The Effect of Particle Gas Composition and Boundary Conditions on Triboplasma Generation: A Computational Study Using the Particle-in-Cell Method

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Abstract—2-D particle-in-cell simulations of free charge creation by collisional ionization of C12 and C60 molecules immersed in plasma for the parameters of relevance to plasma gasification are presented. Our main findings are that: 1) in uniform plasmas with smooth walls, two optimal values that emerge for free electron production by collisional ionization (i.e., a most efficient discharge-condition creation) are the C60: C12 fractions of 10:90 and 80:20 and 2) in plasmas with rough walls, modeled by the comb-like electric field at the boundary, the case of tangential electric field creates significant charge localization in the C12+ and C60+ species, again creating the most favorable discharge condition for triboelectrically generated plasma. The numerical simulation results are discussed with reference to recent triboelectric plasma experiments and are corroborated by suitable analytical models.

Index Terms—Collision processes, partial discharges, simulation, triboelectricity.

I. INTRODUCTION

WITH the depletion of fossil resources and the mounting quantity of waste, energy generation and waste disposal have become very important problems of modern society. Waste-to-energy (WTE) approaches that generate energy as heat or power from waste can provide a balanced solution to both the problems. One of the most promising WTE technologies is associated with the recuperation of energy by transforming nonrecyclable materials through a combination of different high-temperature-involving procedures such as waste gasification and pyrolysis. The advantages of the above thermal techniques over the conventional WTE techniques such as incineration and combustion include higher recycling rates, lower toxic gas emissions, higher energy efficiencies, lower costs, smaller carbon footprints, and reduced environmental impact [1]. Importantly, gasification converts solid waste into a highly fungible synthetic gas (or syngas) very rich in hydrogen and carbon monoxide, which can be converted into clean electricity or other high-value fuels/chemicals, including methanol, synthetic natural gas (SNG), or pure hydrogen [2]–[4].

The use of plasma power has been popular within thermal-waste treatments for its ability to decompose completely the input waste material into a tar-free synthetic gas and an inert, environmentally stable, vitreous material (slag) and to prepare the syngas for efficient electricity production or catalytic transformation [5]. Because of the potential advantages, plasma technologies have been developed for the destruction and removal of various hazardous wastes, such as polychlorinated biphenyls (PCBs) [6], medical waste [7], metallurgical wastes, incineration fly ash [8], and low-level radioactive wastes.

In addition to waste gasification, plasma-assisted combustion is a very active topic of research on its own right, which covers the topic ignition enhancement, ultralean combustion, cool flames, flameless combustion, and controllability of plasma discharge [9].

Currently, in many engineering applications, plasma have been generated by constant current or electromagnetic field, which needs an external electric energy supply. For example, in the existing plasma-gasification technologies, only additions of combustion heat by the waste feedstock or a fuel additive make the process suitable to large waste streams [10]. The main cost of the current plasma-power technologies is associated with the energy required to create artificially significant electromagnetic or electrostatic fields to trigger and sustain gas discharges. For example, for dry air, a few MVm−1 is required to trigger a corona discharge [11]. This is of the same order of magnitude as the maximum power generated by the conventional high-energy particle accelerators. Such a limit is due to the radio-frequency (RF) breakdown phenomenon: when such a large electric field is used in the accelerator cavities, it causes the accelerator to be effectively short-circuited in accordance with the so-called Kilpatrick limit [12]. Notably, this limit is overcome in the novel accelerators that are based...
on plasmas, the so-called *plasma wake field acceleration*, and that can sustain electric fields up to tens of GV m$^{-1}$, without electric short-circuiting [13], [14].

To trigger the discharge, hence creating plasma, the electrodes powered by direct current are typically used [15]. Altogether, the high cost of conventional plasma generators and short working lifetime of electrodes (circa 500 h) encourage researchers to consider other sources of plasma generation that do not need either the external electromagnetic (EM) field or the electrodes.

Triboelectricity, or electricity generation by *mechanical friction*, can provide such an alternative source of plasma generation. An example of triboelectric charging in nature is the ash produced from the volcanic eruption that collides with one another producing significant charging, which is discharged through the lightning strikes.

Triboelectric plasma generation to replace ultimately the expensive direct-current-operated plasma torches can greatly improve the efficiency of modern WTE-gasification schemes while maintaining a very low emission signature.

In a recent laboratory experiment [16], the generation of plasma through a triboelectric effect was reported by impinging a high-speed (150–200 m/s) microjet of deionized water on a dielectric surface. A naturally formed, stable, unconstrained, and topologically coherent triboplasma region in the form of a coherent toroidal structure was obtained in the atmospheric pressure conditions without any external electromagnetic action.

An example of the triboelectric plasma-generation technology is the gasifier apparatus pioneered by LCC Engineering. In this case, a triboplasma region is generated by the collision of ash particles (mostly carbon-based) in a swirling hydrodynamic flow generated by two tangential flow streams at a moderate flow speed (50 m/s) that grazes a serrated surface of an insulated steel wall. A schematic of the LCC Engineering apparatus is shown in Fig. 1. Here, the organic fuel (e.g., chicken farm waste) is supplied from the top of the reactor and dropped through the triboplasma zone in the center (above the "inductor gasifier" on the schematic). Because of a very high temperature in the center of the reactor chamber, the waste is very efficiently decomposed into useful syngas, ash, and a chemically inert slag, with virtually zero emission of toxic gases. Under the effect of particle–wall and particle–particle collisions, spark discharges are triggered, whose intensity grows as the triboelectric self-charging of gas particles increases until a self-sustained localized triboplasma region emerges in the reaction zone (Fig. 2).

Among several fundamental issues understood before the triboelectric plasma generation can be used in real-life gasification applications, the following two questions stand out.

1) According to the preliminary experimental results of Engineering Company Eco-Ardens, gas samples corresponding to successful triboelectric plasma generation are also rich in fullerenes (C60). Fullerenes are nano-sized, football-like, carbon molecules with a low ionization potential in comparison with the hydrocarbons and a large surface-to-volume ratio in comparison with the macroscopic soot particles. They are known to be readily generated in carbon plasmas through a nonequilibrium growth process that involves the dehydrogenation of hydrocarbons, nucleation of large carbon cages, and carbon cage evaporation to produce the small highly symmetry fullerenes such as C60 or C70. The particular question addressed in this work is what is the effect of fullerenes on triboelectric charging of a gas mixture that includes both C60 and the standard carbon (C12) molecules?

2) The triboelectric charge generation is dependent not only on the particulates present but also on the gas particle collisions with the uneven surfaces of the insulated conducting walls. How do the particle interactions with the wall can lead to an intensification of the triboelectric plasma effect?

To address the above questions, we use the state-of-the-art EPOCH, fully kinetic particle-in-cell (PIC) code for solving the kinetic plasma equations with a self-consistent field formulation [17]. To keep the particle in plasma simulations computationally feasible, a 2-D model of the initially neutral carbon particles, C12 and C60, which are immersed into a fully ionized hydrogen plasma, is considered. In this model, the computational domain is covered by an Eulerian computational grid, where the electromagnetic field equations are solved with capturing the characteristic Debye length. Clusters of neutral and charged carbon species as well as free electrons are represented by the Lagrangian particles that collide with including all relevant collision effects. The collision results in a new charge generation, which is analyzed for different concentrations of C60 and different boundary conditions to simulate the effect in the LCC Engineering experiment.
By analyzing the collisional ionization process, it is shown that the rate of the new charge production from collisional ionization between the carbon particles becomes greatly amplified once the concentration of fullerenes added to the gas exceeds a certain threshold value (circa 10% by number per volume fraction). Additional simulations reveal that the introduction of a nonperiodic boundary condition imitating a serrated conducting wall of the experiment leads to a nonuniform concentration of the carbon particles and enhances the collision process, thereby further enabling the increase of electric charge generation in the volume.

II. MODEL AND RESULTS

A. Methods

Particle collisions and ionization are the key phenomena that affect the triboelectric plasma generation. Hence, we briefly discuss how these effects are implemented in the PIC EPOCH model in the following [17]. It can be noted that many PIC models neglect particle collisions over very short (less than grid scale) ranges. At temperatures ($\gtrsim$1 keV) and number densities ($\lesssim$10$^{27}$ m$^{-3}$), the collisional effects in the plasmas are generally considered negligible. This implies that the mean time between the collisions is comparable with the time scales of interest, and the collisionless approximation used in the PIC codes is valid. However, at lower temperatures and/or higher densities, the effect of subgrid scale interactions on the evolution of the system can become nonnegligible.

The maximum temperature in the plasma gasifiers can reach a few 10$^4$ K (recall that 1 keV corresponds to 1.16 $\times$ 10$^7$ K), i.e., of the order if 1 eV. A typical plasma gasifier density is not readily available, but according to the NRL Plasma Formulary, high-pressure arcs have the number densities of 10$^{22}$–10$^{24}$ m$^{-3}$. Hence, the collisional effects for the gasifier setting are important.

A binary collision algorithm, based on the approach of Sento and Kemp, has been implemented in EPOCH. To simplify the momentum-conservation treatment, collisions are calculated in the center-of-momentum reference frame of the two particles. The Lorentz transformations are included in order to evaluate the particles’ momenta in the center-of-momentum frame. This ensures that in the EPOCH, the collision algorithm is fully relativistic. The EPOCH includes a number of ionization models. These account for the different modes by which electrons ionize in the external field (e.g., of an intense laser) and through collisions. To switch on the collisions and the collisional ionization in the EPOCH, an input file is used (input.deck). Four species included in the simulation are electrons, protons, C12, and C60. C12 has two possible ionization energies: 11.26 and 24.38 eV. The particles are immersed into a fully ionized plasma at temperature $T = 10^5$ K, where the number density of the electrons and protons is set to $n = 10^{15}$ m$^{-3}$.

The end simulation time in most runs where there is no electric field forcing applied at the boundary is set to $t_{end} = 11000/\omega_{pe}$ (Figs. 3–7). In the case of driving the electric field at the boundary (Figs. 9–11) to simulate the wall effect, the end time is longer, $t_{end} = 20000/\omega_{pe}$, to make sure that the solution reaches a more-or-less statistically converged state at least for some of the considered forcing regimes.

First, the simulations are performed in a homogenous domain, without accounting for the effect of the uneven wall of the gasifier. The boundary conditions are periodic in the $x$-direction for both the EM fields and the particles and conducting in the $y$-direction for the fields and reflecting for the particles. Different grid-cell and particle-density resolutions,
as well as the domain sizes, are considered (Fig. 3). The simulation results shown in Figs. 4–8 correspond to the grid resolution of \(288 \times 72\) with each cell being 2 Debye length, i.e., \(\Delta = 2\lambda_D\), with \(\lambda_D = V_{th,e}/\omega_{pe}\). The concentration of C12 and C60 species is varied so that the total density stays the same. For example, the C60:C12 fraction of 99:1 means that \(n_{C60} = n \times 99/100\), while \(n_{C12} = n \times 1/100\), with \(n\) being \(n = 10^{15} \text{ m}^{-3}\) number density for both electrons and protons. Similarly, the C60 : C12 fraction of 25 : 75 means that \(n_{C60} = n \times 25/100\), while \(n_{C12} = n \times 75/100\), and so on.

Second, the case of driving the electric field on the bottom wall boundary is considered. The periodic boundary conditions in the \(x\)- and \(y\)-directions for both the EM fields and the particles are imposed. The latter choice is to make sure that driving of the periodic boundary condition to a prescribed forcing field is fully consistent with the governing discretization of Maxwell’s equations. The results of these simulations are shown in Figs. 9–11, which have a grid resolution of \(144 \times 36\), with each cell being \(\Delta = 4\lambda_D\). The increased cell size is in order to have the same size of the computational domain in most simulation cases, with or without the electric field forcing.

**B. Homogeneous Particle-Interaction Problem**

Before presenting the main results, it is important to make sure that the simulation results are not very sensitive with respect to the numerical parameters of the EPOCH model. This means that by altering: 1) the computational domain size; 2) the grid density; and 3) the number of particles per cell (PPC), the obtained solutions remain reasonably unchanged.

We define the following quantity that is a physical measure of free charge creation by collisional ionization:

\[
R(n_e(t)) = \frac{\int_0^{L_x} \int_0^{L_y} (n_e(x, y, t) - n_e(x, y, 0)) \, dx \, dy}{\sqrt{L_x L_y} \times \int_0^{L_x} \int_0^{L_y} n_e(x, y, 0) \, dx \, dy}
\]  

(1)

where \(n_e(x, y, t)\) is the number density of the electrons, and \(L_x\) and \(L_y\) are the grid lengths in the \(x\)- and \(y\)-directions. Because of the normalization by the initial (at \(t = 0\)) number density of the electrons, effectively, \(R(n_e(t))\) gives the percentage of electrons scaled by a scaling factor of \(1/(L_x L_y)^{1/2}\) to compare the solutions obtained for different ensemble sizes corresponding to different numbers of identical computational cells. The scaling factor comes from considering the collision of particles in cells as a random process in terms of the interaction between different cells of the
computational domain similar to the classical diffusion as discussed in the end of this section.

For the numerical parameter sensitivity study, the case of C60:C12 fraction of 50:50 is selected. For the numerical integration in (1), an interactive data language’s (IDL’s) built-in function is used (INT_TABULATED). This function integrates a tabulated set of data \( \{X_i, F_i\} \) on the closed interval \([\text{MIN}(X), \text{MAX}(X)]\), using a five-point Newton–Cotes integration formula. The implementation is based on introducing an auxiliary array in the EPOCH, which contains \( y \)-array with \( x \)-values integrated out. This is followed by the integration of the \( y \)-dependence in order to obtain a single value of \( R(n_e(t)) \) at a given solution time \( t \).

Fig. 3 (top, middle, and bottom) examines the sensitivity of the numerical solution when gradually changing: 1) the domain size; 2) the grid resolution; and 3) the PPC number. In Fig. 3 (top), the grid unit is \( \Delta = 4 \lambda_D \) and the grid size is increasing by an appropriate factor, e.g., 384 × 96 for the solid line. In Fig. 3 (middle), the domain size remains the same, for example, our standard grid resolution 288 × 72 has \( \Delta = 2 \lambda_D \), while 432 × 108 has \( \Delta = (3/4) \lambda_D \), and commensurately, 576 × 144 has \( \Delta = 1 \lambda_D \).

Fig. 3 (bottom) corresponds to the grid resolution of 144 × 36 with \( \Delta = 4 \lambda_D \) and varied PPC. This lower spatial resolution enabled us to access large PPC values while keeping the simulation cost feasible. It can also be noted that in Fig. 3 not all lines go up to the final dimensionless simulation time; this is because all numerical runs have been limited by 10 day (240 h) wall-time. A typical numerical run used circa 144 processing cores connected with Infiniband Interconnect. In the solutions presented in Fig. 3 (middle and bottom), the factor of \( 1/(L_x L_y)^{1/2} \) from (1) corresponds to different grid densities and varied numbers of particles per cell. The solutions for the four highest grid densities from the 288 × 72 resolution and for all PPC numbers are in good agreement with one another. Fig. 3 (top) shows that solutions for different domain sizes are in reasonable agreement with the theoretical scaling depending on the statistical ensemble size.

All in all, this confirms that the suggested simulation results are reasonably nonsensitive to the numerical parameters of the EPOCH model for the parameter range of interest. In all cases, the free charge created by collisional ionization as a function of time has the same functional behavior that can be explained by a simple analytical linear model, as discussed in the end of this section.

In Fig. 4, we explore the effect of different C60:C12 fractions on free electron production. The fractions are as
follows: 0 : 100, 1 : 99, 10 : 90, 25 : 75, 50 : 50, 75 : 25, and 99 : 1. Fig. 4 (top) shows the time evolution of \( R(n_e(t)) \) for these different fractions. One important aspect immediately seen in this panel is that the absence of C60 yields smallest possible free electron production by collisional ionization. Even adding 1% of C60 markedly changes the situation. Fig. 4 (bottom) shows the \( R(n_e(t=t_{\text{END}})) \) solid line with open circles, \( R(n_e(t=0.75\ t_{\text{END}})) \) dashed line with open diamonds, and \( R(n_e(t=0.5\ t_{\text{END}})) \) dashed-dotted line with open triangles.

From the above results, a local optimum for free electron production by collisional ionization (i.e., a most efficient discharge condition) occurs for the C60 : C12 fraction of 10 : 90. There is a second optimum in the flat part of the distribution, which corresponds to the C60 : C12 fraction of about 80 : 20. Fig. 4 (bottom) shows that the two optima are fairly persistent in the data once the simulation time becomes greater than \( t = 0.5\ t_{\text{END}} \).

Interestingly, the optimum corresponding to 10% of C60 is in broad agreement with the preliminary experimental results of the Engineering Company Eco-Ardens who explored the efficiency of triboelectric plasma generation in their gasifier for relatively small (less than 50%) fractions of C60 in the gas mixture.

Fig. 5 shows the effect of different C60 : C12 fractions on C12+ (singly ionized C12) production by collisional ionization. In contrast to free electrons that are always present because of the background plasma field, there are no C12+ at \( t = 0 \).

Hence, instead of a definition similar to (1), we quantify the C12+ production using

\[
N_{\text{C12}}(t) = \frac{\int_0^{L_x} \int_0^{L_y} n_{\text{C12}}(x,y,t) \, dx \, dy}{\sqrt{L_x L_y}} \tag{2}
\]

where \( n_{\text{C12}}(x,y,t) \) is the number density of C12+.

Fig. 5 (top) shows \( N_{\text{C12}}(t) \) for different C60 : C12 fractions. This quantity increases approximately linearly in time, and different C60 : C12 fractions have different growth rates. The difference in growth rates can be more readily seen in Fig. 5 (bottom), where we plot \( N_{\text{C12}}(t=t_{\text{END}}) \) solid line with open circles, \( N_{\text{C12}}(t=0.75\ t_{\text{END}}) \) dashed line with open diamonds, and \( N_{\text{C12}}(t=0.5\ t_{\text{END}}) \) dashed-dotted line with open triangles.

Similar to the free charge distribution (compared to Fig. 4), there are the same two optima corresponding to C60 : C12 fractions of 10 : 90 and 80 : 20. However, the first maximum (C60 : C12 of 10 : 90) is rather flat, while the second peak is more prominent. Fig. 4 (bottom) shows that the broad and sharp maxima are notable in the distribution for all solution times.

It should be noted that the values attained by \( N_{\text{C12}}(t) \) are much smaller than the number density of the electrons in the plasma \( n_e \) as well as the number of free electrons generated by collisions. For example, by making the following substitution \( n_{\text{C12}}(1)(x,y,t) \rightarrow n_e(x,y,t) \) in (1) for the C60 : C12 fraction of 10 : 90 and taking \( t = t_{\text{END}} \), it follows that \( N_{\text{C12}}(t=t_{\text{END}}) = 1.98 \times 10^{14} \), which gives the number of generated free electrons. By comparison with the maximum value of \( N_{\text{C12}} \) [shown in Fig. 5 (top)], the number of free electrons generated by collisions is three orders of magnitude larger.

Fig. 6 demonstrates the effect of different C60 : C12 fractions on the C12++ (doubly ionized C12) production by collisional ionization. \( N_{\text{C12}}(2)(t) \) is defined by (2) but simply replacing \( n_{\text{C12}}(1)(x,y,t) \) by \( n_{\text{C12}}(2)(x,y,t) \) with the latter being the number density of C12++. Three observations follow from Fig. 6 (top):

1) There is no longer monotonous increase of C12++ production by collisional ionization. Instead, the process proceeds in jumps. For example, for the case of C60 : C12 fraction of 10 : 90, represented by a dashed line, there are two jumps at \( t \approx 4500 \) and 6500.

2) The obtained number densities of C12++ are further three order of magnitude smaller than the singly ionized C12 case, e.g., \( N_{\text{C12}}(2)(t=t_{\text{END}}) = 2 \times 10^8 \) for most cases.

3) There are three peaks in the number density distribution of C12++ as a function of the C60 : C12 fraction. In addition to the C60 : C12 fraction of 10 : 90, two new maxima include C60 : C12 fraction of 50 : 50 and another peak at high fullerene concentrations tending to 100 : 0.

Fig. 7 shows the effect of different C60 : C12 fractions on C60+ (singly ionized C60, i.e., fullerene) production by collisional ionization. Broadly speaking, the behavior of C60+ is similar to that of C12+ shown in Fig. 5, except for: 1) the number density values of C60+ are about 30% larger than that of C12+ and 2) the peak at the C60:C12 fraction of 10 : 90 is more clearly pronounced and the broad peak at high fractions of C60:C12 moves to 100 : 0.

To conclude this section, an analytical ionization model is considered, where the C12 and C60 species are lumped together as the two species (ionized and nonionized) of a particle gas immersed in plasma. Following [18], by introducing constant ionization and recombination parameters \( \alpha = \text{const} > 0 \) and \( \beta = \text{const} > 0 \), assuming that the number
of electrons per unit volume is approximately constant, \( n_e = \text{const} > 0 \) (i.e., the change in the electron number density due to ionization is much smaller than the original electron number density in plasma, \( n_e(t)/n_e(0) \approx 1 \)), and denoting the numbers of ionized and nonionized particles by \( n_1 \) and \( n_2 \), respectively, so that \( n_1 + n_2 = n_0 = \text{const} > 0 \), where \( n_0 \)
is the total number of particles per unit volume that is fixed constant, the evolutionary equation for the ionized particles is given by

\[ \frac{dn_1}{dt} = an_2n_2 - \beta n_2n_1. \]  

(3)

Equation (3) is solved in a periodic spatial domain \( 0 \leq x \leq L_x \), and \( 0 \leq y \leq L_y \) with the initial condition: \( n_1(x, y, 0) = 0 \) and under the constraint that \( n_1 + n_2 = n_0 \).

From the integration of (3) over the control volume \( V = \int_0^{L_x} \int_0^{L_y} dx dy \), one obtains

\[ d(n_1)/dt = aN_e(n_2) - \beta N_e(n_1) \]  

(4)

where \( \langle n_2 \rangle = \langle n_0 \rangle - \langle n_1 \rangle \), which is then substituted into (4) to obtain

\[ d(n_1)/dt = aN_e(n_0) - (\beta + a)N_e(n_1) \]  

(5)

where \( N_e \) is the electron number in the considered control volume \( V \), assuming that the nonlinear process in the big volume leads to an appropriate renormalization of the coefficients \( a \) and \( \beta \). By introducing new notations \( n_1 = \langle n_1 \rangle / \langle n_0 \rangle, a = (\alpha + \beta)N_e \), and \( b = a/(\alpha + \beta) \), (5) simplifies to

\[ d(n_1)/dt = -a(n_1 - b). \]  

(6)

Using the initial condition, the solution for the averaged particle number in the control volume \( V \) is

\[ \langle n_1 \rangle(t) = b(1 - \exp(-at)). \]  

(7)

The above analytical solution can be compared with the predictions of the relative change in the electron number density computed using the EPOCH H code, \( R(n_e) = \langle n_e(|t) \rangle - \langle n_e(0) \rangle \rangle / \langle n_e(0) \rangle \rangle \), which essentially coincides with our definition for \( R(n_e) \) from (1). First, note that the number of ionized particles scales with the number of new electrons generated such that

\[ \langle n_1 \rangle = C \cdot \langle n_e(|t) \rangle / \langle n_e(0) \rangle \]  

(8)

where \( C = \text{const} > 0 \). Hence, \( \ln\langle n_1 \rangle = \ln R(n_e) + \ln C \) and, using (7)

\[ d \ln(R(n_e)) = \frac{d \ln(n_1)}{dt} = \frac{a}{1 - \exp(-at)}. \]  

(9)

For initial times \( at \ll 1 \), (9) can be further simplified using the Taylor expansion that leads to the asymptotic solution as follows:

\[ d \ln(R(n_e)) \approx \frac{1}{t}. \]  

(10)

Fig. 8 shows the comparison of the EPOCH solution with the analytical solution (9) and the asymptotic solution (10). In the case of analytical model (9), the value of parameter \( a \) has been adjusted to obtain the best fit with the EPOCH solution.

It can be noted that, despite some noise present in the EPOCH data due to the numerical differentiation, the analytical solutions based on (9) and especially (10) are in good agreement with the numerical solution.

By recalling the need to explain the scale factor of \( 1/(L_xL_y)^{1/2} \) from (1) mentioned when we discuss on Fig. 3 (top), we next explore the effect of the periodic boundary condition for comparison of the simulation results in different domain sizes.

Let us consider the solution of (5) in a large domain, \( V_{NM} = \int_0^{N L_x} \int_0^{M L_y} dx dy \), where \( N, M > 1 \) are the total number of grid cells and the \( x \)- and \( y \)-direction, respectively. The control volume \( V \), considered in the previous analysis, can be treated as a subset of the large domain. The goal is to compare the particle number density solution (7) obtained in the domain \( V \) and the same averaged over the larger domain \( V_{NM} \). To proceed, the large domain is broken down in several over nonoverlapping subvolumes \( V_{kl} = \int_x^{x+1} \int_y^{y+1} dx dy \), where \( 1 \leq k < N \) and \( 1 \leq l < M \). Each subvolume \( V_{kl} \) is equal to \( V \), but, in comparison with the single-volume case, the particle collision processes in separate subvolumes are largely uncorrelated with one another. The particle numbers averaged over each subvolume \( V_{kl} \) satisfy (6). Each quantity can be treated as random variables, whose evolutionary equations can be treated in analogy with the Langevin diffusion

\[ \frac{dn_1}{dt} = -an_1 + [R] \]  

(11)

where \([R] = ab i\) is the generation term that can be interpreted as a random force and the brackets of the volume averaging in the particle number variable are omitted. Here, \( a = (\alpha + \beta)N_e \), where \( N_e \) is the electron number corresponding to the large domain ensemble. In accordance with the well-known solution of the Langevin equation [19], the variance of the ensemble averaged number of the particles grows as

\[ \langle n^2 \rangle(t) = \left[ \langle n^2 \rangle(0) - \text{Amp}(R)^2/(2a) \right] \exp(-2at) + \text{Amp}(R)^2/(2a) \]  

(12)

where \( \text{Amp}(R) = ab \). Hence

\[ \langle n^2 \rangle(t) - \langle n^2 \rangle(0) \sim \sqrt{\langle n^2 \rangle(0) - ab^2/2} \exp(-2at) + ab^2/2. \]  

(13)

At equilibrium, \( \langle n^2 \rangle(t) - \langle n^2 \rangle(0) \rangle = (a/2)^{1/2}b = (N_e/2)^{1/2} \) and the quantity \( \langle n^2 \rangle(t) - \langle n^2 \rangle(0) \rangle / (N_e)^{1/2} = (a/2)(\alpha + \beta)^{1/2} \) should be independent of the size of the considered system, \( \simeq NM \). Using (8) leads to the following scaling of the simulation results for different size periodic domains

\[ R(n_e) / \sqrt{N_e} = \text{const}. \]  

(14)

Fig. 3 (top) shows the simulation results for different domain sizes. It can be noted that the revealed dependence of the ionized particle solution on the domain size is similar to the so-called “shot noise” effect reported in the startup laser problems [20]. Because: 1) \( N_e \propto NM \propto L_xL_y \) and 2) all lines for the different domain sizes in Fig. 3 (top) are tolerably close to each other, the scale factor of \( 1/(L_xL_y)^{1/2} \) from (1) is justified based on our Langevin equation solution.
C. Particle Interaction With Including the Nonhomogenous Wall Condition

The EPOCH results describing how surface roughness affects free charge generation by collisional ionization are presented next. Instead of including actual material rough walls in the simulation, computationally, it is much easier to impose periodic electric field on the domain boundary. Indeed, rough surfaces alter the electric field in the vicinity of the solid boundaries and the imposition of a nonuniform electric field boundary condition along with the periodic condition on the particles is equivalent to considering a small internal volume of the particle domain at some distance away from the material walls. It can be reminded that enforcing of the periodic condition is important for consistency with Maxwell’s equations.

In the EPOCH code, the boundary condition on the electric and magnetic field dynamics is implemented via a subroutine called fields.f90 (see [17] for more details). The following target electric fields at \( y = y_{\text{min}} = 1 \) are considered (\( x \) is tangential to the wall boundary and \( y \) is the normal direction):

1) \( E_x(x,t) = f(x,t) \) and 2) \( E_y(x,t) = f(x,t) \), where

\[
f(x,t) = E_0 \times \left( \exp \left( -\frac{x - 0.2x_{\text{max}}}{x_{\text{max}}/15} \right) + \exp \left( -\frac{x - 0.4x_{\text{max}}}{x_{\text{max}}/15} \right) + \exp \left( -\frac{x - 0.6x_{\text{max}}}{x_{\text{max}}/15} \right) + \exp \left( -\frac{x - 0.8x_{\text{max}}}{x_{\text{max}}/15} \right) \right) \\
\times \left[ 1.0 - \exp \left( -t/(10/\omega_{\text{pe}}) \right) \right]
\]

(15)

and where \( E_0 = 10^7 \text{ V m}^{-1} \).

The boundary condition at \( y = y_{\text{min}} = 1 \) is driven to the target field (15) so that in about \( t_{\text{to}} = 10 \), a steady-state electric field with an amplitude of \( E_0 \) is reached. Such a driving essentially imposes a comb-like electric field with four spikes at the locations of 0.2, 0.4, 0.6, and 0.8 fractions of the computational domain size in the \( x \)-direction.

Fig. 9 shows the simulation results for the case of electric field component normal to the boundary. Fig. 9 (top) demonstrates the electric field \( x \)- and \( y \)-components, and Fig. 9 (bottom) shows the number densities of C12+ (singly ionized C12) and C60+ (singly ionized C60) at the final simulation time \( t_{\text{to}} = 20\,000 \). The length scale units are based on the plasma frequency and the light speed.

In Fig. 9 (top), the electric field gradients are very localized and moderately penetrate in the domain interior. The “hot spots” that emerge in Fig. 9 (bottom) represent the charged ions of the relevant species. These species are relatively rare and more-or-less scattered over the whole domain.

Fig. 10 shows the simulation results for the case tangential to the \( x \)-direction electric field. Fig. 10 (top) shows the electric field \( x \)- and \( y \)- components, and Fig. 10 (bottom) should number the densities of C12+ and C60+ at the final simulation time \( t_{\text{to}} = 20\,000 \). Two important observations from Fig. 10 include the following: 1) \( E_x \) now protrudes into the simulation domain much deeper than in the case of normal electric field driving and the “flames” of the electric field gradient are much wider and 2) the “hot spots” of C12+ and C60+ are clustered in the middle of the simulation domain at \( y = y_{\text{max}}/2 \).

**Fig. 11.** \( R(n_e) \) for the normal and tangential electric field driving cases. Solid line is for the case of normal and dashed for the case of tangential electric field driving.

An important observation from Fig. 11 is that the charge localization effect triggered by the tangential electric field boundary condition means that additional carbon particles, which could be introduced in the “reaction zone” in the center of the computational domain, would further enhance the collisional discharges and lead to a denser triboelectrically created plasma. Indeed, the charge localization effect is the triboelectric plasma generation scenario as suggested by the Engineering Company Eco-Arden’s experimental results (compared with Fig. 2). In these experiments, the additional carbon particles were brought in the triboplasma reaction zone by the pyrolysis products. In comparison with the tangential electric field boundary condition, the normal electric field has no significant effect on the localization of particle charging. Hence, at least for the simulation run times attempted in this study, this other regime is not of interest from the point of view of triboelectric plasma generation.

Let us consider a two-dimensional domain with periodic boundary conditions in the \( x \)- and \( y \)-directions. In comparison with the model considered in the previous section, in the present case, the boundary problem is not homogeneous: the top and bottom boundaries in the \( y \)-direction correspond to conducting walls. On the walls, a periodic variation of the tangential electric field component is imposed \( E_x = E_x(x) \). In accordance with the EPOCH solution (Fig. 10), the electric field penetrates inside the domain and its effect decays away.
from the wall. To proceed with the analytical solution, let us model the effect of the nonhomogeneous electric field on the ionized particle distribution by adding a diffusion term to the linear particle collision model equation (3). At equilibrium, ∂n1/∂t = 0 and the equation for the particle number per unit volume becomes

\[ \alpha_n n_2 - \beta n_1 n_1 + D \frac{\partial^2 n_1}{\partial y^2} = 0 \]  

(16)

where \( D = \text{const} > 0 \). Let us discretize the solution domain into several nonoverlapping bins in the \( y \)-direction, where the coordinates of each bin are \( 0 \leq x \leq L_x, \ h \leq y \leq h + \delta h, \ 0 < h < L_y \). By integrating (16) over each bin volume, one obtains

\[ \alpha N_e (n_2) - \beta N_e (n_1) + \left( D \frac{\partial^2 n_1}{\partial y^2} \right) = 0 \]  

(17)

where the brackets mean averaging over the bin volume. After a rearrangement, using \( n_1 + n_2 = n_0 \) and \( \langle n_1 \rangle = \langle n_1 \rangle / \langle n_0 \rangle \), \( a = (a + \beta) N_e \) and \( b = a/(a + \beta) \), and (17) reduces to

\[ \left( D \frac{\partial^2 n_1}{\partial y^2} \right) = a \langle n_1 \rangle - b. \]  

(18)

By introducing some effective average diffusion coefficient \( D \), the last equation can be integrated to obtain

\[ \langle n_1 \rangle = b + A \exp \left( \frac{a}{\sqrt{D}} y \right) \]  

(19)

where \( A = \text{const} \) is an amplitude parameter to be determined, e.g., from the boundary condition.

At small distances from the bottom wall, \( (a/\sqrt{D})^{1/2} y \ll 1 \), and (19) reduces to be integrated to obtain

\[ \langle n_1 \rangle = C + E y \]  

(20)

where \( C = b = \text{const} \) and \( E = A(1 + (a/\sqrt{D})^{1/2}) = \text{const} \).

To close the model, the slope parameter \( E \) in (20) can be related to the tangential electric field using the particle continuity and the electrostatic force equations as follows. Let us consider the continuity equation for the number of ionized particles in a unit volume at equilibrium

\[ \frac{\partial (u_x n_1)}{\partial x} + \frac{\partial (u_y n_1)}{\partial y} = 0. \]  

(21)

Here, \( u_x \) and \( u_y \) are the effective \( x \)- and \( y \)-velocity components. The particle velocities are driven by the nonhomogeneous electric field. By integrating (21) over the considered control volume close to the wall, to the first order, one obtains

\[ \frac{d \langle n_1 \rangle}{dy} \approx -\langle n_1 \rangle y_0 \frac{\partial (u_x)}{\partial x} / U. \]  

(22)

Equation (22) can be reduced to the form of (20), where brackets correspond to the volume averaging and \( E = -\langle n_1 \rangle y_0 \frac{\partial (u_x)}{\partial x} / U \), which can be treated as constant to the first approximation. Let us further approximate the particle velocity corresponding to their drift away from the wall by a constant value \( u_y = U > 0 \) and consider that the average number of particles does not depend on the \( x \)-coordinate. To evaluate \( \langle \partial (u_x) / \partial x \rangle \) that appears as the slope, \( E \), one can recall that the acceleration exerted on a charged particle due to the electric field is given by

\[ a_x(x, y) = q E_x(x, y) / m \]  

(23)

where \( q \) is the particle charge and \( m \) is its mass, and using the standard kinematic relationships

\[ u_x = \int_0^t a_x(x, y) dt = \int_0^x u_x(x, y) dx \]  

and \[ \frac{\partial u_x}{\partial x} = a_x(x) \]  

(24)

the integration over \([0, x]\), after some rearrangement, leads to

\[ u_x(x, y) = \sqrt{\int_0^x a_x(x, y) dx + u_x^2(0, y)}. \]  

(25)

Hence

\[ \frac{d u_x}{d x} = \frac{1}{2} \frac{a_x(x, y) - a_x(0, y)}{\sqrt{\int_0^x a_x(x, y) dx + u_x^2(0, y)}} \]  

(26)

or

\[ \frac{d u_x}{d x} = \frac{q}{2m} \frac{\Delta E_x}{\int_0^x (q/m) E_x(x, y) dx + u_x^2(0, y)}. \]  

(27)

where \( \Delta E_x = E_x(x, y) - E_x(0, y) \).

Analytical model (20) can be compared with the output of the EPOCH simulations that were provided in the form of the bin-averaged electron number normalized by the peak value \( \langle n_e \rangle(y)/\langle n_e \rangle_{\text{max}} \) as a function of the \( y \)-coordinate. It can be first noted that \( \langle n_e \rangle \) = \( \langle n_e \rangle_{\text{max}} \) and \( (\langle n_e \rangle - \langle n_e \rangle_{\text{max}})/\langle n_e \rangle_{\text{max}} \) ≪ 1 in accordance.

Hence, \( \ln(\langle n_e \rangle(y)/\langle n_e \rangle_{\text{max}}) \propto (\langle n_e \rangle - \langle n_e \rangle_{\text{max}})/\langle n_e \rangle_{\text{max}} \). The latter quantity is compared with (20) in Fig. 12, where the two parameters of the linear model \( C \) and \( E \) were selected from the best fit to the EPOCH data. The good agreement between the fully kinetic plasma solution and the analytical model suggests that the assumptions used in the model are reasonable for the triboelectric plasma-generation regime of interest.
In this article, we present the PIC simulations of free charge creation by the collisional ionization of C12 and C60 particles in plasma for the parameters of relevance to plasma gasification. For plasma simulations, a fully collisional EPOCH model is used and the obtained solutions are reasonably non-sensitive to the numerical parameters such as the grid resolution, the domain size, and the PPC number. There are two regimes considered: with and without excitation of the nonuniform electric field on the boundary. Our main findings are as follows.

1) In uniform plasmas with smooth walls, there appear to be two optimal values of C60: C12 fraction for free electron production by collisional ionization (i.e., a most efficient discharge condition creation): one is 10:90 and the other is 80:20. The first value is in agreement with the experimental results of LCC Engineering who performed gasification tests with relatively low fullerene concentrations.

2) In plasmas with rough walls, modeled by comb-like electric field distribution at the boundary, the case of tangential electric field creates a significant charge localization in the C12+ and C60+ species. This leads to the most favorable discharge condition creation for triboelectrically generated plasma.

3) Linear analytical models are presented for modeling the particle collision process. Predictions of the models are in an encouraging agreement with the numerical simulation results.

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