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

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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 $D(t, n)^4$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 $D(^3\text{He}, p)^4$He (d^3He) reactions in D^3He 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 ''hydroequivalent'') [5], it was shown in Ref. [4] that the d^3He proton and dd neutron yields were about 50% lower than the expected in equimolar D^3He gas-filled capsule implosions. In another study, non-hydro-equivalent DT^3He 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 imploded 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.

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

$$D + T \rightarrow n(14.1\ \text{MeV}) + {}^4_2\text{He}, \qquad Q = 17.6\ \text{MeV}. \quad (2)$$

$$T + T \rightarrow 2n(0\text{–}9.4\,\text{MeV}) + {}^4_2\text{He}, \quad Q = 11.3\,\text{MeV}. \quad (3)$$

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

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

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

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

This expression can be simplified if the reactivity 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 keV for dd/dt, and $T_i = 2$–15 keV for tt/dt [20]). With this condition met, the reactivity 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

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

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

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

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

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 39 794 in which a 2.8 μm thick SiO_2 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 5.0×10^{10} was determined from the spectrum, and a Y_{dt} of 3.9×10^{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 3.7×10^{-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 μm CH capsules filled with DT fuel ($f_T/f_D = 0.63$) at 17.5 atm. A total Y_{DT} of 1.6×10^{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 1.1×10^{-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 μm were used, and in the case of the

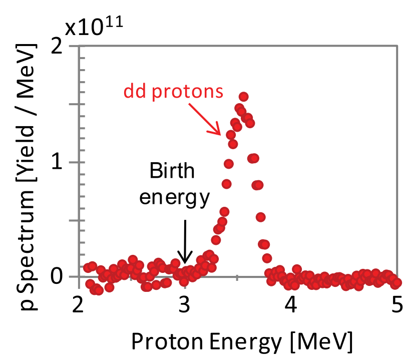

FIG. 1 (color online). Absolute dd proton spectrum measured with the CPS on OMEGA implosion 39794. The proton peak is energy upshifted ~0.5 MeV from its birth energy due to electric fields, as discussed in Ref. [11].

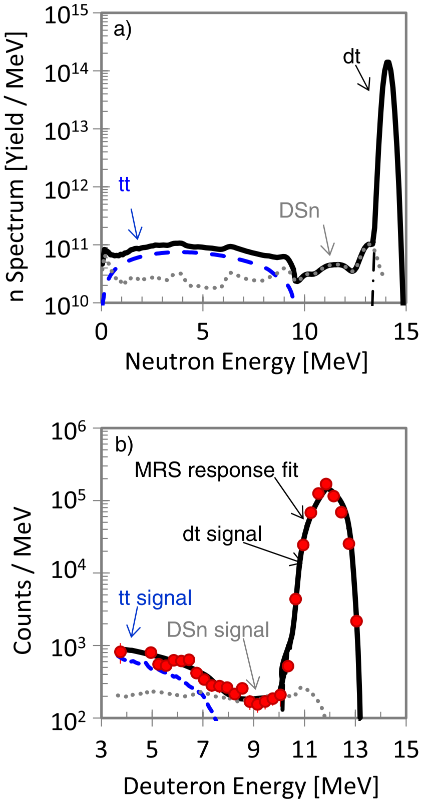

FIG. 2 (color online). (a) Neutron spectrum (black curve) that gives the best fit to the recoil deuteron spectrum measured with the MRS. (b) Recoil deuteron spectrum (red points) measured with the MRS. The spectrum is a convolution of the MRS-response function and neutron spectrum shown in (a). The deuteron peak at ~11.6 MeV is due to the primary *dt* 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 *tt* neutron spectrum dominates at deuteron energies below 7 MeV.

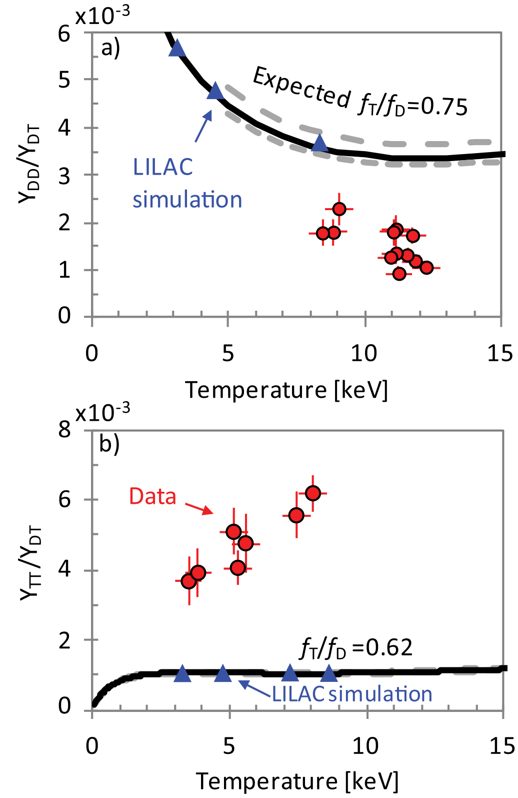

FIG. 3 (color online). (a) Measured Y_{dd}/Y_{dt} yield ratios as a function ion temperature (red points). The solid line represents the expected yield ratio calculated using Eq. (6) for the average initial *DT* fuel mixture ($f_T/f_D \cong 0.75$) used in this study. The grey dashed lines show the variation 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 $f_T/f_D \cong 0.75$). 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 ($f_T/f_D \cong 0.63$) *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).

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 $f_T/f_D \cong 0.75$ for the Y_{dd}/Y_{dt} study and $f_T/f_D \cong 0.62$ 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 these *dt* implosions are shown by the solid black curve. 1D LILAC hydrodynamic simulations [23] were also used to calculate the yield ratios (blue triangles) and the results are in excellent agreement with Eq. (6), as shown in Fig. 3(a). This comparison demonstrates that the observed ratios are significantly lower than expected from known reactivities alone, indicating a lower deuterium fraction in the core than expected. Similarly, Fig. 3(b) illustrates the observed Y_{tt}/Y_{dt} ratios, which are compared to the expected ratio (black solid curve) as a function of ion temperature. The observed reaction yield ratio is anomalously 3 to 6 times higher than predicted at ~4 and ~8 keV, respectively.

The relatively constant tt/dt (and dd/dt) 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 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 preshot 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 d^3He and dt^3He 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 t^3He deuterons using different mixtures of T^3He gas-filled implosions. T^3He deuterons have been observed in previous DT^3He gas-filled implosions [10], but not with the aim of studying possible diffusion effects. Any inferred stratification in t^3He 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^3He 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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