

# Low Energy Neutron Measurements in High Energy Density Plasmas using the National Ignition Facility<sup>\*)</sup>

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Deuterium-tritium-loaded capsules at the National Ignition Facility (NIF) are now regularly producing a *neutron rich high energy density plasma* (nHEDP) with a low-energy “ICF-thermal” neutron density  $> 10^{21}$  neutrons/cm<sup>3</sup>. These low-energy neutrons are produced via multiple scatter off of the highly compressed capsule and therefore provide insight into the confinement time ( $\tau_{\text{confinement}}$ ) of the assembled plasma. Neutrons are formed in the center of the 5 m NIF chamber that is well suited for minimizing “room return” thermal capture. This nHEDP environment is befitting for activation-based measurements of the (n, $\gamma$ ) cross sections responsible for the formation of heavy elements in astrophysical settings. These experiments also offer the first opportunity to search for the effects of nuclear-plasma interaction-induced excited state population on (n,x) reaction rates in a stellar-like plasma environment. Unfortunately, no capability currently exists at the NIF to measure the neutron spectrum in a capsule down to the 100 eV level required to enable these new classes of nuclear-plasma experiments. In this paper we will discuss nHEDP-based neutron capture experiments, compare them to accelerator-based (n, $\gamma$ ) measurements, and discuss the requirements for a NIF-based low energy neutron spectrometer (LENS).

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## 1. Introduction

The deuterium-tritium (DT) fuel in cryogenic capsules at the National Ignition Facility (NIF) is now regularly being assembled to areal densities ( $\rho R_{\text{fuel}}$ ) greater than any ever previously observed at an Inertial Confinement Fusion (ICF) facility [1]. While the primary goal of these experiments is to trap the  $\alpha$ -particle energy from the D+T reaction in the dense fuel in order to achieve thermonuclear ignition, a “side effect” is that a significant fraction of the 14 MeV neutrons from the D+T reaction scatter until they “thermalize” to the keV energies of the assembled fuel. The number and spectral distribution of these ICF-thermal neutrons is an excellent probe of the temperature and plasma confinement time of the cold fuel, potentially providing insight into the entropy of the cold fuel.

In addition to this role as a plasma diagnostic, these low-energy neutrons offer the first opportunity to study the neutron capture (e.g., (n, $\gamma$ )) reactions responsible for the formation of the majority of the elements heavier than iron *in the same sort of high energy density plasma environment in which they are formed in stars*. This neutron-rich high energy density plasma (nHEDP) provides a laboratory capable of exploring the plasma-nuclear processes responsible for the population of excited nuclear levels in astrophysical settings [2]. While the potential for performing (n, $\gamma$ ) astrophysics measurements at ICF facilities has been recognized by the community [3], no neutron-capture products produced in the target assembly had ever been collected from an ICF-based experiment.

However, starting in late 2011, the Solid Radchem Collector (SRC) [4] at NIF has been harvesting radioactive <sup>198</sup>Au and <sup>196</sup>Au produced from (n, $\gamma$ ) and (n,2n) reactions on the ~100 mg of <sup>197</sup>Au in the NIF hohlraum using collec-

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tors that cover a modest ( $\sim 10^{-4}$ ) portion of the total solid angle. The ratio of the  $^{198}\text{Au}/^{196}\text{Au}$  reaction products collected by SRC is correlated with the down-scattered ratio of singly-scattered to un-scattered neutrons as determined using other neutron diagnostics. Model studies, confirmed by experiment, indicate that the dominant source of  $^{198}\text{Au}$  is the capture of scattered neutrons from the compressed capsule rather than lower energy (meV) “room return” neutrons, which arise from scattering off of material in the NIF chamber walls, etc. These results will be presented in greater detail elsewhere [5].

The SRC results open the door to NIF-based neutron activation cross section measurements utilizing mg-sized samples located on the NIF hohlraum. Plans are underway to perform “ride along” ( $n,\gamma$ ) experiments at the NIF in late 2013 using foils mounted on or near the center of the NIF target chamber (TCC). These measurements would complement existing methods of measuring ( $n,\gamma$ ) using prompt  $\gamma$ -calorimetry and pulsed neutron beams [6]. However, these spectrally averaged ( $n,\gamma$ ) cross section measurements require quantitative knowledge of the low-energy portion of the neutron spectrum. While time-of-flight (TOF) spectrometers are the tool of choice for measuring the neutron energy spectrum at NIF [7], their reliance on neutron elastic scattering on hydrogen in the scintillator makes them insensitive to the low momentum imparted by sub-MeV neutrons. Efforts are therefore underway to design a highly segmented low-energy neutron spectrometer (LENS) for NIF. This system, which would be similar to the Large Neutron Scintillator Array (LANSA) spectrometer fielded at NOVA in the 1990s [8], will utilize novel boron and/or lithium loaded scintillators sensitive to low-energy neutrons coupled to modern state-of-the-art digital electronics and photomultiplier tube (PMT) technology.

The  $^{198}\text{Au}$  SRC data provides not only the first evidence that neutron capture experiments can be performed at the NIF, but it also offers a potential platform for examining the effects of nuclear-plasma interactions in neutron-induced nuclear reactions for the first time. In addition to its long-lived  $J^\pi = 2^-$  ground state (6.2 d), the 10.4 h  $^{196}\text{Au}$  metastable state is clearly observable in the NIF data. The  $^{196\text{m}}\text{Au}/^{196\text{g}}\text{Au}$  ratio for Gold collected at the NIF could potentially be used to indicate the interaction of high level density  $^{196}\text{Au}$  “quasi-continuum” states populated  $10^{-15}$  to  $10^{-12}$  s following ( $n,2n$ ) with the nHEDP. This class of nuclear-plasma interactions has never been experimentally accessible before, and its existence would have dramatic consequences on our understanding of the formation of heavy nuclei in astrophysical settings.

## 2. Low-Energy Neutrons as a Plasma Diagnostic

Three conditions are needed to produce a significant low-energy neutron fraction in ICF plasmas:

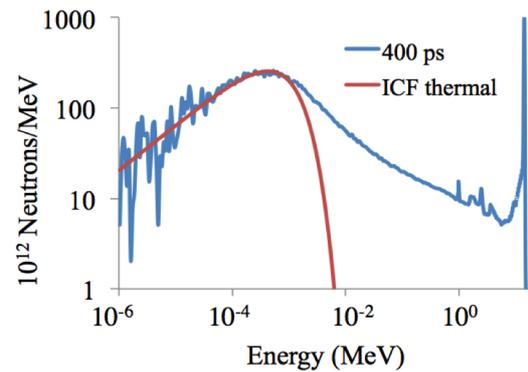


Fig. 1 Hydrodynamic simulations of the neutron spectrum from a typical NIF DT shot (blue line) and an ICF-thermal distribution for  $kT = 850$  eV (red line).

1. A thermonuclear reaction neutron source (e.g., DT, DD, TT, etc.)
2. A high fuel areal density ( $\rho R_{\text{fuel}}$ ) to allow scattering of the neutrons; and
3. A confinement time long enough to allow multiple scattering of neutrons in the cold fuel.

An indirectly driven cryogenic NIF hohlraum+capsule meets these criteria. These capsules, which start with an initial radius of approximately 1 mm, are compressed to a final radius on the order of 30–40  $\mu\text{m}$ . This compression causes the formation of peak DT areal densities in excess of  $1\text{ g/cm}^2$  capable of down-scattering primary 14 MeV neutrons to ICF-thermal energies. Furthermore, the modest yields achieved to date ( $Y_{\text{total}} < 2\text{ kJ}$ ) allow for a primary neutron production in excess of several  $10^{14}$  neutrons and a confinement time in excess of several  $10^{-10}$  seconds.

Figure 1 shows a typical neutron spectrum from advanced “post-shot” hydrodynamic simulations of a NIF shot performed using the HYDRA code [9]. The inputs to these dynamic simulations are adjusted in order to reproduce the data from existing nuclear and x-ray diagnostics. The sub-keV portion of the neutron spectrum includes a broad peak that is well reproduced by a Maxwell-Boltzmann distribution assuming  $kT = 850$  eV (red line). Neutrons with energies below 850 eV have velocities of approximately 400  $\mu\text{m/ns}$ , allowing sufficient time for them to scatter multiple times in the 30  $\mu\text{m}$  cold fuel shell until they are “thermalized” with their local plasma environment. As the plasma expands,  $\rho R_{\text{fuel}}$  becomes too small to scatter the neutrons effectively, thereby limiting the fraction of the neutrons in the “ICF-thermal peak” and making the ICF-thermal neutron fraction a diagnostic for the fuel confinement time ( $\tau_{\text{conf}}$ ).

Hydrodynamic simulations demonstrate this. Figure 2 shows the neutron spectrum, including the low-energy component, for a simulation with conditions comparable to those believed to exist in NIF capsules for confinement times from 400–5000 ps. The fraction of neutrons below

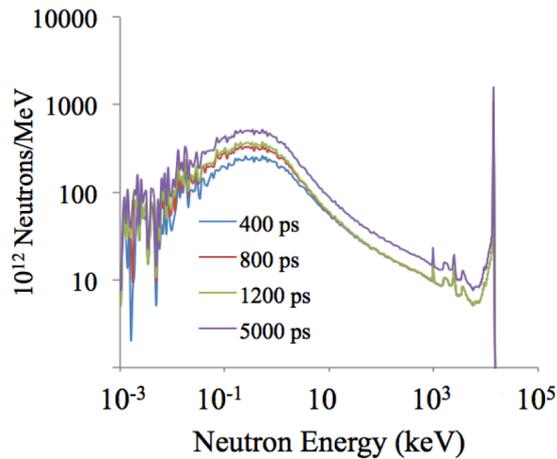


Fig. 2 Hydrodynamic simulations of the neutron spectra from a NIF DT shot for  $\tau_{\text{conf}}$  of 400, 800, 1200 and 5000 ps respectively.

1 keV differs by a factor of nearly 2.5 over this energy range.

### 3. Astrophysical Nucleosynthesis

In addition to their value as a plasma diagnostic,  $(n,\gamma)$  reactions with  $keV$  neutrons are responsible for the formation of almost all of the elements heavier than iron made in astrophysical settings [2]. These reactions occur via both rapid neutron capture in very high-flux environments (the *r-process*) and slow neutron capture in massive and asymptotic giant branch stars throughout their lifetime (the *s-process*). The *s-process* environment exposes “seed” nuclei to neutron densities in excess of  $10^7 \text{ cm}^{-3}$  in stellar plasmas with temperatures from 0.1 - 1.0 billion degrees Kelvin ( $kT \approx 8 - 86 \text{ keV}$ ). These capture processes take place at a rate defined by the neutron capture cross sections ( $\sigma_{(n,\gamma)} \approx 10^{-24} \text{ cm}^2$ ) and the neutron velocity ( $v_n$ ):

$$R_{(n,\gamma)} = \rho_n v_n \sigma_{(n,\gamma)} = 0.1/\text{year}. \quad (1)$$

Neutron capture proceeds along a chain of isotopes for a given element until a radioactive nucleus is formed whose lifetime is comparable to or shorter than the capture rate. If the lifetime is significantly shorter,  $\beta$ -decay will occur. However, if the lifetime of the radioactive product is comparable to the capture rate, a branching takes place where  $\beta$ -decay and neutron capture compete.

Capture on “branch-point” nuclei offers a unique window into the conditions in the interior of massive stars and plays a critical role in the development of stellar models. However, its usefulness is limited by a lack of knowledge of the neutron capture cross section on these radioactive branch point nuclei themselves. This need has been identified by the community [10] and has led to a significant effort to determine these cross sections at astrophysically relevant energies.

Unfortunately,  $(n,\gamma)$  cross sections are difficult to

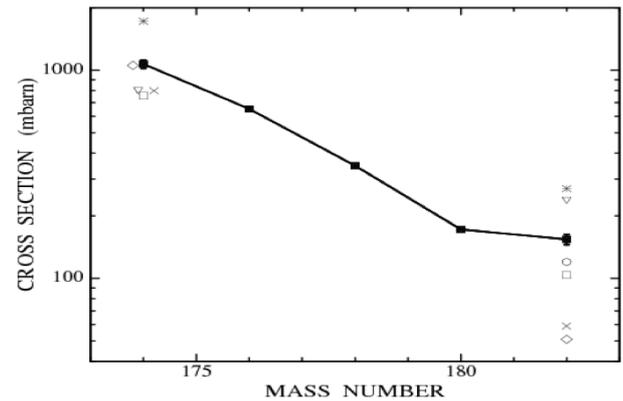


Fig. 3 Modeled and predicted values (open symbols) for neutron capture MACS on  $^{174}\text{Hf}$  and  $^{182}\text{Hf}$  together with those measured by the Karlsruhe group (closed circles) [11].

model accurately. This sensitivity has been most recently demonstrated by Kappeler’s group at Karlsruhe [10, 11], which has been measuring Maxwellian-Averaged Cross Sections (MACS) corresponding to stellar-thermal neutron energy distributions at *s-process* temperatures (e.g., 4 - 25 keV) for more than two decades. The Karlsruhe group informed six leading members of the nuclear reaction modeling community of their intention to measure neutron MACS for two hafnium isotopes,  $^{174}\text{Hf}$  and  $^{182}\text{Hf}$ . Modelers were asked to predict the values for the MACS for these two nuclei prior to their measurement. Figure 3 shows the results of these predictions, which span a factor of 3 for  $^{174}\text{Hf}$  and a factor of 6 for  $^{182}\text{Hf}$ , together with the value measured by the Karlsruhe group. These differences highlight the uncertainties in  $keV$  neutron capture modeling and significantly impair *s-process* modeling efforts.

An additional complication exists with regards to  $(n,\gamma)$  in nHEDP environments. Neutron capture rates are highly sensitive to small changes in the spin of the target nucleus. While nuclei are generally in their ground state in non-plasma environments, a significant portion of them have low-lying states with different spins that can be thermally populated in a nHEDP. The interactions that populate these states, including nuclear excitation by electron capture (NEEC) and nuclear excitation by electronic transition (NEET) have been the subject of extensive work [12, 13] and are the subject of an intense research effort in their own right. The alteration of the  $(n,\gamma)$  cross sections arising from the change in spin due to excited state population is parameterized in *s-process* models via a *stellar enhancement factor* (SEF) that can range from 0.8 - 1.4 times the  $(n,\gamma)$  cross section on the ground state.

In addition to the *s-process*, ICF environments offer the possibility of informing other astrophysical nucleosynthesis processes. One example is the production of the 34 slightly neutron-deficient “*p-process*” nuclei, which are believed to be formed in  $(\gamma,n)$  reactions in hot stellar environments [14]. ICF-based  $(n,\gamma)$  experiments can be used

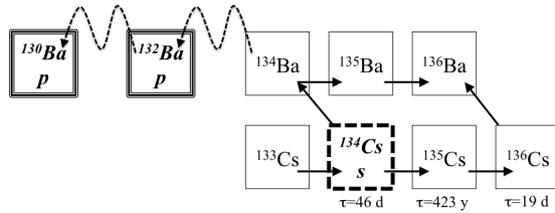


Fig. 4 s- and p-process pathways near cesium ( $Z = 55$ ) and barium ( $Z = 56$ ) adapted from T. Hayakawa *et al.* [14]. Note that branching occurs on  $^{134}\text{Cs}$  where the lifetime is comparable to the inverse capture rate ( $\approx 0.1 - 1 \text{ year}^{-1}$ ). The p-process nuclei  $^{130,132}\text{Ba}$  are formed by two successive  $(\gamma, n)$  reactions.

Table 1 Properties of specific nuclei that could potentially be loaded into ICF capsules in order to determine their neutron capture cross sections.

Nucleus (relevant formation process)	Isotopic abundance or lifetime	Nucleus (relevant formation process)	Isotopic abundance or lifetime
$^{74}\text{Se}$ (p)	0.89%	$^{136}\text{Ce}$ (p)	0.185%
$^{79}\text{Se}$ (s)	290,000 y	$^{138}\text{Ce}$ (p)	0.251%
$^{85}\text{Kr}$ (s)	10.76 y	$^{151}\text{Sm}$ (s)	90 y
$^{78}\text{Kr}$ (p)	0.35%	$^{163}\text{Ho}$ (s)	4570 y
$^{80}\text{Kr}$ (p)	2.25%	$^{170}\text{Tm}$ (s)	128.6 d
$^{130}\text{Ba}$ (p)	0.106%	$^{171}\text{Tm}$ (s)	1.92 y
$^{132}\text{Ba}$ (p)	0.101%	$^{179}\text{Ta}$ (s)	1.82 y
$^{134}\text{Cs}$ (s)	2.065 y	$^{185}\text{W}$ (s)	75.1 d

to set limits on p-process  $(\gamma, n)$  reaction rates through the principle of detailed balance. Figure 4 shows an example of the s-process pathway near barium and cesium, including the branch point at the  $^{134}\text{Cs}$  nucleus ( $t_{1/2} = 46$  days) [Adapted from Ref. 2].

Table 1 lists several illustrative examples of s-process branch point and p-process nuclei that could potentially be incorporated into a NIF capsule for determination of their  $(n, \gamma)$  cross section. The total number of product nuclei that would be formed assuming a sample size of  $10^{16}$  atoms loaded pre-shot, a capture cross section identical to that of  $^{197}\text{Au}$ , and a typical NIF shot with a total neutron yield of  $6 \times 10^{14}$  is approximately  $10^9$ . This number is consistent with the early results obtained to date for  $^{198}\text{Au}$  from the SRC diagnostic [5] when scaled for the SRC solid angle coverage and the relative mass of Au in the hohlraum.

#### 4. Neutron Capture at ICF and Accelerator Facilities

Neutron capture is unlike charged particle induced nuclear reactions in that the lack of a Coulomb barrier causes  $(n, x)$  reaction cross sections to increase with the increas-

ing deBroglie wavelength (decreasing energy) of the neutron. This feature makes activation-based  $keV$   $(n, \gamma)$  cross sections measurements inherently difficult to perform since even a small number of lower-energy “room return” neutrons will have a significantly higher probability to induce activation in the target.

In addition to the issue of room return, many astrophysically-relevant  $(n, \gamma)$  cross sections involve radioactive targets with half-lives on the order of hundreds of days, leading to huge  $\gamma$ -ray decay rates for targets handled in a laboratory. For example, a 1 mg sample of the  $^{134}\text{Cs}$  branch point nucleus has a decay rate of 1 Ci. As a result, efforts to measure  $(n, \gamma)$  cross sections on radioactive nuclei have focused on using high-segmentation arrays to observe the relatively high total  $\gamma$ -ray energy ( $\sim 6 - 9 \text{ MeV}$ ) signal released following capture over the background photons released due to radioactive decay of the target. The Device for Advanced Neutron Capture Experiments (DANCE) spectrometer at Los Alamos is a good example of a calorimetric system [6] designed to perform  $(n, \gamma)$  cross section measurements on radioactive targets. The DANCE spectrometer is an array of 160 highly segmented BaF detectors at the Los Alamos Neutron Science Center (LANSCE). LANSCE provides a “white” neutron source with a flux of  $\approx 10^4 \text{ n/s/cm}^2/\text{keV}$  where the neutron energy is determined via time-of-flight (TOF). The array is located twenty meters away from the neutron source, meaning that only a small fraction of the neutron beam interacts with the target. While DANCE has produced a wealth of nuclear structure information on stable targets, it has yet to measure an astrophysically relevant  $(n, \gamma)$  cross section on an s-process branch point nucleus with a hard  $\gamma$ -ray decay component.

An ICF-driven neutron source provides an alternative approach to measuring  $keV$  neutron-capture cross sections using activation. Nuclei in the inner layer of a NIF capsule are subject to a neutron fluence greater than  $10^{15} \text{ n/cm}^2/\text{keV}$  and a flux of nearly  $10^{25} \text{ n/cm}^2/\text{keV/s}$ . Furthermore, these target nuclides are located in the middle of the NIF chamber, which is essentially a 5 m radius empty sphere surrounded by a thick sleeve of borated concrete material known as “gunnite.” This placement is ideal for minimizing background neutrons from local scattering and “room return” neutrons. However, instead of lower-energy background neutrons, a neutron capture sample in NIF experiences a large fluence of 14 MeV primary neutrons. These neutrons, while sizable in number, have significantly lower capture cross sections compared to  $keV$  neutrons. Furthermore, the contribution of 14 MeV neutrons to capture can be quantified using a NIF capsule referred to as an “exploding pusher” designed to produce few, if any, low energy neutrons.

While NIF as a neutron source compares favorably with existing facilities like LANSCE, as well as proposed future facilities such as FRANZ [15], a number of issues need to be addressed before an ICF-based  $(n, \gamma)$  measure-

ment can take place. Foremost among these is the lack of a well-characterized neutron spectral diagnostic for the low-energy neutrons that dominate the capture process.

## 5. Characteristics of a Low-Energy Neutron Spectrometer for ICF (n, $\gamma$ ) Measurements

A Low Energy Neutron Spectrometer (LENS) sensitive to the low-energy neutron spectrum at an ICF facility would require several features including:

1. High segmentation to increase data bandwidth in a given time interval
2. Location at sufficient distance from the TCC to allow for good energy resolution using time-of-flight (most likely at a 20 m TOF position)
3. The use of neutron detectors inherently sensitive to low-energy neutrons
4. Digital electronic readout to maximize the information contained in the detector trace and to allow “seamless” transition from current mode (where individual neutron hits are not resolved) to pulse mode (where individual hits are resolved and pulse-height thresholds can be employed)

The LANSAs spectrometer fielded at the Nova laser [8] satisfied these requirements. LANSAs was comprised of 960 individual scintillator channels and fielded 20 m from the Nova target. The spectrometer was used to measure primary neutrons directly arising from thermonuclear reactions for ICF shots with yields as low as several  $10^5$  neutrons and operated solely in pulse mode. A NIF-based LENS capable of resolving individual neutron hits in a similar fashion for a  $10^{14}$  neutron yield NIF shot would require an impractically large number of channels. Instead, a NIF-based system would measure the energy of incoming neutrons in current mode for neutrons with energies greater than a critical energy  $E_{\text{crit}}$  given by:

$$\varepsilon_{\text{det}}(E_{\text{crit}}) \times d\Omega_{\text{det}} \times \phi_n(E_{\text{crit}}) \times \Delta t_{\text{crit}} \ll 1. \quad (2)$$

Here  $\varepsilon_{\text{det}}$  is the efficiency,  $d\Omega_{\text{det}}$  is the areal coverage, and  $\Delta t_{\text{det}}$  is the time response of a given LENS detector element and  $\phi_n$  is the neutron flux at the location of the spectrometer. LENS would then “cross-over” to individual pulse mode for neutron energies below  $E_{\text{crit}}$  functioning in a similar manner to LANSAs. For energies below  $E_{\text{crit}}$  LENS could be absolutely calibrated using a non-ICF neutron source with a well-defined neutron flux. A LENS would be calibrated and function in a manner similar to the existing NIF NTOF system for energies above  $E_{\text{crit}}$ . Early results using the new NIF NTOF system near the “south pole” of the target chamber are consistent with this current to pulse mode transition occurring for  $t > 4 \mu\text{s}$ .

The ideal location for the LENS at NIF would be in one of the existing long-baseline (20 m) neutron time-of-

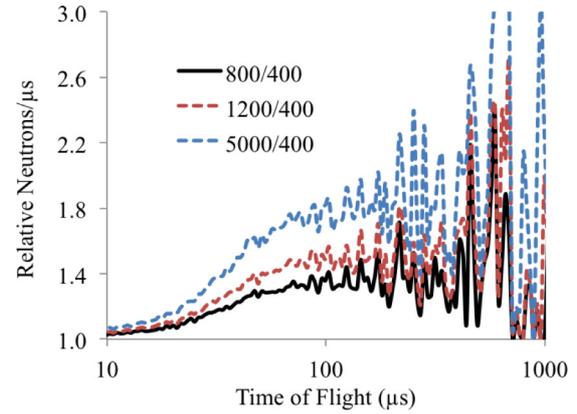


Fig. 5 Hydrodynamic simulations showing the ratio of the number of neutrons/ $\mu\text{s}$  arriving 20 m from TCC following a NIF DT shot for  $\tau_{\text{conf}}$  of 800 ps (black), 1200 ps (red) and 5 ns (blue) relative to  $\tau_{\text{conf}} = 400$  ps. Note that 100  $\mu\text{s}$  corresponds to a neutron energy of  $\approx 200$  eV.

flight positions on either the equator ( $90^\circ$ ,  $174^\circ$ ) or the alcove ( $116^\circ$ ,  $316^\circ$ ) off of the NIF target chamber (position given in spherical coordinates). Consider for example a 256-element LENS array located 20 m from TCC at NIF that is tasked with observing the late-time (low energy) neutron spectrum. Figure 5 shows the relative number of neutrons observed in such a LENS as a function of TOF for the modeled NIF DT shots with values of  $\tau_{\text{conf}}$  shown in Fig. 2 above. The ratios differ by nearly 50% for a neutron TOF of 100  $\mu\text{s}$ , corresponding to  $E_n \approx 200$  eV.

Fast organic crystal scintillators have been used quite successfully in current mode at NIF for the detection of neutrons with energies  $> 140$  keV [16]. However, these scintillators are not well suited to the task of detecting low-energy neutrons due to their reliance on elastic scattering of hydrogen to convert the neutron kinetic energy to light. This issue can be rectified through the addition of a dopant that leads to the production of charged particles with a large, positive  $Q$ -value and cross section at low  $E_n$ . Two frequently employed dopants are lithium and boron. These nuclei are highly sensitive to low energy neutrons through the  ${}^6\text{Li}(n,t)\alpha$  and the  ${}^{10}\text{B}(n,t)2\alpha$  reactions. Furthermore, Li-doped glass has been employed at the GEKKO laser facility [17], and boron-loaded scintillators, such as BC-454 from Saint-Gobain are commercially available.

## 6. LENS Testing at the LBNL 88-Inch Cyclotron

The LENS prototype system and its various candidate scintillator materials can be tested and calibrated using a variety of accelerator-based neutron sources. One such facility is the thick-target deuteron break-up neutron source at the Lawrence Berkeley National Laboratory (LBNL) 88-Inch Cyclotron. This facility was used in the study and

testing of the new bibenzyl-based scintillator developed for NIF [16]. Deuteron beams with currents up to ten  $\mu\text{A}$  are made incident upon a thick target where up to 10% of the beam is fragmented into its constituent protons and neutrons. The protons and unreacted deuterons stop in a thick water-cooled target, while the neutrons travel through approximately 2 m of concrete collimation plus an additional two to eight meters of air before entering the experimental area. The cyclotron beam consists of individual pulses with a width  $\geq 1$  ns and a pulse spacing on the order of 100 ns allowing their energy to be determined using TOF. The resulting neutron energy spectrum is broad (typically several MeV), but centered slightly below half of the beam energy and is well described in the literature [17, 18]. Recent activation measurements have confirmed the published neutron spectrum, and the first testing of candidate scintillator materials for LENS is underway.

## 7. Conclusion

ICF facilities present a unique environment for studying neutron capture in plasma environments due to the presence of a significant low-energy neutron component arising from the multiple scatter of neutrons on the highly compressed hydrogen fuel. These low-energy neutrons offer a direct measurement of the confinement time of the assembled plasma and open the possibility of astrophysically relevant  $(n,\gamma)$  cross section measurements. However, all of these ICF-based neutron studies require a quantitative understanding of the low-energy neutron spectrum. An effort is underway to develop an appropriate low-energy neutron spectrometer for use at NIF following the approach pioneered at NOVA by Nelson and Cable [8] using the intense neutron source at the LBNL 88-Inch Cyclotron [19].

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