# Experimental and numerical study of ferromagnetic enhanced inductively coupled Cl<sub>2</sub>/Ar plasma source

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**Abstract:** A low frequency (~100 kHz) ferromagnetic enhanced inductively coupled plasma (FMICP) source with  $Ar/Cl_2$  gas mixture is studied in order to obtain a large volume  $(2 \cdot 10^5 \text{ cm}^3)$  of dense  $(10^{10}-10^{13} \text{ cm}^3)$  uniform plasma at low pressures (0.1-10 Pa). A simplified model for large scale FMICP is developed and presented. New data on plasma parameters of FMICP were are obtained experimentally and numerically.

**Keywords:** Ferromagnetic enhanced inductively coupled plasma, argon/chlorine plasma, plasma etching, 450 mm wafers.

## 1.Introduction

Radio-frequency induction discharges are one of the most important types of gas discharges for many scientific and practical applications [1]. This electrodeless method of generating a low-temperature plasma with a high concentration of ions and radicals over a wide range of pressures  $(0.1-10^5 \text{ Pa})$  creates unique possibilities for carrying out a wide range of plasma-chemical reactions and processes and made it possible to create new technologies in lighting engineering (electrodeless lamps), plasma chemistry (RF induction plasma torches), and (plasma-chemical microelectronics etching and deposition). The high frequency of generation of inductive discharges (of the order of 1-10 MHz) and low power factor of ICP coil resulting in a low power transfer efficiency stimulates the search for new ways of efficient generation of an inductively coupled plasma in the lowfrequency range. The simplest way to achieve this is to use ferromagnetic materials to improve the magnetic coupling between plasma and ICP coil. At present, this type of ICP sources is termed as ferromagnetic enhanced inductively coupled plasma source or FMICP (the term was proposed by Valery Godyak), that was firstly used in the 1960s to create an electrodeless argon laser [2] and a prototype of an electrodeless fluorescent lamp [3], with the power of 40 W, with the frequency of 200-400 kHz.

In the paper [4], a new approach to the generation of FMICP, a so called distributed FMICP, was proposed. This plasma source has one main chamber with sideinjected plasma produced with multiple ferromagnetic core antennas. The plasma radial profile in the main chamber is governed by plasma and metastable atom diffusion from side discharges as well as ionization caused by fast electrons generated there that are able to penetrate deeply into the main chamber. This approach was proposed to produce large volumes of homogeneous plasma and to solve the problem of scaling RF induction discharge devices for the future 450 mm standard of semiconductor industry.

In [5], a method for controlling the spatial distribution of plasma density by imposing an auxiliary RF induction discharge on the central region of the distributed lowpressure (5 mTorr) argon FMICP was proposed. An auxiliary RF induction discharge was also used to control the spatial uniformity of the distributed FMICP discharge at elevated argon pressures of 600 mTorr [6]. Thus, the possibility to generate large volumes of "dense" plasma efficiently with the use of distributed FMICP, and the ability to control radial distributions of plasma parameters by means of an auxiliary discharge were clearly demonstrated. A conclusion was made about the prospects of using FMICP for large-scale plasma processing [7].

However, for plasma treatment of semiconductor materials it is necessary to introduce halogen-containing gases into the plasma. The addition of an electronegative plasma-forming gas will radically change the plasma parameters of the distributed FMICP, which creates new challenges for researchers. The aim of this paper is to reveal the main problems and analyse the particularity of generating distributed FMICP in the atmosphere of electronegative plasma-forming gas. For this purpose, a large volume  $(2 \cdot 10^5 \text{ cm}^3)$  of dense  $(10^{10} - 10^{13} \text{ cm}^{-3})$ uniform plasma at low pressures (0.1-10 Pa) is experimentally obtained (see Section 2). A global model for plasma parameters of FMICP in argon/chlorine mixture is developed and presented in Section 3. The results of the numerical modeling are presented in Section 4. Finally, the discussion and conclusions are presented in Section 5.

## 2.Experimental setup

In Figure 1, a scheme of experimental setup for the study of a distributed FMICP is shown. The discharge chamber is made of stainless steel. The internal diameter of the main gas discharge chamber (1) is 700 mm. On the sides of the main gas discharge chamber there are eight U-shaped gas discharge tubes (2) with an internal diameter of 50 mm. The U-shaped tubes and the main chamber are dielectrically insulated. Each U-tube has a ferrite core (3) with a primary winding (ICP coil) connected to a power source with a frequency of 100 kHz. Each U-shaped tube has an inlet (4) to supply a plasmaforming gas, which can flow through the open ends of the U-shaped tubes into the main chamber. An additional inlet for the plasma-forming gas is located in the centre of the top flange of the setup (not shown in the figure).

In this paper, to control the uniformity of the plasma density profile inside the gas discharge chamber, two auxiliary FMICP sources with a frequency of 100 kHz (located on the top flange of the main discharge chamber) are proposed to be used instead of a 13.56 MHz RF ICP source [5]. This permits us to increase the power factor of the ICP coil and eliminate the capacitive coupling between plasma and the coil [7]. All these FMICP sources form a common spatial distribution of the plasma inside the main gas discharge chamber. In the setup there are ports for optical and probe methods of plasma diagnostics. The device is designed to study the processes that determine the formation of spatial distribution of plasma parameters in order to obtain large volumes of homogeneous plasma. A substrate (450 mm in diameter) will be placed inside the main chamber to investigate the processes of ion-plasma etching of various materials.



Fig. 1. Experimental setup. 1 - Gas-discharge chamber, 2 - U-shaped sections, 3 - Ferrite cores with primary winding, 4 - Fungal seals, 5 - Plasma-forming gas supply,

6 - Windows for optical diagnostics of plasma, 7 - Side

flanges, 8 - Flanges for connecting vacuum gauges,

 9 - Flange for vacuum pumping, 10 - Flanges for access to the substrate holder, 11 - The substrate holder.

## 3.Model

Plasma parameters in a large scale cylindrical discharge chamber of radius R = 35 cm and height L = 50 cm are investigated. For this purpose, a global (volume averaged) model of chlorine/argon discharge adopted after works [8,9] is used. Previously, this model was used for plasma characterisation in moderate scale chambers with of radius R = 10 cm and height L = 10 cm. FMICP chamber under investigation has the volume 60 times larger, i.e.  $2 \cdot 10^5$  cm<sup>3</sup>.

The model takes into account the following features of experimental plasma source. Through the vacuum valve a constant flow rate Q of neutral particles is introduced into the chamber (Cl<sub>2</sub> and Ar). The density of neutral particles is assumed to be uniform over the space. With the adjustment of the pressure at the outlet of the chamber the gas pumping is controlled and the working gas pressure p is regulated, which is defined as the sum of the partial pressures of all the components of the plasma gas.

In addition to electrons in the discharge, chlorine molecules in the ground state of Cl<sub>2</sub> (v = 0), vibrationally excited chlorine molecules Cl<sub>2</sub> (v = 1–3) with excitation threshold  $\varepsilon_{ex} = 0,07-0,21$  eV, chlorine atoms in the ground state Cl (3p<sup>5 2</sup>P), negatively charged chlorine ions Cl<sup>-</sup> ( $\varepsilon_{aff} = 3,6$  eV), positively charged chlorine ions Cl<sup>-</sup> ( $\varepsilon_{i} = 13,0$  eV) and Cl<sub>2</sub><sup>+</sup> ( $\varepsilon_{i2} = 11.5$  eV), argon atoms in the ground state Ar (3s<sup>2</sup> 3p<sup>6</sup>), metastable states of argon atoms Ar<sub>m</sub> (1s<sup>5</sup> and 1s<sup>3</sup>) ( $\varepsilon_m = 11.6$  eV), radiation-related levels of Ar<sub>r</sub> (1s<sup>4</sup> and 1s<sup>2</sup>) ( $\varepsilon_r = 11.7$  eV), highly excited argon atoms Ar (4p) ( $\varepsilon_{ex} = 13.2$  eV) and positive Ar<sup>+</sup> ( $\varepsilon_{Ar,I} = 15,8$  eV) are considered.

The balance equation for particles of type X can be written as:

$$\frac{dn^{(X)}}{dt} = \sum_{i} R_{G,i}^{(X)} - \sum_{i} R_{L,i}^{(X)}, \qquad (1)$$

where  $R_{G,i}^{(\chi)}$  and  $R_{L,i}^{(\chi)}$  are the rates of generation and loss of X particles. In this paper, various processes of loss and generation of particles in a discharge are considered, which includes reactions between electrons and gas particles, between two (or more) gas particles, recombination of neutral particles on the chamber wall and neutralization of positively charged ions (for all neutrals and positive ions). The supply of molecular chlorine Cl<sub>2</sub> and argon Ar to the chamber and the pumping of gas particles from the chamber are also taken into account. The reaction rates  $R^{(X)}$  were calculated as the product of the reactant densities and the rate constant of the corresponding process, k:

$$R_{G,i}^{(\chi)} = k \times \prod_{i} n_{r,i}$$
(2)

where  $n_{r,i}$  is a density of *i*-th reactive gas.

Since the discharge is assumed to be electrically quasineutral:

$$n_e = [Ar^+] + [Cl^+] + [Cl_2^+] - [Cl^-],$$
(3)  
where [X] – the density of particles X.

The reactions rate constants depend on the electron energy as follows:

$$k(T_e) = (2e/m_e)^{1/2} \int_{\varepsilon_{th}}^{\infty} \sigma(\varepsilon) \varepsilon^{1/2} f(\varepsilon) d\varepsilon, \qquad (4)$$

where  $\varepsilon_{th}$  is the energy threshold and  $\sigma(\varepsilon)$  is the crosssection of the process. Sets of cross-sections for Cl<sub>2</sub>, Cl and Ar are taken from the works [8,9]. It is assumed that electrons have a Maxwell energy distribution function, and the electron collision velocity coefficients are calculated using analytical equations for the electron temperature range of 0.01 <  $T_e$  < 10 eV.

On the walls of the discharge tube, the loss and birth of particles occurs due to the recombination of chlorine radicals and argon atoms and the neutralization of positive ions ( $Ar^+$ ,  $Cl^+$ ,  $Cl_2^+$ ). Diffusion losses of Cl atoms in the reactor are estimated using the effective loss factor [8-10]:

$$K_{Cl,wall} = \left(\frac{\Lambda_{Cl}^2}{D_{Cl}} + \frac{2V(2 - \gamma_{rec})}{A\nu_{cl}\gamma_{rec}}\right)^{-1},\tag{5}$$

where  $D_{Cl}$ ,  $v_{Cl}$  and  $\gamma_{rec}$  are diffusion coefficient, thermal velocity and recombination coefficient of neutral chlorine

atoms on the wall, respectively. V and A are volume and surface area of the chamber wall, and  $\Lambda_{Cl}$  is an effective diffusion length of atoms of neutral chlorine [10]:

$$\Lambda_{Cl} = \left( \left(\frac{\pi}{L}\right)^2 + \left(\frac{2.405}{R}\right)^2 \right)^{-1/2},$$
 (6)

The rate of positive ions loss i+ due to their flow on the walls is given by the expression:

$$K_{i^+,wall} = u_{Bohm}^{i^+} \frac{A_{eff}}{V},\tag{7}$$

where  $u_{Bohm}^{i^+} = (kT_e/m_i)^{1/2}$  is a Bohm's speed,  $A_{eff}$  is an effective ion loss area

$$A_{eff} = h_L A_L + h_R A_R = 2\pi (R^2 h_L + RLh_R),$$
(8)

 $h_L$  and  $h_R$  are the ratios of ion densities at the edge and at the center. Expressions for  $h_L$  and  $h_R$  in a wide range of pressures and electronegativity parameters are obtained in the works [8-10]. It is assumed that vibrationally excited chlorine molecules Cl<sub>2</sub> (v > 0) and excited argon atoms  $Ar_{mb}$   $Ar_r$  and Ar (4p) are all deactivated on the surface of the chamber, i.e.

$$Cl_2(v) + wall \rightarrow Cl_2(v-1)$$
 (9)

and

$$lr^* + wall \to Ar. \tag{10}$$

It is supposed that the quenching of excited particles on the walls is similar to the diffusion losses of neutral atoms on the wall (5). The quenching probability on the walls  $\gamma_Q$ , used in equation (5) instead of being equal to  $\gamma_{rec}$ , is assumed to be equal to one for quenching both Cl<sub>2</sub> (v) and Ar\*.

The gas supply of  $Cl_2$  and Ar was carried out separately. To maintain cleanliness, the gas in the discharge chamber was usually replenished by simultaneously emitting gas into the chamber and evacuating gas out of the chamber during operation. Gas transfer in the chamber is taken into account in the  $Cl_2/Ar$ model, assuming that the reactions:

 $[Source] \rightarrow \{Cl_2(v=0), Ar\},$  (11) which contribute to obtaining Cl<sub>2</sub> (v = 0) and Ar molecules at a rate given by  $R = 4,48 \times 10^{17} Q_{\{Cl_2,Ar\}}/V$ , where  $Q_X$  is the flow of particles of the type X into the chamber in sccm, V is the chamber volume, and the multiplier converts sccm to [particles/s]. Dilution with argon is carried out by changing the ratio from  $Q_{Ar}$  to  $Q_{Cl2}$ , and, consequently, the rate of Cl<sub>2</sub> and Ar while maintaining a fixed total flow rate of  $Q_{Cl2} + Q_{Ar}$ .

The evacuation of gas from the chamber is taken into account in the same way, that is, with the reactions:

 $\{Cl_2, Cl, Cl^+, Cl_2^+, Ar, Ar^*, Ar^+\} \rightarrow [Drain],$  (12) which contribute to the loss of each gas particle, with the exception of negative Cl<sup>-</sup> ions, which are usually absent in the near-wall areas of the chamber, therefore, are not pumped out. The coefficient estimation for the reaction (12) is determined by the expression:

$$k_{p_0} = 1.27 \times 10^{-5} \frac{Q_{Cl_2} + Q_{Ar}}{p_0 V},$$
(13)

where  $p_0$  is the output pressure in Torrs and factor converts sccm to Torr m<sup>3</sup> s<sup>-1</sup>.

The power balance equation, which equates absorbed power to power loss due to elastic and inelastic collisions of electrons and the energy carried by the flow of charged particles to the wall, is given as:

$$\frac{a}{dt}\left(\frac{3n_e \kappa I_e}{2}\right) = \frac{a}{dt}\left(\frac{3p_e}{2}\right) = \frac{1}{V}(P_{abs} - P_{loss}).$$
(14)  
Energy loss (per volume unit):

 $P_{loss} = P_C + P_{+,wall} + P_{e,wall} + P_{-,wall}$ , (15) where  $P_C$  is the energy lost by electrons in collisions with neutrals in the volume,  $P_{+,wall}$ ,  $P_{e,wall}$ ,  $P_{-,wall}$  the loss of kinetic energy on the walls by fluxes of positive ions, electrons and negative ions.

The energy losses in elastic and inelastic collisions are equal:

$$P_{C} = eV n_{e} \left( \sum_{X} \left( n^{X} K_{iz}^{X} \varepsilon_{iz}^{X} + \sum_{k}^{N_{ex}} n^{X} K_{ex,k}^{X} \varepsilon_{ex,k}^{X} + n^{X} K_{el}^{X} \frac{3m_{e}}{m^{X}} T_{e} \right) \right), \quad (16)$$

where  $\varepsilon_{iz}^{X}$  (eV) and  $K_{iz}^{X}$  (m<sup>3</sup>s<sup>-1</sup>) – energy and ionization rate constant for particles of type X,  $\varepsilon_{ex,k}^{X}$   $\mu$   $K_{ex,k}^{X}$  are the energy and ionization rate constant of *k*-th process of exciting particles of a type X,  $K_{el}^{X}$  - elastic scattering rate constant of X particles.

## 4. Results

The numerical calculation of plasma parameters of FMICP were performed for different values of the power absorption rates  $P_{abs}$ , discharge current  $I_d$ , and argon  $Q_{Ar}$  and molecular chlorine  $Q_{Cl2}$  flow rates. These parameters can be maintained manually during the experiments. As a result of calculations, the plasma content, i.e. the densities of neutral, excited and charged particles, as well as the electron temperature  $T_e$  were calculated.



Fig. 2. Charged particle density dependences on argon  $Q_{Ar}$  and chlorine  $Q_{Cl2}$  flow rates for small reactor,  $P_{abs} = 1 \text{ kW}, p = 1 \text{ Pa}.$ 

Plasma parameters for two scales FMISPs, i.e.  $R_I = 10$  cm, height  $L_I = 10$  cm and  $R_2 = 35$  cm, height  $L_2 = 50$  cm, were calculated. The dependencies of densities of electrons  $n_e$ , positive chlorine ions  $n_{Cl+}$ , negative chlorine ions  $n_{Cl+}$ , positive molecular chlorine ions density  $n_{Cl+}$ 

and argon ions are presented in Fig. 2 for small reactor and in Fig. 3 for large reactor. With an increase of argon flow rate  $Q_{Ar}$ , the density of argon ions and electrons increases while the density of chlorine ions decreases. The electric quasi-neutrality condition is fulfilled. The absolute values of the densities of electrons and chlorine positive ions have approximately the same values ( $10^{12}$  cm<sup>-3</sup>) for small and large reactors at  $P_{abs} = 1$  kW and 10 kW, correspondingly.



Fig. 3. The charged particle density dependences on argon  $Q_{Ar}$  and chlorine  $Q_{Cl2}$  flow rates for large reactor,  $P_{abs} = 10$  kW, p = 1 Pa.



Fig. 4. The electron temperature  $T_e$  dependencies on argon  $Q_{Ar}$  and chlorine  $Q_{Cl2}$  flow rates for small reactor case (solid line) and large reactor case (dashed line), p = 1 Pa.

Fig. 4 presents the electron temperature dependencies on argon  $Q_{Ar}$  and molecular chlorine  $Q_{Cl2}$  flow rates for the conditions presented in Fig. 3 and Fig. 4. In large reactor, the electron temperature is lower than that in small reactor due to lower absorbed power density, i.e. the ratio  $P_{abs}/V$ . However, these conditions are enough to produce dense plasma for plasma processing.

### 5.Conclusion

A simplified global model of FMICP in Ar/Cl<sub>2</sub> mixture has been developed. The model is based on the solution of the balance equations for neutral and excited active argon/chlorine plasma species together with the energy balance equation and the quasi-neutrality condition. The Maxwellian electron distribution function is assumed.

The numerical calculations of plasma parameters of FMICP were performed for different scales of the discharge reactor, different values of the absorbed power  $P_{abs}$ , and argon  $Q_{Ar}$  and molecular chlorine  $Q_{Cl2}$  flow rates. As a result of calculations, the plasma content, i.e. the densities of neutral, excited and charged particles, as well as the electron temperature  $T_e$  were calculated.

The results show that the plasma densities have approximately the same values  $(10^{12} \text{ cm}^3)$  for small and large reactors at  $P_{abs} = 1 \text{ kW}$  and 10 kW, correspondingly. The electron temperature decreases with the chlorine dilution that is the common feature for this type of discharges.

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