An investigation on neutron induced reactions on stable CNO isotopes (Postprint)

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Abstract

The neutron induced reactions on stable Carbon, Nitrogen, and Oxygen isotopes are investigated by using the Talys1.4 toolkit with the default parameters. The neutron incident energy covers a range from 0.20 MeV to 85.00 MeV. For 12C and 14N, the Talys1.4 results agree with the experimental data, while the parameters should be adjusted for 16O. Some En windows are found by comparing the main channels of n + C/N/O reactions, which induce element change. In these En windows, a specific element is activated to a different one while leaving the other element atoms unchanged. The results will facilitate the research of doping effects in organic materials by using neutron activation technique.

Full Text

Preamble

An Investigation on Neutron Induced Reactions on Stable CNO Isotopes

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Neutron induced reactions on stable Carbon, Nitrogen, and Oxygen isotopes are investigated using the Talys1.4 toolkit with default parameters. The neutron incident energy covers a range from 0.20 MeV to 85.00 MeV. For ^{12}C and ^{14}N, the Talys1.4 results agree with experimental data, while parameters should be adjusted for ^{16}O. Energy windows are identified by comparing the main channels of n + C/N/O reactions that induce elemental change. In these energy windows, a specific element can be activated to a different one while leaving other elements unchanged. These results will facilitate research on doping effects in organic materials using neutron activation techniques.

Keywords: Neutron activation, Doping, Organic material, C/N/O activation
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Introduction

Doping is an important method for modifying material properties. For example, substituting certain atoms in organic semiconductors can greatly promote charge transport and stability [1, 2]. The conventional doping method is chemical synthesis, which introduces new elements into the original material. However, neutron activation represents an alternative important doping method that can directly change one element to another, either immediately or through the decay of unstable isotopes produced by activation. In this case, different doping effects can be expected compared to chemical doping.

It is well known that different nuclei have different threshold energy values for specific channels in neutron induced reactions [3]. For materials containing different elements, these varying threshold energies provide an opportunity for selective activation if the neutron energy is carefully chosen in experiments.

In this article, neutron induced reactions on stable carbon, nitrogen, and oxygen isotopes are investigated using the Talys toolkit. The energy windows between different isotopes are analyzed. The theoretical framework is briefly discussed in Sec. II. Results are presented in Sec. III, and a summary is given in Sec. IV.

II. Methods

The optical model can describe neutron induced reactions well across a wide range of incident energies. In Talys1.4, ECIS-06 is implemented as a subroutine to handle optical model calculations [4]. The description of Talys1.4 and its implemented functions can be found in the user manual [5, 6].

In statistical models for predicting cross sections, nuclear level densities are used at excitation energies where discrete level information is unavailable or incomplete. Several models are implemented in Talys to describe level density, ranging from phenomenological analytical expressions to tabulated level densities derived from microscopic models. The constant temperature and Fermi-gas model is set as the default parameter for the low excitation energy region, while the Fermi-gas model is used in the high excitation energy region [6].

Due to the existence of threshold energies for different channels in neutron induced reactions on a nucleus, a given channel can only occur if the incident neutron energy exceeds its threshold. This potentially provides energy windows where only one channel is active while others are prohibited. In this work, the incident energy for calculated reactions ranges from 0.20 MeV to 85.00 MeV, and the default parameters in Talys1.4 are adopted. The main reaction channels include (n, np), (n, p), (n, α), (n, 2n) and (n, γ). Calculated results are compared with measured data extracted from the EXFOR library provided by the National Nuclear Data Center (NNDC) [7]. All natural abundance data and isotope half-life values are taken from Wikipedia [8].

III. Results and Discussion

A. Neutron Induced Reactions on Carbon Isotopes

The element C has two stable isotopes, ^{12}C and ^{13}C, with natural abundances of 98.93% and 1.07%, respectively. Only the n + ^{12,13}C reactions are calculated.

1. n + ^{12}C reactions The ^{12}C(n, p)^{12}B, ^{12}C(n, α)^{9}Be, and ^{12}C(n, 2n)^{11}C channels have been measured previously. ^{12}B is an unstable nucleus that decays to ^{12}C via electron emission with a half-life of 20.20 ms. ^{9}Be is a stable isotope. ^{11}C is also unstable, decaying to ^{11}B via positron emission with a relatively long half-life of 20.334 min. Thus, the ^{12}C(n, 2n)^{11}C and ^{12}C(n, np)^{11}B channels are the main channels that result in elemental changes, both producing the same final product, ^{11}B.

The results are plotted in Fig. 1 [Figure 1: see original paper].

First, for the ^{12}C(n, α)^{9}Be channel, the Talys1.4 results are consistent with measured results when $E < 11.00$ MeV [9], but differ significantly from results measured by Stevens et al. [10] in the range 18.65 MeV $< E < 21.46$ MeV. In the energy range 20.00 MeV $< E < 60.70$ MeV, the Talys1.4 results are also much larger than those calculated by Dimbylow et al. [11]. Above $E > 11.00$ MeV, the results for the ^{12}C(n, α)^{9}Be channel show large discrepancies, suggesting that further experiments should be performed for systematic understanding.

Second, for the ^{12}C(n, p)^{12}B channel, measured results by Kreger et al. [12] and Rimmer et al. [13] coincide when $E < 16.00$ MeV, but differ at higher energies. The Talys1.4 results agree well with measured results when $E < 16.00$ MeV, but overestimate the measured results by Kreger et al. [12] in the range 16.00 MeV $< E < 22.00$ MeV. The calculated results for ^{12}C(n, p)^{12}B in the range 20.00 MeV $< E < 60.70$ MeV by Dimbylow et al. [11], which also uses the optical model, favor the measured results by Kreger et al. [12]. Meanwhile, Talys1.4 predicts similar results for the ^{12}C(n, p)^{12}B and ^{12}C(n, np)^{11}B reactions when $E < 20.00$ MeV.

Third, for the ^{12}C(n, 2n)^{11}C channel, which has been measured by many groups, the measured data agree well when $E < 27.00$ MeV. In the range 27.00 MeV $< E < 40.00$ MeV, the results can be divided into two groups: the upper group measured by Welch et al. [14], Anders et al. [15], and Kim et al. [16]; and the bottom group by Brill et al. [17], Uno et al. [18], Brolley et al. [19], and

Soewarsono et al. [20]. The calculated results by Dimbylow et al. [11] prefer the upper group results when E < 35.00 MeV. Kim et al. [16] measured results in the energy range from 55.00 MeV to 64.00 MeV. The Talys1.4 calculated results largely overestimate the measured ones when E < 50.00 MeV, but agree with the measured results by Kim et al. [16]. The calculated results by Dimbylow et al. [11] underestimate the measured results by Kim et al. [16].

The threshold energies of the $^{12}C(n, \alpha)^{9}Be$, $^{12}C(n, p)^{12}B$, $^{12}C(n, np)^{11}B$ and $^{12}C(n, 2n)^{11}C$ channels increase progressively, with values of approximately 6.18 MeV, 13.64 MeV, 14.89 MeV and 20.30 MeV, respectively. The $^{12}C(n, \gamma)^{13}C$ channel occurs across the entire energy range but with much lower probability compared to other channels.

2. n + ^{13}C reactions No measured data for neutron induced reactions on ^{13}C were found. Only the Talys1.4 calculated results are plotted in Fig. 2 [Figure 2: see original paper]. The threshold energy values increase for the (n, α), (n, 2n), (n, p) and (n, np) channels, with values of 4.13 MeV, 5.33 MeV, 13.64 MeV and 16.50 MeV, respectively. The (n, γ) channel has no lowest energy threshold. The (n, γ) and (n, p) channels have relatively low probabilities compared to other channels. Across the entire calculated energy range, when E < 15.00 MeV, the main channel is $^{13}C(n, \alpha)^{10}Be$, in which ^{10}Be decays to ^{10}B by electron emission with a very long half-life of 1.39×10^{6} years; when E > 15.00 MeV, the dominant channel is $^{13}C(n, np)^{12}B$, in which ^{12}B mainly decays to ^{12}C with a half-life of 20.20 ms.

B. Neutron Induced Reactions on Nitrogen Isotopes

The element N has two stable isotopes, ^{14}N and ^{15}N, with natural abundances of 99.64% and 0.36%, respectively. Only the n + $^{14,15}N$ reactions are calculated.

1. n + ^{14}N reactions The calculated channels for the n + ^{14}N reaction are $^{14}N(n, np)^{13}C$, $^{14}N(n, p)^{14}C$, $^{14}N(n, \alpha)^{11}B$, $^{14}N(n, \gamma)^{15}N$, and $^{14}N(n, 2n)^{13}N$ reactions. The nuclei ^{13}C, ^{15}N, and ^{11}B are stable, while ^{14}C and ^{13}N are unstable. ^{13}N decays to ^{13}C by positron emission with a half-life of 9.965 min. The (n, np), (n, p), (n, 2n), and (n, α) channels are the main ones that result in elemental changes.

The results of the n + ^{14}N reaction are plotted in Fig. 3 [Figure 3: see original paper]. For clarity, the results are shown in different panels. The $^{14}N(n, \alpha)^{11}B$ and $^{14}N(n, p)^{14}C$ channels have small threshold energies that are very similar. The $^{14}N(n, \alpha)^{11}B$ channel has been measured by different groups [21-25], and $^{14}N(n, p)^{14}C$ has also been measured [21, 23-25]. For both channels, data measured by different groups are consistent. The Talys1.4 results agree with measured data at low incident energies but overestimate the measured results as E increases.

For the $^{14}N(n, 2n)^{13}N$ channel, measured results [26-30] are consistent when E

< 19.00 MeV. When E > 24.00 MeV, measured results by Brill et al. [17] and Yashima et al. [31] are relatively consistent, and calculated results by Dimbylow et al. [11] also agree with measured data but have relatively large errors. The threshold energy predicted by Talys1.4 is E 11.00 MeV, but the Talys1.4 cross sections overestimate measured results across the entire energy range, increasing rapidly with E and reaching a maximum at E = 24.00 MeV, then decreasing when E > 24.00 MeV. Since ^{13}N decays to ^{13}C, the final product is the same as the ^{14}N(n, np)^{13}C channel.

The ^{14}N(n, np)^{13}C channel has a low threshold energy of about 6.00 MeV, and the cross section increases rapidly with E , peaking at about E = 14.00 MeV. The threshold energies increase in the order of (n, α), (n, np), and (n, 2n), with values of about 0.13 MeV, 5.67 MeV and 11.27 MeV, respectively. The (n, γ) and (n, p) channels occur across the entire E range and form peaks around 19.00 MeV and 9.50 MeV with wide widths, but their cross sections are very small compared to other channels.

2. n + ^{15}N reactions No measured data for the n + ^{15}N reaction were found. Only the Talys1.4 calculated results are plotted in Fig. 4 [Figure 4: see original paper]. The calculated results for the (n, np) and (n, α) channels have almost the same values when E < 12.00 MeV, and the (n, np) channel has much larger values than the (n, α) channel. Meanwhile, the cross sections of the (n, p) and (n, 2n) channels only show small differences when E > 17.00 MeV. The (n, np), (n, α), (n, p), and (n, 2n) channels have relatively similar threshold energy values of about 8.57 MeV, 8.09 MeV, 9.55 MeV and 11.56 MeV, respectively. The cross sections of the (n, γ) channel are much smaller compared to other channels. When E > 40.00 MeV, large fluctuations are found in the results of the (n, p) and (n, γ) channels.

C. Neutron Induced Reactions on Oxygen Isotopes

The element O has three stable isotopes, ^{16}O, ^{17}O, and ^{18}O, with natural abundances of 99.75%, 0.0038%, and 0.205%. Only the n + ^{16},^{18}O reactions are calculated.

1. n + ^{16}O reactions In Fig. 5 [Figure 5: see original paper], the results of the n + ^{16}O reactions are plotted. The channels include ^{16}O(n, α)^{13}C, ^{16}O(n, p)^{16}N, ^{16}O(n, 2n)^{15}O, ^{16}O(n, np)^{15}N, and ^{16}O(n, γ)^{17}O. ^{16}N decays to ^{16}O by electron emission with a half-life of 7.13 s. Thus, the main channels that cause elemental change are ^{16}O(n, α)^{13}C and ^{16}O(n, np)^{15}N.

For the ^{16}O(n, α)^{13}C channel, the Talys1.4 calculated results are consistent with those measured by Johnson et al. [32] and Seitz et al. [33], except for those by Divatia et al. [34] when E is smaller than 5.00 MeV. When E > 7.00 MeV, measured results by Dickens et al. [35], Bormann et al. [36], and Dandy et al. [37] are consistent, but the Talys1.4 calculated results are unable to reproduce the measured data well.

For the ^{16}O(n, p)^{16}N channel, measured results by Martin et al. [38], Bormann et al. [39], and Seeman et al. [40] agree well. Measured results by Subashi et al. [41] and DeJuren et al. [42] also agree with those results but show relatively large differences. The Talys1.4 calculated results largely underestimate the measured results but show similar trends.

For the ^{16}O(n, 2n)^{15}O channel, measured results by Yashima et al. [33] and Brill et al. [17] are in different energy ranges, but for the overlapping E range, the results show some differences. The Talys1.4 results overestimate the measured ones when E $<$ 40.00 MeV but underestimate them when E $>$ 40.00 MeV.

The ^{16}O(n, np)^{15}N and ^{16}O(n, γ)^{17}O channels have not been measured. For the ^{16}O(n, np)^{15}N channel, the probability increases rapidly above the threshold energy of 10.57 MeV and reaches a maximum value around E = 20.00 MeV. The ^{16}O(n, γ)^{17}O channel occurs across the entire E range but has much smaller values.

The Talys1.4 calculated threshold energies of the (n, α), (n, p), (n, np) and (n, 2n) channels for ^{16}O are 2.36 MeV, 10.25 MeV, 10.57 MeV and 16.65 MeV, with peaks forming at around 10.00 MeV, 14.50 MeV, 26.00 MeV and 28.00 MeV, respectively.

2. n + ^{18}O reactions The main channels covered by the n + ^{18}O reactions are ^{18}O(n, α)^{15}C, ^{18}O(n, np)^{17}N, ^{18}O(n, p)^{18}N, ^{18}O(n, 2n)^{17}O, and ^{18}O(n, γ)^{19}O. ^{17}N can decay to ^{16}O and ^{17}O with a half-life of 4.173 s; ^{18}N can decay to ^{18}O, ^{14}C, or ^{17}O with a half-life of 622 ms, all of which are stable nuclei (^{14}C has a very long half-life).

In Fig. 6 [Figure 6: see original paper], the calculated results for the channels are plotted. The thresholds of the (n, α), (n, 2n), (n, p), and (n, np) channels are 5.29 MeV, 8.49 MeV, 13.85 MeV and 14.49 MeV, and the peaks form at around 10.00 MeV, 17.00 MeV, 18.00 MeV and 49.50 MeV, respectively. When E $>$ 33.00 MeV, the yield of the (n, p) channel shows large fluctuations with E .

D. Comparison Between Main Channels of n + C/N/O Isotopes

The differences between threshold energies of different channels make it possible to change one element to another. To specifically change elements in organic materials, the incident neutron energy should be selected to match the energy window, as illustrated in the results above. Comparing the thresholds of main channels that induce elemental changes will clarify these energy windows.

For ^{12}C, both the final products of ^{12}C(n, np)^{11}B and ^{12}C(n, 2n)^{11}C are ^{11}B, since ^{11}C decays to ^{11}B via positron emission with a half-life of 20.30 ms. The threshold energy of ^{12}C(n, α)^{9}Be is about 6.18 MeV. When E $>$ 7.00 MeV, ^{12}C can be changed to ^{9}Be. The threshold energy of ^{12}C(n, np)^{11}B is about 14.89 MeV. When E $>$ 16.00 MeV, ^{12}C can be changed to both ^{9}Be and ^{11}B.

This provides practical application of neutron activation on C isotopes. Chemically synthesized compounds in which a C atom is substituted by a B atom demonstrate a novel molecular engineering concept for organic semiconductors [43].

For ^{14}N, the most important channel is (n, 2n), which occurs at very low neutron incident energy. Since the final yields in the n + ^{14},^{15}N reactions are mainly carbon isotopes, we do not discuss the energy window for these channels.

For ^{16}O, the main channels are ^{16}O(n, α)^{13}C and ^{16}O(n, np)^{15}N when E $<$ 15.00 MeV, producing ^{13}C and ^{15}N, respectively. There is also an energy window between these two channels in the range from 4.00 MeV to 11.00 MeV. At higher E , the ^{16}O(n, 2n)^{15}O channel opens. ^{15}O decays to ^{14}N by emitting a positron with a half-life of 70.60 s, meaning ^{16}O is ultimately changed to ^{14}N.

For better understanding of the energy windows, the energy above the Talys-calculated threshold for each channel is plotted in Fig. 8 [Figure 8: see original paper]. The numbers from 1 to 24 represent the channels, and the letters from a to w represent the threshold energy values, which are also listed as follows (units in MeV): 1, E = 0 for the ^{14}N(n, p)^{14}C and (n, γ) channels of C/N/O; 2, ^{14}N(n, α)^{11}B (a = 0.13); 3, ^{16}O(n, α)^{13}C (b = 2.36); 4, ^{13}C(n, α)^{10}Be (c = 4.13); 5, ^{18}O(n, α)^{15}C (d = 5.29); 6, ^{13}C(n, 2n)^{12}C (e = 5.33); 7, ^{14}N(n, np)^{13}C (f = 5.67); 8, ^{12}C(n,α)^{9}Be (g = 6.18); 9, ^{15}N(n, α)^{12}B (h = 8.09); 10, ^{18}O(n, 2n)^{17}O (i = 8.49); 11, ^{15}N(n, np)^{14}C (j = 8.57); 12, ^{15}N(n, p)^{15}C (k = 9.55); 13, ^{16}O(n, p)^{16}N (l = 10.25); 14, ^{16}O(n, np)^{15}N (m = 10.57); 15, ^{14}N(n, 2n)^{13}N (n = 11.27); 16, ^{15}N(n, 2n)^{14}N (o = 11.56); 17, ^{13}C(n, p)^{13}B (p = 13.635); 18, ^{12}C(n, p)^{12}B (q = 13.645); 19, ^{18}O(n, p)^{18}N (r = 13.85); 20, ^{18}O(n, np)^{17}N (s = 14.49); 21, ^{12}C(n, np)^{11}B (t = 14.89); 22, ^{13}C(n, np)^{12}B (u = 16.49); 23, ^{16}O(n, 2n)^{15}O (v = 16.65); 24, ^{12}C(n, 2n)^{11}C (w = 20.30). Although the energy windows are clearly shown in Fig. 8, careful analysis is required when neutrons are used to activate specific compounds containing different elements.

IV. Conclusion

In this article, neutron induced reactions on stable C, N, and O isotopes are investigated using the Talys1.4 toolkit, which calculates reactions within the optical model framework. On one hand, it is found that for ^{12}C and ^{14}N, the Talys1.4 results agree with experimental data, while for ^{16}O, parameters in Talys1.4 should be adjusted for better prediction. On the other hand, a systematic comparison among the main channels of n + C/N/O reactions that induce elemental change is performed to identify energy windows among the original C, N, and O stable isotopes (considering the final production of each channel, i.e., direct change or indirect change through decay to a different elemental isotope). In these energy windows, specific elements can be activated to different ones while leaving other elements unchanged. These results may help study material modification using neutron induced doping techniques, such as in organic materials like organic semiconductors.

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