Observation of interspecies ion separation in inertial-confinement-fusion implosions

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Abstract - We report direct experimental evidence of interspecies ion separation in direct-drive, inertial-confinement-fusion experiments on the OMEGA laser facility. These experiments, which used plastic capsules with D$_2$/Ar gas fill (1\% Ar by atom), were designed specifically to reveal interspecies ion separation by exploiting the predicted, strong ion thermo-diffusion between ion species of large mass and charge difference. Via detailed analyses of imaging x-ray-spectroscopy data, we extract Ar-atom-fraction radial profiles at different times, and observe both enhancement and depletion compared to the initial 1\%-Ar gas fill. The experimental results are interpreted with radiation-hydrodynamic simulations that include recently implemented, first-principles models of interspecies ion diffusion. The experimentally inferred Ar-atom-fraction profiles agree reasonably, but not exactly, with calculated profiles associated with the incoming and rebounding first shock.

Interspecies ion separation has been proposed as a potential fusion-yield degradation mechanism in inertial-confinement-fusion (ICF) implosions [1–3]. Several ICF experimental campaigns [4–11] have reported yield or yield-ratio anomalies where interspecies ion separation within the hot spot is a possible explanation. Meanwhile, first-principles analytic theories for multi-ion-species diffusion [1,2,12–16] have been developed, some of which have been implemented into ICF implosion codes [17,18], enabling us to quantitatively assess the magnitude of interspecies ion separation and yield degradation in ICF implosions. In addition, ion-Fokker-Planck [19,20] and particle-in-cell kinetic simulations [3,21,22] are being used to address this problem, and are particularly appropriate for more-kinetic scenarios, e.g., exploding pushers, hotspot formation in ignition capsules, hohlraum plasmas, or near steep gradients at shock fronts and material interfaces. We do not address the question here of whether interspecies ion separation substantially affects the yield in ignition-class implosions on the National Ignition Facility [23], but point out that this and related work will help establish a validated capability to answer the question in a quantitative manner.

The purpose of this work is to complement and expand upon prior ICF experimental campaigns that relied on yield or yield-ratio anomalies as evidence for species separation. Here, based on direct-drive ICF implosions on the OMEGA laser facility [24], we provide direct, spatially resolved experimental evidence of interspecies ion separation via detailed analyses of imaging x-ray-spectroscopy data, which are less sensitive to other potential causes of yield degradation in an ICF implosion. In this work: 1) we exploit recently developed analytic theory [14,15] of interspecies ion diffusion to design an ICF implosion that maximizes interspecies ion diffusion, such that it is observable via x-ray diagnostics; 2) rather than focusing on separation between fuel species, e.g., D/\textsuperscript{3}He or D/T, we use D and a trace amount of Ar, where both the choice of Ar itself and its pre-fill concentration are chosen to maximize the expected interspecies diffusion coefficient; and 3) we aim for a more-collisional implosion so that interpretations using the recently formulated interspecies-ion-diffusion theories are appropriate. The main results in this letter are the first direct identification of interspecies ion separation in an ICF implosion based on the analyses of spatially resolved x-ray-spectroscopy data, observation
of spatially non-uniform Ar-concentration enhancement and depletion (compared to the spatially uniform pre-fill of 1% Ar by atom) during the implosion, and reasonable agreement of the experimentally inferred Ar-atom-fraction profiles with radiation-hydrodynamic simulations that model interspecies ion diffusion from first principles.

We briefly discuss the theory that guided these experiments. The diffusive mass flux of the lighter ions relative to the center of mass in a plasma with two ion species can be written as [14,15]

\[ \vec{i} = -\rho \nabla T_i + k^{(i)}_T \nabla \log T_i + k^{(c)}_E \nabla \log \rho + k^{(e)}_T \nabla \Phi + \frac{e k^{(e)}_E}{T_i} \nabla \Phi + \frac{e k^{(c)}_E \nabla \log T_e}{T_i}, \]

where \( \rho \) is the total mass density, \( D \) the classical diffusion coefficient, \( c \equiv p_D/(\rho_D + p_{Ar}) \) for ion thermo-diffusion, \( k^{(i)}_T \), electron thermo-diffusion, \( k^{(c)}_T \), baro-diffusion, \( k_E \), and electro-diffusion, \( k_E \), assuming charge state \( Z_{Ar} = 18 \). (b) We shot four target types: 15 \( \mu \)m/3 atm and 13 \( \mu \)m/5 atm, with 1% or 0.1% Ar by atom.

Thus, we focused our attention on an implosion design with large \( \nabla T_i \) and \( f_{Ar} \approx 1\% \) to maximize ion thermo-diffusion. Furthermore, we wanted relatively high \( T_i \) and \( D \) while keeping the Ar-D mean free path much smaller than the hot-spot size for most of the implosion. Although the theory [14–16] is not strictly correct within a shock front, it should nevertheless capture the qualitative, leading-order effects on interspecies ion separation during the passage and rebound of the first incoming shock.

We performed 1D HYDRA [32] simulations of direct-drive OMEGA implosions with a 1 ns square pulse to arrive at the capsule designs in fig. 1(b). Table 1 summarizes the as-shot laser and target parameters and neutron-based measurements. To evaluate the amount of expected species separation, we also performed pre-shot 1D simulations using xRAGE [17] with its recently implemented two-ion-species transport model [16]. All our pre-shot xRAGE results showed relative deviations in \( f_{Ar} \) of \( \geq 20\% \), in some cases much larger, from the initial values (\( f_{Ar} = 0.01 \) or 0.001) over much of the implosion, giving us confidence that we could resolve the species separation based on error bars achieved in previous analyses of imaging x-ray-spectroscopy data.

The primary diagnostics are two x-ray monochromatic imagers (MMI) [33] with quasi-orthogonal views (mounted on TIMs 3 and 4, where TIM stands for a “ten-inch manipulator” diagnostic port), a streaked x-ray spectrometer (TIM 1), and a time- and space-integrated, absolutely calibrated x-ray spectrometer (TIM 2). Standard neutron diagnostics and full-aperture backscatter systems were also fielded. The MMIs used 10 \( \mu \)m diameter pinholes, setting the spatial resolution at \( \geq 10 \mu \)m, and recorded data on x-ray framing cameras between 3.3 and 5.5 keV around the time of first-shock convergence. Each camera recorded four frames per shot at different times; frame trigger times are given in the figure captions. We obtained analyzable MMI data for several shots with initial \( f_{Ar} \approx 1\% \) (table 1), and used the Ar-He(3.68 keV), Ly\( \beta \) (3.94 keV), and Ly\( \gamma \) (4.15 keV) lines in our spectroscopic analyses. Shots with initial \( f_{Ar} \approx 0.1\% \), predicted to have much reduced ion thermo-diffusion, provided weaker spectral lines.

The key objective and result of this work is the experimental inference of \( f_{Ar} \) vs. radius \( r \). Deviation from the spatially uniform target-pre-fill value constitutes proof of
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interspecies ion separation. Figures 2 and 3 illustrate the key steps of the MMI data processing and analyses required to arrive at $f_{Ar}(r)$. MMI data processing to obtain narrow-band images (fig. 2(b)) is based on the method of refs. [29,31], and [34]. Extraction of electron density $n_e$ and temperature $T_e$ vs. $r$ are based on the emissivity-analysis method of ref. [28]. Figure 2(a) shows one frame of MMI data after it has been converted from film density to intensity (arbitrary units). The photon-energy dependence of the MMI data is corrected using streaked and absolutely calibrated spectral data\(^\text{1}\). Figure 2(b) shows the narrow-band images constructed from the data in fig. 2(a), and fig. 2(c) shows the space-integrated spectrum from fig. 2(a) before continuum subtraction.

The next step is to extract $n_e$ and $T_e$ radial profiles. Abel inversion is applied to each narrow-band image, e.g., fig. 2(b), giving argon emissivity (with continuum subtracted) vs. $r$ for four radial zones of approximately 10 $\mu$m width (set by both the size of MMI pinholes and spectral signal-to-noise considerations for each zone), at the time of the particular MMI frame. The time is approximately known based on the experimental trigger time of each frame of the x-ray framing camera, but due to cable delays and the finite time of the sweep across the camera’s photocathode strips, we regard the quoted times as having an uncertainty $\sim$ 50 ps. The Ar-He$\beta$ and Ly$\beta$ emissivity profiles are analyzed to provide $n_e$ and $T_e$ vs. $r$ (fig. 3(a)) based on comparisons with detailed atomic-physics models. The underlying atomic database was generated via the fully relativistic path in the Los Alamos suite of codes RATS, ACE, and GIPPER [35,36], followed by the collisional-radiative code ATOMIC [36–38]. The resulting NLTE (non-local thermal equilibrium) spectral data were used in the intensity and emissivity analyses of the MMI data (figs. 3, 4(a), (b)), as well as in the forward modeling of space-resolved spectra and narrow-band images by the FESTR spectral post-processing code [39,40]. Error bars for $T_e$ are obtained based on the greater of the $T_e$ resolution (100 eV) in the atomic database mentioned above or the standard deviation of $T_e$ (e.g., average of 63 eV for the four radial zones for shot 78199, TIM 3, frame 2) as determined from the theoretical emissivity ratio of Ly$\beta$/He$\beta$ at four different values of $n_e$ ($5 \times 10^{21}$, $8 \times 10^{21}$, $1 \times 10^{24}$, and $2 \times 10^{24}$ cm$^{-3}$) in our regime of interest. Error bars for $n_e$ are obtained based on the greater of the uncertainties in the $T_e$ (100 eV) and $n_e$ solutions (1–2 $\times 10^{23}$ cm$^{-3}$, depending on the density) in the database, or the standard deviation in $n_e$ (e.g., average of $1.1 \times 10^{23}$ cm$^{-3}$ for the four radial zones for shot 78199, TIM 3, frame 2) from assuming slightly higher and lower values of $n_e$ in the central zone (e.g., $4 \pm 1 \times 10^{23}$ cm$^{-3}$ for shot 78199, TIM 3, frame 2), which is derived from the line broadening of each of the three Ar lines from the space-integrated spectrum.

The argon density $n_{Ar}$ is determined using $n_{Ar} = n_u/F_u(T_e, n_e)$, where $n_u$ and $F_u$ are the upper-level number densities and fractional populations, respectively, of the particular line transition being used [41]. $F_u(T_e, n_e)$ is retrieved from the atomic database based on the previously determined $n_e(r)$ and $T_e(r)$. Since the spectral lines are sufficiently optically thin [28], $n_u$ (arbitrary

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\(^\text{1}\)The MMI data is corrected as follows. First, the streaked x-ray-spectroscopy data (SSCA) is corrected by matching the time-integrated SSCA data to the space- and time-integrated, absolutely calibrated x-ray-spectroscopy data (XRS2). Then the space-integrated MMI spectrum is corrected to match the corrected SSCA data at the time of the MMI data.
The deuterium density in the innermost to outermost radial zone for shot 78199, initial pre-fill value from several different shots and times. Results shown are the averages from the Ar-Heβ, Lyβ, and Lyγ lines. The non-uniform spatial profiles of fAr and their deviation from the initial pre-fill value from several different shots and times constitute proof of interspecies ion separation between the D and Ar, which is the main result of this letter.

To build confidence in our conclusions, we apply the same analysis procedure to synthetic Ar-Heβ, Lyβ, and Lyγ images and space-resolved spectra. The synthetic data are generated using the FESTR code [40], using the data in fig. 3 and assuming 1D spherical symmetry, for two cases: i) nAr as shown in fig. 3(b) and ii) spatially uniform fAr ≈ 0.01 with equal nT + nAr as in i). In both cases, we extract fAr(r) in good agreement with the fAr(r) in the synthetic data. This demonstrates that our analysis accurately extracts fAr(r) and is also able to distinguish between species separation vs. no species separation.

We also note some limitations of our analysis. Firstly, our analysis assumes 1D spherical symmetry, introducing possible errors if the implosion has significant 2D and/or 3D structure. While our implosions appear fairly round, and quasi-orthogonal MMI views at similar times yield similar results (e.g., shot 78197, blue and orange curves in fig. 4(b)), small-scale non-uniformities are clearly visible (e.g., fig. 2(b)). We assessed the effect of non-uniformities for shot 78199 (TIM 3, frame 2) by dividing the narrowband images of fig. 2(b) into three wedges of 120° each and doing independent analysis on each wedge to obtain ne and Te profiles; the standard deviations of ne(r) and Te(r) across the three wedges are similar to the error bars already considered (as shown in fig. 3) and thus would not substantially increase the final error bars shown in fig. 4. Future work should employ both 2D [31] and 3D [42] reconstruction techniques to further assess both 2D and 3D effects on our analysis. Secondly, unaccounted-for CH shell mix into the hot spot affects the species balance in the quasi-neutrality condition used in our analysis by causing overestimates of the inferred nD, which affects the inferred fAr. However, we have estimated that the local CH fraction in the hot spot must exceed an unrealistically large value ~ 10% before it could affect our conclusion of D/Ar separation presented in figs. 4(a) and (b). Finally, our use of the known initial amount of Ar in the fuel region to infer absolute nAr from ne (arbitrary units) means that nAr is overestimated if Ar leaks into the shell. The latter could be a possible explanation for why fAr(r) < 1% everywhere for shot 78197 (fig. 4(b)), keeping in mind that fAr(r) is extracted only from regions with visible, analyzable Ar spectral emission. Resolution of the shell/gas-mix issues requires further work, including the use of N-species ion-diffusion models that can model both the D/Ar separation and gas/shell mix.

Next, we interpret our results via a post-shot, 1D simulation of shot 78199 using xRAGE [17] with a two-ion-species transport model [16]. Figure 4(c) shows fAr(r) for shot 78199, revealing that fAr is reduced ahead of the incoming first shock and enhanced behind it (green and red curves of fig. 4(c)). After shock reflection (at ≈ 1.18 ns and ≈ 87% of neutron bang time in the simulation), fAr is enhanced throughout much of the hot spot (up to ≈ 20 μm) through neutron bang time.
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