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Response function generation for the transmutation of ^{129}I by high energy neutron and proton interactions in the MCNPX Monte Carlo code

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Abstract For long-term use of the nuclear energy program, control and management of nuclear waste generated in a reactor are essential. In recent decades, the transmutation of long-lived fission products (LLFPs) into some stable or short-lived nuclei by accelerator driven systems (ADS) has been investigated and developed as one of the methods to reduce the radioactive contamination of spent fuel. In MCNPX code, the interaction tally calculates the transmutation rate of nuclei for which tabulated data exist, but does not score in the model physics regime. To circumvent this problem, this study proposes a response function that is consistent with the physics models used in the MCNPX code. This is used to estimate the transmutation rate of ^{129}I in an ADS containing protons and spallation neutrons with energies up to 1500 MeV. To generate this response function, a very thin ^{129}I target is used, the small size of the target allows attenuation and energy loss to be ignored. To do so, for neutrons, 168 energy bins from 10 MeV to 1500 MeV, and for protons, 120 energy bins from 1 MeV to 1500 MeV are considered.

1 Introduction

Nuclear waste generated in a reactor contains a wide range of highly toxic products with long-lived, and the management of these LLFPs is one of the challenges of using nuclear energy. Due to the mobility of some of these LLFPs, their direct underground disposal is not acceptable because, in case of leakage, these radionuclides may contaminate underground water channels or food chains. Therefore, alternative methods are needed to reduce the hazard of these LLFPs. One such approach is the transmutation of these LLFPs by the accelerator-driven system (ADS) into some stable or short-lived radionuclides [1–4]. In the transmutation process, one isotope is converted to another isotope by receiving or losing one (or multiple) nucleon(s). This process can reduce the activity, mass, volume, heat load or radiotoxicity of nuclear waste [5, 6]. Diverse studies have been performed on the transmutation of LLFPs by ADS [7–11]. In this work, by generating the transmutation response function, we make it possible for the computation of the transmutation rate for ^{129}I at high energies in MCNPX code. ^{129}I is the longest-lived radioactive fission product, with a half-life of 1.57×10^7 years.

In this study, the MCNPX code was used due to its availability, transport of charged particles, robust physics models and the ability to run in analog mode. MCNPX code has two ways of calculating cross sections: it can use tabulated data when the data exists or it can use physics models to calculate the cross sections "on the fly" [12]. In the physics regime of interest here, MCNPX model interactions use a combination of the cascade excitation model (CEM), the nuclear optical model and the Los Alamos version of the quark gluon string model (LAQGSM) [13, 14].

In MCNPX, the Interaction tally (FM tally multiplier card) calculates the transmutation rate of nuclei for which tabulated data exist. The tabulated data generally only exist for a limited energy range. For ^{129}I, the ENDF/B-VII.1 libraries are only evaluated up to 20 MeV. If the particle's energy is in the physics model regime (i.e., above the energy cutoff for the tabulated data), the interaction tally will not score any contributions, and the warning "FM tallies do not score in model physics regime" is given. For example, in the case of ^{129}I that neutrons cross-section library evaluated up to 20 MeV, neutrons with energies above 20 MeV will not contribute at all to the tally [12, 15, 16].

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2 Response function generation

The number of transmutations that occur in a pure target is equal to the number of neutrons and protons lost in nuclear interactions, because in the bombardment of ^{129}I with proton particles, any interaction leads to transmutation. To run the system in analog mode, the cutoff energy of the neutrons must be zero and for the protons less than the energy required to overcome the Coulomb barrier. In this mode, the MCNPX code tracks every particle until its death and prints a table of the number of neutrons and protons lost in the output file.

By proton bombardment of ^{129}I, based on some reactions such as (p, n), (p, α), (p, np), (p, 2np), (p, d), (p, ^{3}He), (p, 2p), (p, γ), (p, 2n), (p, 3n), (p, t) and (p, nα) radionuclides with shorter lives such as ^{128}I (half-life 24.99 min), ^{129m}Xe (half-life 8.88 days), ^{127}Xe (half-life 36.35 days), ^{127}Te (half-life 9.35 h), ^{127m}Xe (half-life 62.9 s), ^{125m}Te (half-life 57.40 days) and ^{127m}Te (half-life 106.1 days), stable isotopes such as ^{129}Xe, ^{125}Te, ^{130}Xe, ^{128}Xe, ^{127}I and ^{126}Te are produced. As you can see, the longest half-live radionuclide ^{127m}Te (half-life 106.1 days) is much shorter than ^{129}I; so in the case of ^{129}I, any nuclear interaction other than scattering will transform the nucleus into a shorter-lived nuclide [17].

The mentioned method of counting lost particles in the MCNPX output file can be used only for pure targets, if we use spallation materials such as lead or a reflector such as graphite to increase the transmutation rate causes a problem, because MCNPX does not distinguish between particle losses to different nuclei; so some particle losses have occurred in other material such as lead and graphite that would not result in transmutations of ^{129}I.

In order to estimate the number of transmutations of iodine nuclei by particles whose energy is higher than the MCNPX's tabulated data, the transmutation response function must be generated. To measure the average response function, for neutrons, 168 energy bins from 10 to 1500 MeV, and for protons, 120 energy bins from 1 to 1500 MeV, are considered. The energy considered in each bin is the upper limit of energy for that bin, and the lower limit is the energy considered in the previous bin. In order to regardless of the attenuation and the coulombic energy loss, a monodirectional point source of particles and a short and very thin cylindrical ^{129}I target were modeled. Then, for each energy bin, we run a separate MCNPX deck. (Approximately 500 decks or more have been run for this study.)

The response value $\sigma(E)$ is given by Equation [13]:

$$\sigma(E) = \frac{n_{\text{intr}}}{nps \cdot z \cdot N} \tag{1}$$

where n_{intr} is the number of interactions in the MCNPX output file, nps is the number of particles run, **z** is the target thickness is cm, and **N** is the atomic density in atoms/cm^3.

At different energies, we need to choose different sizes of targets. A suitable target thickness is one in which the number of interactions with the target is less than 10% but more than 1% of the total incident particles. To check whether the attenuation is insignificant or not, you could try using two surface tallies, one at the entry and one at the exit of the target. The F1 and F11 tallies at the entrance and exit of your target should be close to the same. The F11 at the exit will be slightly lower, by the amount that has interacted, so if 1% of particles interact, the exit tally will be 99% of the entrance. If the exit is much lower, then the attenuation is significant and should be accounted for. To do so, many geometries have been studied, for example, in the energy bin of 1500 MeV, the target of ^{129}I was considered a cylinder with a thickness of 0.001 cm. In addition, an energy cutoff card can be used to eliminate the interaction of particles whose energy is outside the range of each energy bin [13].

When the transmutation response function was obtained for each energy bin, based on Eq. (2), En and EMn cards can be used to enter the values of the transmutation response function to MCNPX deck and create a custom tally to calculate the transmutation rate at high energies for ^{129}I in the region containing target material:

$$\text{Transmutation rate} = \sum_{p}^{\text{particles}} \sum_{E}^{\text{bins}} \Phi(p, E)\sigma(p, E) \times V \times N \tag{2}$$

where $\sum_{p}^{\text{particles}}$ is a sum over all particles, $\sum_{E}^{\text{bins}}$ is the sum over all energy bins tallied for particle type p, $\Phi(p, E)$ is the flux of particle p within energy bin E, $\sigma(p, E)$ is the response function value for particle p and energy bin E, V is the volume of material, and N is the atom density of ^{129}I [13].

For ^{129}I, radiative capture (n, γ) is the main mechanism for transmutation below 10 MeV, which leads to the production of ^{130}I with 12.36 h half-life. MCNPX by the 102 capture tally will tally (n, γ) reactions for the nuclide specified in the range for which has tabulated cross-section data; therefore, for energies higher than 10 MeV, a custom tally can be used for all other non-scattering nuclear interactions. (Scattering does not lead to transmutation.) The final value of the response function can be found in Appendix, Tables 2 and 3.

3 Results and discussion

Experiments performed on the ^{129}I transmutation by proton beam mostly calculated the transmutation rate of ^{129}I by neutrons produced in spallation reactions induced by protons in the target, not the transmutation rate of ^{129}I by the proton itself. In the study presented by [18], the cross section is calculated for some of the proton-induced reactions leading to ^{129}I transmutation. The response function presented in our study calculates the total transmutation rate, but in [18], the cross section of many of the reactions that lead to the transmutation of ^{129}I is not calculated, such as the reactions that lead to the transmutation of iodine to ^{129m}Xe, ^{127}Te, ^{127m}Xe, ^{125m}Te, ^{127m}Te, ^{129}Xe, ^{125}Te, ^{130}Xe, ^{128}Xe, ^{127}I, ^{126}Te, ^{127m}Te, etc., so it is not possible to accurately compare the results. The total cross section calculated in 660 MeV for iodine is 0.496 b, which is certainly less than the value calculated for the total cross section in 660 MeV of this study, which includes all the reactions that lead to iodine transmutation. In addition, in experiments in which the transmutation rate of ^{129}I is calculated by the produced neutrons, the transmutation rate is mainly calculated for limited reactions such as (n, γ), not for the total transmutation rate that the sum of all the reactions that lead to ^{129}I transmutation. The transmutation response function by neutron presented in this study calculates the total ^{129}I transmutation rate (other than radiative capture and scattering, scattering does not lead to transmutation), and it is not possible to compare it with the experimental results, which are mainly calculated for specific channels. With these interpretations, it is not possible to make an accurate comparison with the experimental results.

To validate, the results were tested by two methods for energies lower and higher than 200 MeV. Tabulated data for the interactions of protons with ^{129}I up to 200 MeV are available in the JENDL/ImPACT-18 [19] and TENDL-2019 [20] libraries, but the format of these libraries is ENDF, to analyze the number of transmutations obtained from the custom tally and the FM tally of these libraries in MCNPX, was needed to convert their format to ACE by NJOY code. The NJOY nuclear data processing system is a modular computer code used for converting evaluated nuclear data in the ENDF format into libraries useful for applications calculations, and the ACER module prepares libraries in ACE format for the Los Alamos continuous-energy Monte Carlo MCNP codes [21, 22].

Figures 1 and 2 show the generated custom cross section in this study with the cross sections in the JENDL and TENDL libraries, and Table 1 contains the number of proton-induced transmutations caused by 10,000 protons in a ^{129}I target at different energies using custom tally and FM tally of TENDL and JENDL libraries which both indicate a good agreement in the results.

For validation at energies higher than 200 MeV, the custom tally was used to estimate the number of transmutations in a pure ^{129}I target, then the number of lost protons in the MCNPX output file was counted and the results were compared. The number of

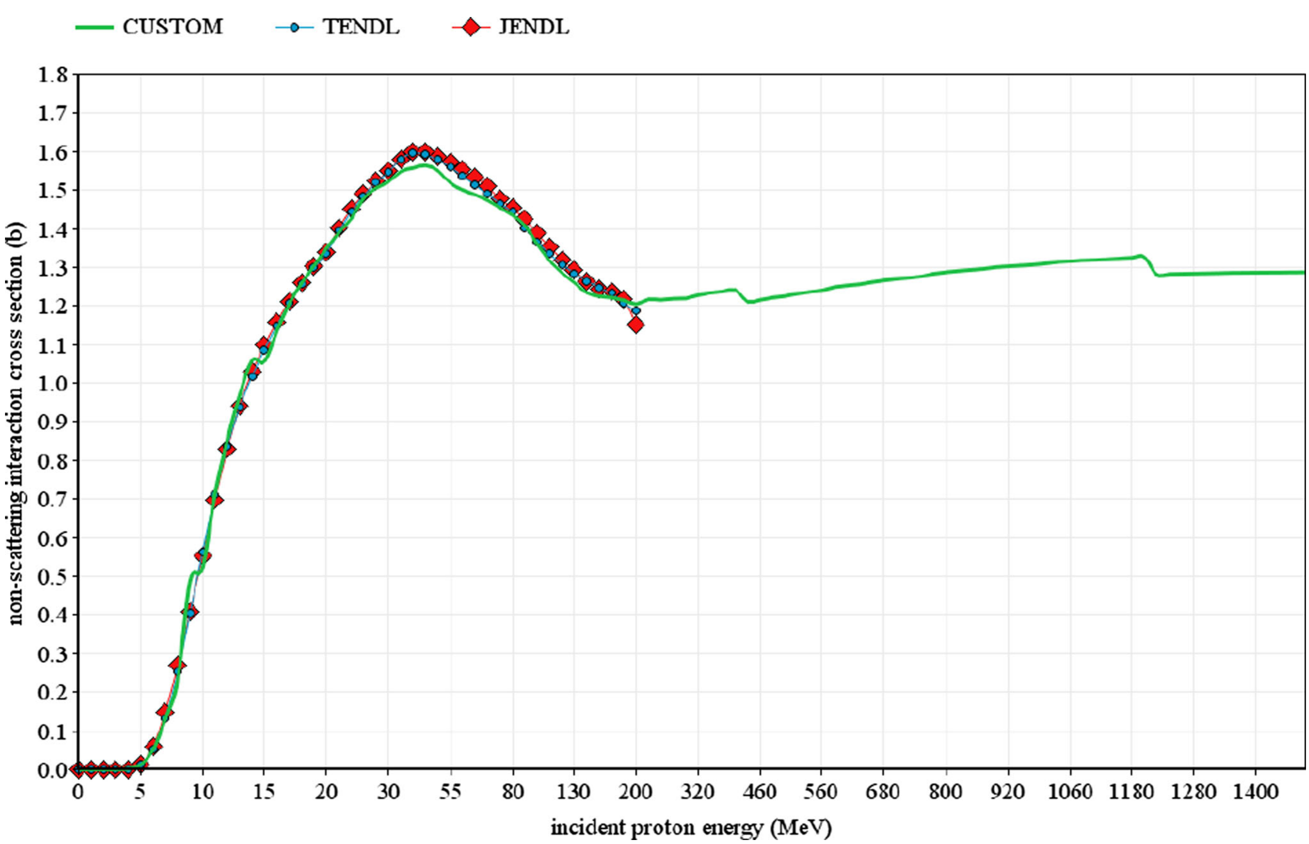

Fig. 1 Interaction cross section vs. energy using TENDL and JENDL and the response function generated in MCNPX code

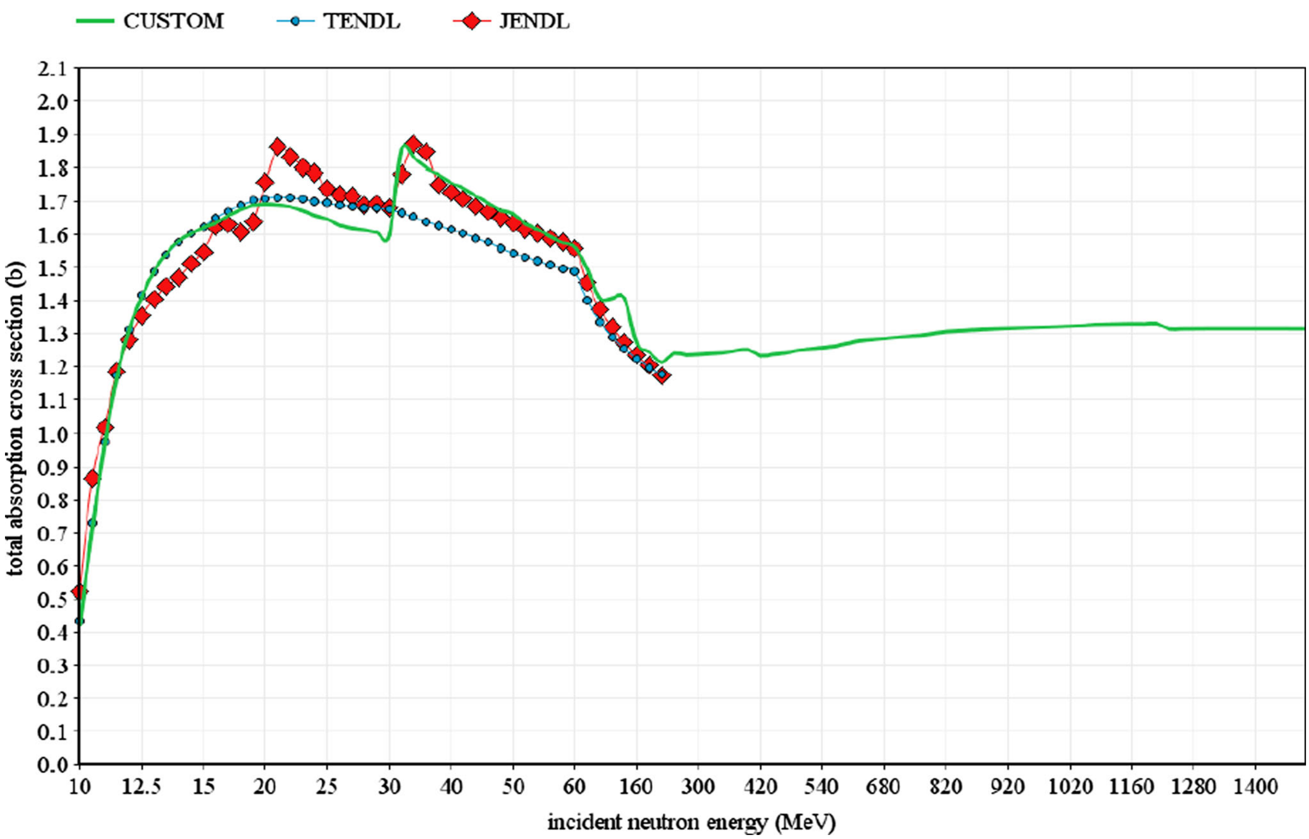

Fig. 2 Total neutron absorption cross section vs. energy using TENDL and JENDL and the response function generated in MCNPX code

Table 1 Number of proton-induced transmutations of ^{129}I with 10,000 incident protons

Energy (MeV)	JENDL	CUSTOM	TENDL
20	35	35	35
40	184	181	184
60	401	394	401
80	656	648	658
100	942	930	939
120	1247	1227	1240
150	1728	1701	1725
180	2226	2213	2235
200	2561	2565	2580

proton-induced transmutations obtained for energies 300 MeV, 900 MeV and 1500 MeV with 10,0000 incident protons are 44,672, 124,033 and 143,928, respectively, for the custom tally, and 43,776, 122,269 and 144,133 for counting the number of protons lost in transmutation in the output file. The results show that the error is about 2%, which is considered acceptable.

4 Conclusion

A light water reactor (LWR), 1 GWh (e), discharges about 21 tons of radioactive fuel every year, of which 760 kg are fission products [23]. One way to manage LLFPs is to transmute them to some stable or shorter-lived radionuclides by an accelerator-driven system (ADS). The MCNPX code can be used to simulate the transmutation rates of these LLFPs, but the tabulated data used in this code is not available for high-energy protons and neutrons. This study proposes a transmutation response function that is consistent with the physics models used in the MCNPX code. The generated custom tally can be input into MCNPX and used for the estimation of the transmutation rate of ^{129}I by an accelerator-driven system with energies up to 1500 MeV.

For energies lower than 200 MeV, the results were compared with data from the JENDL and TENDL libraries. The difference in values is very small and can be caused by inherent errors in the simulation codes. As you can see, even the data from the JENDL and TENDL libraries are slightly different. As well as, using cross-section data from other libraries that are not always consistent with the models used in MCNPX can cause differences in the transmutation rates.

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Declarations

Conflict of interest The authors declare that they have no conflict of interest.

Data availability All data generated or analyzed during this study are included in this published article (and its supplementary information files).

Appendix

See Table 2 and 3.

Table 2 Response function value for transmutation by protons

E (MeV)	σ (*E*) (barn)	*E* (MeV)	σ (*E*) (barn)	*E* (MeV)	σ (*E*) (barn)	*E* (MeV)	σ (*E*) (barn)
1	1.0042$E-14$	90	1.4096	720	1.27286	1420	1.28674
2	5.13542$E-8$	100	1.3606	740	1.27632	1440	1.28693
3	2.862$E-5$	110	1.3171	760	1.27927	1460	1.28718
4	0.00125	120	1.2861	780	1.28338	1480	1.28734
5	0.01201	130	1.2607	800	1.28758	1500	1.28743
6	0.04983	140	1.2346	820	1.29074		
7	0.12987	150	1.2251	840	1.29339		
8	0.2314	160	1.2215	860	1.29609		
9	0.4883	180	1.2146	880	1.29820		
10	0.5204	200	1.2048	900	1.30044		
11	0.7105	220	1.2169	920	1.30270		
12	0.8514	240	1.21573	940	1.30477		
13	0.9683	260	1.21864	960	1.30675		
14	1.0589	280	1.21943	980	1.30834		
15	1.0563	300	1.22359	1000	1.30967		
16	1.1342	320	1.22771	1020	1.31161		
17	1.2059	340	1.23398	1040	1.31406		
18	1.2589	360	1.23748	1060	1.31598		
19	1.3042	380	1.24146	1080	1.31813		
20	1.3489	400	1.24458	1100	1.31909		
22	1.3897	420	1.21057	1120	1.32079		
24	1.4256	440	1.21085	1140	1.32288		
26	1.4782	460	1.21536	1160	1.32416		
28	1.5052	480	1.22133	1180	1.32492		
30	1.5212	500	1.22529	1200	1.32742		
35	1.5481	520	1.23153	1220	1.28167		
40	1.5572	540	1.23609	1240	1.28250		
45	1.5638	560	1.24026	1260	1.28317		
50	1.5504	580	1.24471	1280	1.28385		
55	1.5184	600	1.24893	1300	1.28448		
60	1.5004	620	1.25282	1320	1.28507		
65	1.4882	640	1.25594	1340	1.28546		
70	1.4739	660	1.26142	1360	1.28594		
75	1.4552	680	1.26610	1380	1.28619		
80	1.4353	700	1.26995	1400	1.28648		

Table 3 Response function value for neutrons other than radiative capture or scattering

E (MeV)	σ (E) (barn)	*E* (MeV)	σ (*E*) (barn)	*E* (MeV)	σ (*E*) (barn)	*E* (MeV)	σ (*E*) (barn)
10	0.4181	13.5	1.5427	35	1.8145	260	1.236522
10.1	0.4763	13.6	1.5530	36	1.8002	280	1.236925
10.2	0.5318	13.7	1.5606	37	1.7906	300	1.238360
10.3	0.5921	13.8	1.5677	38	1.7799	320	1.240904
10.4	0.6503	13.9	1.5752	39	1.7675	340	1.243532
10.5	0.7077	14	1.5827	40	1.7569	360	1.246144
10.6	0.7615	14.1	1.5834	41	1.7477	380	1.249522
10.7	0.8155	14.2	1.5884	42	1.7395	400	1.253078
10.8	0.8656	14.3	1.5919	43	1.7261	420	1.234561
10.9	0.9130	14.4	1.5981	44	1.7173	440	1.238333
11	0.9619	14.5	1.6032	45	1.7057	460	1.242564
11.1	1.0049	14.6	1.6074	46	1.6959	480	1.246534
11.2	1.0445	14.7	1.6109	47	1.6845	500	1.250381
11.3	1.0849	14.8	1.6146	48	1.6731	520	1.254573
11.4	1.1284	14.9	1.6178	49	1.6639	540	1.258746
11.5	1.1683	15	1.6226	50	1.6639	560	1.262923
11.6	1.2027	16	1.6357	51	1.6414	580	1.267068
11.7	1.2307	17	1.6581	52	1.6337	600	1.271106
11.8	1.2587	18	1.6753	53	1.6238	620	1.275159
11.9	1.2866	19	1.6895	54	1.6166	640	1.278832
12	1.3147	20	1.6931	55	1.6065	660	1.282566
12.1	1.3353	21	1.6904	56	1.5955	680	1.286165
12.2	1.3518	22	1.6848	57	1.5859	700	1.289579
12.3	1.3684	23	1.6724	58	1.5763	720	1.29285
12.4	1.3897	24	1.6579	59	1.5707	740	1.296045
12.5	1.4106	25	1.6474	60	1.5629	760	1.298935
12.6	1.4270	26	1.6286	80	1.4965	780	1.301703
12.7	1.4428	27	1.6202	100	1.4075	800	1.304335
12.8	1.4589	28	1.6144	120	1.3442	820	1.306787
12.9	1.4760	29	1.6075	140	1.3121	840	1.309004
13	1.4898	30	1.5988	160	1.2718	860	1.311112
13.1	1.5004	31	1.8672	180	1.2457	880	1.313219
13.2	1.5122	32	1.8552	200	1.2141	900	1.315092
13.3	1.5231	33	1.8422	220	1.242008	920	1.316714
13.4	1.5333	34	1.8291	240	1.237498	940	1.318173

E (MeV)	σ (*E*) (barn)	*E* (MeV)	σ (*E*) (barn)
960	1.319515	1240	1.314865
980	1.320905	1260	1.315197
1000	1.322057	1280	1.315466
1020	1.323473	1300	1.315753
1040	1.325013	1320	1.315827
1060	1.326182	1340	1.315912
1080	1.327443	1360	1.315942
1100	1.328303	1380	1.315958
1120	1.329269	1400	1.315928
1140	1.330187	1420	1.315921
1160	1.330873	1440	1.315891
1180	1.330569	1460	1.315800
1200	1.332054	1480	1.315719
1220	1.314485	1500	1.315558

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