

$^{124}\text{Xe}(n,\gamma)^{125}\text{Xe}$ and $^{124}\text{Xe}(n,2n)^{123}\text{Xe}$ measurements for National Ignition Facility

Megha Bhike^{1,2,a}, Nurin Ludin³ and Werner Tornow^{1,2}

¹Department of Physics, Duke University, Durham, North Carolina 27708, USA

²Triangle Universities Nuclear Laboratory, Durham, North Carolina 27708, USA

³University of Denver, Denver, Colorado, 80208, USA

Abstract. The cross section for the $^{124}\text{Xe}(n,\gamma)^{125}\text{Xe}$ reaction has been measured for the first time for neutron energies above 100 keV. In addition, the $^{124}\text{Xe}(n,2n)^{123}\text{Xe}$ reaction has been studied between threshold and 14.8 MeV. The results of these measurements provide sensitive diagnostic tools for investigating properties of the inertial confinement fusion plasma in Deuterium-Tritium (DT) capsules at the National Ignition Facility (NIF) located at Lawrence Livermore National Laboratory.

Introduction

Nuclear reactions play an important role as diagnostic tools to understand the complicated physics governing the inertial confinement fusion (ICF) plasma. Currently, a consistent theory of stopping powers of charged particles in ICF plasmas does not exist. A prerequisite for the full understanding of the neutron flux distribution at National Ignition Facility (NIF) requires the spectral characteristics of the NIF neutron spectrum. For this purpose, fast neutron reactions are envisioned as an important component. Efforts are underway at NIF to accurately measure the neutron energy distribution obtained in DT shots employing 192 powerful lasers, which deposit up to 500 TW of peak power (1000 times more than the US uses at any one moment) and up to 1.85 MJ of UV light on the DT pellet.

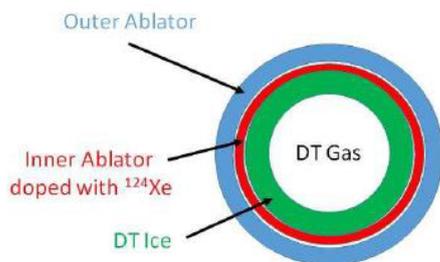


Figure 1. Schematic of the Deuterium-Tritium capsule used in laser shots at the National Ignition Facility.

A sketch of a typical DT pellet is shown in Fig. 1. The radius of the pellet is of the order of 400 μm , and the DT gas pressure is of the order of 20 atm. Radiochemical

^a Corresponding author: megha@tunl.duke.edu

tracer nuclei are bonded to the inside of the ablator (Be or CH), which completely burn away when hit by the 192 lasers. About 10^{15} atoms of the isotope(s) of interest are loaded into the inner-most layer of the ablator shell, which in turn surrounds a layer of DT ice and a sphere of DT gas. Noble gases are considered to be the best dopants. For many reasons, ^{124}Xe became the primary choice of capsule dopant. In the presence of DT neutrons ^{124}Xe undergoes both (n, γ) and (n,2n) reactions leading to ^{125}Xe and ^{123}Xe respectively. Following a NIF shot, the xenon gas is collected with Radiochemical Analysis of Gaseous Samples (RAGS) [5], and subsequently the two isotopes ^{125}Xe and ^{123}Xe are analyzed using gamma ray spectrometry. One reason for choosing ^{124}Xe as capsule dopant is its evaluated [6] and predicted extremely large (n, γ) cross-section. For example, the capture cross section of the neutron-rich isotope ^{136}Xe is predicted to be of the order of 1 mb at 1 MeV, while evaluations predict the capture cross section of ^{124}Xe to be a factor of 500 greater. Experimental data for the $^{124}\text{Xe}(n,\gamma)^{125}\text{Xe}$ reaction does not exist above 30 keV neutron energy. Furthermore, for the (n,2n) reaction experimental data do not exist, except for three conflicting measurements at 14.5 MeV [2-4].

The threshold for the $^{124}\text{Xe}(n,2n)^{123}\text{Xe}$ reaction is 10.569 MeV, and its cross section increases with energy in the neutron energy region of interest. Therefore, this reaction is sensitive to the primary 14 MeV DT neutrons. The energy threshold for $^{124}\text{Xe}(n,\gamma)^{125}\text{Xe}$ reaction is zero and therefore, this reaction is sensitive to neutrons below 10 MeV for which the $^{124}\text{Xe}(n,2n)^{123}\text{Xe}$ channel is not open. Other reaction products do not produce nuclei of interest (noble gases). A NIF shot on a 2.1 mm diameter

spherical glass shell filled with a 1:1 DT mixture and a small amount of ^{124}Xe was performed as early as 2011 for commissioning of RAGS. In order to provide experimental data and to guide model calculations, cross-section measurements have been performed for the $^{124}\text{Xe}(n,\gamma)^{125}\text{Xe}$ reaction in the neutron energy range 0.4 to 7.5 MeV. Additionally, cross-section data for the $^{124}\text{Xe}(n,2n)^{123}\text{Xe}$ reaction were obtained between 11.3 and 14.8 MeV. In the following the experimental approach is described and the results are presented in comparison to existing model calculations.

Experimental Setup and Analysis

Mono-energetic neutrons were obtained via the $^3\text{H}(p,n)^3\text{He}$ reaction ($Q=-0.764$ MeV) between 0.37 and 3.8 MeV, via the $^2\text{H}(d,n)^3\text{He}$ reaction ($Q = +3.269$ MeV) between 4.5 and 14.5 MeV, and finally at 14.8 MeV via the $^3\text{H}(d,n)^4\text{He}$ reaction ($Q=+17.589$ MeV). The charged-particle beams were accelerated by the tandem accelerator of the Triangle Universities of Nuclear Laboratory (TUNL). Typically proton and deuteron beam currents on target were between 1.5 μA and 3.5 μA . Fig. 1 shows a schematic view of the experimental setup for the $^{124}\text{Xe}(n,\gamma)^{125}\text{Xe}$ measurement using the $^2\text{H}(d,n)^3\text{He}$ reaction. It consists of a 3 cm long cell pressurized to 3 atm of high-purity deuterium gas. A 6.5 μm Havar foil separates the gas from the accelerator vacuum.

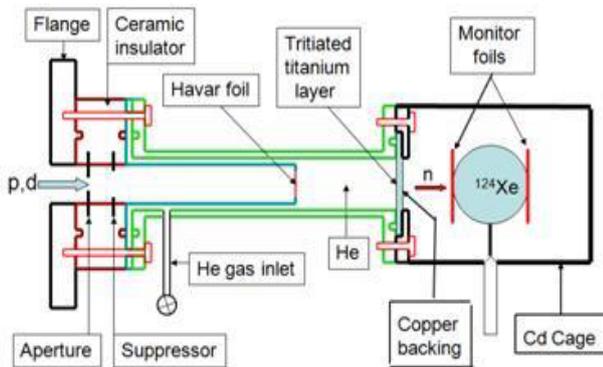


Figure 1. Schematic view of the experimental arrangement for the $^{124}\text{Xe}(n,\gamma)^{125}\text{Xe}$ and $^{124}\text{Xe}(n,2n)^{123}\text{Xe}$ cross-section measurements using the $^2\text{H}(d,n)^3\text{He}$ reaction.

The ^{124}Xe gas (enriched to 99.99%) of 2.6968 g was contained in a stainless steel sphere with inner diameter of 20.0 mm and wall thickness of 0.6 mm, corresponding to a pressure of about 120 atm. The distance between the end of neutron production target and the centre of the ^{124}Xe sphere was typically 19 mm for measurements of the $^{124}\text{Xe}(n,\gamma)^{125}\text{Xe}$ reaction cross section, and 25 mm for the $^{124}\text{Xe}(n,2n)^{123}\text{Xe}$ reaction cross section.

For the (n,γ) measurements the $^3\text{H}(p,n)^3\text{He}$ reaction was used to produce neutron beams at 0.37, 0.86, 1.86 MeV, while the $^2\text{H}(d,n)^3\text{He}$ was employed at 4.48, 5.31, 6.31 and 7.25 MeV. Indium or gold monitor foils were attached to the upstream and downstream side of the sphere for neutron fluence determination. The sphere-foil assembly was enclosed by a cover made of 0.5 mm thick cadmium to efficiently absorb room-return thermal neutrons. Neutron irradiation times varied between 1.5

and 4 hours, depending on the predicted cross section. The neutron flux was monitored with a BC-501A liquid scintillator positioned at 0° relative to the incident charged-particle beam at a distance of 2.9 m from the neutron source. The tritiated titanium target used in connection with the $^3\text{H}(p,n)^3\text{He}$ reaction consists of 2.1 Ci of ^3H loaded into a 2.2 mg/cm^2 thick layer of titanium of 16 mm diameter, which in turn is evaporated onto a 0.4 mm thick copper backing. The tritiated target described above was also used for the $^3\text{H}(d,n)^4\text{He}$ reaction to produce 14.8 MeV neutrons. In this case lower energy neutrons are not produced due to the low incident deuteron energy. After irradiation, the ^{124}Xe filled sphere and the monitor foils were γ -ray counted using a well shielded 60% High-Purity Germanium (HPGe) detector of known efficiency. The data-acquisition hardware consisted of a Canberra amplifier, the Multiport II system and the associated GENIE 2K software, all provided by Canberra [7]. The yield of the γ -ray line at 188.418 keV resulting from the decay of ^{125}Xe with $I_\gamma=53.8\%$ and $T_{1/2}=16.9$ h was recorded as a function of time. An identical, but empty stainless steel sphere was irradiated to prove that there was no γ -ray line, which could interfere with the line of interest. The peak areas of the γ -ray lines of interest were obtained using the peak fitting software TV [8].

Above the deuteron energies of 2.225 MeV, the deuterons can break up on the entrance collimator, the Havar foil or the tantalum beam stop, resulting in lower energy neutrons than the primary neutrons of interest from the $^2\text{H}(d,n)^3\text{He}$ reaction. Auxiliary measurements were performed with the deuterium gas pumped out, leaving all other parameters unchanged. The incident deuteron beam charge deposited on the deuterium gas cell was used for normalization purposes for these two types of experiments. In case of the $^3\text{H}(p,n)^3\text{He}$ reaction, once the proton energy exceeds about 3 MeV, corresponding to primary neutron energy greater than 2.2 MeV, the primary neutrons are accompanied by low-energy neutrons originating from (p,n) reactions on the titanium and copper backing. Therefore, at $E_n = 2.73$ and 3.61 MeV, auxiliary measurements were performed with an untritiated but otherwise identical tritium target.

For the measurements of the $^{124}\text{Xe}(n,2n)^{123}\text{Xe}$ cross section the $^2\text{H}(d,n)^3\text{He}$ reaction was used. Data were obtained between 11.5 and 14.5 neutron energy in 1 MeV energy steps. The irradiation time was 1 hour. Gold foils were attached to the upstream and downstream side of the ^{124}Xe containing sphere for neutron fluence determination. During the irradiation, the Cd cover was not used. The yield of the 148.9 keV ($I_\gamma = 48.9\%$) γ -ray line from the decay of ^{123}Xe ($T_{1/2} = 2.08$ h) was used for the cross-section determination. The empty stainless steel sphere was irradiated at 12.5 MeV to check whether the 148.9 keV energy region is contaminated by background events which would interfere with the gamma-ray line of interest.

The neutron flux ϕ was determined from the monitor reactions using the activation formula,

$$\phi = \frac{A\lambda}{N\sigma\epsilon I_{\gamma}(1 - e^{-\lambda t_i})e^{-\lambda t_d}(1 - e^{-\lambda t_m})} \quad (1)$$

where the induced activity A is the total yield in the photopeak, λ is the decay constant of the residual nucleus, N is the number of target nuclei, σ is the cross-section of monitor reaction, ϵ is the photopeak efficiency for the γ -ray energy of interest, I_{γ} is its branching ratio, t_i is the irradiation time, t_d is the decay time between the end of irradiation and the beginning of off-line counting, and t_m is the measuring time. The $^{115}\text{In}(n,n')^{115m}\text{In}$ reaction was used for incident neutron energies between 0.85 and 7.31 MeV. The $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reaction was used at 0.37 MeV. Finally the reaction $^{197}\text{Au}(n,2n)^{196}\text{Au}$ was employed at energies above 9 MeV. The correction factor for variation in the neutron flux was found to be negligible, because the beam intensity was constant throughout the irradiations. Monte-Carlo calculations were performed to obtain the mean neutron energy and its associated energy spread. The same Monte-Carlo code was used to account for the tight geometry of the experimental setup which causes the average of the neutron fluences deduced from the two monitor foils to deviate slightly from the neutron fluence seen by the Xe cell. The activation formula given in Eqn 1. was also employed to obtain σ from the measured activities as described in [9].

Results

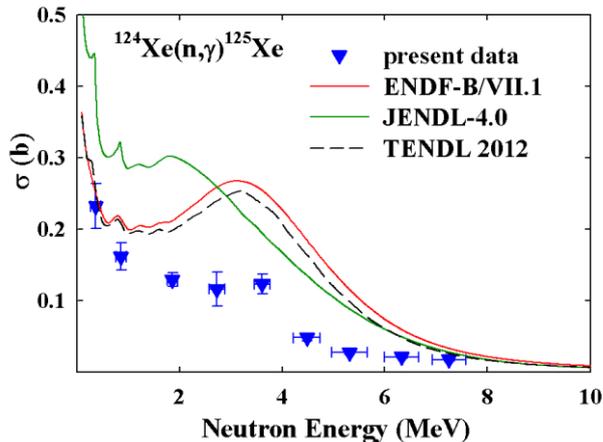


Figure 2. $^{124}\text{Xe}(n,\gamma)^{125}\text{Xe}$ cross-section data obtained in the present work in comparison to the evaluations ENDF/B-VII.1 [6], and JENDL-4.0 [10], and the model calculation TENDL-2012 [11].

Preliminary results for the $^{124}\text{Xe}(n,\gamma)^{125}\text{Xe}$ are shown in Fig. 2 along with predictions from various evaluations and the model calculation TENDL-2012. None of the evaluations are in agreement with the experimental data, although the order of magnitude is reasonably well predicted. The preliminary cross-section results for the $^{124}\text{Xe}(n,2n)^{123}\text{Xe}$ reaction are shown in Fig. 3, in comparison to the existing data around 14.5 MeV and the model calculation TENDL-2012 and evaluations.

excellent agreement with the datum of Sigg *et al.* [2] while the datum of Kondaiah *et al.* [3], and especially the data of Bazan *et al.* [4] gives larger cross-section values. Focussing on the evaluations, we observe that our data are in better agreement with ENDF/B-VII.1 than with TENDL-2012 but they deviate considerably from the JENDL-4.0 evaluation.

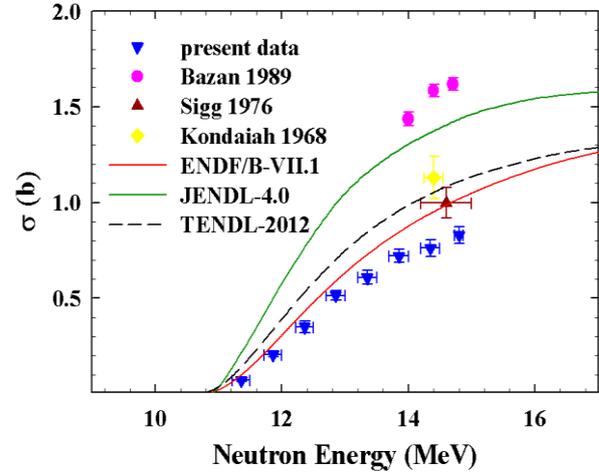


Figure 3. $^{124}\text{Xe}(n,2n)^{123}\text{Xe}$ cross-section data in comparison to evaluations [9-11] and previous data [2-4].

In summary, the $^{124}\text{Xe}(n,\gamma)^{125}\text{Xe}$ and $^{124}\text{Xe}(n,2n)^{123}\text{Xe}$ cross-section data obtained in the present work provide for the first time an accurate basis for interpreting measurements of the $^{125}\text{Xe}/^{123}\text{Xe}$ intensity ratio performed at NIF in laser shots on ^{124}Xe loaded DT capsules.

Acknowledgements

We thank A. P. Tonchev, LLNL, M. Bosewell, LANL, S. W. Finch, Duke University, and M. E. Gooden, North Carolina State University. This work was supported partially by the US Department of Energy, Office of Nuclear Physics, under Grant No. DE-FG02-97ER41033, and by the National Nuclear Security Administration under the Stewardship Science Academic Alliance Program through the US department of Energy Grant No. DE-NA0001839.

References

1. D. A. Shaughnessey, C. Cerjan, K. J. Moody, L. Bernstein, R. Hoffman, M. A. Stoyer, R. Fortner, and D. Schneider, Lawrence Livermore National Laboratory Report No. LLNL-TR-472995 (2011)
2. R. A. Sigg, and P. K. Kuroda, Nucl. Sci. And Eng. **60**, 235 (1976)
3. E. Kondaiah, N. Ranakumar, and R. W. Fink, Nucl. Phys. A **120**, 337 (1968)
4. F. Bazan, LLNL Internal Report UCRL-53929, **162** (1989)

5. D. A. Shaughnessy, C. A. Velsko, D. R. Jedlovec, C. B. Yeamans, K. J. Moody, E. Tereshatov, W. Stoeffl, and A. Riddle, *Rev. Sci. Instrum.* **83**, 10D917 (2012)
6. M. B. Chadwick *et al.*, *Nucl. Data Sheets* **112**, 2887 (2011)
7. <http://www.canberra.com>
8. J. Theuerkauf *et al.*, Program TV, Institute for Nuclear Physics, University of Cologne, 1993
9. M. Bhike and, W. Tornow, *Phy. Rev. C* **89**, 031602 (R) (2014)
10. K. Shibata *et al.*, *Nucl. Sci. Technol.* **48**, 1 (2011)
11. A. J. Koning *et al.*, *Nucl. Sci. Technol. Suppl.* **2**, 1161 (2002)