Plasma Barodiffusion in Inertial-Confinement-Fusion Implosions: Application to Observed Yield Anomalies in Thermonuclear Fuel Mixtures

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The observation of large, self-generated electric fields ($\gtrsim 10^9$ V/m) in imploding capsules using proton radiography has been reported [C. K. Li *et al.*, Phys. Rev. Lett. **100**, 225001 (2008)]. A model of pressure gradient-driven diffusion in a plasma with self-generated electric fields is developed and applied to reported neutron yield deficits for equimolar D^3He [J. R. Rygg *et al.*, Phys. Plasmas **13**, 052702 (2006)] and $(DT)^3He$ [H. W. Herrmann *et al.*, Phys. Plasmas **16**, 056312 (2009)] fuel mixtures and Ar-doped deuterium fuels [J. D. Lindl *et al.*, Phys. Plasmas **11**, 339 (2004)]. The observed anomalies are explained as a mild loss of deuterium nuclei near capsule center arising from shock-driven diffusion in the high-field limit.

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Inertial-confinement-fusion (ICF) capsule implosions are typically modeled as charge-neutral, average-atom, single-component fluids despite their underlying plasma nature [1]. As a result, plasma-related phenomena arising from self-generated electric ($\gtrsim 10^9$ V/m) and magnetic ($\cong 1$ MG) fields may be overlooked in some instances. Although such fields have been recently observed with ≈ 15 MeV proton radiography [2,3], their implications for ICF target performance in general, and upcoming ignition tuning campaigns on the National Ignition Facility (NIF) [4] in particular, have not been established to date.

A potential venue for gauging the importance of plasma fields on ICF capsule performance may arise in the recently reported neutron yield anomaly when ^3He is mixed with deuterium (D_2 or DD) fuel [5] in direct-drive ICF implosions at the OMEGA laser facility [6]. Compared with the performance of pure D_2 [4] and deuterium-tritium (DT) [7] fuels, the observed neutron yields are found to fall short of simulation predictions by nearly a factor of 2 for equimolar D^3He and $(DT)^3He$ atomic mixtures. A number of explanations have been proffered, ranging from equation-of-state anomalies for binary mixtures to atomic mix, preheat, and fuel stratification, but no dominant mechanism has been satisfactorily identified [5,7]. Another example of underperforming ICF implosions is when the fuel is doped with trace amounts of argon to enhance x-ray self-emission for diagnostic imaging of the fuel [8]. Of current interest are planned ignition tuning efforts on the NIF that will rely on ternary isotopic mixtures of hydrogen (THD) fuel. In this Letter we consider the effects of barodiffusion [9], adapted to include the presence of plasma electric fields, as a candidate explanation for the observed neutron yield anomalies in ICF implosions of mixed fuels.

Pressure and temperature gradients generally give rise to component separation in an initially homogeneous gaseous or fluid mixture; see Fig. 1. The one-dimensional mass diffusional flux of the lighter species component in the x direction in the absence of viscous momentum transfer is given by $i_1 = -\rho D(d\alpha/dx + k_p d\ln P/dx + k_T d\ln T/dx) = -i_2$, where ρ is the total mass density, D is the diffusion coefficient, $\alpha \equiv \rho_1/\rho$ is the mass concentration ratio of the lighter ("1") component, P is the total kinetic pressure, T is the temperature, $k_p D$ is termed the barodiffusion coefficient, k_T the thermal diffusion ratio, and i_2 is the mass diffusion flux of the heavier ("2") ion species [9]. The barotropic diffusion ratio k_p is determined by the thermodynamic properties of the mixture, whereas k_T is a function of the ionic or atomic interactions. We ignore the role of k_T in the following analysis, which is often justified when the pressure and temperature gradient scale lengths are comparable [10]. To derive k_p, we first write for the equilibrium ion number density distributions: $n_j \propto \exp(-m_j g x/k_B T - Z_j e\Phi/k_B T)$, where m_j is the ion mass of species $j = 1, 2$, g is the (uniform) acceleration, k_B is Boltzmann's constant, Z_j is the jth ion charge state, $-e$ is the electron charge, and Φ is the electrostatic potential. Adopting ideal equations of state, including the electron pressure, assuming equal ion and electron (uniform) temperatures, and setting the diffusional flux to zero for steady-state equilibrium, $d\alpha/dx + k_p d\ln P/dx = 0$, obtains

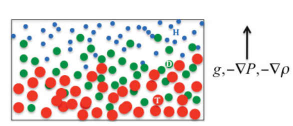

FIG. 1 (color). Schematic of acceleration-driven barodiffusion of a hydrogen (blue), deuterium (green), and tritium (red) fuel mixture.

$$k_p = \alpha(1-\alpha)(m_2-m_1)\frac{[\frac{\alpha(1+Z_1)}{m_1}+\frac{(1-\alpha)(1+Z_2)}{m_2}][1-\frac{eE}{g}\frac{(Z_2-Z_1)}{(m_2-m_1)}]}{\alpha(1+Z_1)+(1-\alpha)(1+Z_2)-\frac{eE}{m_1 g}[\alpha(1+Z_1)Z_1+(1-\alpha)(1+Z_2)Z_2(\frac{m_1}{m_2})]}, \quad (1)$$

where $E = -\nabla\Phi$ is the electric field. To proceed further we consider two cases: (i) an accelerating, isothermal (plasma) atmosphere with a large length-scale, self-generated electric field $eE/m_1 g = O(1)$, and (ii) $eE/m_1 g \gg 1$ as in a shock front.

For an accelerating, isothermal atmosphere in steady state, e.g., an imploding ICF capsule, the self-generated electric field follows first from electron momentum balance (after neglecting the electron inertia): $E = -\nabla P_e/en_e$, where P_e is the total electron pressure and n_e is the total electron number density. But $P_e = P\sum_{j=1,2} Z_j n_j/\sum_{j'=1,2}(Z_{j'}+1)n_{j'}$ for an ideal gas mixture, giving

$$\frac{eE}{m_1 g} = \frac{\alpha Z_1 + (1-\alpha)Z_2}{\alpha Z_1(1+Z_1)+(1-\alpha)Z_2(1+Z_2)m_1/m_2}, \quad (2)$$

where gradients in Z_j are neglected and $\nabla P = -\rho g$ is satisfied. If $Z_1 = Z_2 \equiv Z$, Eqs. (1) and (2) reduce to $k_p = \alpha(1-\alpha)(m_2-m_1)[\alpha/m_1 + (1-\alpha)/m_2](1+Z)$. For all isotopes of hydrogen, the inclusion of ionization increases the barodiffusion by 2× over the nonplasma case ($Z_j = 0$). For D^{3}He, Eqs. (1) and (2) give k_p identically zero, suggesting that barodiffusion in this electric field regime cannot explain the (≅2×) yield deficit in an equimolar D^{3}He fuel compared with a DD or DT fuel.

For the ternary hydrogen isotopic fuel mixtures (THD) proposed for ignition tuning on the NIF [4], we estimate the amount of depleted hydrogen (H) from barodiffusion based on Eqs. (1) and (2). First, we show that barodiffusion ($\propto\nabla\ln P \cong -g/C_s^2$, where C_s is the sound speed) is often comparable in magnitude to or exceeds concentration gradient-driven diffusion ($\propto\nabla\alpha$) in ICF implosions. The latter term is strictly diffusive, scaling as $\alpha/\sqrt{D\tau_{\text{imp}}}$, where $\tau_{\text{imp}} \cong 2R_f/C_s$ is an average implosion time and R_f is the fuel radius at deceleration onset. The diffusion coefficient is derived from Fokker-Planck theory: $D = 9\sqrt{\pi/2}\lambda_{De}^2\omega_{pe}/(G\ln\Lambda)$, where $\lambda_{De} = (k_B T/4\pi n_e e^2)^{1/2}$ is the Debye length, $\omega_{pe} = (4\pi n_e e^2/m_e)^{1/2}$ is the plasma frequency, G is the plasma parameter (or reciprocal number of electrons in a Debye sphere), and $\ln\Lambda$ is the Coulomb logarithm. Forming the ratio η of $|\nabla\alpha|$ to the barodiffusion source term $k_p d\ln P/dx$, gives $\eta = \sqrt{\rho[\text{g/cm}^3]\ln\Lambda/R_f[\text{cm}]}\times 0.21\alpha/k_p g[10^{17}\ \text{cm/s}^2]$. Typically, this ratio is $\approx O(1)$ for hydrogen fuels ($Z = 1$), suggesting that barodiffusion may be important over ICF implosion time scales. A possible deficit of hydrogen near the center of the fuel after deceleration onset ($g > 0$) arising from barodiffusion alone then follows: $i_1 \cong -\rho D k_p d\ln P/dx \cong \rho k_p g D/C_s^2$. The mass fraction of lighter ions $\Delta M_1/M_1$ leaving the fuel volume over the time τ_{imp} scales as $3i_1\tau_{\text{imp}}/\alpha\rho R_f \cong 6i_1/\alpha\rho C_s = 0.032k_p g[10^{17}\ \text{cm/s}^2]\times T[10^4\ \text{eV}]\mu_1^{5/2}/\alpha\rho[100\ \text{g/cm}^3]\times\ln\Lambda$, where μ_1 is the ratio of (light) ion mass to the proton mass. Evaluating for a HT mixture with $T = 7$ keV, $\rho = 20$ g/cm^3, $\ln\Lambda = 7$ when averaged over the inner fuel between deceleration onset and ignition, gives $\alpha = 0.13$ and $k_p = 0.25$, and an ensuing H loss fraction of several percent. For a HD mixture with $\alpha = 0.85$ ($k_p = 0.23$) the relative H mass loss is a scant 0.5%. For a DT mixture with $\alpha = 0.02$ ($k_p \cong 0.02$) the deuterium mass loss is nearly 10%. Interestingly, an equimolar mixture of D and T for ignition capsules gives a mass loss fraction of deuterium of nearly 5%.

In the regime $m_2 \gg m_1$ and $Z_2 \gg Z_1$, potentially significant barodiffusion at small high-Z (dopant) concentrations ($\alpha \cong 1$) is admissible near a mixed-material interface. Figure 2 shows the variation of k_p with α for two cases of interest in ICF: (1) mixing of DT fuel with a Au inner shell in a proposed ignition double-shell target [11] and (2) diagnostic use of trace amounts of argon dopant in a deuterium fuel for elevated x-ray self-emission for imploded core imaging [8]. For double shells, deceleration onset ($g > 0$) may lead to increased barodiffusion of the DT fuel within the Au shell, potentially resulting in higher threshold ignition temperatures. For the case of argon dopants, the DD implosion database for indirect drive is generally marked by ≈3× lower neutron yields than for undoped DD implosions, despite only 0.05 atm of argon out of 10 atm of DD fuel ($\alpha \cong 0.95$) [8]. Interestingly, Fig. 2 shows that ionization and electric fields significantly reduce k_p from the (un-ionized) gaseous case ($Z = 0$), except at small $1-\alpha$ where a predicted maximum is in the vicinity of the experimental conditions. An estimate of $\Delta M_1/M_1$ for this case ($T \cong 1$ keV, $\rho \cong 1$ g/cm^3, $\mu_1 = 2$, $\ln\Lambda \cong 6$) with $k_p \cong 1.2$ [cf. Fig. 2] gives <4%, too small to account for the ≈3× neutron deficit in this electric field regime [case (i)].

We now consider the second case ($eE/m_1 g \gg 1$) germane to a shock front. According to recent data [2] and analysis [12], the self-generated electric field at a shock front arising from electrons diffusing ahead of the ions [10,13] [and not from an underlying acceleration as in case (i)] is on the order of 10^9–10^{10} V/m. Compared with typical peak implosion accelerations of 10^{17} cm/s^2, the ratio $eE/m_1 g$ is on the order of 10^2–10^3 over the shock front. In this limit Eq. (1) simplifies to

$$k_p = \alpha(1-\alpha)(Z_2-Z_1)\left[\frac{\alpha\frac{(1+Z_1)}{m_1}+(1-\alpha)\frac{(1+Z_2)}{m_2}}{\alpha\frac{Z_1(1+Z_1)}{m_1}+(1-\alpha)\frac{Z_2(1+Z_2)}{m_2}}\right]. \quad (3)$$

We note that k_p is nonzero with equal constituent masses

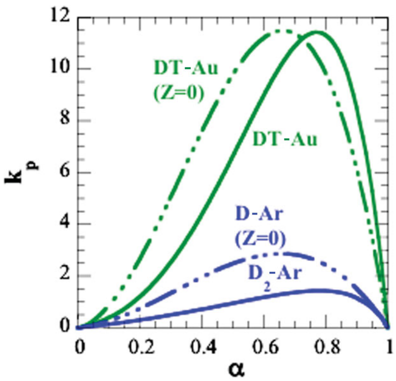

FIG. 2 (color). Barodiffusion ratio k_p versus light ion mass fraction α for a DT-Au mixture with $Z_1 = 1$, $Z_2 = 20$ (green) and a D-Ar mixture with $Z_1 = 1$, $Z_2 = 16$ (blue) based on Eqs. (1) and (2); dot-dashed curves denote cases with no ionization ($Z_1 = Z_2 = 0$).

($m_1 = m_2$). For the case of DD or DT fuel mixtures, k_p is vanishingly small [$Z_1 = Z_2 = 1$; see Eq. (1)], whereas k_p approaches a maximum value of $(\sqrt{2}-1)^2$ at $\alpha = 2-\sqrt{2}$ for D^3He fuels. This strong ionization-dependent scaling [Eq. (3)] provides an intriguing possibility for explaining the observed anomalous behavior of D^3He compared with standard DD or DT fuels, provided the pressure gradient-driven diffusion is strong enough to form a deficit of deuterium near the capsule center. Such necessarily strong pressure gradients are associated with shock fronts, and a rearward deficit of the lighter ion species can result when the shock is nonstationary (and nonplanar) [14] as in ICF.

The preferential diffusion of the lighter constituent in a binary mixture across a shock front is well known [10]. Here, we estimate the depletion of deuterium at the fuel center due to pressure gradient-driven diffusion by (an outgoing) weak shock transit of the fuel [15]; see Fig. 3. The diffusional flux due to barodiffusion alone readily follows: $i_1 \approx \rho D k_p (\Delta P/P)/\Delta x \approx \rho D k_p (\Delta P/P)^2/\ell_{\rm MFP} \approx \rho C_s k_p (\Delta P/P)^2$, where $\Delta x \approx \ell_{\rm MFP} P/\Delta P = \ell_{\rm MFP}[{\rm M}/({\rm M}^2-1)](\gamma+1)/2\gamma$ is the shock width, $\ell_{\rm MFP}$ is the post-shock ion-mean-free path [$\approx O(10\ \mu{\rm m})$ for $T \cong 3$ keV, $\rho \cong 0.1\ {\rm g/cm^3}$ following shock convergence at the center], M is the Mach number, and $\gamma = 5/3$ is the ratio of specific heats [16,17]. The mass of deuterium that has barotropically diffused scales as $i_1 \times 4\pi(\Delta x)^2\tau$, where τ is the (return) shock-transit time over a shock width Δx. The fraction of diffused deuterium ΔM_1 over the enclosed fuel mass M_1 is

$$\frac{\Delta M_1}{M_1} \cong \frac{3k_p}{\alpha {\rm M}^3}\frac{4\gamma^2({\rm M}^2-1)^2}{(\gamma+1)^2}. \qquad (4)$$

For an equal D^3He mixture by number density ($\alpha = 0.4$), $k_p = 0.15$ [cf. Eq. (3)], ${\rm M} \cong 1.2$ from radiation-hydrodynamics simulations, and we estimate from Eq. (4) that $\approx 20\%$ of the deuterium (by mass) at the center of the fuel could be displaced radially outward from barodiffusion. In the limit $\alpha \rightarrow 0$, the leading order α dependence cancels out in Eq. (4), but the outward diffusion of D through the ^3He over a shock-transit time $\Delta x/{\rm M}C_s$ is limited by increased collisions with ^3He ions; i.e., the diffusion length for D over a shock-transit time is nearly $2\times$ shorter than for $\alpha = 1$. The impact of fuel depletion on neutron production can be readily assessed.

A suite of D_2 and D^3He fuel mixtures is considered hydrodynamically equivalent when the number densities (including free electrons) and the ion mass densities are preserved [5]. These two conditions set requirements on the fill (partial) pressures of D_2 and ^3He and the deuterium ion fraction $f_D \equiv n_D/(n_D + n_{^3{\rm He}})$. The hydrodynamically equivalent neutron yield is proportional to $f_D^2/(3-f_D)^2$, while the proton yield scales as $f_D(1-f_D)/(3-f_D)^2$ [5]. In addition, the thermonuclear cross section is a strong function of the temperature, so that the value of the central (peak) temperature largely dictates the neutron yield under similar levels of atomic mix between the shell and fuel materials. The reported ($\cong 2\times$) neutron yield deficit for D^3He is maximized for $f_D \approx 0.5$ [5,7]. Referring back to Eq. (4), $\Delta f_D/f_D = (\Delta M_1/M_1)[1+\alpha(m_2/m_1-1)]^{-1}$ leads to a nearly $\cong 1.5\times$ reduction in neutrons and a departure from hydrodynamic equivalence, representing a large fraction of the observed degradation.

For the D^3He protons, a departure from hydrodynamic equivalence is also seen [5], though considerably milder (by $>2\times$) than for the neutron data and mostly restricted to a higher value of f_D ($\cong 0.8$, giving $\alpha \cong 0.73$). This latter feature may be mostly explained by noting that the proton yield has a theoretical maximum at $f_D = 0.6$ [5], so that a 15%–20% deficit in deuterium at capsule center arising from shock-induced barodiffusion actually *increases* the proton production due to a more optimal helium abundance. Using the fact that $k_p = 0.155$ and the Mach number is elevated by $\approx 5\%$ at this concentration ($\bar{Z} = 1.2$, $\bar{\mu} = 2.2$), an increase in proton yield by more than 31% over strict hydrodynamic equivalence is expected near $f_D \simeq 0.8$.

A final application of the diffusion model in the high-field limit is towards the aforementioned neutron deficit

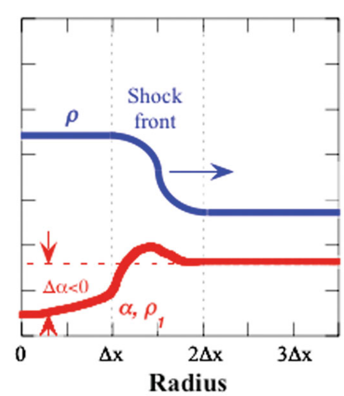

FIG. 3 (color). Schematic of light ion species concentration α (red) and total density ρ (blue) versus radius normalized to the shock-front thickness Δx for an outwardly propagating shock.

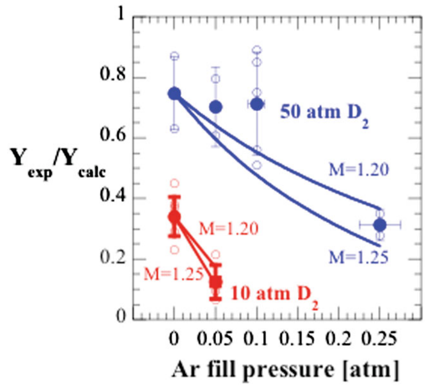

FIG. 4 (color). Ratio of measured to calculated neutron yield in the absence of fuel-pusher mix (''clean'') versus argon fill pressure for 50 atm D_2 fill (blue) and 10 atm fill (red) in plastic capsules [8] (solid points denote average over individual shots shown as open circles; 10 atm data as shown are limited to nominally smooth surface finishes with root-mean-square amplitudes ≤15 nm). Solid curves denote barodiffusion model for indicated Mach numbers M and average argon ionization state $Z_2 = 16$.

observed for argon-doped DD implosions in indirect drive. For these doped fuels, Eq. (3) gives $k_p = 0.52$ for $\alpha \cong 0.95$ and He-like Ar: $Z_2 \cong 16$. The fractional change in depleted deuteron density is $\approx -29\%$ (41%) and the corresponding decrease in neutron yield is ≈2× (3×), using M ≅ 1.2 (1.25) in Eq. (4). Thus, shock-driven barodiffusion is consistent with the routinely observed neutron yield deficit (≅3×) in indirect-drive implosions with argon-doped fuel [8]. A further data set with argon dopant exists for low-convergence implosions (50 atm DD fill) where the dopant level was varied from 0.0–0.25 at. %. Figure 4 shows the trend of the data and the model agreement based on shock-induced barodiffusion.

In summary, a diffusion model based on pressure gradients and adapted to include plasma electric fields and ionization states is developed. A dependence on the plasma electric field is generally found—with two limits of general interest to the study of ICF. The first refers to large length-scale electric fields generated by the electron pressure gradient in an imploding (accelerating), quasi-isothermal capsule. Ionization effects are found to enhance the barodiffusion coefficient by 2× for all (binary) hydrogen isotope fuel mixtures, leading to the potential for elevated fractionation of the fuel. The second case considers the consequences of electric fields generated within a shock front (outwardly) traversing the capsule fuel. The large pressure gradient across a shock front is a natural candidate for driving barodiffusion of the lighter fuel species, e.g., H or D, compared with T, He, or Ar. The model is found to simplify in this case, predicting diffusion even for a binary mixture of equal mass ions, but with differing ionization states. This result directly pertains to a recently observed anomaly where a 50:50 mixture ($f_D = 0.5$) of D^3He was found to produce half the expected neutron yield compared with hydrodynamically equivalent fuels of DD [5] and DT [7]. Between DT and D^3He, the only physical difference is the higher ion charge state ($Z_2 = 2$) of ^3He, which the model naturally distinguishes. Applying the model to an estimate of deuterium depletion at the fuel center for D^3He presents a scenario where much of the yield degradation can be explained. The corresponding surplus of ^3He near capsule center leads to a locally higher electron density (and pressure) after shock flash, potentially resulting in lower fuel compressions and larger x-ray image sizes, as reported in Ref. [7]. The analysis is applied to the argon-doped DD implosion database for indirect drive and is found to largely account for the observed ≅3× neutron deficit. A final application of the model may be found in planned ignition tuning campaigns on the NIF using hydrogen isotope mixtures (THD) where the potential for plasma-mediated isotopic species fractionation exists.

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[15] Prior to shock arrival at the origin, the fuel mixture may not yet be fully ionized and therefore possibly not subject to the plasma effects described herein. In this manner, the neutron yield contribution from shock convergence at the origin or ''shock flash'' is arguably less affected [7].
[16] For the weak shocks ($M - 1 \ll 1$) under consideration here, we are justified in dropping the term proportional to the concentration gradient in Eq. (1), $d\alpha/dx \approx (\Delta P)^3$, compared with the barotropic term, $dP/dx \approx (\Delta P)^2$ [10].
[17] For low Mach-number shocks in a plasma, the shock width is considerably larger than for a fluid shock, scaling as $\ell_{\rm MFP}\sqrt{m_i/m_e}$, where m_i (m_e) is the ion (electron) mass [13].