Influence on argon metastable atoms and on electron densities of
dust formation in argon + 6% acetylene CCP plasmas

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Basic properties and application of dusty plasmas (plasmas containing nano- to micro-sized particles) have been discussed by many authors in the literature \cite{1}. Dusty plasma models predict important changes in plasma characteristics (electrons temperature and density, as well as on excited species and radicals densities) when nanoparticles are formed, or introduced in a rare gas plasma.

In the present work, we have studied the behaviour of metastable atoms and electron densities ($N_e$) in the discharge and in the afterglow of a 0.1 mbar, 8 sccm flow rate argon plus 6% $\text{C}_2\text{H}_2$, symmetrical CCP discharge (30 cm electrodes diameter, 7 cm gap). The 13.56 MHz RF power was ranging from 10 to 80 W. An external cavity Diode Laser (DL) tuned on argon line (1s$^5$-2p$^7$) at 772.38 nm was used to measure the absolute density of Ar* metastable atoms by optical absorption and to record their axial distribution with LIF experiments, respectively. The electron density $N_e$ was measured with a 26.5 GHz µ-wane interferometer. The gas temperature, $T_g$ has also been deduced from the Doppler width of the 772.38 nm absorption line profile \cite{2}. In all conditions, $T_g$ was found to be at room temperature with 15 W of RF power and it increased up to 360 K in the bulk of the plasma when dust particles have been formed at 50 W of RF. To study the kinetics of formation and destruction of these species, the RF power was square wave modulated at 100 Hz. At fixed RF power, four different situations have been studied: 1- pure argon plasma (Ar); 2- argon/acetylene plasma before dust formation (Ar/$\text{C}_2\text{H}_2$); 3- argon/acetylene plasma after dust particles have been formed and grown to reach a steady-state (Ar/$\text{C}_2\text{H}_2$/dust); 4- after step 3, the plasma is maintained running but the acetylene flow is stopped. This way, dust particles remain inside a “pure” argon plasma (Ar/dust). Figure 1 shows at 20 W RF power the time evolution in the bulk plasma of Ar* metastable and electron densities for the four plasma conditions.

In pure argon, the steady-state Ar*($^{3}\text{P}_2$)=$2\times10^{16}$ m$^{-3}$ and $N_e$=$4\times10^{15}$ m$^{-3}$ densities are reached during the discharge and the decay time in the afterglow is about 3 ms for Ar*. Increasing the RF power to 80 W leads to an almost factor of 4 enhancement of the electron density but it has almost no influence on the steady-state density and the decay time in the afterglow of Ar* atoms. The production, by e-impact excitation from the ground state and the destruction, by stepwise ionization of Ar* atoms, balance each other, resulting in an electron density independent Ar* metastable density.

When 0.5 sccm of $\text{C}_2\text{H}_2$ is added to the feed gas, the Ar* density and its decay time in the afterglow are both reduced by a factor of about 20. This diminution is due to the quenching of metastable atoms by $\text{C}_2\text{H}_2$, whose rate coefficient is $k_q$=$5.6\times10^{16}$ m$^{-3}$s$^{-1}$ \cite{3}. However, the measured decay time of Ar* (a few 100s of µs) indicates that $\text{C}_2\text{H}_2$ must be highly dissociated by the plasma and its dissociation rate is about 95% at 20 W and 95% at 80 W. But the steady-state electron density remains almost unchanged whereas its decay in the afterglow becomes much faster, because the attachment of electrons to acetylene and to radicals formed by its dissociation.

A few minutes after having started $\text{C}_2\text{H}_2$ flow, nanoparticles are formed (Ar/$\text{C}_2\text{H}_2$/dust). This leads to an almost 50 times enhancement of the steady-state density of metastable atoms and a decrease by an order of magnitude of $N_e$. The decay of Ar* atoms in the afterglow becomes much slower, indicating an even higher degree of dissociation of
C₂H₂ in this plasma than before dust formation. At 80 W, the dissociation rate of C₂H₂, deduced from the decay rate in the afterglow of Ar⁺ density, reaches about 99%. Despite the diminution of electron density and a higher destruction rate of Ar⁺ atoms, compared to the pure argon plasma, the steady-state density of Ar⁺ is surprisingly superior to its value in pure argon. The origin of this enhancement of Ar⁺ density are: first, in the presence of dust particles the plasma becomes more resistive and the RF power is mostly dissipated in the bulk plasma [4]. Second, the excitation zone which was near the electrodes in the wave riding α mode, becomes more homogeneous with dusts. Using LIF technique, we have observed his modification of the axial distribution of Ar⁺ density. But he most astonishing is the strong enhancement of Nₑ in the afterglow, which indicates an electron production mechanism in the afterglow.

When the C₂H₂ flow is stopped, nanoparticles remain trapped between electrodes for several minutes and the behaviours of Ar⁺ and Nₑ are again modified. The steady-state Ar⁺ density increases even more and becomes about an order of magnitude larger than in pure argon and the decay rate in the afterglow of metastable atoms becomes comparable to its value in the pure argon plasma. The origin of this large difference relays on the enhancement of Tₑ [5] combined to the spatially homogeneous excitation of Ar⁺ atoms in γ regime, associated to the presence of dust. In pure argon, in which the excitation is in α regime, excitation zones are localized near the electrodes. This behaviour has also been observed by LIF experiments. Different mechanisms responsible for these important modifications in Ar⁺ and Nₑ behaviour will be discussed. The preliminary results reported in Ref. [6] indicate that under our experimental conditions, the de-excitation of metastable atoms on the surface of dust particles and ions, under the impact of Ar⁺ metastable atoms.


Fig. 1 Variation during the RF pulse (0-5 ms) and in the afterglow of Ar⁺(3P₂) density (left) and electron density (right) at 20 W of RF power in pure argon (black), Ar + C₂H₂ (red), Ar + C₂H₂ with dust (green) and Ar + dust (blue) at the reactor centre.