ELECTRONIC SUPPLEMENTARY INFORMATION (ESI)

Time-resolved Image Velocimetry and 3D Simulations of Single Particles in the New Conical ICP Torch

Xiaoman Guo, ^{ab} Sina Alavi, *a Elham Dalir, a Jingmin Dai, b and Javad Mostaghimi *a

^a Department of Mechanical and Industrial Engineering, University of Toronto, Toronto, M5S 3G8, Canada

^b Department of Automatic Measurement and Control Engineering, Harbin Institute of Technology, Harbin, 150001, P. R. China

* Corresponding authors: salavi@mie.utoronto.ca, mostag@mie.utoronto.ca, mostag@mie.utoronto.ca, mostag@mie.utoronto.ca, mostag@mie.utoronto.ca, mostag@mie.utoronto.ca, mostag@mie.utoronto.ca)

Table of Content:

S1. Governing Equations and Boundary Conditions

S1. Governing Equations and Boundary Conditions

A three-dimensional (3D) model was developed using the ANSYS-FLUENT software to simulate the ICP and compare the performance of the new and Fassel torches in terms of fluid flow, heat transfer, electromagnetic field effects, trajectory of particles, and so on. Assuming the ideal gas fluid flow to be Newtonian, in a steady-state condition, and laminar, the Navier-Stokes equations can be written as follows:

$$\frac{\partial}{\partial x}(\rho v_x) + \frac{\partial}{\partial y}(\rho v_y) + \frac{\partial}{\partial z}(\rho v_z) = 0$$

- Momentum:

~1

$$\rho\left(v_{x}\frac{\partial v_{x}}{\partial x}+v_{y}\frac{\partial v_{x}}{\partial y}+v_{z}\frac{\partial v_{x}}{\partial z}\right)=-\frac{\partial p}{\partial x}+\frac{\partial}{\partial x}\left(\mu\frac{\partial v_{x}}{\partial x}\right)+\frac{\partial}{\partial y}\left(\mu\frac{\partial v_{x}}{\partial y}\right)+\frac{\partial}{\partial z}\left(\mu\frac{\partial v_{x}}{\partial z}\right)-F_{d,x}+F_{L,x}$$

$$\rho \left(v_x \frac{\partial v_y}{\partial x} + v_y \frac{\partial v_y}{\partial y} + v_z \frac{\partial v_y}{\partial z} \right) = -\frac{\partial p}{\partial y} + \frac{\partial}{\partial x} \left(\mu \frac{\partial v_y}{\partial x} \right) + \frac{\partial}{\partial y} \left(\mu \frac{\partial v_y}{\partial y} \right) + \frac{\partial}{\partial z} \left(\mu \frac{\partial v_y}{\partial z} \right) - F_{d,y} + \rho g_y + F_{L,y}$$
 S3

$$\rho\left(v_x\frac{\partial v_z}{\partial x} + v_y\frac{\partial v_z}{\partial y} + v_z\frac{\partial v_z}{\partial z}\right) = -\frac{\partial p}{\partial z} + \frac{\partial}{\partial x}\left(\mu\frac{\partial v_z}{\partial x}\right) + \frac{\partial}{\partial y}\left(\mu\frac{\partial v_z}{\partial y}\right) + \frac{\partial}{\partial z}\left(\mu\frac{\partial v_z}{\partial z}\right) - F_{d,z} + F_{L,z}$$

Also, assuming the viscous dissipation to be negligible, the energy equation can be written as:

$$\rho\left(v_{x}\frac{\partial h}{\partial x} + v_{y}\frac{\partial h}{\partial y} + v_{z}\frac{\partial h}{\partial z}\right) = \frac{\partial}{\partial x}\left(\frac{k\,\partial h}{c_{p}\partial y}\right) + \frac{\partial}{\partial z}\left(\frac{k\,\partial h}{c_{p}\partial z}\right) + \sum_{i}\left(\frac{\partial}{\partial x}\left(\rho h_{i}D_{i,m}\frac{\partial Y_{i}}{\partial x}\right) + \frac{\partial}{\partial y}\left(\rho h_{i}D_{i,m}\frac{\partial Y_{i}}{\partial y}\right) + \frac{\partial}{\partial z}\left(\rho h_{i}D_{i,m}\frac{\partial Y_{i}}{\partial z}\right)\right) + Q_{JH} - Q_{R}$$
S5

In these relations, ρ , μ , k, and c_p are the density, dynamic viscosity, thermal conductivity, and specific heat capacity, respectively. Also, p, v, and h are the pressure, velocity, and enthalpy, respectively, while x, y, and z denote the components of the coordinate system as shown in Figure S1. The gravitational acceleration g_y was also assumed to be toward the negative y direction. The sink terms $F_{d,x}$, $F_{d,y}$, and $F_{d,z}$ also account for the momentum exchange between the continuous phase (i.e., plasma) and the discrete phase (i.e., graphite particles injected into the plasma).

In addition, the species transport equation was included in the model to account for the influence of surrounding air on argon plasma. The conservation equation for species can be written as follows:

$$\rho\left(v_{x}\frac{\partial Y_{i}}{\partial x}+v_{y}\frac{\partial Y_{i}}{\partial y}+v_{z}\frac{\partial Y_{i}}{\partial z}\right)=\frac{\partial}{\partial x}\left(\rho D_{i,m}\frac{\partial Y_{i}}{\partial x}\right)+\frac{\partial}{\partial y}\left(\rho D_{i,m}\frac{\partial Y_{i}}{\partial y}\right)+\frac{\partial}{\partial z}\left(\rho D_{i,m}\frac{\partial Y_{i}}{\partial z}\right)$$

where Y_i is the mass fraction of the i^{th} species and $D_{i,m}$ is the diffusion coefficient of the i^{th} species in the mixture. The diffusion coefficient of each species (here only air and argon) was calculated using the kinetic theory for which the Lennard-Jones parameters were taken from Hirschfelder et al.¹ To account for the transport of enthalpy due to diffusion of species, additional terms were added to the energy equation. These terms appear in Equation S5 in the form of a summation including the enthalpy h_i and mass fraction Y_i of various species. Aside from density which was calculated based on the ideal gas law, the other temperature-dependent properties (i.e., c_p , k, and μ) were taken from Boulos et al.² and attributed to each computational cell using the mass-weighted mixing law. As shown in Figure S1, a mass-flow-inlet boundary condition (B.C.) with a constant temperature was applied to the (carrier, outer, and intermediate) gas inlet ports of the torch. It can be seen that inlet port for the outer gas (and intermediate gas for the Fassel torch) is modeled according to reality to be able to introduce the gas tangentially and simulate the swirl effects inside the torch. The torch walls were considered to have a no-slip condition, while the pressure-outlet B.C. was applied to all the free boundaries of the computational domain. All the B.Cs. are schematically demonstrated in Figure A1.



Figure S1. Schematics of the 3D computational domain and boundary conditions for simulating the ICP torch.

The Maxwell equations were added to FLUENT in the form of user-defined functions (UDF) and scalars (UDS). For this purpose, the displacement currents associated with the oscillating magnetic field were neglected and the plasma was assumed to be globally neutral. Therefore, the Maxwell equations can be written as follows:

$$\nabla \cdot E = 0 \tag{S7}$$

$$\nabla \cdot B = 0$$
 S8

$$\nabla \times E = -\frac{\partial B}{\partial t}$$

$$\nabla \times B = \mu_0 (J_{coil} + J_{ind})$$
S10

$$V \times B = \mu_0 (J_{coil} + J_{ind})$$
S10
S11

where E, B, and μ_0 are the electric field intensity vector, magnetic flux density vector, and vacuum magnetic permeability, respectively. Also, J_{coil} denotes the vector of current density applied to the load coil as a source term, whereas J_{ind} is the vector of current density induced in the plasma by the load coil. The values of electrical conductivity σ of the plasma at various temperatures were taken from Boulos et al.² Since inside the torch tube is dominantly filled with argon, the effect of air on the electrical conductivity was neglected. For solving the Maxwell equations, Mostaghimi and Boulos ³ suggested the vector potential method based on the following expression:

 $J_{ind} = \sigma E$

$$B = \nabla \times A$$
 S12

wherein A is the magnetic vector potential. Assuming that the scalar potential is zero due to absence of an electrostatic charge in the plasma, the electric field can be expressed as the time derivative of vector potential (i.e., $E = -\partial A/\partial t$). Therefore, by combining Equations S9 through S12, and using vector identities, we can write:

$$\nabla^2 A - \mu_0 \sigma \frac{\partial A}{\partial t} = -\mu_0 J_{coil}$$

If it is assumed that A is characterized by a sinusoidal time variation with frequency f, we can write the following relation for A using complex notation:

$$A = \tilde{A} e^{i\omega t}$$
 S14

where \mathbf{A} is the phasor and $\omega = 2\pi \mathbf{f}$. In a 3D coordinate system, the phasor will have 3 spatial components as $\mathbf{A} = \mathbf{A}(\mathbf{A}_x, \mathbf{A}_y, \mathbf{A}_z)$. Therefore, using Equation S14, Equation S13 can be rewritten as:

$$\frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2} + \frac{\partial^2 A}{\partial z^2} - i(\mu_0 \sigma \tilde{\omega} A) = -\mu_0 \tilde{J}_{coil}$$
S15

Using this method, time, t, is effectively removed from the electromagnetic field equations. Considering that the phasor has x, y, and Z components and that each component has both the real and imaginary parts (i.e.,

 $\mathbf{\tilde{A}} = (\mathbf{A}_{x,re}, \mathbf{A}_{y,re}, \mathbf{A}_{z,re}) + i(\mathbf{A}_{x,im}, \mathbf{A}_{y,im}, \mathbf{A}_{z,im})$ with $i^2 = 1$), Equations 15 is in fact 6 equations which should be separately solved.

To account for the effects of Lorentz forces, source terms $F_{L,x}$, $F_{L,y}$, and $F_{L,z}$ were added to the momentum equations (Equations S2 – S5) in the ^x, ^y, and ^z directions, respectively. Two more source terms were added to the energy equation (Equation S6) to account for the effects of Joule heating Q_{JH} and radiative losses Q_R . Assuming the plasma to be optically thin and in a local thermal equilibrium (LTE) condition, radiative losses were approximated using the net emission coefficients for pure argon.² The values of Joule heating and Lorentz forces were calculated as follows:

$$Q_{JH} = \frac{1}{2}\sigma(\tilde{E} \cdot \tilde{E}^*)$$
 S16

$$F = (F_{L,x}, F_{L,y}, F_{L,z}) = \frac{1}{2}\sigma R[\tilde{E} \times \tilde{B}]$$
S17

The superscript $\square *$ denotes the complex conjugate and R shows the real part of the complex variable.

As shown in Figure S1, the load coil was considered to be helical to study the associated asymmetrical effects on the shape of plasma and particle trajectory. To solve Equation S15, \tilde{J}_{coil} should be provided to the program as an input. However, it would be more practical to provide a value for power instead of current. Since power is not present as a variable in Equation S15, this equation

was modified by dividing both sides to the magnitude of current density J_{coil} (i.e., $J_{coil} = \sqrt{J_{coil,x}^2 + J_{coil,y}^2 + J_{coil,z}^2}$) and rewritten as:

$$\frac{\partial^2 a}{\partial x^2} + \frac{\partial^2 a}{\partial y^2} + \frac{\partial^2 a}{\partial z^2} - i(\mu_0 \sigma \tilde{\omega} a) = -\mu_0 (\tilde{J}_{coil} / J_{coil})$$
S18

where $a = A/J_{coil}$. The term J_{coil}/J_{coil} would be essentially a unit vector revolving around the coil axis while being tangent to the coil turns at any given point. Using this change of variable, the electric field intensity can also be expressed as:

$$\tilde{E} = -i\omega J_{coil} \tilde{a}$$
 S19

Using this relation and based on Equation S16, the following expression can be written for Joule heating:

$$Q_{JH} = \frac{1}{2}\sigma(\tilde{E} \, \tilde{.E}^{*}) = \frac{1}{2}\sigma\omega^{2}(\tilde{A} \, \tilde{.A}^{*}) = \frac{1}{2}\sigma\omega^{2}J_{coil}^{2}(\tilde{a} \, \tilde{.a}^{*})$$
S20

In each computational cycle, the values of ^a were first obtained by solving Equation S18. Based on the extended field approach,^{4, 5} the computational domain was taken large enough to be able to set the vector potential equal to zero on all the outer boundaries of the domain. As a result, the far-field boundary conditions for Equation S18 could be written as $\tilde{a} = 0$ (Figure S1). Next, the values of \tilde{a} were integrated over all the computational cells to calculate the coil current density J_{coil} , using the following relation:

$$J_{coil} = \sqrt{\frac{Q_{coil}}{\frac{1}{2}\omega^2 \sum \left[\sigma(\tilde{a} \cdot \tilde{a}^*)\forall\right]}}$$
S21

in which Q_{coil} is the applied power to the coil—provided as input to the program by user—and \forall is the volume of each computational cell. The value of J_{coil} found in this manner could then be used to calculate electric field intensity, magnetic flux density, and, eventually, the values for Lorentz forces and Joule heating source terms. Based on a mesh independence study, it was found that a mesh resolution equivalent to ~ 1000 cells/mm³ is sufficient and optimum for all the calculations. With this resolution, a typical CPU time for each case was around 1 - 3 days using 4 threads of an Intel Core i7-6700 processor in parallel in addition to 16 GB of RAM.

References

- 1. J. O. Hirschfelder, C. F. Curtiss, R. B. Bird and M. G. Mayer, Molecular theory of gases and liquids, Wiley New York, 1954.
- M. I. Boulos, P. Fauchais and E. Pfender, *Thermal plasmas: fundamentals and applications*, Springer Science & Business Media, 2013.
- 3. J. Mostaghimi and M. I. Boulos, Plasma Chem Plasma Process, 1989, 9, 25-44.
- 4. D. Bernardi, V. Colombo, E. Ghedini and A. Mentrelli, *The European Physical Journal D-Atomic, Molecular, Optical and Plasma Physics*, 2003, 27, 55-72.
- 5. S. Xue, P. Proulx and M. I. Boulos, J. Phys. D: Appl. Phys., 2001, 34, 1897.