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 ($\approx 10^9$ 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$^3$He [J. R. Rygg et al., Phys. Plasmas 13, 052702 (2006)] and (DT)$^3$He [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 ($\approx 10^9$ V/m) and magnetic ($\approx 1$ MG) fields may be overlooked in some instances. Although such fields have been recently observed with $\approx 15$ 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 $^3$He 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$^3$He and (DT)$^3$He 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 $i_i = -\rho D (\partial \alpha / \partial x + k_p d \ln P / \partial x + k_T d \ln T / \partial x) = -i_2$, where $\rho$ is the total mass density, $D$ is the diffusion coefficient, $\alpha = \rho_1 / \rho$ 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 theionic 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: $n_j \approx \exp(-m_j g x / k_B T - Z_j e \Phi / k_B T)$, 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 $j$th ion charge state, $-e$ is the electron charge, and $\Phi$ 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, $\partial \alpha / \partial x + k_p d \ln P / \partial x = 0$, obtains

FIG. 1 (color). Schematic of acceleration-driven barodiffusion of a hydrogen (blue), deuterium (green), and tritium (red) fuel mixture.
\[ k_p = \alpha (1 - \alpha) (m_2 - m_1) \frac{[\alpha(1+Z_1) + (1-\alpha)(1+Z_2)] [1 - e^E \frac{m_1}{m_2}] \left( \frac{Z_2 - Z_1}{m_2} \right)}{\alpha(1 + Z_1) + (1 - \alpha)(1 + Z_2) - \frac{e^E}{m_1} \left( \frac{\alpha(1 + Z_1) Z_1 + (1 - \alpha)(1 + Z_2) Z_2}{m_2} \right)} \]  

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): \( E = -\nabla P_e/\epsilon n_e \), 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

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

where gradients in \( Z_j \) are neglected and \( \nabla P = -\rho g \) is satisfied. If \( Z_1 = Z_2 = Z \), 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 \( 2 \times \) over the nonplasma case (\( Z_j = 0 \)). For \( ^3\text{D}^3\text{He} \), Eqs. (1) and (2) give \( k_p \) identically zero, suggesting that barodiffusion in this electric field regime cannot explain the \( (2 \times) \) yield deficit in an equimolar \( ^3\text{D}^3\text{He} \) 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 \( \propto \nabla \ln P \equiv -g/C_2^2 \), where \( C_1 \) 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 \( \alpha / \sqrt{D/C_1} \), where \( D_{\text{imp}} \equiv 2R_{\text{C}} / \alpha \) is an average implosion time and \( R_{\text{C}} \) is the fuel radius at deceleration onset. The diffusion coefficient is derived from Fokker-Planck theory: \( D = 9\sqrt{\pi} \lambda_D^3 \omega_{pe} / (G \ln \Lambda) \), where \( \lambda_D = (k_B T / 4\pi n_e e^2)^{1/2} \) 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 \( \eta \equiv |\nabla \alpha| \) to the barodiffusion source term \( k_p d \ln P / d x \), gives

\[ \eta = \sqrt{g / \ln \Lambda} [\ln \Lambda / R] [g / \ln \Lambda] \times 0.21 \alpha / k_p g [10^{17} \text{ cm} / \text{s}^2]. \]  

Typically, this ratio is \( = O(1) \) 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: \( i_1 \equiv -\rho D k_p d \ln P / d x \equiv \rho k_p \rho D / C_1^2 \). The mass fraction of lighter ions \( \Delta M_1 / M_1 \) leaving the fuel volume over the time \( \tau_{\text{imp}} \) scales as \( 3i_1 \tau_{\text{imp}} / \alpha P_{\text{f}} = 6i_1 / \alpha P_{\text{f}} = 0.032 (k_B g [10^{17} \text{ cm} / \text{s}^2] \times [T / 10^4 \text{ eV}] \mu_1 / \alpha P_{\text{f}} [10^{10} \text{ g} / \text{cm}^3] \times \ln \Lambda) \), where \( \mu_1 \) is the ratio of (light) ion mass to the proton mass. Evaluating for a HT mixture with \( T = 7 \text{ keV} \), \( \rho = 20 \text{ g} / \text{cm}^3 \), \( \ln \Lambda = 7 \) when averaged over the inner fuel between deceleration onset and ignition, gives \( \alpha = 0.13 \) and \( k_p = 0.25 \), and an ensuing H mass 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 \( \alpha = 0.02 \) \( (k_p = 0.02) \) the deuterium mass loss is nearly 10%. Interestingly, an equimolar mixture of D and T for ignition capsules gives a mass loss fraction of deuteron 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 \( (\approx 1) \) is admissible near a mixed-material interface. Figure 2 shows the variation of \( k_p \) with \( \alpha \) 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 implored 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 \( \approx 3 \times \) lower neutron yields than for undoped DD implosions, despite only 0.05 atm of argon out of 10 atm of DD fuel \( (\alpha = 0.95) \) [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 \( (T = 1 \text{ keV} , \rho = 1 \text{ g} / \text{cm}^3, \mu_1 = 2, \ln \Lambda = 6) \) with \( k_p = 1.2 \) [cf. Fig. 2] gives <4%, too small to account for the \( \approx 3 \times \) 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 \( 10^9 - 10^{10} \text{ V/m} \). Compared with typical peak implosion accelerations of \( 10^{12} \text{ cm} / \text{s}^2 \), 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

\[ k_p = \alpha (1 - \alpha) (Z_2 - Z_1) \frac{\alpha (1 + Z_1) + (1 - \alpha) (1 + Z_2)}{\alpha (Z_1 + 1) Z_1 + (1 - \alpha) Z_2} \]  

We note that \( k_p \) is nonzero with equal constituent masses.
diffusion. In the limit of the fuel could be displaced radially outward from barotropically diffused scales as follows:

Here, we estimate the depletion of deuterium at the fuel center due to pressure gradient-driven diffusion by (an outwardly propagating shock) is well known \[10\]. The preferential diffusion of the lighter constituent in a binary mixture across a shock front 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 inward deficit of the lighter ion species can result when the shock is nonstationary (and nonplanar) \[14\] as in ICF.

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A suite of \(D_2\) and \(^3\)He 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 \(^3\)He and the deuterium ion fraction \(f_D = n_D/(n_D + n_{3\text{He}})\). 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 \((\approx 2\times)\) neutron yield deficit for \(^3\)He is maximized for \(f_D = 0.5\) \[5,7\]. 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 \(\approx 1.5\times\) reduction in neutrons and a departure from hydrodynamic equivalence, representing a large fraction of the observed degradation.

For the \(^3\)He protons, a departure from hydrodynamic equivalence is also seen \[5\], though considerably milder (by \(\approx 2\times\)) than for the neutron data and mostly restricted to a higher value of \(f_D\) \((\approx 0.8\), giving \(\alpha \approx 0.73\)). 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 \(f_D \approx 0.8\).

A final application of the diffusion model in the high-field limit is towards the aforementioned neutron deficit of the deuterium being displaced radially outward from barodiffusion. In the limit \(\alpha \rightarrow 0\), the leading order \(\alpha\) dependence cancels out in Eq. \(4\), but the outward diffusion of \(D\) through the \(^3\)He over a shock transit time \(\Delta x/MCs\) is limited by increased collisions with \(^3\)He 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.

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observed for argon-doped DD implosions in indirect drive. For these doped fuels, Eq. (3) gives \( k_p \approx 0.52 \) for \( \alpha \approx 0.95 \) and He-like Ar: \( Z_2 \approx 16 \). The fractional change in depleted deuterium density is \( \approx -29\% \) and the corresponding decrease in neutron yield is \( \approx 2\times (3\times) \), using \( M \approx 1.2 \) (1.25) in Eq. (4). Thus, shock-driven barodiffusion is consistent with the routinely observed neutron yield deficit (\( \approx 3\times \)) 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 fields generated by the electron pressure gradient across a shock front is a natural candidate 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 \( 2\times \) 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\(^3\)He was found to produce half the expected neutron yield compared with hydrodynamically equivalent fuels of DD [5] and DT [7]. Between DT and D\(^3\)He, the only physical difference is the higher ion charge state (\( Z_2 = 2 \)) of D\(^3\)He, which the model naturally distinguishes. Applying the model to an estimate of deuterium depletion at the fuel center for D\(^3\)He presents a scenario where much of the yield degradation can be explained. The corresponding surplus of D\(^3\)He 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 \( \approx 3\times \) 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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