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HECA Research Conference - November 15th 2022

Location: Griffith College Dublin

Simulations of Plasma Species during liquid-plasma interaction in Two-Phase Flow at Atmospheric Pressure

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Agenda

- Introduction to two-phase flow and PlasmaStream system
- Coupled Fluid-droplet model for PlasmaStream atmospheric pressure jet system
- Results of Fluid-droplet model
- Liquid precursors (Water, TEOS and HMDSO) in He-N_2 gas mixture
- Effect of distinct liquid precursors and gas flow rates
- Distribution of charged species in APP
- Comparison of Modelling and Experimental size distributions
- Conclusions

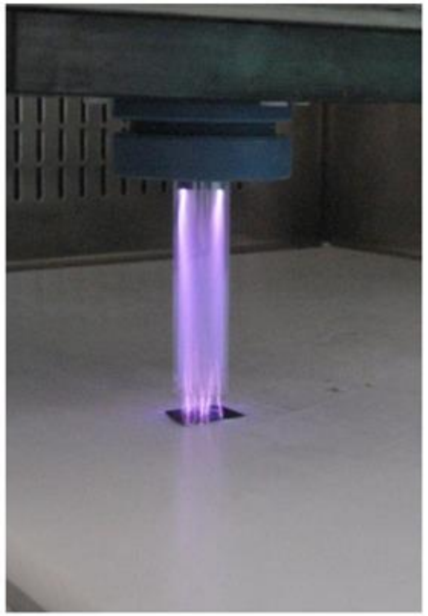
Introduction to Atmospheric Pressure Plasmas

PSAJD System

- Understand the characteristics of liquid-plasma interaction in the ***PlasmaStream atmospheric pressure jet deposition system (PSAJD)*** during downward transport and find the operating conditions under which it will be useful for various applications, such as medicine, tissue engineering and surface deposition of materials.

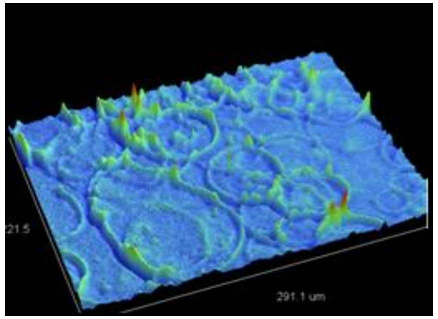
- How to solve this problem using numerical modelling as well as computer programming?

- Which mathematical model would be most appropriate to describe the essential characteristics of plasma in the PlasmaStream System?

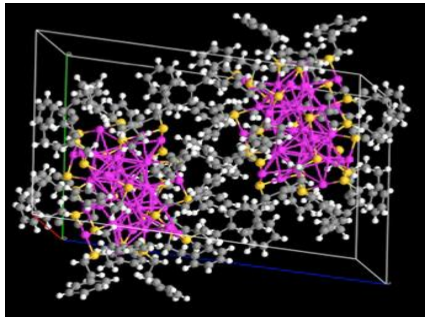


Plasma stream atmospheric pressure jet deposition system

Coalescence of Droplets

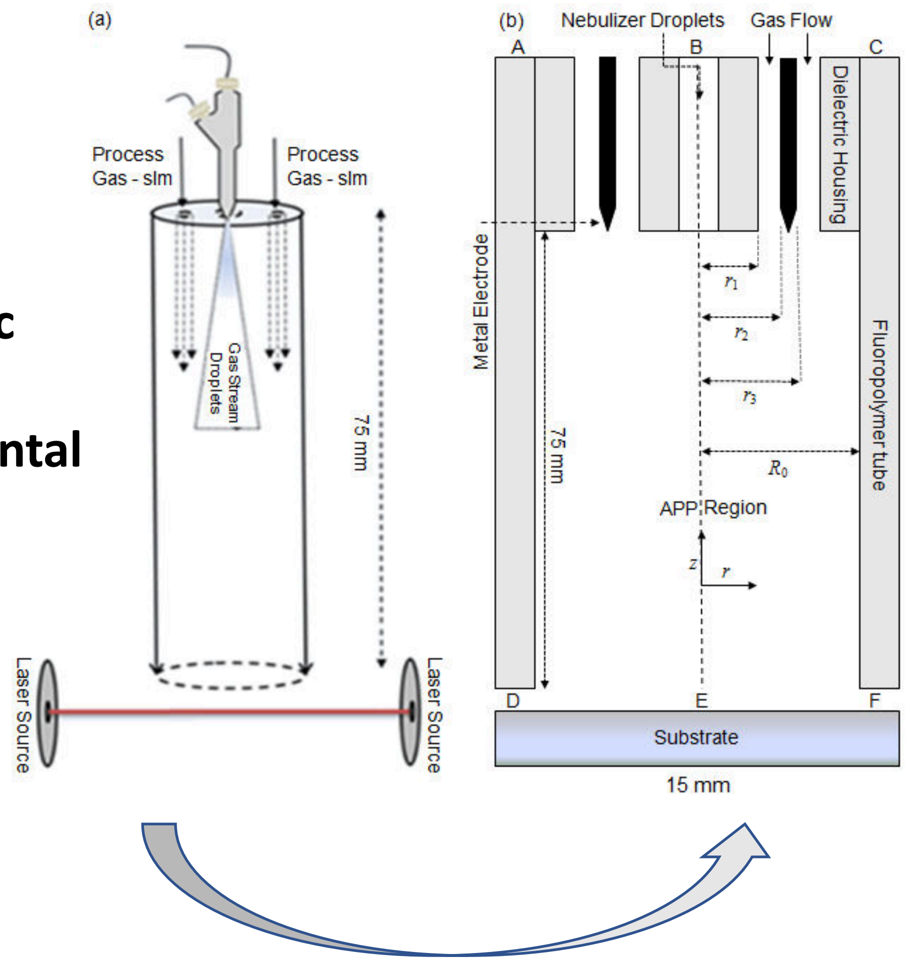


Nucleation of Nanoparticles



Mathematical Model for Plasma Chamber

Schematic view of experimental chamber



- Fluid Model for Atmospheric Pressure Plasma
- Stochastic Liquid Droplet Model for transport of droplets in the PlasmaStream system
- Challenge is to model two-phase flow using an approach to couple both models in order to describe the important characteristics of APP for industrial applications.

Fluid Model Equations for Plasma

Continuity equation for discharge species

$$\frac{\partial n_\varepsilon(r,z,t)}{\partial t_2} + \vec{\nabla}.\left(\vec{\Gamma}_\varepsilon(r,z,t) + \varepsilon n_e \vec{u}(r,z,t)\right) = S_\varepsilon$$

Species Flux is given by momentum balance equation using D-D approximation as

$$\vec{\Gamma}_{sp}(r,z,t) = \mathrm{sgn}(q_{sp})\mu_{sp}\vec{E}n_{sp} - D_{sp}\vec{\nabla} n_{sp}$$

Electron mean energy is evaluated from the following equation as

$$\frac{\partial n_\varepsilon(r,z,t)}{\partial t_2} + \vec{\nabla}.\left(\vec{\Gamma}_\varepsilon(r,z,t) + \varepsilon n_e \vec{u}(r,z,t)\right) = S_\varepsilon$$

where $n_\varepsilon = n_e\bar{\varepsilon}$

The electric field is calculated by the Poisson's equation as

$$\vec{\nabla}.(\varepsilon_0 \vec{E}(r,z,t)) = -\sum_{sp} q_{sp} n_{sp}(r,z,t)$$

The Dirichlet and Von Neumann Symmetry Boundary Conditions can be expressed as

$$V_a = \left.\begin{cases} V_0 \sin(2\pi f t) & \text{at driven electrode} \\ 0 & \text{at grounded electrode} \end{cases}\right\} \quad \frac{\partial n_p(x,y,z,t)}{\partial x} = \frac{\partial n_p(x,y,z,t)}{\partial z} = 0, \quad \frac{\partial V_a(x,y,z,t)}{\partial x} = \frac{\partial V_a(x,y,z,t)}{\partial z} = 0$$

The flux of discharge species at the electrode and grounded substrate can be considered as

$$\mathbf{\Gamma_p}.\mathbf{n} = (2\beta - 1)\,\mathrm{sgn}(q_p)\mu_p \mathbf{E}_p^{eff}.\mathbf{n}n_p(x,y,z,t) + \frac{1}{2}v_{th,p}n_p(x,y,z,t)$$

$$\mathbf{\Gamma_e}.\mathbf{n} = -(2\beta - 1)\mu_e \mathbf{E}.\mathbf{n}\, n_e(x,y,z,t) + \frac{1}{2}v_{th,e}n_e(x,y,z,t) - 2(1-\beta)\sum_p \gamma_p \boldsymbol{\Gamma}_\mathbf{p}.\mathbf{n}$$

$$\mathbf{\Gamma_\varepsilon}.\mathbf{n} = -(2\beta - 1)\frac{5}{3}\mu_e \mathbf{E}.\mathbf{n}n_\varepsilon(x,y,z,t) + \frac{2}{3}v_{th,e}n_\varepsilon(x,y,z,t)$$

$$\mathbf{\Gamma_n}.\mathbf{n} = \frac{1}{2}v_{th,n}n_n(x,y,z,t)$$

$$\beta = \begin{cases} 1 & \mathrm{sgn}(q)\mu\mathbf{E}.\mathbf{n} > 0 \\ 0 & \mathrm{sgn}(q)\mu\mathbf{E}.\mathbf{n} \le 0 \end{cases}$$

The discharge current density is evaluated by the following relation as

$$j = \frac{e}{d}\int_0^d (\Phi_i - \Phi_e)dx + J_D, \text{ where } J_D = \varepsilon_0 \frac{\partial V_a}{\partial t}$$

Coupled Fluid-Droplet Model

Discrete parcel method for the solution of droplet equation

- **Eulerian Method** (For APP phase)
- **Lagrangian Method** (For parcels of liquid precursor droplets)

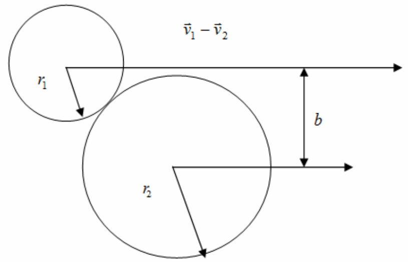
Motion of a single parcel of droplets

$$\left.\begin{aligned}
\frac{d\vec{x}_p}{dt_1} &= \vec{u}_{\mathrm{p}} \\
\frac{d\vec{u}_p}{dt_1} &= \vec{F} \\
\frac{dr_p}{dt_1} &= \left(\frac{\lambda_{\mathrm{g}}}{\rho_1 c_{\mathrm{p_g}}}\right)\left(\frac{Nu_{\mathrm{g}}}{2r_{\mathrm{p}}}\right)\left(\frac{Y_{V_{\mathrm{s}}} - Y_V}{1 - Y_{V_{\mathrm{s}}}}\right) \\
\frac{dT_p}{dt_1} &= \left(\left(\frac{3\lambda_1 Nu_1}{2\rho_1 c_1 r_{\mathrm{p}}^2}\right) + 3\frac{dr_p/dt_1}{r_{\mathrm{p}}}\right)\left(T_{\mathrm{s}} - T_{\mathrm{p}}\right)
\end{aligned}\right\}$$

Set of equations for the modelling of plasma gas as

$$\left.\begin{aligned}
&\frac{\partial \rho_{\mathrm{g}}}{\partial t_1} + \rho_{\mathrm{g}} \vec{\nabla} . \vec{u}(r,z,t) = S \\
&\frac{\partial(\rho_{\mathrm{g}} \vec{u}(r,z,t))}{\partial t_1} = \vec{\nabla} P + \vec{\nabla} . \Gamma_{\mu} \nabla \vec{u}(r,z,t) + S_{\mathrm{M}} \\
&\frac{\partial(\rho_{\mathrm{g}} h)}{\partial t_1} = \vec{\nabla} . \frac{k}{C_{\mathrm{p}}} \vec{\nabla} h + \vec{j} . \vec{E} + S_{\mathrm{E}} \\
&\rho_{\mathrm{g}} \frac{\partial Y}{\partial t_1} = \vec{\nabla} . D_{\mathrm{AB}} \vec{\nabla} Y + S
\end{aligned}\right\}$$

b (Impact parameter) $< b_{cr}$ (Critical impact value)



$$b_{cr}^2 = (r_1 + r_2)^2 E_{coal}$$

Two types of collision processes are considered for droplets

- ***Grazing*** (droplets change positions and velocities after grazing)
- ***Coalescence*** (two droplets merge into single droplet)

Impact parameter $b = (r_1 + r_2)\sqrt{YY}$

Critical impact parameter $b_{\mathrm{crit}} = (r_1 + r_2)\sqrt{E_{\mathrm{coal}}}$

$b < b_{\mathrm{crit}}$

Development of Numerical Solvers

- **Successive Over Relaxation (SOR)** Solver for Poisson Equation
- **Modified Strongly Implicit (MSI)** Solver for Plasma Species (electrons, ions, excited species and dimers)
- **Alternating direction implicit (ADI)** solver for electron mean energy
- ***C++*** programming language is used to write the code for the implementation of these solvers in order to simulate liquid-plasma interaction.
- ***MATLAB/ OriginLab*** employed to plot the graphs for the simulations.
- ***Input data*** is based on the reaction rates of various chemical processes.
- ***Output*** generated data is stored as tsv (tab separated format).

```
Array3D<real> SevendiagonalSolver(const Array3D<real> &ae1,
                                  const Array3D<real> &aw1,
                                  const Array3D<real> &af1,
                                  const Array3D<real> &ac1,
                                  const Array3D<real> &an1,
                                  const Array3D<real> &as1,
                                  const Array3D<real> &ab1,
                                  const Array3D<real> &At1)
```

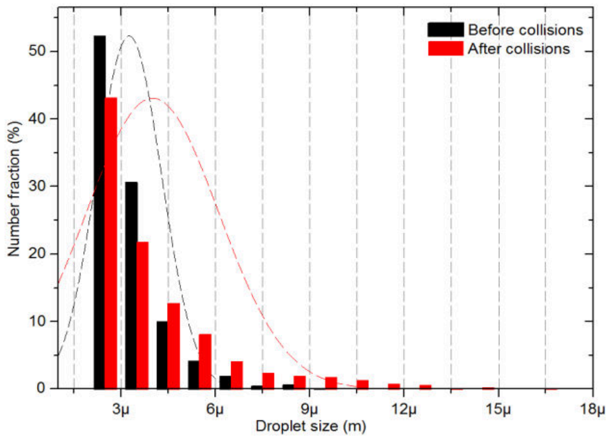
```
Array<real> TridiagonalSolver(const Array<real> &a,
                              const Array<real> &b,
                              const Array<real> &c,
                              const Array<real> &r)
```

```
LinearInterpolator::LinearInterpolator(const Array<real> &x,
                                       const Array<real> &y)
```

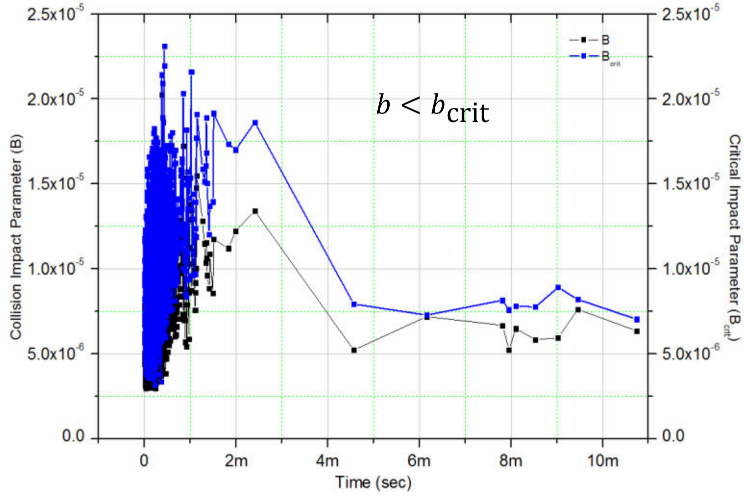
Spatio-temporal Distributions

Fluid-droplet Model using H_2O

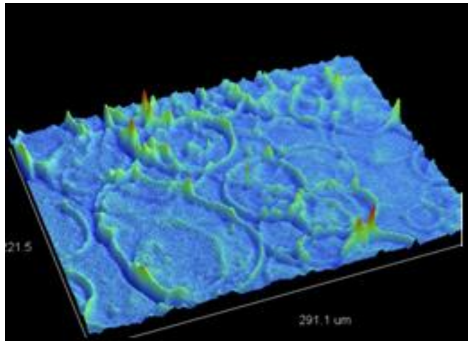
Droplet count and size distributions at 0.4 msec after coalescence of droplets



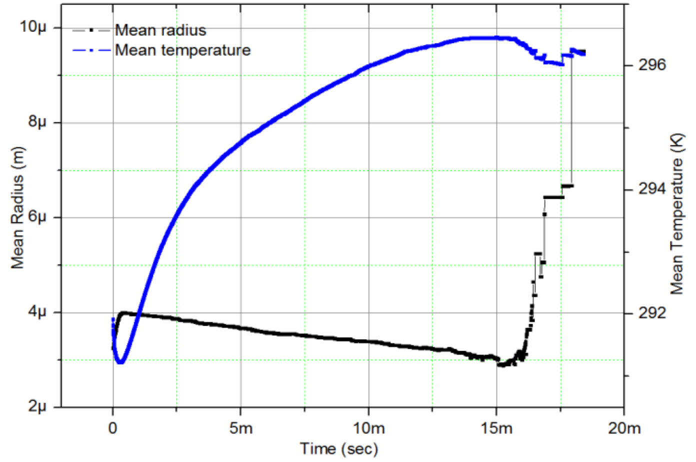
Coalescence events during downward travelling



Initial velocity distribution of droplets is not available, we applied different initial velocities of droplets to understand the behaviour of water droplets in APP.



Temporal profiles of mean radius and temperature

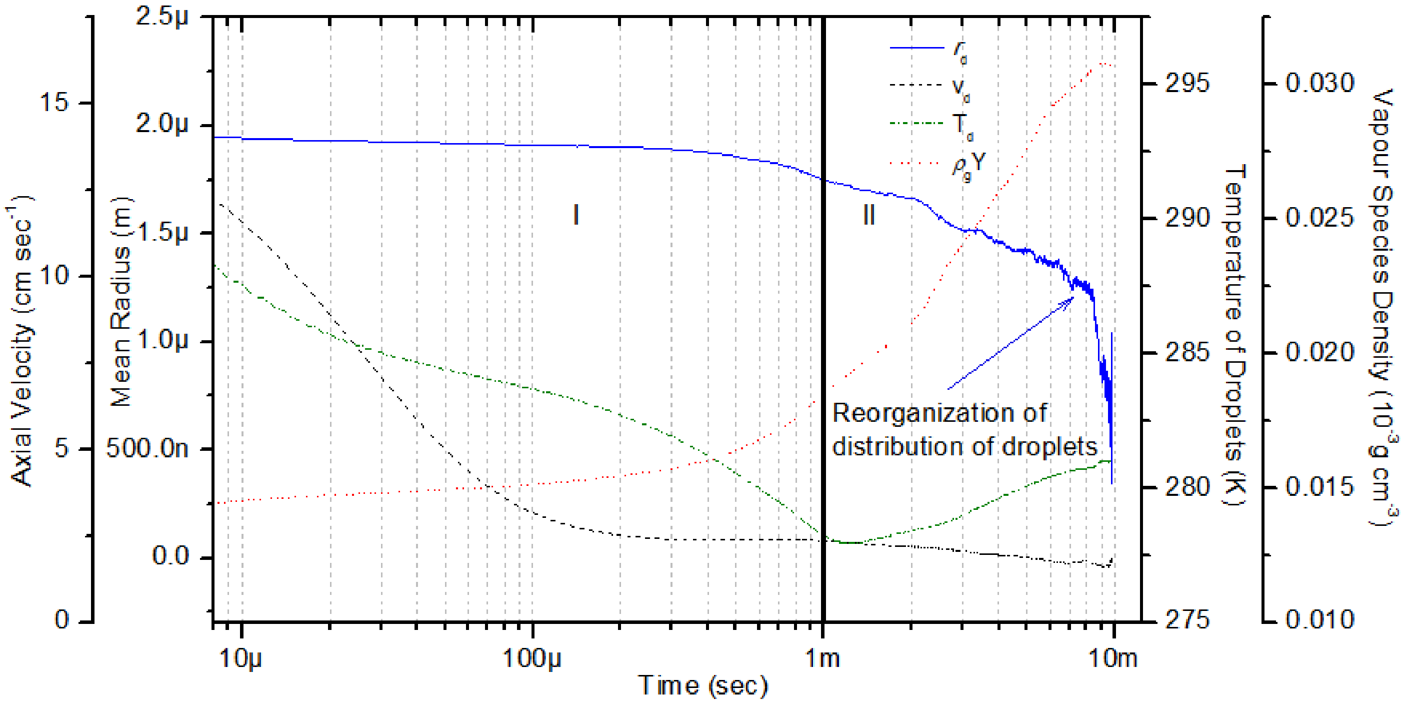


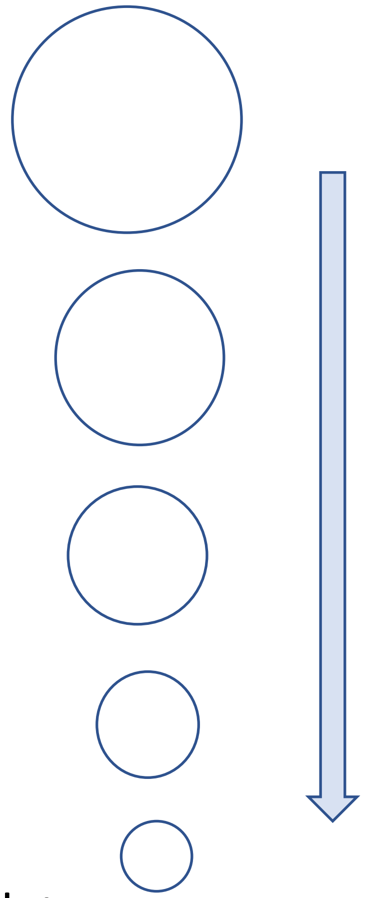
Droplet Flow Rate: **100 μL min^{-1}**
Gas Flow Rate: **5 L min^{-1}**

V_{appl} = -13.5 kV, f = 20 kHz in He-N_2 gas

Spatio-temporal Distributions

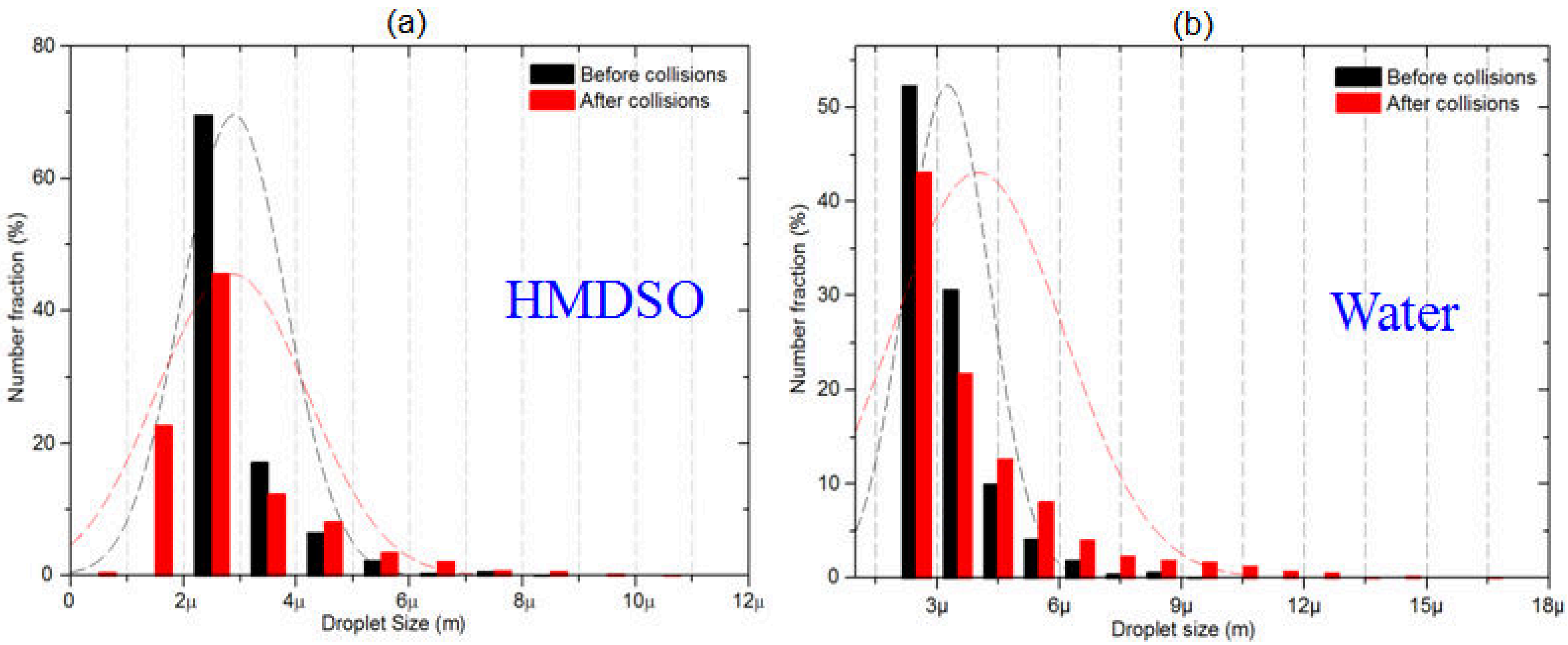
Fluid-droplet Model using HMDSO





- Average profiles of droplet radius (**solid line**), droplet axial velocity (**dashed line**), droplet temperature (**dash-dotted line**) and mean vapour species density (**dotted**) of parcels using HMDSO precursors at flow rate = 100 mL/min, gas flow rate = 5 L/min , f = 20 kHz and V_0 = -13.5 kV.

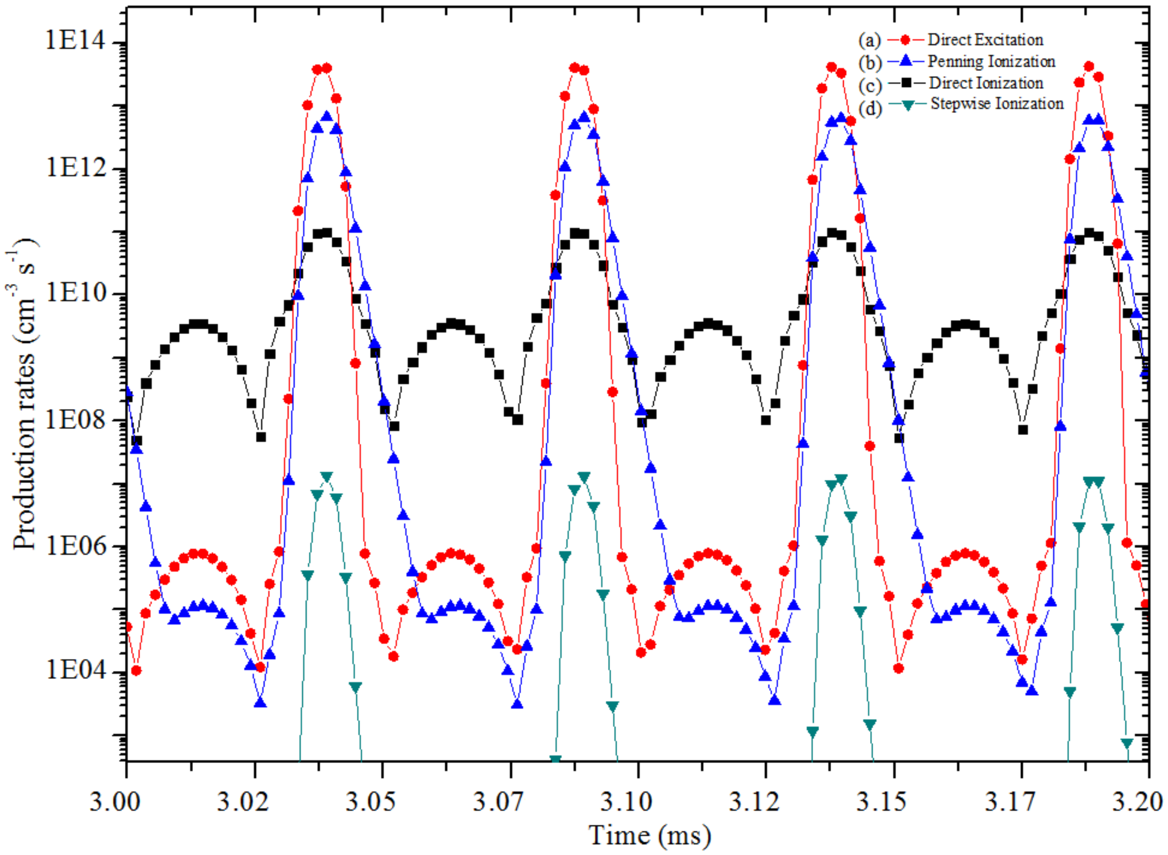
Coalescence of Droplets in APP



- Size distribution of the droplets before collisions (black bars at 0 msec) and after collisions (red bars at 1 msec) for (a) **HMDSO** and (b) **Water** at precursor flow rate = 100 μL min^{-1} and gas flow rate = 5 L min^{-1}, f = 20 kHz and V_0 = −13.5 kV in He-N_2 gas mixture.
- **Behaviour of coalescence is dominant in Water as compared to HMDSO.**

M. M. Iqbal, M. M. Turner, "Investigations of Droplet-Plasma Interaction using Multi-Dimensional Coupled Model", Contrib. Plasma Phys., Volume 55, Issue 9, pages 627 – 642, October 2015.

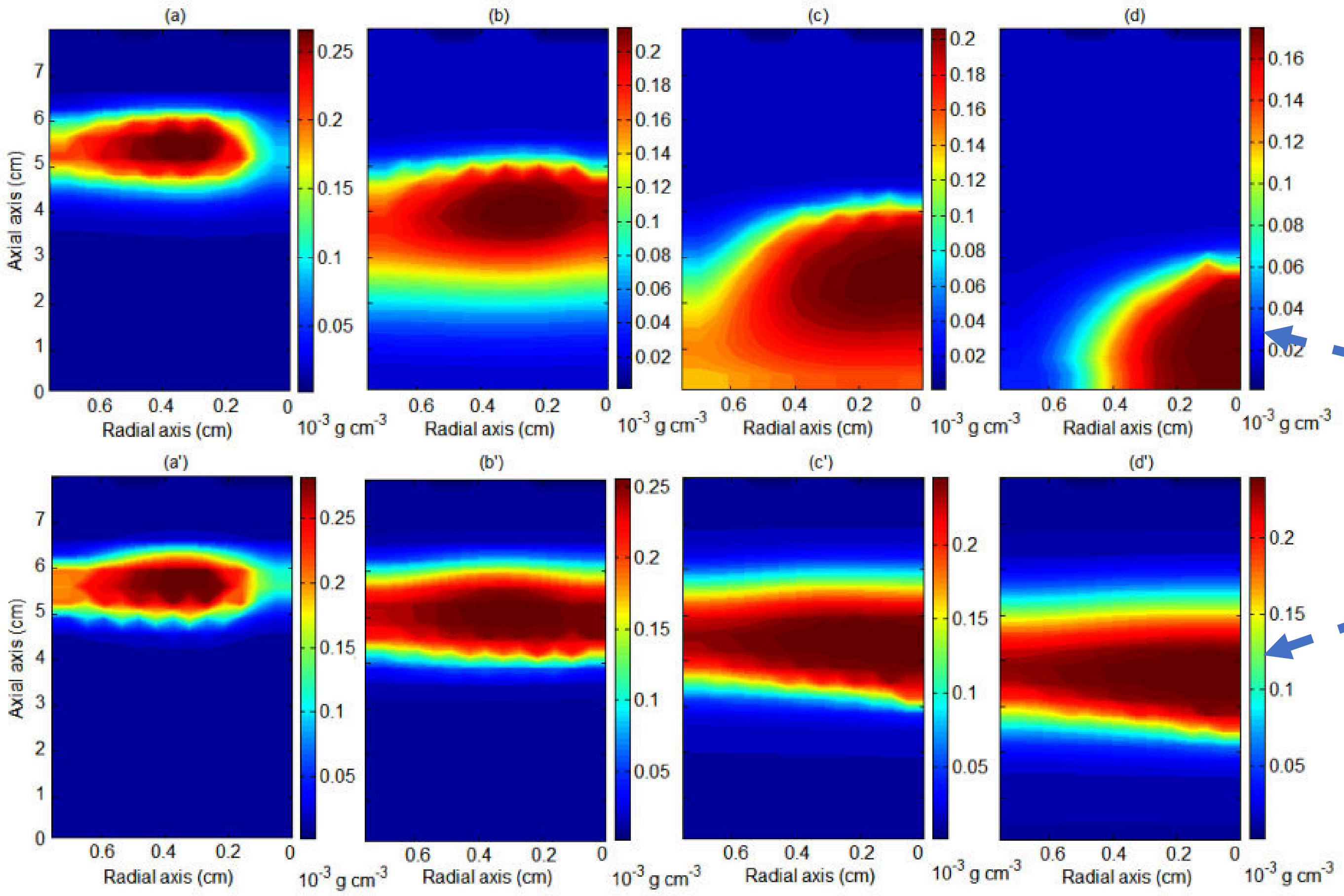
Penning Ionization Role in Liquid-Plasma



Penning ionization has been recognized as dominant as compared other ionization processes.

- Temporal evolution of line-averaged distribution of **Direct excitation (DE), Penning ionization (PI), Direct ionization (DI)** and **Stepwise ionization (SI)** rates using water droplets at precursor flow rate = 50 μL min^{-1} and gas flow rate = 5 L min^{-1}, f = 20 kHz and V_0 = −13.5 kV

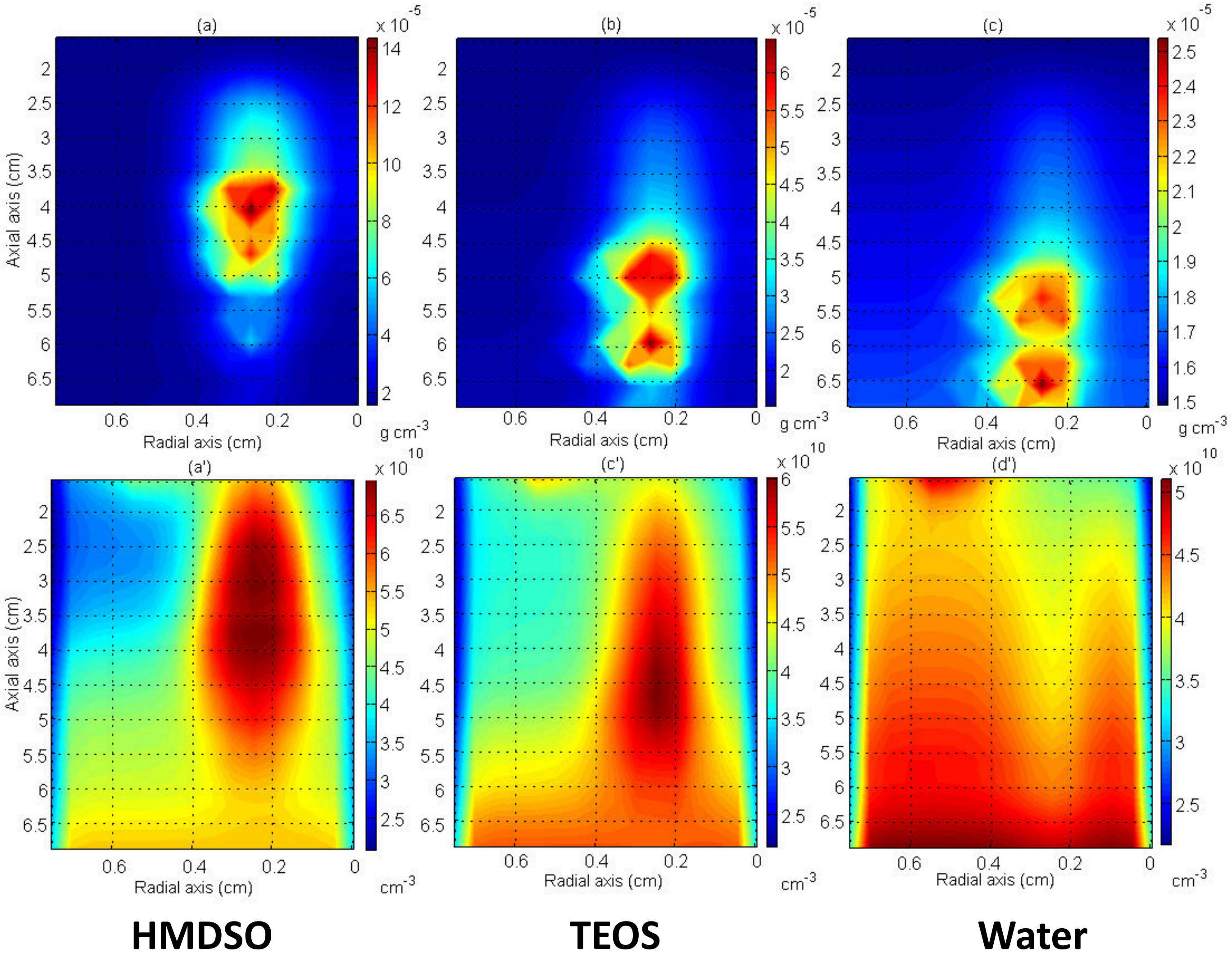
Evaporation in APP and Gas Mixture



Download pull and evaporation of droplets is more stronger in the case of APP at the top row as compared to the He-N_2 gas mixture at the bottom row.

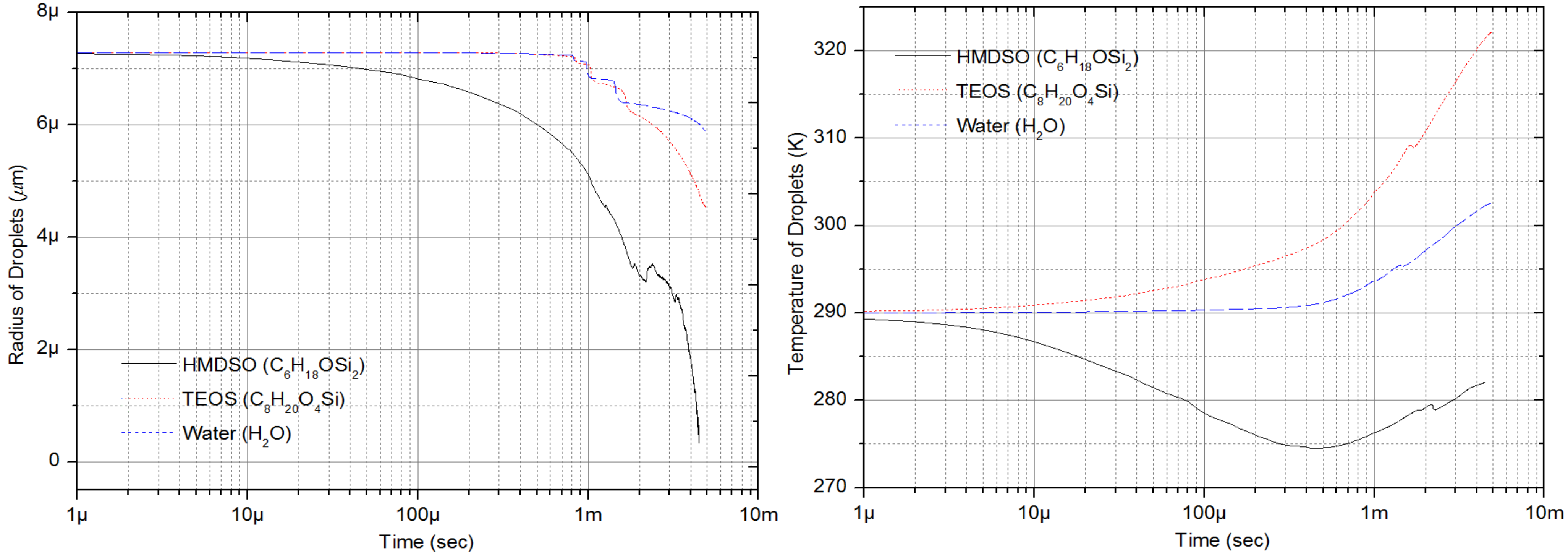
- Comparison of vapour species density in two different mediums**, i) Atmospheric pressure plasma (top row)**, **ii) He-N_2 gas mixture (bottom row)** for HMDSO droplets at precursor flow rate = 500 µL min^{-1}, gas flow rate = 5 L min^{-1}, f = 20 kHz and V_0 = −13.5 kV.

Effect of various Liquid Precursors



Muhammad M. Iqbal, Charlie. P. Stallard Denis P. Dowling and Miles M. Turner, Plasma Processes and Polymers, Volume 12, Issue 11, pages 1256 – 1270, November 2015.

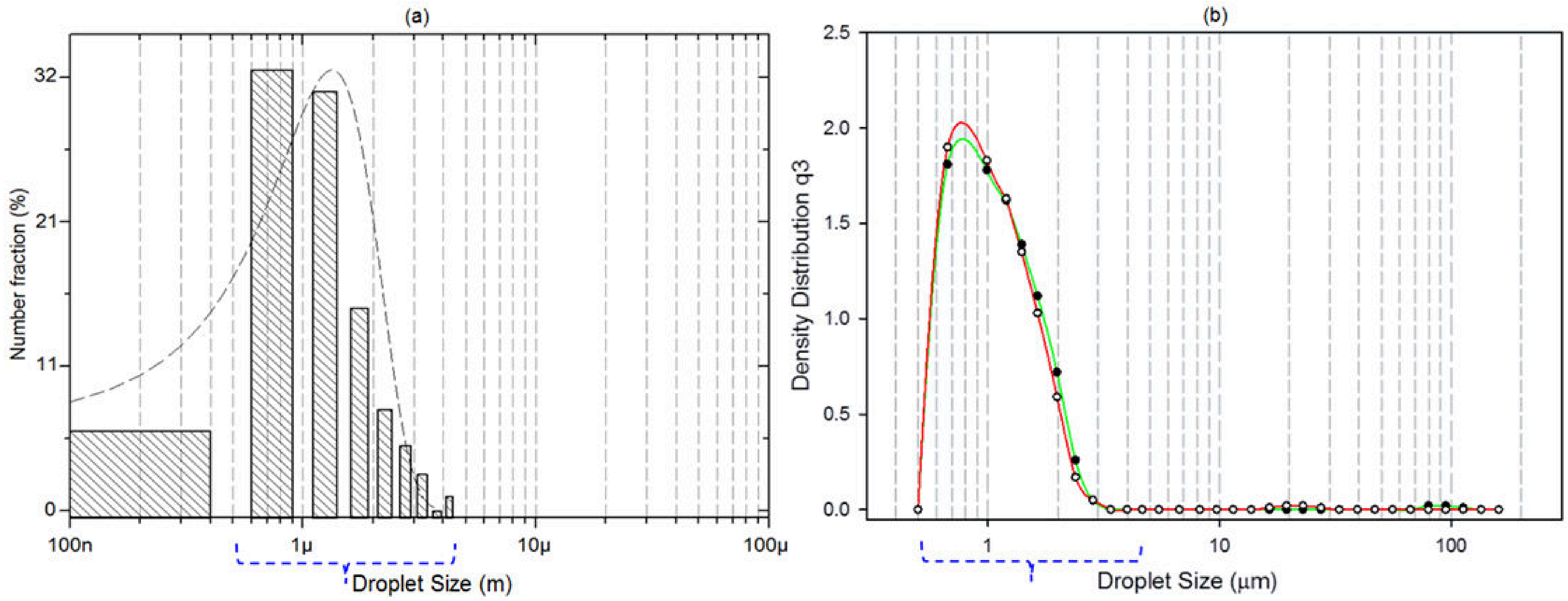
Effect of Different Liquid Precursors



- **Temporal profiles of mean radius and temperature of droplets in parcels for Hexamethyldisiloxane (HMDSO), Tetraethylorthosilicate (TEOS) and Water**

Precursor flow rate = 100 μL/min, gas flow rate = 5 L/min, f = 20 kHz and V_{appl} = -13.5 kV.

Comparison of Model and Experiment



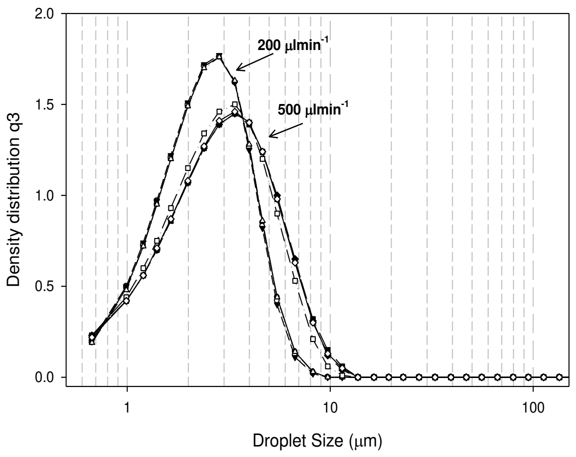
Comparison of (a) fluid-droplet model and (b) experimental observations of size distribution of droplets using HMDSO precursor droplets at flow rate = 100 mL min^{-1} and gas flow rate = 5 L min^{-1} , f = 20 kHz and V_0 = -13.5 kV.

Muhammad M. Iqbal, Charlie. P. Stallard Denis P. Dowling and Miles M. Turner, Plasma Processes and Polymers, Volume 12, Issue 11, pages 1256 - 1270, November 2015.

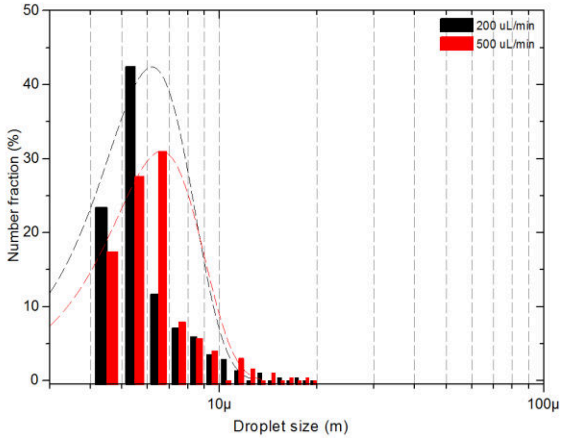
Effect of Different Precursor Flow Rates

Initial droplet size distributions at 200 and 500 µL/min using laser diffraction particle size analysis technique

Experimental observations

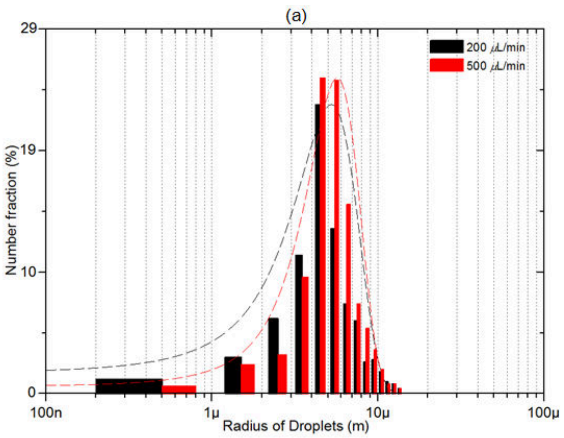


Fluid-droplet model simulations

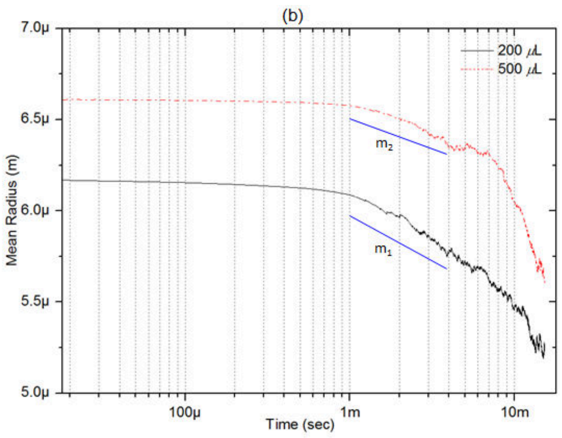


Droplet size distributions after 14 msec during downward flight of HMDSO droplets

Size distribution of droplets



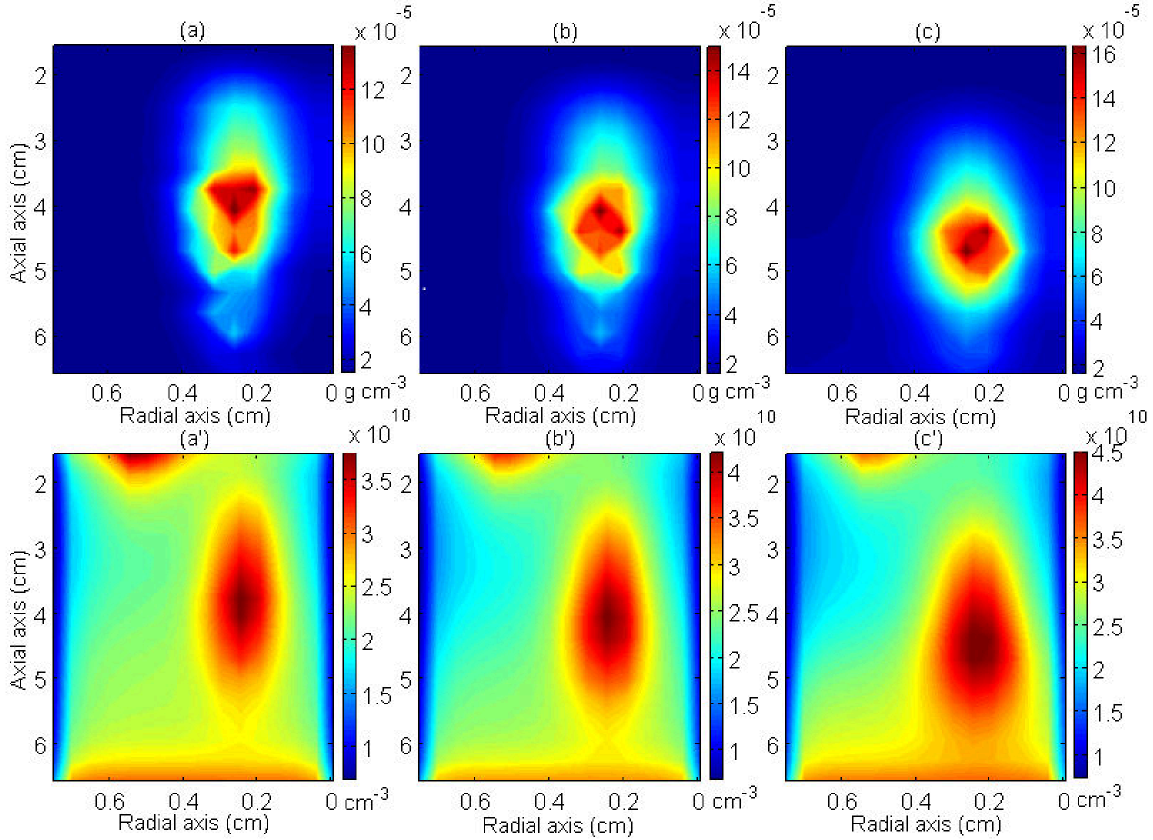
Temporal profiles of mean radius



Gas flow rate = 5 L/min, *f* = 20 kHz and Vappl = 13.5 kV in He-N2 gas mixture

Influence of Gas Flow Rates

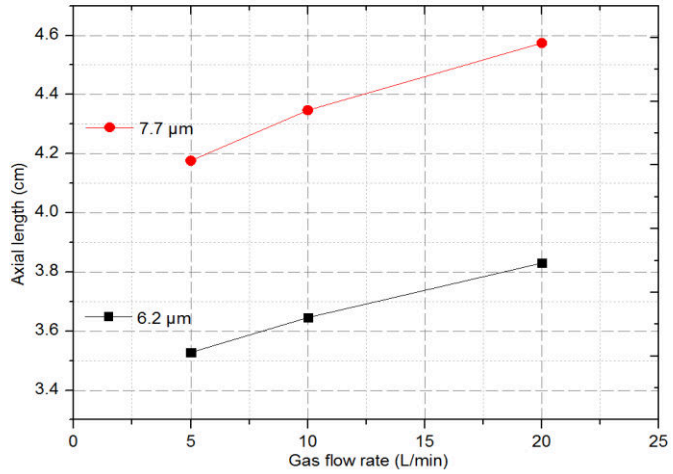
5 L/min **10 L/min** **20 L/min**



Electron density is shifted from ~ 3.5×10^9 to 4.5×10^9 cm^{-3}

Variation in the vapour species density for different gas flow rates (5, 10 and 20 L min^{-1}) at approximately 2 msec

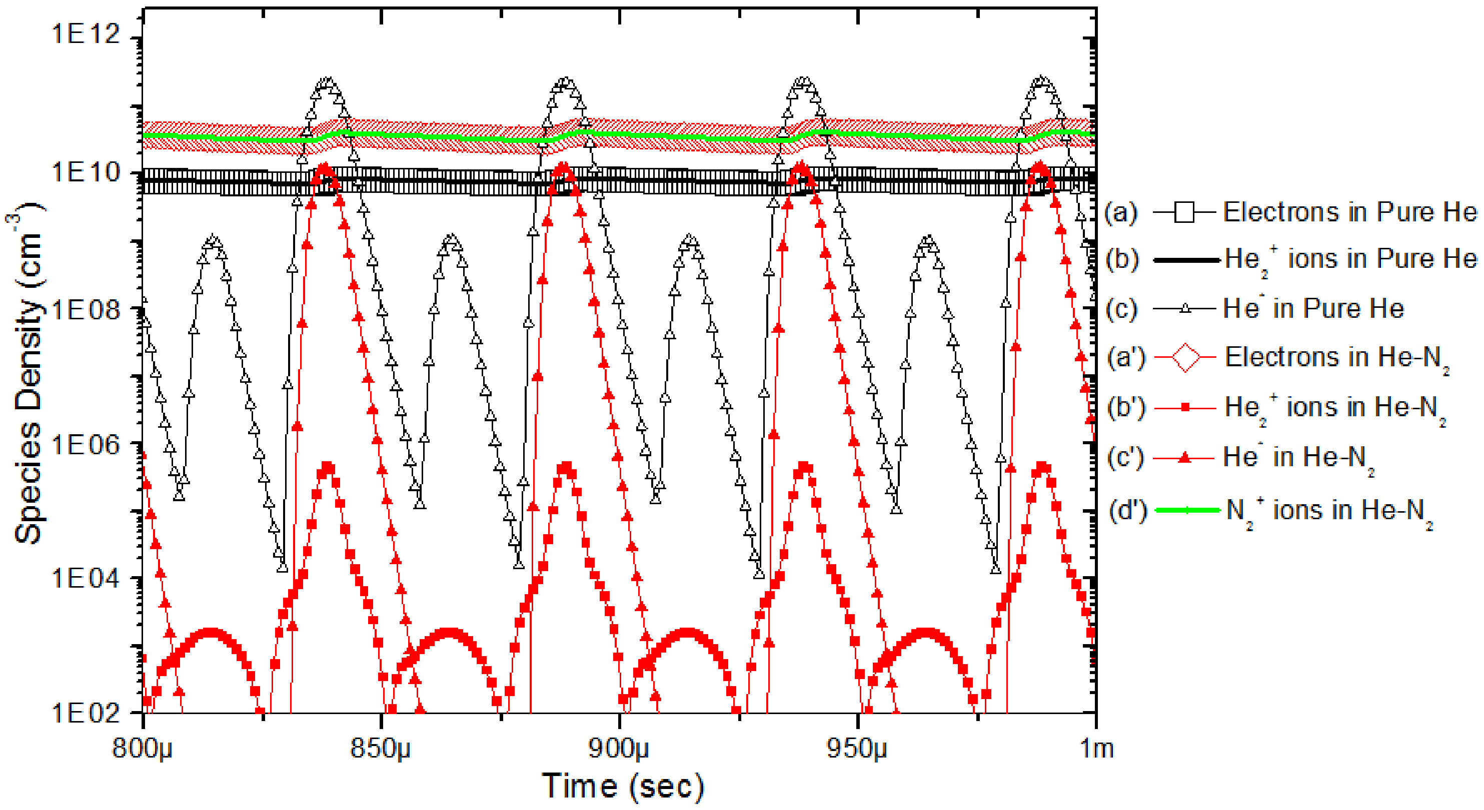
Examples of two different radii of droplets



HMDSO precursor flow rate =100 µL/min, *f* = 20 kHz and Vappl = 13.5 kV.

Line-averaged Density Distributions

Plasma Discharge Species



Comparison of line-averaged species density (electrons, He_2^+ ions, N_2^+ ions and He^*) in the pure Helium and $He\text{-}N_2$ gas mixtures using HMDSO droplets at precursor flow rate = 100 μL min^{-1}, gas flow rate = 10 L min^{-1}, f = 20 kHz and V_0 = −13.5 kV.

M. M. Iqbal and M. M. Turner, Contrib. Plasma Phys. 55, No. 9, 627 – 642 (2015) / DOI 10.1002/ctpp.201500048.

Conclusions

- The findings demonstrated that numerical modeling and simulations can deepen our knowledge of the fundamental characteristics of APP.
- The stationary state is obtained for these results to provide validity, and the entire code is developed in C++ utilizing a variety of solvers.
- Computer simulations can raise the level of understanding for the complex 2-phase flow problem that uses fluid modeling and a stochastic liquid droplet model.
- To demonstrate the validity of modeling and programming, the simulation results are contrasted with experimental findings provided by UCD in the Precision Strategic Research Cluster project.
- The entire research completed during Post doctoral research in Precision Strategic Research Cluster project at DCU.
- Numerous data science projects and modules are created and taught at CCT employing programming and algorithmic skills at the graduate and postgraduate levels.

Resources/ References

- **Acknowledgement:** “This material is based upon works supported by the Science Foundation Ireland under Grant No.08/SRC/I1411.”

- Muhammad M Iqbal, M Tetraethyl orthosilicate Turner, Book chapter, "Interaction and Transport of Liquid Droplets in Atmospheric Pressure Plasmas (APPs)" published in August 2022, DOI: 10.5772/intechopen.105010, In book: Fundamental Research and Application of Droplet Dynamics [Working Title], License CC BY 3.0.
- Muhammad Iqbal, Oral Presentation, "Engaging Students with Authentic Assessment", CCT Teachmeet (2022), CCT College Dublin.
- M. M. Iqbal, M. M. Turner, “Investigations of Droplet-Plasma Interaction using Multi-Dimensional Coupled Model”, Contrib. Plasma Phys., Volume 55, Issue 9, pages 627–642, October 2015.
- Muhammad M. Iqbal, Miles M. Turner, Plasma Processes and Polymers, Volume 12, Issue 10, pages 1104–1116, October 2015.
- Muhammad M. Iqbal, Charlie. P. Stallard Denis P. Dowling and Miles M. Turner, Plasma Processes and Polymers, Volume 12, Issue 11, pages 1256–1270, November 2015.