.nima.2020.164095.
- [20] A. J. Koning and D. Rochman, *Modern nuclear data evaluation with the TALYS code system*, Nucl. Data Sheets **113**, 2841 (2012), doi:10.1016/j.nds.2012.11.002.
- [21] M. Herman, R. Capote, M. Sin et al., EMPIRE-3.2 Malta modular system for nuclear reaction calculations and nuclear data evaluation; User's manual, Tech. rep., Brookhaven National Laboratory (2013), doi:10.2172/1108585.

- [22] V. A. Kudryavtsev, P. Krawzcun and R. Bocheva, *Calculation of neutron production in* (α, n) *reactions with SOURCES4*, in 8th topical, workshop on low radioactivity techniques, (arXiv preprint) doi:10.48550/arXiv.2211.02080.
- [23] J. K. Bair and J. Gomez del Campo, *Neutron yields from alpha-particle bombardment*, Nucl. Sci. Eng. **71**, 18 (1979), doi:10.13182/NSE71-18.
- [24] E. B. Norman, T. E. Chupp, K. T. Lesko, P. J. Grant and G. L. Woodruff, ${}^{19}F(\alpha, n)$ thick target yield from 3.5 to 10.0 MeV, Appl. Radiat. Isot. 103, 177 (2015), doi:10.1016/j.apradiso.2015.04.018.
- [25] G. J. H. Jacobs and H. Liskien, Energy spectra of neutrons produced by α particles in thick targets of light elements, Ann. Nucl. Energy 10, 541 (1983), doi:10.6100/IR104719.
- [26] W. A. Peters et al., A kinematically complete, interdisciplinary, and co-institutional measurement of the ${}^{19}F(\alpha, n)$ cross section for nuclear safeguards science, Idaho technical report INL/EXT-16-38791 (2016), doi:10.2172/1263500.
- [27] V. A. Kudryavtsev, P. Krawzcun and R. Bocheva, *Calculation of neutron production in (α, n) reactions with SOURCES4*, IDM2022, Vienna, Austria (2022), https://indico.cern.ch/event/922783/contributions/4892800/attachments/2481355/ 4264640/alphan-sources4-VK-IDM2022.
- [28] G. N. Vlaskin, Y. S. Khomyakov and V. I. Bulanenko, Neutron yield of the reaction (α, n) on thick targets comprised of light elements, At. Energy 117, 357 (2015), doi:10.1007/s10512-015-9933-5.
- [29] T. Murata, H. Matsunobu and K. Shibata, *Evaluation of the* (α, xn) *reaction data for JENDL/AN-2005*, Jpn. At. Energy Agency, JAEA-Research 2006-052 (2006), doi:10.11484/jaea-research-2006-052.
- [30] G. V. Gorshkov and O. S. Tsvetkov, Be, B, C, O, F, Na, Mg, Al and Si, neutron yields from an (α, n) reaction under the effect of thorium and uranium α -particles and their decay products, At. Energiya 14, 550 (1963).