Observation of persistent species temperature separation in inertial confinement fusion mixtures

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The injection and mixing of contaminant mass into the fuel in inertial confinement fusion (ICF) implosions is a primary factor preventing ignition. ICF experiments have recently achieved an alpha-heating regime, in which fusion self-heating is the dominant source of yield, by reducing the susceptibility of implosions to instabilities that inject this mass. We report the results of unique separated reactants implosion experiments studying pre-mixed contaminant as well as detailed high-resolution three-dimensional simulations that are in good agreement with experiments. At conditions relevant to mixing regions in high-yield implosions, we observe persistent chunks of contaminant that do not achieve thermal equilibrium with the fuel throughout the burn phase. The assumption of thermal equilibrium is made in nearly all computational ICF modeling and methods used to infer levels of contaminant from experiments. We estimate that these methods may underestimate the amount of contaminant by a factor of two or more.
Inertial confinement fusion (ICF) involves the compression of capsules containing deuterium and tritium (DT) with the goal of achieving self-sustaining thermonuclear burn. The performance of capsule implosions is limited by the introduction of contaminant material that mixes with the fuel through interfacial instabilities and engineering features. We use the term mixing broadly, to include the introduction of contaminant (non-hydrogen) material into the fuel region of a capsule implosion. This includes the limiting cases of contaminant that is atomically mixed with the fuel as well as contaminant “chunks” that penetrate the fuel region as macroscopic fingers, jets, or “meteors”. This contaminant degrades yield by enhancing conductive and radiative losses, as well as displacing fuel. ICF experiments have recently achieved an alpha-heating regime, in which fusion self-heating is the dominant source of yield, by reducing the susceptibility of implosions to the instabilities that inject contaminant mass.

It has been argued that material mixed into hot, compressed fuel equilibrates rapidly with the hot spot due to rapid inter-particle collisions, so that the equilibration time can be neglected. This assumption is essential to estimates of mix mass from X-ray emission that are frequently used in the literature as well as our understanding of how contaminant degrades yield. It also implies that chunks of contaminant rapidly atomize, which has not been demonstrated. In the literature, the impact of mixing has focused on radiative losses associated with hot contaminant. Nevertheless, by effectively increasing the surface area of the hot spot and introducing large temperature gradients, cold contaminant enhances conductive losses, which can be just as significant. Whether conductive or radiative losses are dominant depends on the local hot spot conditions, quantified by the temperature and areal density (see, e.g., ref. 12) and the conduction losses will be amplified by the existence of cold chunks of contaminant. It is therefore crucially important to understand the amount of both cold and hot contaminant in fuel regions.

Species temperature separation arises for several reasons. Hot spot ignition designs for the National Ignition Facility (NIF) require carefully timed shocks to heat the fuel layer, and hence the ablator, less than the low-density vapor. In general, temperature separation arises from different specific heats of initially separated materials, causing hydrogenic fuel to heat more than contaminant. Thermal equilibration between separated species can occur via electron thermal conduction (ion conduction is a factor of $m_i/m_e$ smaller) or local collisions after atomically mixing—i.e., diffusion or convection—and the latter process is much faster. The timescale at which a hot $T_e = 1$ keV mixing region with a 50$\mu$m extent equilibrates by electron thermal conduction can be estimated, assuming full ionization and using Spitzer’s conductivity, as $\tau \approx \frac{P_f C_{\nu e} e^{-1}}{\sqrt{\pi}} \approx 1.6 \text{ ns}$. Therefore, thermal equilibration between initially separate species is dependent on the development of atomic mixing, which varies depending on implosion details. Simulations resolving the mixing processes in warm OMEGA implosions indicate that atomic mixing continues to develop through the burn phase. For more general hydrodynamically unstable flows, one can apply theory from ref. 12 to estimate the time $t^*$ for the flow to achieve a mixing transition, in which the flow drives a rapid development of atomic mixing:

$$t^* \geq 6 \delta \nu^{-1} 100^2 C_{\nu e}^{1/2} \text{Re}^{-1/2},$$

where $\text{Re} = 1.6 \times 10^5$, $\delta$ is the experimental enclosure lengthscale, $\nu$ is the characteristic velocity, and $C_{\nu e}$ is a diffusion layer coefficient (here, $C_{\nu e} = \sqrt{15}$). For high-yield NIF capsule implosions, $t^* \approx 6-12 \text{ ns}$, comparable to implosion timescales.

Since many instabilities arise late in the implosion, chunks of contaminant have the potential to remain unmixed and hence unequilibrated with the fuel during the burn phase.

We report the results of experiments and highly detailed and resolved 3D simulations that provide evidence that for a wide range of initial conditions, mixing regions in inertially confined plasmas—i.e., regions where contaminants have mixed with hydrogenic fuel—maintain chunks of contaminants that do not achieve thermal equilibrium (for both ions and electrons) with the fuel during ICF implosions. We will demonstrate that as a consequence, models used to estimate contaminant mass through enhanced X-ray bremsstrahlung emission may underestimate the amount of contaminant by a factor of two or more. Given the physical processes involved, a corollary of our observation is that atomic mixing between fuel and contaminant develops faster than equilibration through conduction and that both processes can be slower than the implosion. This observation is critically important to ICF because of the strong sensitivity of contaminant bremsstrahlung radiative emission, the dominant energy loss mechanism, and DT thermonuclear reactivity to temperature variations. The power of bremsstrahlung emission

$$P_{\text{br}} \propto \sqrt{T_e^6},$$

where $T_e$ is the electron temperature and $T_i$ is the ion temperature, with $3 \leq \alpha \leq 7$ for ICF-relevant temperatures. The introduction of cold contaminant into the hot spot also enhances conductive losses, due to the presence of temperature gradients inside the hot spot as well as enhancing the surface area of the hot spot boundary. Temperature separation has previously been observed in experiments designed to maximize kinetic effects as well as between species in fuel regions in more hydrodynamic implosions. The primary advancement of our work is that we consider conditions in compressed mixing regions containing both carbon and fuel, conditions that have been observed to minimize kinetic effects. Our results suggest that there is margin to improve the prospects for ignition by improving target quality to reduce the magnitude of imperfections that seed these instabilities or by improving tolerance to these asymmetries.

Results

Description of experiments and context. We report the results of novel separated reactant ICF experiments designed to help to understand the behavior of contaminant mass during ICF implosions, along with detailed and highly-resolved three-dimensional (3D) simulations that are in good agreement with the experimental data. Our experiments explore the limiting case where chunks of contaminant are present in the initial conditions, allowing the maximum possible time to atomically mix and equilibrate with the fuel. Our experiments involve the compression of capsules (Fig. 1b) containing deuterated open-cell foams whose pores are filled with hydrogen and tritium, and we control the initial conditions by varying the foam pore sizes. Contaminant and fuel are initially physically separated and heat to different temperatures due to this separation as well as their different specific heats. As the implosion progresses, hydrodynamic processes lead to atomic mixing of these materials, resulting in a complex distribution of materials containing regions of pure chunks of contaminant or fuel and atomically mixed regions. As a result of the process of atomic mixing, fuel and contaminant in atomically mixed regions locally achieve thermal equilibrium. Our experiments uniquely address the persistence of chunks and whether these chunks remain in thermal equilibrium with the fuel. Furthermore, our results indicate that the observed phenomena can be explained by appealing to well-known physical processes—hydrodynamic instabilities, which lead to mixing and local equilibration,
combined with thermal conduction, which brings materials into thermal equilibrium over larger scales—and the time-scales over which they operate. These timescales have previously only been addressed theoretically.

We will demonstrate reactant temperature separation in experiments by contradiction. Assuming the reactants are in thermal equilibrium, $T_{\text{DT}} = T_{\text{DD}}$, and the DT/DD yield ratio is determined by the extent to which the reactants are atomically mixed. However, our experimental data lie above this theoretical maximum, so this assumption cannot be correct. To further support this conclusion, reactant temperature separation is directly observed in simulations that are in agreement with the experimental observables. We have evaluated several alternative explanations, including kinetic effects, anomalously high ion temperatures, target mischaracterization, and complete atomic mixing, and these cannot plausibly explain the data.

The observation of species temperature separation in ICF mixing regions has important consequences for our understanding of implosions. Models used to estimate contaminant mass through enhanced X-ray bremsstrahlung emission assume thermal equilibrium between contaminant and DT\textsuperscript{2-6}. Such models underpredict the amount of contaminant by overestimating the emission of the contaminant. Due to the difficulty of probing the relevant timescales and length-scales, our understanding of ICF implosions relies on simulations, which nearly universally assume mixtures to be in thermal equilibrium through modeling choices. Only with significant time on today’s largest supercomputers is it possible to accurately simulate the inherently 3D hydrodynamics\textsuperscript{22} and coupled physics at the sub-micron scales where mixing occurs. Therefore, this has only been done a few times\textsuperscript{7,14,19,23,24}. Instead, mixing is either ignored, enforced in initial conditions, or modeled with a dynamic mix model, e.g., Reynolds-Averaged Navier-Stokes (RANS). As presently implemented, these force mixtures into thermal equilibrium (i.e., they assume negligible equilibrium timescales regardless of how chunky the mixture is). RANS models can be coupled to reaction rate equations\textsuperscript{25} as for reactive flows\textsuperscript{36,27}, e.g., flames, detonations, and combustion, where equilibrium timescales are much shorter than reaction timescales. This works in the absence of reactivity fluctuations\textsuperscript{28}.

However, our results indicate that fluctuations are present and persistent in ICF mixing layers, so that these methodologies are incomplete in the context of ICF. In ICF simulations, temperature gradients cause deviations between DT and DD reaction burn-rate times\textsuperscript{11} and DD reaction burn-weighted ion temperatures (BWTI), yet larger deviations are observed experimentally\textsuperscript{29}. Thermal fluctuations caused by complicated material distributions with unequilibrated contaminants could contribute to this discrepancy. This could also explain why RANS simulations consistently require unphysical adjustments to reproduce the results of separated reactants experiments\textsuperscript{30,31}. Additionally, thermal fluctuations could seed velocity gradients that broaden the neutron spectrum, resulting in artificially high measured ion temperatures\textsuperscript{32}.

Another open question in ICF has been whether hydrodynamic approximation of the true system is sufficient to capture the evolution of contaminant in an ICF hot-spot. Our experiments, which involve complex and evolving material distributions, provide a test bed, and our high-resolution 3D simulations, which are necessary to test this, capture the experimental observables, providing evidence that the hydrodynamic approximation remains valid.

To test the impact of our observation, we performed 2D simulations\textsuperscript{9,33,34} of a high-yield implosion\textsuperscript{11} including the fill tube, a source of chunks of contaminant. In simulation, 160 ng of contaminant mass is injected into the hot spot, and its temperature remains 2.5 keV lower than that of the DT. However, applying experimental methods for calculating contaminant mass using continuum emission from ref.\textsuperscript{5,6} to simulated X-ray self-emission produces an estimate of 80 ng. The discrepancy arises because the model assumes the contaminant is radiating at the temperature of the fuel, which is equivalent to approximately doubling the contaminant emissivity. This effect will also have implications for interpretation of the line emission from contaminant mass. For example, temperature fluctuations such at this will likely be associated with density fluctuations, which have been demonstrated to significantly affect emissivity\textsuperscript{35}.

Our experiments, performed as part of Los Alamos National Laboratory’s (LANL) MARBLE campaign\textsuperscript{36}, are separated reactants experiments in which deuterium resides in contaminant material instead of fuel, so DT reactions occur only as a consequence of atomic mixing between the contaminant and the fuel. Previous separated reactants experiments\textsuperscript{14,30,31,37-41} included deuterium in the shell, whereas our capsules contain open-cell deuterated polyurethane (CH\textsubscript{0.5}D\textsubscript{0.5}) foams\textsuperscript{42-44} whose pores are filled with hydrogen and tritium (HT). This represents the limiting case in which chunks of pre-mixed material exist in
the hot spot before it forms and are given the maximum possible time to atomic mixing and equilibrate with the fuel. Foams are engineered with fixed macro-pores sizes of 30, 50, and 90 μm, in addition to the sub-micron pores that exist throughout the open-cell foam matrix. The macro-pores seed hydrodynamic instabilities, primarily the Richtmyer-Meshkov instability, which induce mixing between the foam and HT. The RMI is known to exhibit sensitivity to and memory of initial conditions both in fluid and plasma regimes. Larger pores are expected to produce mixing that is more “chunky” (and hence produce a lower DT/DD yield ratio) whereas smaller pores produce more atomic mix (hence a higher DT/DD ratio). Ten capsule implosions were directly driven at the Laboratory for Laser Energetics’ (LLE) OMEGA laser facility with an 1Ns square pulse using 26kJ of laser energy; another 12 implosions were performed on a separate shot day that produced consistent results. In simulations, conditions in the burn region are similar to those observed in mixing regions in simulations of current high-yield implosions.

**Simulation details.** Simulations used LANL’s xRAGE, applying methodology for simulating direct drive implosions detailed in ref. 14. xRAGE features adaptive mesh refinement (AMR), making it ideal for resolving the foam pore structure. xRAGE has previously been used to successfully model experiments involving foams. Simulations included shell thickness variations and the target mount. Example initial conditions are visualized in Fig. 1d. The use of 3D was necessitated by foam geometry and hydrodynamically unstable flows. To resolve mixing processes, simulations used a maximum resolution of 0.25 μm. This resolves the Kolmogorov lengthscale. Simulations included shell thickness variations and the target mount. Example initial conditions are visualized in Fig. 1d. The use of 3D was necessitated by foam geometry and hydrodynamically unstable flows. To resolve mixing processes, simulations used a maximum resolution of 0.25 μm. This resolves the Kolmogorov lengthscale. Simulations included shell thickness variations and the target mount. Example initial conditions are visualized in Fig. 1d. The use of 3D was necessitated by foam geometry and hydrodynamically unstable flows. To resolve mixing processes, simulations used a maximum resolution of 0.25 μm. This resolves the Kolmogorov lengthscale. Simulations included shell thickness variations and the target mount. Example initial conditions are visualized in Fig. 1d. The use of 3D was necessitated by foam geometry and hydrodynamically unstable flows. To resolve mixing processes, simulations used a maximum resolution of 0.25 μm. This resolves the Kolmogorov lengthscale.

Several companion experiments were performed to evaluate simulations, including capsule implosions with deuterated methane, deuterated foams with and without hydrogen gas, and CH foams and deuterium gas. Cylindrical experiments were also performed to enable X-ray radiography of shock transit through various foams and pore collapse. For capsule implosions, simulations accurately captured experimental yield and bang time trends. For cylindrical experiments, simulations accurately captured shock and pore dynamics.

**Comparison between simulations and experiments.** The primary observable in experiments is the DT/DD neutron yield ratio, which measures the amount of atomic mixing. Experimental results, including yield ratios and DT burn-weighted T_e (BWTI) from 10 shots, are presented along with results from 3D simulations in Fig. 3. Yield ratios are normalized by

\[
\text{Average 1D simulated yield ratio} = \frac{\text{Average 1D simulated yield ratio}}{\text{Shot 1D simulated yield ratio}} \tag{2}
\]

to account for variation of the laser pulse, fill pressure, and ablator thickness. The experimental results exhibit no significant dependence on pore size. Apparent temperatures from 3D simulations, including fluid motion corrections, are plotted in Fig. 3b and are in good agreement with experiment. If we assume the reactants are in thermal equilibrium, observed yield ratios are consistently higher than the theoretical maximum. We calculate the maximum yield ratio (the uniform atomic mixed limit) assuming thermal equilibrium by dividing the DT reaction rate formula by the DD reaction rate formula and noting that atomic mix maximizes the former and minimizes the latter, hence maximizing the ratio. The resulting theoretical maximum is given by

\[
\frac{2 n_T \sigma_{DT}(T) - n_D \sigma_{DD}(T)}{n_D \sigma_{DD}(T)} = 74 \pm 18, \tag{3}
\]

where n_D and n_T are the number of deuterons and tritons, respectively. This estimate is also consistent with 1D simulations where atomic mixing is enforced in the initial conditions. Our
The HT temperatures are consistently higher than the CH0.5D0.5 temperature. For comparison, we include results that would have been produced by the 3D simulations if the reactants were in thermal equilibrium (labeled “Equilibrated Tion”). We calculated this by integrating the reaction rate equation spatially and temporally over the burn region using the instantaneous burn-weighted Ti to calculate the DT and DD reactivities rather than the local ion temperatures. The resulting yield ratios are below the theoretical maximum and decrease with increased pore size, dominated by reactant spatial distribution.

Simulated mass-weighted temperatures are in Fig. 4 (results for Ti are similar). In all cases, the gas heats more than the foam matrix. The foam temperature peaks later than the HT temperature due to convective heating: vortical flows atomically mix hot HT with colder foam material, heating the latter. The level of separation between temperatures is proportional to pore sizes: simulations with smaller pores exhibit more atomic mixing, enabling local temperature equilibration. The 30 μm pore simulation exhibits the most atomic mix, as quantified by the spatial distribution of HT mass, whereas the 50 and 90 μm pore simulations demonstrate decreasing levels of atomic mix. This behavior is visualized in Fig. 5, where we visualize material distributions at bang time, where it is possible to observe the persistence of larger scale regions of pure HT in implosions with larger pores.

MARBLE experiments underway at the NIF have demonstrated similar trends to the data presented above. With more laser power available to drive larger capsules, it is possible to obtain more implosion data, including X-ray imaging and BWTIs for both reactions. Results exhibit an increase in BWTI with increased pore size consistent with the results in this paper. Results also indicate a small increase in DT/DD yield ratio with increased pore size, consistent with temperature separation between reactants.

Discussion
We evaluated alternative hypotheses for why experimental yield ratios lie above the theoretical maximum, including kinetic effects, anomalous temperatures highly discrepant from the computed values, mischaracterization of targets and/or fill...
resulting in anomalously high tritium concentrations relative to deuterium, and the possibility that the reactants are atomically mixed before bang time. None of these things can plausibly explain the experimental data. Unmodeled kinetic effects are unlikely to have a large enough effect to explain the results because of the high ion densities and carbon presence. As a result, the Knudsen number inside the burn region, which quantifies the validity of continuum approximations, can be estimated as $K_n = \frac{\lambda}{L} \approx 0.001$, where $\lambda$ is the ion mean free path and $L$ is a physical length scale (here, the temperature gradient scale length is used). Nevertheless, kinetic simulations of small regions under conditions relevant to MARBLE implosions show increases to DT fusion rates from non-Maxwellian distributions and relative drifts between species, and decreases from Knudsen-layer losses. These have a complex integrated effect on the results that is currently being explored. Furthermore, these results are not inconsistent with our conclusions. Discrepant temperatures cannot explain the results since, by setting $T_i = 11.6$ KeV into the theoretical maximum given by Eq. (3) to maximize $\sigma_{\text{DT}}(T_i) / \sigma_{\text{DD}}(T_i)$, the resulting yield ratio, 105, is still too low to explain the experimental data. Target mischaracterization would require density or pressure deviations greater than five times the measurement error (quoted in the Methods section) to achieve the experimental results. Finally, if the reactants were completely atomically mixed at bang time, the experimental yields would have to fall within the estimate given in Eq. (3) (which is also consistent with simulations where atomic mix is enforced). However, the data lie well above this estimate.

In summary, we have performed novel separated reactants experiments along with detailed high-resolution 3D simulations, which indicate that species temperature separation is present and persistent at conditions relevant to mixing regions in high-yield ICF experiments. In experiments, we infer this from the ratio of DT/DD neutron yields, which is higher than can be explained when reactants are in thermal equilibrium. Experimental results, including yield ratios, are in agreement with simulations in which species temperature separation is directly observed. Furthermore, simulations and plasma theory indicate that separations persist throughout the burn phase. These results indicate that the amount of contaminant mass in the hot spots of high-yield implosions is routinely underestimated, since methods for estimating this mass assume thermal equilibrium between contaminant and fuel. This suggests that there is significant margin for improving the performance of capsule implosions through decreasing the magnitude of capsule asymmetries or adjusting designs to make them more robust to hydrodynamic instabilities.

### Methods

**Targets.** Capsules consisted of deuterated open-cell polystyrene ($\text{CH}_3\text{D}_n$) foams with 430 $\mu$m radius surrounded by polystyrene (CH) shells with 24 $\mu$m thickness (see Fig. 1). For the present experiments, foams were produced with fixed macro-pore sizes of 30, 50, and 90 $\mu$m. The process for producing the engineered pore foams was developed in ref. 54. The engineered pore foams are constructed by placing hollow spherical SiO$_2$ particles of the desired pore size in an aerogel precursor. Once the matrix has polymerized, the SiO$_2$ particles are chemically leached out. The matrix foam is prepared such that the final bulk density is approximately 30 mg cm$^{-3}$. The matrix itself also includes sub-micron pores. The engineered pores were characterized using X-ray microscale tomography to determine the distribution of pore sizes and void fractions. The void fractions for all foams was measured to be 49 ± 2%. The atomic compositions of the foam were measured using carbon 13 nuclear magnetic resonance spectroscopy, indicating that the foams are 50 ± 1% carbon, 25.5 ± 1% hydrogen, and 24.5 ± 1% deuterium. Density measurements were made gravimetrically using machined samples of the foams with known volumes, and variations among different samples were evaluated from transmission measurements using a narrow-band X-ray source. The bulk density for all foams was measured to be 33.0 ± 0.5 mg cm$^{-3}$. The foams are machined into 860 $\mu$m-diameter spheres and placed inside machined Rexolite step-jointed hemi-shell halves, which were then glued and machined into 900 $\mu$m-diameter spheres, which were then coated with 3 $\mu$m of glow discharge polymer (GDP). This procedure produced capsules with average shell thicknesses ranging from 22.0 to 26.2 $\mu$m and shell thickness variances ranging from 0.5 to 8.7 $\mu$m.

**Experiments.** Experiments were performed at the University of Rochester’s OMEGA laser facility. Capsules were imploded in a direct-drive configuration using all 60 beams of the OMEGA laser, delivering 25 kJ of energy in a 1 ns square pulse. The laser energy delivery was within 3% of the requested value for all shots. Based on the availability of capsules, the implosions included three capsules with 30 $\mu$m pores, five capsules with 50 $\mu$m pores, and two capsules with 90 $\mu$m pores. The capsules were diffusion filled with a requested 10 atm of HT gas, which occupies both the foam macro-pores and the sub-micron pores in the matrix. Due to leakage, the fill pressure at shot time varied from 6.2 to 8.6 atm. Because of the large amount of carbon in the hot spot, which gives the fuel a large heat capacity and allows for significant radiative losses, the experimental yields were very low compared to typical gas-filled capsules imploded on OMEGA, producing yields of only 1.3 × 10$^{14}$ to 2.3 × 10$^{15}$ DT neutrons and 7.8 × 10$^{14}$ to 2.2 × 10$^{17}$ DD neutrons. Because of this, the neutron diagnostic data were too noisy to extract typically reported quantities such as the time-dependent neutron production rate. Bang times (the time of peak neutron production) and BWTI were manually extracted from the data instead of using typical automated processes. An example of the analysis of a neutron time of flight (nTOF) signal is shown in Fig. 6.

**Simulations.** Simulations were performed using Los Alamos National Laboratory’s (LANL) xRAGE radiation-hydrodynamics code. xRAGE solves the Euler and coupled equations in an Eulerian reference frame utilizing square/cubic cells and features AMR, which makes it ideal for performing simulations with a resolved pore structure. The hydrodynamics solver is a custom approximate Godunov-type solver for the Euler equations similar to that of Harten-Lax-van Leer. The radiation transport equations are solved using a multi-frequency gray diffusion approximation. Simulations employ LANL OPLIB opacity data through the TOPS code and LANL SESAME tabular equation of state data. Electron and ion thermal conductivities are based on the formulae of Lee and More with modifications. Laser ray-tracing is performed using the Mazzini laser package, developed by the University of Rochester’s Laboratory for Laser Energetics, which has been integrated into the xRAGE code.
times. In the case of the cylindrical simulations, experiments accurately predicted shock propagation and pore dynamics.

**Data availability**

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Author contributions
B.M.J performed the 3D post-shot simulations and simulation data analysis. R.C.S executed the experiments. J.M.S. performed the pre-shot experimental design simulations as well as simulations of experiments relevant to validating our ability to model these experiments. T.J.M. conceived the idea for these experiments. Y.K. and T.J.M. executed experiments used to validate our ability to model these experiments. T.C., C.E.H., M.N.L., and D.W.S. contributed to target fabrication. K.C.H. and B.M.P. performed measurements to obtain target characterization necessary for simulations. C.F. and V.Yu. performed neutron data analysis for the experiments. B.M.H., R.C.S., J.M.S., A.R.C., and M.D. assisted in understanding the experimental and simulation results as well as performing or analyzing similar experiments that contributed to these experiments. B.J.A. and M.R.D. planned and helped to execute MARBLE experiments. All of the authors contributed to the writing of the manuscript.
Competing interests
The authors declare no competing interests.

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