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An Experimental and Simulation Study of Larger Volume Micro discharge for the Realization of Microplama Based Reactor Applicable to Fuel Reforming and Material Processing

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ABSTRACT

Microhollow cathode discharge (MHCD) and microhollow cathode sustained discharge (MCSD) are two particular types of microdischarge configurations that produce stable glow plasmas at high pressures. Larger volume of higher density diffuse plasma in argon at various pressures is generated experimentally by MCSD in a split third electrode configuration. This enlarged volume microplasma serves as a source of high temperature electrons, ions and other excited species. Micro plasma reactor technology either for fuel reforming or material processing is based on using the energy of the high temperature electrons and other charged particles. Owing to their inherent difficulty in conventional diagnostics a detailed study on MHCD or MCSD relies on numerical simulations. This work describes the experimental procedure of generating MCSD with split third electrodes, realization of microreactor based on this enlarged volume microdischarge and numerical simulation of MCSDusing a 2D fluid model to determine the properties of this microdischarge.

Keywords: Microplasma, Microreactor, MHCD, MCSD, Split electrode.

1. Introduction

Plasma state of a matter is considered as the fourth state of matter, which has some unique properties to distinguish it from three other states solid, liquid and gases. Interestingly, much of the visible matter in the universe, viz., stars, all visible interstellar matter, is in the plasma state comprising 99% of the universe, both by mass and by volume. Since its first discovery in 1879 by Sir William Crookes [1], plasma has been produced in a variety of discharge configuration with different mechanism including dielectric barrier (DBD) discharge, microwave discharge, radio frequency (RF) discharge, direct current (DC) glow discharge etc.In a laboratory, one of the simplest ways to produce plasma is applying an electric field to a neutral gas. Plasma discharges can be categorized mainly of two parts thermal plasma and non-thermal plasma. Thermal plasmas are the plasmas where electron temperature *Te*, ion temperature *Ti*and neutrals temperature T is inthermal equilibrium and electron temperature is considered as almost the same with ions and neutrals temperature. In non-thermal plasma, electrons temperature *Te*is much higher than the ions temperature and neutrals temperature. High pressure arc discharges are the common example of the thermal plasma and low pressure glow discharges are the example of non-thermal plasma or cold plasma. New types of plasma discharges namely microhollow discharges has attracted great interest among the researchers. The properties of microdischarges fall in between arc discharges and glow discharges. Like arc discharges, it can be operated at high pressure but the electron temperature is much higher than the other particles which make it more likely to be a non-thermal glow discharges. For that reason this microdischarge is referred as 'high pressure glow discharges' [2]. Non equilibrium glow discharges can be achieved by various approaches such as microhollow cathode discharge (MHCD) [3] .Three most important characteristics of the MHCD are - high pressure, high energy electrons and larger surface to volume ratios. High surface-to-volume ratio in MHCD hole imparts excellent thermal management and mixing characteristics that help maintain homogeneous, isothermal reacting volumes for fuel reforming. Indeed, novel uses of MHCD have been proposed in enormous application such as propellant gas preheating for microplasma thruster [4], ignition assistance [5], hydrogen generation for fuel cell power [6], hydrocarbon reforming, source of UV and eximer radiation[7] etc . The use ofMHCD has also been explored as flow reactor by Hsu and Graves [8], where they have shown that flowing of molecular gases through the MHCD was found to induce chemical modifications by molecular decomposition processes.The volume of the MHCD is very small to use in industrial purposes. To increase the volume of the high pressure glow discharge, researchers has developed another discharge namely microhollow cathode sustained glow discharges where the discharge is sustained by the plasma cathode which act as a source of electron in the discharge. This kind of large volume discharge can be used for fuel reforming and gas treatment purpose as well. Conventional technique for fuel reforming and gas treatment process includes steam reforming by oxidations. Thermal oxidation requires heating of the ambient gas and raises the capital cost for cooling system.Main advantage of non-thermal plasma is the presence of high energy electrons which reacts with the fuel particles and causes significant reforming. Residence time for the gas is very short in case of nonthermal plasma with low power requirement. Our experimental study approaches to the enlargement of the high pressure hollow cathode sustained glow discharges by employing split electrodes replacing single planar third electrode(Fig.1.1) and eventually applies this large

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volume glow in fuel reforming and gas treatment processes. As a microplasma reactor either for fuel reforming or material processing, microdischarges serve as sources of high energy electrons, radicals and ions. In order to improve the results in all these application fields, a good insight into the discharge processes is desirable. So it is very important to know the properties of plasma parameters such as electron density, atomic and molecular ion densities, excited species (metastables) densities, electron temperature, gas temperature etc.

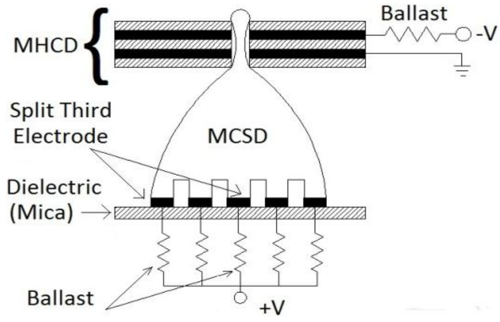

Fig.1.1MCSD with split third electrode

However, the reduced dimensions, high operating pressure, and high power density of microdischarges make conventional diagnostics of the microplasma for experimental characterization very complicated and challenging. Given the experimental challenges, computer simulations provide a valuable alternative for studying microdischarges. While MHCD devices have been studied numerically over the recent decade, understanding of the fundamental mechanisms of MCSD that generate larger volume microplasmas has lagged. Early works paid little attentions to the MCSD with single third electrode configurationand plasma properties like electron, ion and excited particles densities in MCSD with single planar third electrode are reported in a very few simulation studies carried out by Pitchford and collaborators [9]. However, they didn't mention about gas temperature and electron temperature in their studies which are also crucial parameter for microchemical reactor [2]. Here numerical investigationof MCHD and its sustained discharge MCSD in argon with split third electrodes(Fig.1.1)is also attempted to estimate the discharge properties by using a two-dimensional axis-symmetric, self-consistent multi-species, multi-temperature, continuum (fluid) model. Predicted results are compared to available experimental and numerical data.

2. Experimental Setup and Numerical Modeling

Fig.2.1shows the schematic diagram of the experimental setup which consists of vacuum system, electrical measurement system, optical measurement system and gas flow system.Vacuum system consists of vacuum chamber, vacuum pump, pressure sensor. Discharge setup was placed inside the vacuum chamber. Electrical measurement system consists of high voltage dc power supply, electrical circuits, and digital multiple purpose multimeter. Optical measurement system consists of digital single lens reflex camera which is placed firmly on the steady stand, as macro lenses are very sensitive and small displacement may cause change the magnification of the different discharges. We have used 105 mm Macro lens with lowest F number 2.8 for proper focusing. During capturing the pictures dark environment was provided to see the actual visual characteristic of the plasma discharge and physical scale length was measured before capturing the pictures. Gas flow system consists of gas chamber, gas flow tubes, gas flow meter.

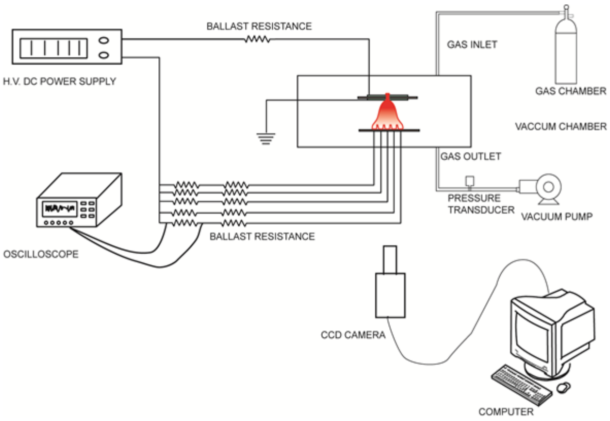

Fig.2.1 Schematic diagram of the experimental setup

Figure 1.1 presents a schematic diagram of MCSD configuration with a split third electrode and the corresponding discharge structure. MHCD layer placed on the top of the system consists of Nickel electrodes of 0.2mm thickness pasted on both sides of 0.2mm thick mica dielectric material. A cylindrical hole of 0.3mm diameter is drilled mechanically through the three layers of MHCD configuration. The third electrode is placed 5mm away from the MHCD configuration. The third electrode which is placed 5mm away from the MHCD layer is split into 5 rectangular pieces. Each split electrode has a surface area of 0.5 mm × 3 mm and is separated by 2 mm. Total span of the third electrode is about 10mm wide.

The number of split electrodes and the separation between them can be adjusted so that a total span of discharge region can be controlled accordingly. The sharp edges of each split electrode is slightly rounded in order to avoid a high local electric field which can cause a non-uniform glow discharge on the third electrode surface. One of the MHCD electrodes facing with the third electrode is always grounded as shown in Fig. 1.1 while the voltages on the top-layer electrode and the third electrode are biased with an opposite polarity of maximum 2.5 KV provided by dc power supply. The third split electrodes are connected with an individual 100 KΩ ballast resistor and 220Ω current-view resistor in series. Current through each split electrode is measured by a digital oscilloscope. The entire setup is placed in a vacuum chamber which provides desired ambient pressure and the chamber is replenished in every run. A high speed CMOS camera (Photron SA3 120K) and digital hand-held camera are used to visualize discharge structures.Numerical simulation of non-equilibrium glow discharge plasma of an MCSD is performed with the VizGlow plasma modeling tool [10]. The model is based on a 2-D axisymmetric, self-consistent, continuum fluid description of the plasma. Species conservation, electron energy, and gas energy equations are solved in the gas, while Poisson's equation for the electrostatic potential is solved in the gas and in the dielectrics. A detailed description of the governing equations solved is described in Ref. [10]. An MCSD is generated in argon at pressures of 60 Torr and 300 Torr. The argon species involved in the simulation include electrons (e), atomic and dimer ions (AR$_+$and AR$_{2+}$), and atomic and dimer metastables (ARm and AR2m). The reactions used inthis model with rate coefficient data from Ref. [11].

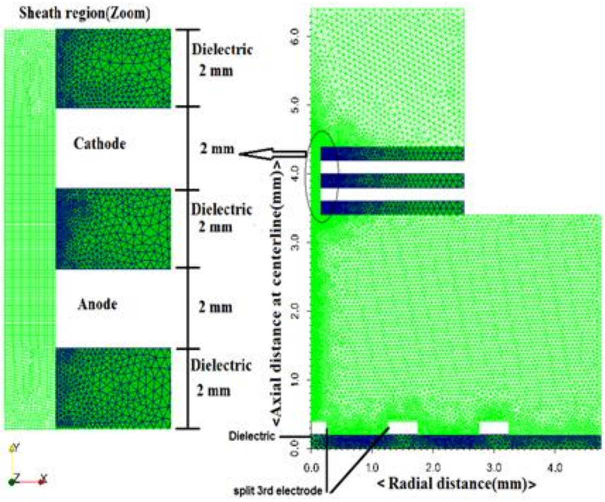

Fig.2.2 Schematic of the MCSD with split third electrode and computational mesh.

Fig.2.2 shows the computational mesh and the cylindrically symmetric geometry of the MCSD with split multiple third electrodes used in this study. The same experimental geometry is used for numericl simulation. The dimensions are identical to the dimensions of the experiment shown in Figure 1.1 except the dimensions of the boundary of the computational domain.The mesh consists of about 28,765 cells of which about 22,917 are in the gas subdomains and about 5,848 are in the dielectric subdomains. To obtain a stable numerical solution, first the MHCD is run with a time step of 5x10^{-12} s till steady state is reached with the third electrodes turned off. Then, the third electrodes are tuned on and a smaller time step of 2x10^{-12} s is found to be necessary to reach a combined steady state for the MCSD

3. Result and Discussion

The optical images of MCSD at different pressures with split third electrodes captured by high speed digital camera have been demonstrated. The pressure effect is illustrated in Fig.3.1. For anode third electrode case at 60 Torr pressure, MCS discharge starts at very low third electrode current connecting all 5 split electrodes with MHCD. MHCD current was fixed at 2mA and third electrode current increases from lower value to higher value. At 7mA of third electrode anode current, suddenly the glow disappears at the gap between MHCD and third electrode. In this case discharge appears at the MHCD cathode side and photogenic glow like discharge appears from each split electrodes. At the current level more than 7mA, this glow from the third electrodes again connect to the MHCD and large volume bulk discharge form in the gap between MHCD and split third electrodes. For anode split third electrode cases, denser plasma glow discharge forms in all current levels at 300Torr cases. At 1mA of anode current discharge connects to three electrodes and at 6mA of current it connects to all the 5 split electrodes. With increasing anode current, denser and brighter plasma forms in the gap without some major changes in the structure. At very high applied electric field this glow like discharge will transfer to the arc regime.However as shown in the Fig. 3.1, it is clear that much larger volume of MCS discharge can be produced by employing a split third electrode than the single third electrode case as presented in Ref. [12]. In addition to the enlarged discharge volume it is expected to be possible with more number of finer electrodes that plasma volume, density and its location can be controlled by addressing the desired split electrodes or by changing an arrangement of the split electrodes. However with a single third electrode this controllability of the discharge is very limited and only plasma density can be adjusted by MCSD current.Fig.3.2-3.9 shows Spatial and axial profile of electrostatic potential, Electron, monomer ion (AR$^+$), dimer ion (AR$_2^+$), and metastable species (AR^m and AR$_2^m$) population at 60 Torr and 300 Torr respectively. The potential profile (Fig.3.2) explains that the MCSD is a positive column expanded outside the MHCD hole due to a weak electric field in the gap between the MHCD and the third electrode.

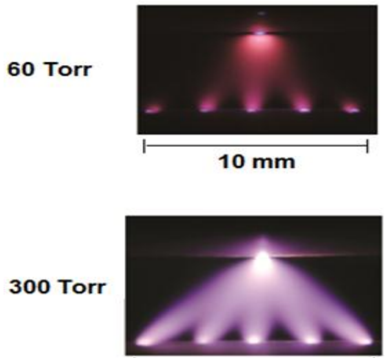

Fig. 3.1 MCSD with 10 mm span 5 split electrodes biased as at 60 Torr and 300 Torrpressure.

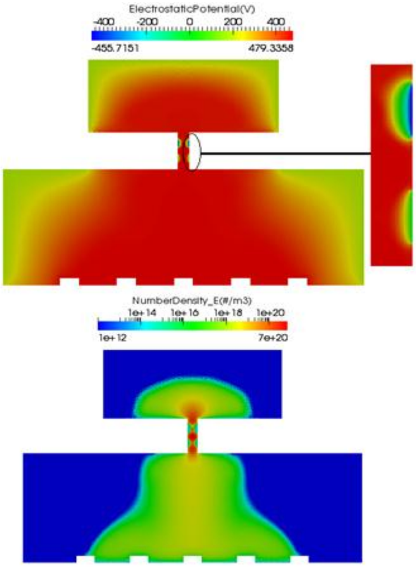

Fig. 3.2 Spatial profile for electrostatic potentialand electron number density at P=60 Torr

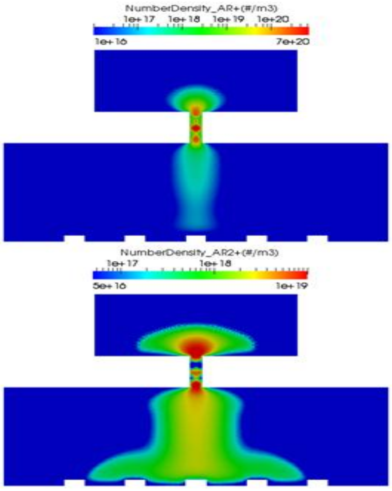

Fig.3.3 Spatial profile for monomer ion (AR^+) and dimer ion (AR_2^+)number density at 60 Torr

The sheath area is confined inside the MHCD hole because of the dielectric layer on the top surface. Electric field is more extended between MHCD hole and third electrode in split electrode case which is more favorable for larger expansion of plasma species. The peak density is as high as$1\times10^{20}m^{-3}$ at 60 Torr near the exit plane of the MHCD and on-axis due to the expansion of the plasma leaving the MHCD. The main mechanism of creation of electrons, in the zone of radial expansion of the MCSD, is the step-wise ionization, and the losses of electrons are via diffusion in radial direction and dissociative recombination.The highest monomer ion (AR^+) density is about ~$1x10^{20}$ m^{-3}, peak dimer ion(AR_2^+) density is about ~$1x10^{19}$ m^{-3}, peak value of metastable atom (AR^m) density is about (~$10^{21}m^{-3}$) and peak dimer metastable (AR_2^m) density is ~10^{17} about occur along exit hole of MHCD on the centerline the MCS discharge at pressure p=60 Torr.

Significant presence of excited species (AR^m and AR_2^m) is useful in the sense that they are considered as important constituent for plasma assisted ignition [9].At 60 Torr, peak value of bulk temperature (Fig.3.5) is 1422K occurs along the centerline of the cathode region in the MHCD and negligible bulk temperature (peak value 320K) is observed in the MCSD. It shows consistency in a recent measurements of the gas temperature in Ar/O_2 mixtures in the MCS region where limited gas temperature is observed [13]. Peak electron temperature (Fig.3.6) is observed in the cathode sheath region within the hollow, which is in the order of several tens of eV. Other parts of the discharge maintain the electron temperature in the order of ~1 eV. In an experimental observations of the emission from high-lying excited states of ionic species in MDs [14], such high electron temperatures are expected, which is in again good agreement with our results.Experimental result shows that plasma is extended over five split electrodes (Fig.3.1) without ensuring which species are responsible for plasma expansion. From simulation it is clearly seen that electron is the most responsible particle for extended plasma near third split electrodes. Dimer ion and metastable atom also partially contribute to the expansion of larger volume plasma near split third electrode.Plasma densities are constant in the middle of axis for a particular species at both 60 Torrand 300 Torr (Fig.3.7). It is observed that number densities of all plasma species on the axis of MCSD increased at higher pressure (Fig.3.8-3.9) due to comparatively stronger electric field.Electrons are likely to diffuse more along the dielectric surface outside the MHCD hole at higher pressure. Other interesting phenomena is that metastable species especially dimer metastable has been significantly expanded near split third electrode with an increase in number densities at higher pressure. Peak values of electron, atomic and molecular ion, atomic and molecular excited particles number densities exceed $3x10^{20}m^{-3}$, $2x10^{20}m^{-3}$, $1x10^{20}m^{-3}$, $2x10^{21}$ m^{-3} and $5x10^{18}$ m^{-3} respectively at 300 Torr.

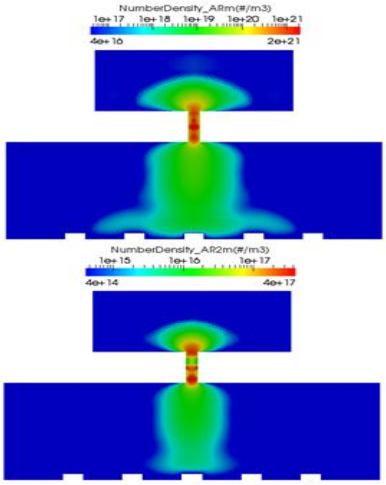

Fig.3.4 Spatial profile for metastable species (AR^m, AR_2^m) number density at 60 Torr

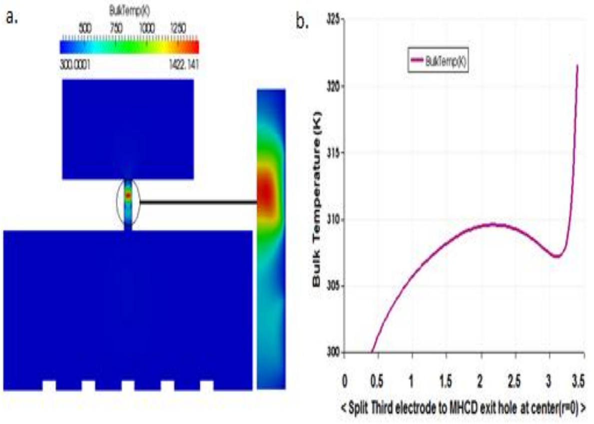

Fig.3.5 Spatial profile for a) bulk temperature and b)corresponding axial profile in the sustained discharge at P=60 Torr

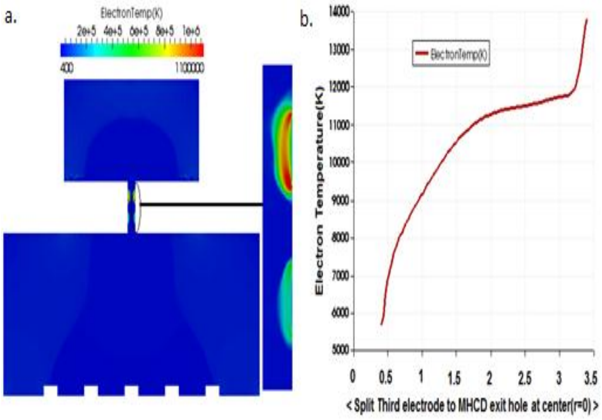

Fig.3.6 Spatial profile for a) electron temperature at 60 Torr and b)corresponding axial profile in the sustained discharge at 60 Torr

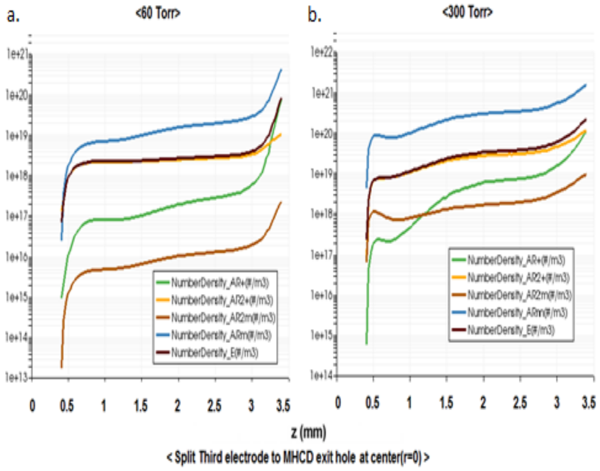

Fig.3.7 Axial distribution of a)different plasma species number density in MCSD at 60 Torr and b) different plasma species number density in MCSDat 300 Torr.

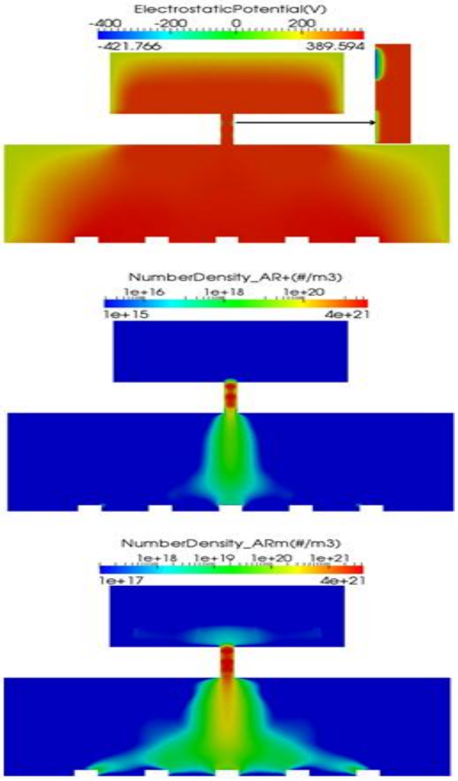

Fig.3.8 Spatial distribution of potential and AR^+ and AR^m number densities for MCSD with split third electrodes at 300 Torr.

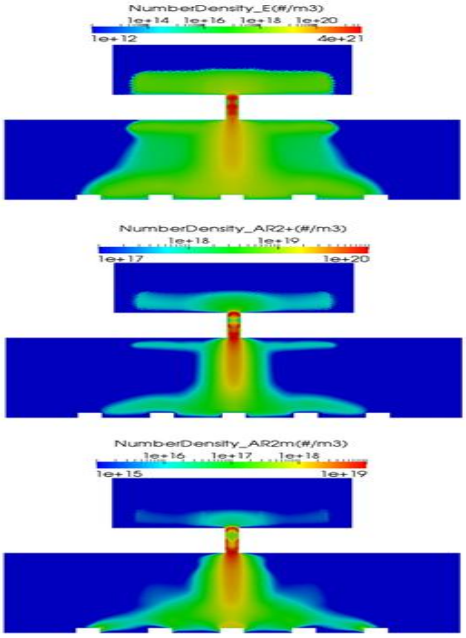

Fig.3.9 Spatial distribution of electron, AR_2^+ and AR_2^mnumber densities for MCSD with split third electrodes at 300 Torr.

4. Conclusion

In this article, a unique approach has been demonstrated that a larger volume glow discharge at atmospheric pressure is possible with MCSD configuration which has split third electrodes. This large volume non equilibrium plasma discharge can be used as a reactor to reform fuel and gases. Simulation results are presented for MCSD with split third electrode which shows that expansion of plasma is larger than that of MCSD with single planar third electrode cases and confirms the species which are responsible for extended plasma near split third electrode which is not clear from experiment. Charged species densities of order ($10^{19} \sim 10^{21}$) m^{-3} and metastable species densities of order ($10^{18} \sim 10^{21}$) $m^{-3}$are predicted in the microhollow cathode discharge (MHCD) for the conditions investigated. In the sustained discharge (MCSD), charged species densities of order ($10^{16} \sim 10^{20}$) m^{-3} and metastable species densities of order ($10^{16} \sim 10^{21}$) m^{-3} are predicted for the conditions investigated.

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NOMENCLATURE

AR^{+},AR^{m}	: Molecular ion of Argon
AR_2^{+},AR_2^{m}	: Dimer ion of Argon
eV	: electron temperature unit, electron volt
P	: Pressure, Torr
T	: Temperature, K