Observations of Multiple Nuclear Reaction Histories and Fuel-Ion Species Dynamics in Shock-Driven Inertial Confinement Fusion Implosions

H. Sio\textsuperscript{1}, J. A. Frenje\textsuperscript{1}, A. Le\textsuperscript{2}, S. Atzeni\textsuperscript{3}, T. J. T. Kwan\textsuperscript{2}, M. Gatu Johnson\textsuperscript{1}, G. Kagan\textsuperscript{2}, C. Stoeckl\textsuperscript{4}, C. K. Li\textsuperscript{1}, C. E. Parker\textsuperscript{1}, C. J. Forrest\textsuperscript{4}, V. Glebov\textsuperscript{4}, N. V. Kabadi\textsuperscript{1}, A. Bose\textsuperscript{1}, H. G. Rinderknecht\textsuperscript{4}, P. Amendt\textsuperscript{5}, D. T. Casey\textsuperscript{5}, R. Mancini\textsuperscript{6}, W. T. Taitano\textsuperscript{2}, B. Keenan\textsuperscript{2}, A. N. Simakov\textsuperscript{2}, L. Chacón\textsuperscript{2}, S. P. Regan\textsuperscript{4}, T. C. Sangster\textsuperscript{4}, E. M. Campbell\textsuperscript{4}, F. H. Seguin\textsuperscript{1}, and R. D. Petrasso\textsuperscript{1}

\textsuperscript{1}Plasma Science and Fusion Center, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA
\textsuperscript{2}Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA
\textsuperscript{3}Dipartimento SBAI, Università degli Studi di Roma “La Sapienza,” Via Antonio Scarpa 14, 00161, Roma, Italy
\textsuperscript{4}Laboratory for Laser Energetics, Rochester, New York 14623, USA
\textsuperscript{5}Lawrence Livermore National Laboratory, Livermore, California 94551, USA
\textsuperscript{6}Physics Department, University of Nevada, Reno, Nevada, 89557, USA

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

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1 Plasma Science & Fusion Center, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139 USA
2 Los Alamos National Laboratory, Los Alamos, NM 87545
3 Dipartimento SBAI, Università degli Studi di Roma “La Sapienza”, Via Antonio Scarpa 14, 00161, Roma, Italy
4 Laboratory for Laser Energetics, Rochester, New York 14623
5 Lawrence Livermore National Laboratory, Livermore, California 94551
6 Physics Department, University of Nevada, Reno, Nevada, 89557, USA

Fuel-ion species dynamics in DT\(^3\)He-gas-filled Inertial Confinement Fusion implosion is quantitatively assessed for the first time using simultaneously measured D\(^3\)He and DT reaction histories. These reaction histories are measured with the Particle X-ray Temporal Diagnostic, which captures the relative timing between different nuclear burns with unprecedented precision (~10 ps). The observed 50±10 ps earlier D\(^3\)He reaction history timing (relative to DT) cannot be explained by average-ion hydrodynamic simulations, and is attributed to fuel-ion species separation between the D, T, and \(^3\)He ions during shock convergence and rebound. At the onset of the shock burn, inferred \(^3\)He/T fuel ratio in the burn region using the measured reaction histories is much higher as compared to the initial gas-fill ratio. As T and \(^3\)He have the same mass but different charge, these results indicate that the charge-to-mass ratio plays an important role in driving fuel-ion species separation during strong shock propagation.

The goal of Inertial Confinement Fusion (ICF) at the National Ignition Facility (NIF) is to produce thermonuclear fusion in the laboratory by imploding a spherical target filled with light-ion fuel [1]. Recent experimental results reported increasing disagreements with average-ion-fluid simulations as ICF implosions become more kinetic [2, 3], as well as indications of ion species separation and thermal decoupling [4]. However, these and other experimental results [5–9] thus far have all relied on time-integrated nuclear observables such as yields and reaction temperatures. In experiment [10], Ar concentration change in a D-Ar mixture was observed using temporally and spatially resolved X-ray measurements; however, this technique cannot be applied to fuel-ions such as D, T, or \(^3\)He.

In contrast with previous studies that relied on time-integrated measurements, this work presents the first time-resolved observation of fuel-ion species dynamics in ICF implosions using DT and D\(^3\)He reaction histories. These reaction histories were measured with the Particle X-ray Temporal Diagnostic (PXTD) [11], which captures the relative timing between these reaction histories with unprecedented precision (~10 ps). These time-resolved measurements are contrasted with average-ion DUED [12] and multi-ion LSP [13] simulations. It is shown that the differential timing between reaction histories is a new manifestation of multi-ion dynamics, and that the difference between measured DT and D\(^3\)He reaction histories is consistent with rapidly changing fuel-ion composition caused by a strong shock in the central gas of an ICF target. Whereas previous ICF experiments in this plasma regime reported reasonable agreements with average-ion simulations using burn-averaged nuclear quantities [2], time-resolved reaction rates in this work clearly show differences between measurements and average-ion simulations not captured by time-integrated measurements.

Average-ion hydro simulations are essential for understanding and interpreting ICF implosions. For implosions with burn-averaged ion-ion mean free paths smaller than the burn radius, these simulations generally capture the implosion behaviors and burn-averaged quantities (yields, temperatures) [2], although not one-dimensional quantities like nuclear burn profiles [14]. Multi-ion dynamics, which are expected to impact and modify plasma conditions during the shock phase of ICF implosions [15–17], are approximately simulated in an ad hoc fashion in average-ion-fluid codes with additional physics models [18], as well as in kinetic-ion codes [19–22].

This experiment uses an exploding pusher platform [23], which is simple and ideal for studying the multi-ion dynamics during the shock phase in any ICF implosion. The reason for this is that shock phase plasma conditions (temperature, density, ion-ion mean-free-path, shock strength) are similar in all these implosions [2]. These exploding pusher targets are 860 \(\mu\)m in diameter with a 2.7-\(\mu\)m-thick SiO\(_2\) shell. The gas-fill density is 2.2 mg/cc, with an atomic fuel composition of 49.6% D, 49.7% \(^3\)He, and 0.7% T. These targets are driven symmetrically by sixty laser beams at the OMEGA laser facility [25] with a total energy of 14.4 kJ using a 0.6-ns square pulse shape.
These low-convergence (convergence ratio between 3 to 5), shock-driven implosions are not affected by hydrodynamic instabilities and mix [3]. On the other hand, SiO$_2$ from the shell could conceivably have been mixed into the gas from shock breakout across the fuel-shell interface. This scenario is very unlikely, as the absolute X-ray emissions measured by the Hard X-Ray Detector (HXRD) [26] confirm an X-ray emission from a clean, mix-free D$_3$He fuel. The effect of spherically converging shock instability is expected to be negligible, as estimated using the analytical and numerical work by Gardner et al. [27]. Shock front deviation over the mean shock radius ($\delta r/r$) in this model increases with an amplitude given by $r^{-0.72}$ (for an ideal gas) as the shock converges. Given that laser illumination non-uniformity on target is less than 2% [29], and that initial local shock speed scales as the 1/3 power of the local laser intensity [30], the initial difference in local shock speed is $\sim$0.7%. At shock convergence of 20, $\delta r/r$ is only $\sim$6%, or a $\delta r$ of 1 $\mu$m. Shock collapse and rebound further reduce $\delta r/r$ perturbations. In addition, experiments [28] with initial shock perturbations up to 14% show no difference in nuclear yields or timing during the shock phase.

The primary measurements in this experiment are the absolute DT and D$_3$He reaction histories, which are simultaneously measured with the PXTD. This is done by measuring the time-arrival histories of the monoenergetic 14.1-MeV DT-n and 14.7-MeV D$_3$He-p as they escape the implosion. As all measurements are made with the same diagnostic, the relative timing uncertainty between the DT and D$_3$He reaction histories is $\sim$10 ps (versus $\sim$40-50 ps, with the standard method of cross-timing between two standalone diagnostics). This innovation is crucial to capturing the relative timing between different nuclear burns with sufficiently high precision to enable meaningful comparison between measurements and simulations.

The time period probed by the measured reaction histories in these ICF implosions is referred to as the shock phase. Nuclear yields are produced when the strong shock rebounds from the center of the implosion. Fig.1 is a Lagrangian diagram from an average-ion DUED simulation for shot 82615, showing the trajectories of fluid elements as a function of time. The shell trajectory (black data) as measured by an X-Ray Framing Camera (XRFC) [31] agrees with the simulated shell trajectory. The measured DT and D$_3$He reaction rates provide information during the shock phase on the relative temporal differences between D, T, and He temperature and density profiles through the following relations:

$$Y_{D3He}(t)/s = \int n_D n_{3He}(\sigma v)_{D3He}(T_i,D,T_i,3He) dV \quad (1)$$

$$Y_{DT}(t)/s = \int n_D n_T(\sigma v)_{DT}(T_i,D,T_i,T) dV \quad (2)$$

where $n$ is the ion number density, $T_i$ is the ion temperature, and $\langle \sigma v \rangle$ is the Maxwellian-averaged reactivity. In the average-ion framework, $n_D$, $n_T$, and $n_{3He}$ are related by the initial gas-fill ratio, and the ion temperatures for all three ions ($T_i,D$, $T_i,T$, $T_i,3He$) are the same.

As time-resolved and time-integrated measurements are repeatable within uncertainty for four implosions (see Table I), shot 82615 is used as a representative shot in this manuscript. Experimentally, the D$_3$He bang time

<table>
<thead>
<tr>
<th>Shot</th>
<th>Gas-fill (atm)</th>
<th>Laser (kJ)</th>
<th>Bang time (ps)</th>
<th>Yield (kJ)</th>
<th>Ti (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>82613</td>
<td>D$_2$[4.9]T$_2$[0.07]He[9.3]</td>
<td>14.3</td>
<td>755</td>
<td>809</td>
<td>4.0 $\times 10^{10}$</td>
</tr>
<tr>
<td>82614</td>
<td>D$_2$[4.9]T$_2$[0.07]He[9.7]</td>
<td>14.2</td>
<td>800</td>
<td>841</td>
<td>4.9 $\times 10^{10}$</td>
</tr>
<tr>
<td>82615</td>
<td>D$_2$[4.9]T$_2$[0.07]He[9.8]</td>
<td>14.2</td>
<td>780</td>
<td>831</td>
<td>5.2 $\times 10^{10}$</td>
</tr>
<tr>
<td>82616</td>
<td>D$_2$[4.9]T$_2$[0.07]He[9.8]</td>
<td>14.1</td>
<td>840</td>
<td>875</td>
<td>4.0 $\times 10^{10}$</td>
</tr>
</tbody>
</table>

*TABLE I. Experimental parameters and key observables. The absolute bang-time uncertainty (relative to laser start) is 50 ps. The relative bang-time uncertainty (between D$_3$He and DT) is 10 ps. The uncertainties for the D$_3$He-p, DT, and DD yields are 20%, 5%, and 5%, respectively. The uncertainties for the D$_3$He-p, DT, and DD burn-averaged temperatures are 1.5 keV, 0.5 keV, and 0.5 keV, respectively.*
is 50±10 ps before the DT bang time (Fig. 2a), and is contrasted to the reaction histories as simulated by the average-ion hydrodynamic code DUED (Fig. 2b). The DUED simulation used an electron flux limiter of 0.07, and included ion viscosity [32]. The DUED simulation also used a multi-group diffusive treatment of radiation transport and an equation-of-state [36]. For shot 82615, the DUED-simulated D³He, DT, and DD yields are 5.4×10¹⁰, 3.7×10¹¹, and 7.0×10¹⁰, respectively, comparable to the measured yields in Table I. The DUED-simulated burn-averaged D³He, DT, and DD temperatures are 12.7, 11.0, and 10.4 keV, respectively, in good agreement with the measured temperatures. However, the average-ion DUED simulation cannot explain the relative timing between the measured D³He and DT reaction histories, showing only a 10-ps timing difference between the two histories in the simulation. The DUED-simulated DT and D³He burnwidths are also more narrow as compared to the PXTD measurements.

In comparison with the average-ion simulation, a significantly higher D³He reaction rate is observed relative to DT at the onset of the shock burn. This is observed on all four shots. Higher-than-expected ion temperature early-in-time in the fuel cannot explain this observation, as it would have also lead to higher burn-averaged D³He and DT temperatures, contrary to the measured burn-averaged temperatures.

The effect of different D, T, and ³He temperatures cannot be ignored, but the effect is small according to DUED simulation post-processed with a multi-Ti model. Using this model, energy is partitioned to the D, T, and ³He ions according to their masses, and temperatures are equilibrated using local plasma conditions. As expected from the short ion-ion thermalization time (Fig. 3a), the higher temperatures of the T and ³He ions have a small impact on the reaction yields, and most importantly, have no effect on the timing of the reaction histories (Fig. 3b) [33]. However, the measured timing difference is consistent with ion species separation driven by sharp pressure gradients at the shock front [37] in the implosion. Explicitly calculating the ion diffusive flux (which depends on the charge and mass of the ion species) using expressions from [38] and gradients from the DUED simulation shows the T ions are lagging behind the D and ³He ions during shock convergence, consistent with the PXTD measurements. As the shock propagates radially inward, the diffusion coefficients at the shock front are ~2×10³ μm²/ns, and the normalized pressure gradients (∇P/P) are ~0.2 μm⁻¹. These terms lead to an ion diffusive flux of ~300 μm/ns in the shock frame. The dominant terms driving the D and ³He ions forward relative to the T ions are from the ion pressure gradient (baro-diffusion, which accelerates the lighter D ions ahead) and the electron pressure gradient (electro-diffusion, which accelerates the higher-charge ³He ions ahead).

As temperature effects are demonstrably small, the observed difference between the measured reaction histories is attributed to fuel-ion-species separation between the D, T, and ³He ions. To infer the level of separation in the burn region needed to explain the measured reaction histories, the ratio of Eq. 1 and Eq. 2 is approximated as:

\[
\frac{Y_{D^3He}(t)/s}{Y_{DT}(t)/s} \approx \frac{\langle n_{D^3He} \rangle \langle \sigma v \rangle_{D^3He}}{\langle n_T \rangle \langle \sigma v \rangle_{DT}}
\]

(3)
where \( n \) is ion number density for the different ion species and \( \langle \sigma v \rangle \) is the Maxwellian-averaged reactivity for the different reactions. This approximation for the instantaneous \( \text{D}^3\text{He}/\text{DT} \) yield ratio is valid if the ion temperature variance over the burn region is small [9], which is the case for these hydrodynamic-like implosions. Calculating Eq.(3) and the exact ratio explicitly in simulation shows that this approximation introduces less than 20% uncertainty.

The measured \( \text{D}^3\text{He} \) and DT reaction histories in Fig.2a are used to obtain the instantaneous \( \text{D}^3\text{He}/\text{DT} \) yield ratio in Fig.4a. Using Eq.(3), the burn-averaged \( \text{D}^3\text{He}/\text{T} \) fuel ratio in the burn region as a function of time (Fig.4b) is inferred from the measured instantaneous \( \text{D}^3\text{He}/\text{DT} \) yield ratio. The horizontal purple-dashed line indicates the initial \( \text{D}^3\text{He}/\text{T} \) gas-fill ratio. The reactivity ratio is extracted from the average-ionic simulation, and constrained by measured DD, DT, and \( \text{D}^3\text{He} \) ion temperatures. At the onset of the shock burn, the inferred \( \text{D}^3\text{He}/\text{T} \) fuel ratio in the burn region is much higher as compared to the initial \( \text{D}^3\text{He}/\text{T} \) gas-fill ratio in all implosions. The relaxation of \( \langle n_{\text{D}^3\text{He}} \rangle / \langle n_T \rangle \) toward initial gas-fill ratio is partially a consequence of the burn region expanding outward and encompassing a larger fraction of the fuel volume as the shock rebound.

As a check on this inference using Eq.3, we can instead assume that the fuel-ion ratio is fixed, and from that infer the ion temperature history needed to explain the measured reaction histories. However, this temperature history assuming constant fuel-ion ratio would have also lead to 20% higher burn-averaged \( \text{D}^3\text{He} \) and DT temperatures, which contradicts the measured burn-averaged \( \text{D}^3\text{He} \) and DT temperatures (see Table I). That is, the assumption of constant fuel-ion ratio is inconsistent with the measured reaction histories and measured burn-averaged temperatures.

It is also insightful to illustrate the D, T, and \( \text{D}^3\text{He} \) density profiles evolution in an ICF implosion using a simulation code that treats the D, T, and \( \text{D}^3\text{He} \) population separately. The Particle-in-Cell (PIC) code LSP is used to simulate OMEGA shot 82615, treating the D, T, \( \text{D}^3\text{He} \), and \( \text{SiO}_2 \) ion species as kinetic. The electrons are treated as a fluid. More details on the LSP simulation method can be found in [40] and [41]. The LSP simulation is initiated at \( t = 0.55 \text{ ns} \) using initial conditions from a hydrodynamic simulation shortly after the shock breaks out from the shell (see Fig.1). The LSP-simulated implosion trajectory agrees with DUED simulation and XRFC data (Fig.1).

110 ps later after LSP initialization at \( t = 0.66 \text{ ns} \) (Fig.5), fuel-ion species separation has already developed between the D, T, \( \text{D}^3\text{He} \) ions. As the shock rebounds from the center (\( t = 0.76 \text{ ns} \)), the temperature profiles are centrally peaked. The fusion reactivities’ dependence on temperature weights the DT and \( \text{D}^3\text{He} \) reaction profiles toward the center of the implosion. However, because the T ion number density profile is skewed toward the outer volume of the fuel that is not yet heated by the rebounding shock, the \( \text{D}^3\text{He} \) yield is higher than DT yield at this early time relative to average-ionic simulation, leading to an earlier \( \text{D}^3\text{He} \) reaction history relative to DT (see Fig.6) that is consistent with the PXTD measurements in Fig.2. The LSP-simulated \( \text{D}^3\text{He} \) burnwidth is consistent with the PXTD measurements, while the LSP-simulated DT burnwidth is notably wider. Qualitatively, this LSP simulation clearly demonstrates how fuel-ion species separation that developed during shock propagation and re-

FIG. 4. (color online) a) instantaneous \( \text{D}^3\text{He}/\text{DT} \) yield ratio for shot 82615. In c), the inferred \( \langle n_{\text{D}^3\text{He}} \rangle / \langle n_T \rangle \) (black data) is plotted. The purple-dashed line marks the initial \( \text{D}^3\text{He}/\text{T} \) fuel ratio. Uncertainties in the data are indicated by the shaded regions.

FIG. 5. (color online) LSP-simulated ion number density profiles at \( t = 0.66 \text{ ns} \) (shock converging) and \( t = 0.76 \text{ ns} \) (shock rebounding). The D, T, and \( \text{D}^3\text{He} \) ion number densities are plotted in blue, green, and red, respectively. The number density profile for the T ions has been scaled by 50 for clarity. The vertical black-dashed line denotes the fuel-shell interface.
bound manifest as a timing differential between reaction histories. In addition, the effects of non-Maxwellian distribution (fast-ions, etc) have been captured by the LSP simulations.

In summary, the timing difference between measured DT and D3He reaction histories in hydrodynamic-like implosions cannot be explained by average-ion simulations, and is attributed to ion species separation between the D, T, and 3He ions during shock convergence and rebound. At the onset of the shock burn, the $^{3}\text{He}/T$ fuel ratio in the burn region inferred from the measured reaction histories is much higher as compared to the initial $^{3}\text{He}/T$ gas-fill ratio, in contrast with average-ion simulations. As T and 4He have the same mass but different charge, these results indicate that the charge-to-mass ratio plays an important role in driving fuel-ion species separation during strong shock propagation. It is unclear how these multi-ion effects affect implosion performance during the deceleration and compression phase, as existing experimental results [5–9] have been mixed. The next step in addressing this open question is to determine, using time-resolved data measured with the PXTD diagnostic, whether fuel-ion species separation developed in the shock phase persists into and affects performance during the compression phase in ablatively-driven ICF implosions with both shock and compression burns.

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FIG. 6. (color online) LSP-simulated DT (blue) and D3He (red) reaction histories for shot 82615. The magnitude of the D3He reaction history is scaled by a factor of 3.1 for clarity. The absolute magnitude of the LSP-simulated reaction histories is notably lower than the measurements, in part because of the reduced laser coupling in the simulation [41].

* hsio@mit.edu

Two other 1-D radiation-hydrodynamic codes, LILAC [35] and HYADES [34], are used to simulate these implosions, with similar results. DUED has also been extensively benchmarked against LILAC and HYADES in ref [2].

The effective DT and D<sup>3</sup>He fusion temperatures for two Maxwellian ion populations are given by [20] $T_{fus,DT} = (T_{i,D} + m_D T_{i,T})/(m_D + m_T)$ and $T_{fus,D3He} = (m_{3He} T_{i,D} + m_D T_{i,3He})/(m_D + m_{3He})$, respectively, where $m$ is the ion mass and $T_i$ is the ion temperature for each ion species.

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S. Atzeni et al., Laser Part. Beams 4, 393 (1986).


As the laser pulse is truncated once the LSP simulation begins, 30% less laser energy is coupled to the target in the LSP simulation. The reduced laser coupling is not the cause of the timing difference between the DT and D<sup>3</sup>He reaction histories, as hydrodynamic simulations ran with full laser coupling and reduced laser coupling have the same timing difference.