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Evidence for Stratification of Deuterium-Tritium Fuel in Inertial Confinement Fusion Implosions

Plasma Science and Fusion Center, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA

Laboratory for Laser Energetics, University of Rochester, Rochester, New York 14623, USA

D. P. McNabb, P. A. Amendt, R. N. Boyd, and J. R. Rygg
Lawrence Livermore National Laboratory, Livermore, California 94550, USA

H. W. Herrmann and Y. H. Kim
Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

A. D. Bacher
Department of Physics, Indiana University, Bloomington, Indiana 47405, USA

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Measurements of the $D(d, p)T$ ($dd$) and $T(t, 2n)^4$He ($tt$) reaction yields have been compared with those of the $D(t, n)^3$He ($dt$) reaction yield, using deuterium-tritium gas-filled inertial confinement fusion capsule implosions. In these experiments, carried out on the OMEGA laser, absolute spectral measurements of $dd$ protons and $tt$ neutrons were obtained. From these measurements, it was concluded that the $dd$ yield is anomalously low and the $tt$ yield is anomalously high relative to the $dt$ yield, an observation that we conjecture to be caused by a stratification of the fuel in the implosion core. This effect may be present in ignition experiments planned on the National Ignition Facility.

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In laser-driven inertial confinement fusion (ICF), spherical capsules are compressed and heated to high enough temperatures and densities for fusion reactions to occur [1,2]. The fusion products from these reactions carry information about the core, and can be used to diagnose the underlying implosion physics. For instance, measurements of fusion products from the $D(d, p)T$ ($dd$) and $D(^3$He, $p)^4$He ($dt$) reactions in $D^3$He gas-filled capsule implosions at OMEGA [3] have shown anomalous yield behavior, indicating that aspects of the underlying physics governing an ICF implosion are not completely understood [4]. Based on scaling of $D_2$ implosions with the same mass and particle density (which are “hydro-equivalent”) [5], it was shown in Ref. [4] that the $d^3$He proton and $dd$ deuteron yields were about 50% lower than the expected in equimolar $D^3$He gas-filled capsule implosions. In another study, non-hydro-equivalent $DT^3$He gas-filled capsule implosions have shown anomalous $D(t, n)^4$He ($dt$) reaction yield behavior that was ~50% lower than expected [6]. A third study showed indirect-drive experiments, with trace Ar dopants in $D_2$ capsule implosions, which also had observed $dd$ yields that were 30%–50% of expectation [7,8]. The results from these studies suggest species diffusion effects, as proposed by Amendt et al. [7,9]. These effects, which are to the best of our knowledge not included in simulations, appear to measurably degrade the nuclear yields of lighter ion species in an ICF implosion.

Generalizing these observations from the different elemental mixtures (different $Z$) of Refs. [4,6,7] to include isotopic mixtures of the same $Z$ (e.g., $Z = 1$ for hydrogen, deuterium, tritium), is of fundamental interest. The latter case is particularly important, as it is directly relevant to current ignition experiments at the National Ignition Facility (NIF), where different mixtures of $H$, $D$ and $T$ are being utilized.

To address questions about ion diffusion in $DT$ implosions, this Letter reports on measurements of the $dd$ and $tt$ reaction yields ($Y_{dd}$ and $Y_{tt}$) and how they contrast to the measured $dt$ reaction yield ($Y_{dt}$). Spherical thin-glass ($SiO_2$) and thick-CH capsules were filled with $DT$ gas, and imploled using 23–30 kJ of energy delivered by the OMEGA laser in 1-ns square laser pulses. The absolute $dd$ proton spectrum was measured by two magnet based charged particle spectrometers (CPSs) [10,11], and the $dt$ neutron yield and ion temperature were measured with the suite of neutron time-of-flight (nTOF) detectors [12]. The absolute $dt$ and $tt$ neutron spectra were measured with the magnetic recoil spectrometer (MRS), discussed in detail in Refs. [13–15]. The different reactions utilized in this study are summarized below.

$$D + D \rightarrow T + p(3.0 \text{ MeV}), \quad Q = 4.0 \text{ MeV}. \quad (1)$$

$$D + T \rightarrow n(14.1 \text{ MeV}) + ^4_2\text{He}, \quad Q = 17.6 \text{ MeV}. \quad (2)$$
\[ T + T \rightarrow 2n(0-9.4 \text{ MeV}) + \frac{4}{3}\text{He}, \quad Q = 11.3 \text{ MeV}. \quad (3) \]

As shown by Eqs. (1) and (2), the \(dd\) reaction produces a 3 MeV proton and a triton, and the \(dt\) reaction produces a 14.1 MeV neutron and an alpha particle. At ICF relevant conditions, the \(tt\) reaction produces two neutrons and an alpha particle [Eq. (3)]. The spectrum of these \(tt\) neutrons can be described by a 3-body continuum that is modified by the \(n-n\) and \(n\)-alpha final-state interactions [16]. The details of the \(tt\) neutron spectrum are further discussed in Refs. [17–19].

The yield \((Y_{12})\) for the different reactions is determined by integrating the spectrum of the \(dd\) protons, \(tt\) neutrons, and \(dt\) neutrons. To relate \(Y_{12}\) to the conditions in an ICF implosion, the reaction yield can be expressed as

\[
Y_{12} = \int \frac{f_1 f_2}{1 + \delta_{12}} \rho(t, t)^2 \langle \sigma v \rangle_{12} d\tilde{r} dt, \quad (4)
\]

where \(\langle \sigma v \rangle\) is the Maxwellian averaged reaction rate, \(f_1\) and \(f_2\) are the atomic fractions of the reactants, \(\rho\) is the fuel mass density, \(\bar{n}\) is the average reactant mass, and the Kronecker delta \(\delta_{12}\) accounts for double counting of identical reactants [2]. Using Eq. (4), the reaction yield ratio \(Y_{11}/Y_{12}\) can be expressed as

\[
Y_{11}/Y_{12} = \frac{1}{2} \int \frac{f_1 f_2 \rho(t, t)^2 \langle \sigma v \rangle_{11} d\tilde{r} dt}{f_1 f_2 \rho(t, t)^2 \langle \sigma v \rangle_{12} d\tilde{r} dt}. \quad (5)
\]

This expression can be simplified if the reaction ratio for the two reactions is slowly varying within the ion temperature \(T_i\) range of the reacting fuel, which is the case for the \(Y_{dd}/Y_{dt}\) and \(Y_{tt}/Y_{dt}\) measurements discussed in this Letter \((T_i = 9–18 \text{ keV for } dd/dt\) and \(T_i = 2–15 \text{ keV for } tt/dt\) [20]). With this condition met, the reaction ratio can be removed from the integral in Eq. (5). In addition, as hydrodynamic models of an ICF implosion often assume that the reactant density ratio \(f_1/f_2\) is spatially and temporally constant during the implosion (although, we will show this to be inconsistent with the data herein), \(f_1\) and \(f_2\) can also be removed from the integrals. Now, the integrals cancel and the reaction yield ratio can be expressed as

\[
Y_{11}/Y_{12} = \frac{1}{2} \frac{f_1}{f_2} \langle \sigma v \rangle_{11}/\langle \sigma v \rangle_{12}. \quad (6)
\]

Using the known reactivities (obtained from the ENDF/B-VII.0 database [20]) for the \(dd\), \(dt\), and \(tt\) reactions in the temperature range specified above, the expected \(Y_{dd}/Y_{dt}\) and \(Y_{tt}/Y_{dt}\) ratios in a DT implosion are given by

\[
Y_{dd}/Y_{dt} \cong 2.6 \times 10^{-3} (f_T/f_D)^{-1} \quad (T_i = 9–18 \text{ keV}). \quad (7)
\]

\[
Y_{tt}/Y_{dt} \cong 1.7 \times 10^{-3} (f_T/f_D) \quad (T_i = 2–15 \text{ keV}). \quad (8)
\]

Here, \(f_T\) is the fraction of tritium in the core, \(f_D\) is the deuterium fraction of the core, and \(Y_{tt}\) is the \(tt\) reaction yield, which is half of the \(tt\) neutron yield because the 3-body branch emits two neutrons per reaction. Unless otherwise specified, all yields described herein refer to the reaction yield.

To test this prediction, \(Y_{dd}\) can be determined in a DT implosion from the measured \(dd\) proton spectrum; an example is shown in Fig. 1. This spectrum was obtained for OMEGA shot 39794 in which a 2.8 \(\mu\)m thick SiO2 capsule filled with 20 atm of DT gas \((f_T = 0.39, f_D = 0.56,\) and trace hydrogen impurity, or \(f_T/f_D = 0.69\) [21]) was imploded. A \(Y_{dd}\) of \(5.0 \times 10^{10}\) was determined from the spectrum, and a \(Y_{dt}\) of \(3.9 \times 10^{13}\) and \(T_i\) of 11.8 keV were determined from the nTOF measurement. This results in a yield ratio of \(Y_{dd}/Y_{dt} = (1.3 \pm 0.2) \times 10^{-3}\) for this implosion, which is about a factor of 3 lower than \(3.7 \times 10^{-3}\) predicted by Eq. (7).

Similarly, \(Y_{tt}\) can be determined in a DT implosion from the measured \(tt\) neutron spectrum; an example is shown in Fig. 2. Figure 2(a) shows the neutron spectrum that best describes the recoil deuteron spectrum measured by the MRS [Fig. 2(b)]. This spectrum was obtained by integrating nine nominally identical OMEGA implosions (shots 55074–55083) [22], using 16 \(\mu\)m CH capsules filled with DT fuel \((f_T/f_D = 0.63)\) at 17.5 atm. A total \(Y_{DT}\) of \(1.6 \times 10^{14}\) and a burn averaged ion temperature of 5.3 keV were measured with the nTOF detector. As shown by Fig. 2, the neutron spectrum consists of a \(dt\) neutron component, a \(tt\) neutron component, and a down-scattered neutron (DSn) component. The shape and magnitude of the DSn component are determined by the differential cross sections for the \(n-d, n-t, n-c,\) and \(n-h\) elastic and inelastic scattering. From this neutron spectrum, a yield ratio of \(Y_{tt}/Y_{dt} = (4.1 \pm 0.5) \times 10^{-3}\) was determined, which is more than a factor of 3 larger than \(1.1 \times 10^{-3}\) predicted by Eq. (8).

As an extension of the above study, the \(Y_{dd}/Y_{dt}\) and \(Y_{tt}/Y_{dt}\) ratios were determined for several series of different types of capsule-implosions, resulting in different burn averaged ion temperatures \((T_i)\). In the case of the \(Y_{dd}/Y_{dt}\) study, thin-glass capsules with thicknesses in the range of 2.4–3.7 \(\mu\)m were used, and in the case of the

![FIG. 1 (color online). Absolute \(dd\) proton spectrum measured with the CPS on OMEGA implosion 39794. The proton peak is energy upshifted \(\sim 0.5\) MeV from its birth energy due to electric fields, as discussed in Ref. [11].](image-url)
Y_{tt}/Y_{dt} study, thin-glass capsules with thicknesses of 3.8 μm and CH (or CD) capsules with thicknesses of 10, 16, and 20 μm were used. The initial DT fuel mixture was on average f_T/f_D = 0.75 for the Y_{dd}/Y_{dt} study and f_T/f_D = 0.62 for the Y_{tt}/Y_{dt} study. The differences in the initial f_T/f_D are due to tritium decay and refueling of the DT inventory at OMEGA. The observed Y_{dd}/Y_{dt} data are shown in Fig. 3(a) as a function of ion temperature. For comparison, the expected yield ratios calculated using Eq. (6) for the average initial DT fuel mixture (f_T/f_D = 0.75) used in this study. The grey dashed lines show the variation in the initial f_T/f_D due to tritium decay between these shots, which were obtained over a two-year period. 1D hydrodynamic simulations using LILAC (blue triangles) are also shown (for constant f_T/f_D = 0.75). The results show a suppression of the Y_{dd}/Y_{dt} yield ratio, indicating a lower deuterium fraction in the core than expected.

Anomalously high Y_{tt}/Y_{dt} yield ratio is consistent with a lower deuterium fraction in the core than expected, as suggested by the results in Fig. 3(a). Anomalously 3 to 6 times higher than predicted at ~4 and ~8 keV, respectively.

The relatively constant t/t (and d/d) reactivity ratio over the range of observed temperatures, and the consequential insensitivity to complex time-evolving density and temperature profiles strongly suggests this yield anomaly is due to a change in the reacting-fuel fractions f_T/f_D induced by deuterium leaving the center of the implosion. These anomalies, which are stronger for Y_{tt}/Y_{dt} than for Y_{dd}/Y_{dt}, could be caused by the combined effect of the centrally peaked temperature profile and stratified fuel species (recall the yield ratio is insensitive to the temperature profile only if f_T/f_D is fixed). As the temperature profile is peaked at the center of the

FIG. 2 (color online). (a) Neutron spectrum (black curve) that gives the best fit to the recoil deuteron spectrum measured with the MRS. (b) Recoil deuteron spectrum (red points) measured with the MRS. The spectrum is a convolution of the MRS-response function and neutron spectrum shown in (a). The deuteron peak at ~11.6 MeV is due to the primary d neutrons. The width of this peak is primarily due to the MRS resolution. A DSn component is also observed in the deuteron-energy range 8–10 MeV, where other neutron sources are absent. The t neutron spectrum dominates at deuteron energies below 7 MeV.

FIG. 3 (color online). (a) Measured Y_{dd}/Y_{dt} yield ratios as a function ion temperature (red points). The solid line represents the expected yield ratio calculated using Eq. (6) for the average initial DT fuel mixture (f_T/f_D = 0.75) used in this study. The grey dashed lines show the variation in the initial f_T/f_D due to tritium decay between these shots, which were obtained over a two-year period. 1D hydrodynamic simulations using LILAC (blue triangles) are also shown (for constant f_T/f_D = 0.75). The results show a suppression of the Y_{dd}/Y_{dt} yield ratio, indicating a lower deuterium fraction in the core than expected. (b) Measured Y_{tt}/Y_{dt} reaction yield ratios as a function of ion temperature (red points). The solid line represents the expected yield ratio for the (f_T/f_D = 0.63) DT fuel mixture used in this study. The anomalously high Y_{tt}/Y_{dt} is consistent with a lower deuterium fraction in the core than expected, as suggested by the results in Fig. 3(a).
compressed core, where the fuel is tritium rich, the effective $tt$ reactant temperature relative to $dt$ and $dd$ is higher. Because the reactivity is a strong function of temperature, this will further enhance $Y_{tt}/Y_{dt}$ and likewise suppress $Y_{dd}/Y_{dt}$. Therefore, these anomalous yield ratios indicate that $f_T/f_D$ has changed but cannot be directly used to infer $f_T/f_D$ without a self-consistent model of the density change in the core. However, implosion temperature and density profiles simulated using LILAC can be used to estimate the $\langle f_T/f_D \rangle$ (averaged over the $DT$ burn region) required to produce the observed yield ratios in Fig. 3. This work suggests that $\langle f_T/f_D \rangle$ has been increased by ~40%–70% above its preset value during the implosion. More detailed estimates for $\langle f_T/f_D \rangle$ will be the subject of further study.

Some insight as to when stratification begins, may come from the fact that anomalous yield behavior is observed (Fig. 3) in two different implosion types: thin-shell (2.4–3.8 μm SiO$_2$) shock-driven “exploding-pusher” and thick-shell (10–20 μm CH or CD) ablatively driven implosions. In both the exploding-pusher and ablative implosions, the laser launches a strong-shock that significantly heats the gas and produces a “shock yield” after rebounding off the center. By the time the shock yield is produced, the shell is mostly ablated away for the exploding-pusher case and no additional yield is produced. However, for the ablatively driven case much of the shell remains, which continues imploding inward until stagnation, where $pdV$ work heats the gas producing an additional “compression yield.” As both of these implosion types show an apparent change in $f_T/f_D$ in the core, this implies that the change begins relatively early in the implosion process, shortly after the first shock breaks out of the shell and certainly by the time the shock rebounds off the center and the shock yield is produced. This stratification may continue between the shock and compression-yield for the ablative implosions, but without temporal measurements of the relative yields, this cannot yet be definitively established.

A possible mechanism for the fuel stratification is plasma baro-diffusion, recently proposed by Amendt et al. [7,9] to explain the previously mentioned $dt^3$He and $dr^3$He yield anomalies, which causes lighter ions to diffuse away from the implosion center (and the heavier nuclei into the center). We propose future experiments with the aim of studying this possible mechanism and determining the role of the mass and charge of the fuel constituents. The first is to observe $^3$He$^3$He protons (which have recently been observed for the first time in ICF implosions [24]), produced in different mixtures of $^3$He$^4$He gas-filled implosions. This combination will feature same $Z$ but different constituent masses, which will directly complement this study but at $Z = 2$. A second experiment would be to measure $r^3$He deuterons using different mixtures of $T^3$He gas-filled implosions. $T^3$He deuterons have been observed in previous $DT^3$He gas-filled implosions [10], but not with the aim of studying possible diffusion effects. Any inferred stratification in $r^3$He would then be isolated to the difference in charge as these constituents feature the same mass. A third experiment would look at the $dd$ yield in hydro-equivalent THD fuel mixtures, analogous to the previously mentioned $D^3$He study [4].

In summary, the $dd$ and $tt$ reaction yields are anomalously low and high, respectively, when compared to the $dt$ reaction yield. We hypothesize that this discrepancy is caused by a stratification of the fuel, which causes $f_T/f_D$ to increase at the center of the compressed core, an effect that becomes stronger with higher temperature. The anomaly is larger for $Y_{tt}/Y_{dt}$ than for $Y_{dd}/Y_{dt}$, which may be the result of the combined effect of the temperature profile and stratified fuel. This stratification of the fuel may be driven by plasma baro-diffusion [7] of the fuel ions, which pushes the lighter ions from the imploding ICF core. The implications of these anomalous yields have bearing on other $dt$ experiments in ICF including the ignition experiments planned on the NIF, potentially resulting in a more restrictive ignition threshold [25,26] and reduced $dt$ yield.

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[5] Hydrodynamic behavior was predicted to be the same for implosions with the same mass density, total particle density, and equation of state, despite having different fuel compositions.
[22] Nine implosions were integrated to obtain better statistics because the neutron detection efficiency is relatively low when compared to charged particle measurements.

Nominally identical shell thickness, diameter, fill pressure, and laser profiles result in very similar nuclear-burn-averaged ion temperatures and $DT$ yield. In this series, the shot-to-shot ion temperature varied by 5% and the $DT$ yield varied by 28%.