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Experimental investigation of electron heating modes in capacitively coupled radio-frequency oxygen discharge

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Abstract

A Langmuir probe has been used to investigate electron heating mechanisms in a capacitively coupled oxygen discharge over a wide pressure range (50 – 800 mTorr) at a fixed applied power (200 W). Evidence presented here from experimentally obtained electron energy distribution functions (EEDFs) illustrates discharge transition from collisionless (stochastic) to collisional (ohmic) dominant regime with increasing oxygen pressure. The discharge exhibited a bi-Maxwellian EEDF in the collisionless regime, dominated by stochastic heating whereas Druyvesteyn-like EEDFs in the collisional dominant regime. Moreover, in the transition between these two regimes, parameters such as electron density, effective electron temperature and electron-neutral collision frequency exhibited significant variations.

Keywords: capacitively coupled plasmas, oxygen discharge, Langmuir probe, electron energy distribution function, electron heating

1. Introduction

Radio-frequency (rf) capacitively coupled plasma (CCP) discharges are a key technology in plasma applications for dry etching [1-3] and thin film deposition [4-7]. CCP's are a widespread method for generating technical plasmas where electron populations are heated via modes which are strongly pressure dependent. Technologically relevant plasmas are typically electronegative in nature, however, to date, the pressure dependence of electron behaviour in such important discharges has not been extensively studied experimentally, and is therefore a key motivation for this work. This endeavour is particularly timely as critical dimension in semiconductor devices approach the atomic scale [8] with increasing demand for understanding & control of plasma & surface dynamics.

Variations in gas pressure in an rf discharge can lead to a marked change in the electron heating regime [9]. At moderate to higher pressures electron populations heated via an externally applied rf electric fields deposit energy to other gas constituents through direct collisions. This mechanism of electron heating is often referred as collisional or ohmic where electron collisions resemble resistance heating of a current passing through a conductor. When a discharge is sustained through collisional dominant heating, it is commonly referred as alpha (α) mode which most significantly occurs at moderate to higher pressures with low applied powers/voltages. At large applied power, where significant fraction of power is absorbed by ions which accelerate across the sheath by the large electric field and release secondary electrons, by ion impact on the powered electrode, which can dominate the ionization balance with the discharge is said to be in gamma (γ) mode. In the lowest pressure capacitive discharges, stochastic or collision-less heating becomes important as the electron mean free path extends towards the dimensions of the reactor. Here, electrons can gain additional kinetic energy via momentum transfer from the oscillating plasma boundary sheath [9-11]. This mechanism is key to sustain many low pressure discharges by heating a fraction of the electron population to relatively high energies. Another electron heating mode demonstrated in electronegative

46 discharges and some molecular discharges is called the drift-ambipolar mode [12, 13] in which electrons are
47 accelerated by strong drift and ambipolar fields at the discharge centre and sheath edges.

48 The Langmuir probe [14] is the most widely used diagnostic tool for investigating behavior of charged species
49 within a plasma which can be used to understand electron heating. There are numerous reports on the kinetic
50 behavior of electrons using Langmuir probes in various discharges [15-20] and in particular capacitively-coupled
51 plasmas [2, 9-11, 21-28], which is of interest here. Perhaps surprisingly given their widespread use in industrial
52 processes there have been few reports of the kinetic behavior of electrons in oxygen plasmas [25, 26, 29].
53 Gudmundsson *et al* [30] simulated the evolution of the electron heating mechanism in a capacitively coupled oxygen
54 discharge as a function of pressure using 1-D object oriented particle-in-cell Monte Carlo collision and found ohmic
55 heating to dominate at the plasma bulk at low pressure (10 mTorr) whereas at higher pressures (50-500mTorr),
56 electron heating occurs mainly in the sheath regions. A detailed investigation using a Langmuir probe was carried
57 out recently by Kechkar *et al* [31] who measured electron density, effective electron temperature and electron
58 energy probability function (EPPF) in a capacitively coupled oxygen discharge and showed that any changes in the
59 electron heating mechanism would cause variation in the EPPF measured in the plasma bulk. Importantly that work
60 revealed the threshold power for discharge transition into the γ - mode to be 50 W. Interestingly, with an increase in
61 pressure, the discharge was shown to transit from collisionless to collisional regime at low power (30 W) whereas at
62 higher power (200 W) the transition was from collisionless to γ -mode. However, the pressure variation studies on
63 electron plasma parameters in capacitive oxygen discharge were limited to 200 mTorr due to probe limitations used
64 in that work. The primary aim of this work is to overcome those probe limitations by altering the position of
65 components of probe assembly in the Langmuir probe and measure the variation of plasma parameters such as
66 electron density (n_e), effective electron temperature (T_{eff}) and electron energy distribution function (EEDF) over a
67 wide pressure range for an extended correlation with electron heating mechanism which in turn can further improve
68 the understanding of processing plasmas.

69 Moreover, transitions in the electron heating mechanism in rf discharge have been shown to be correlated with
70 current-voltage characteristics such as the phase shift angle between current and discharge power. Lisovsky *et al*
71 [32-35] have demonstrated for capacitive discharges that the behavior of these global current-voltage characteristics
72 varies with the electron heating mode. Behavior of these current-voltage characteristics can be monitored using an
73 inline impedance probe positioned below the driven electrode.

74 In order to gain fundamental insight into key plasma processes, detailed measurements of the pressure
75 dependence of electron populations in capacitively coupled discharges at various is clearly important. To this end,
76 the time averaged EEDF, n_e and T_{eff} were determined for a wide range of pressures (50-800 mTorr) at fixed power
77 (200 W) applied rf power using a Langmuir probe in an oxygen capacitively couple plasma.

78

79 **2. Experimental tools and diagnostics**

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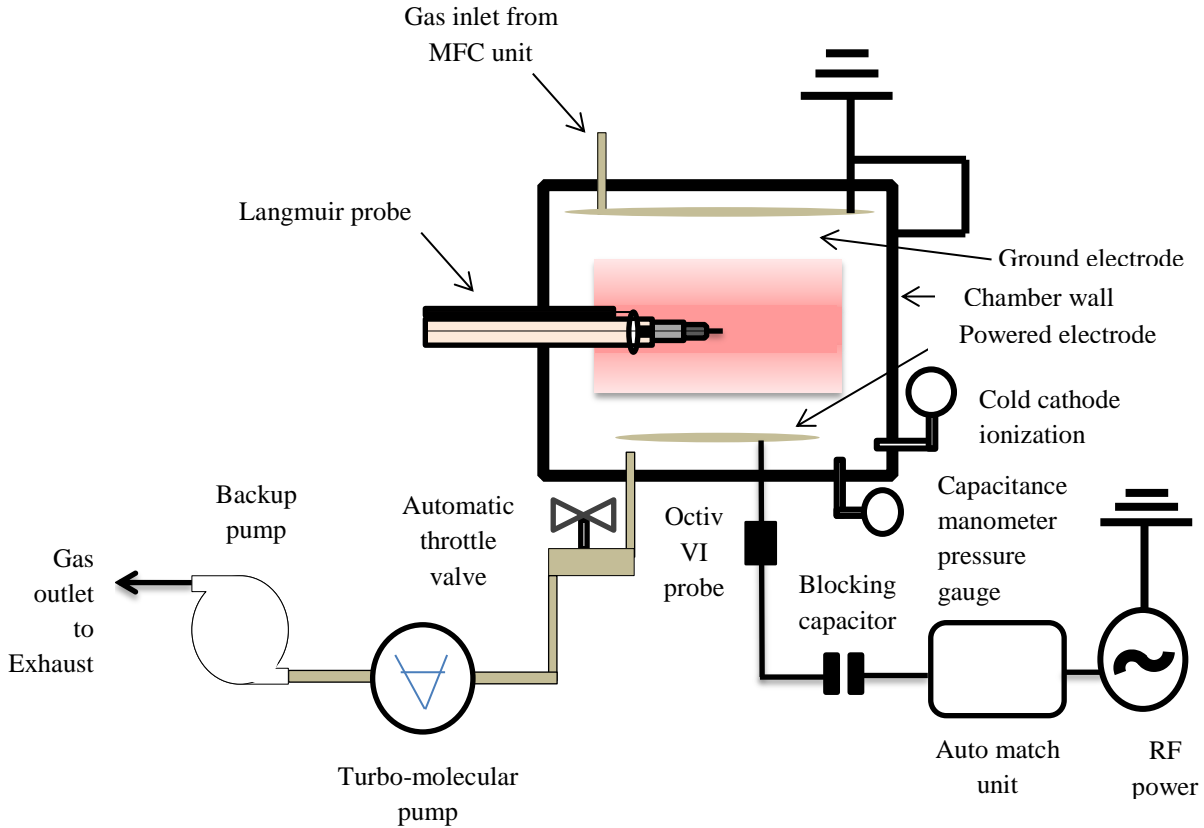
81 In this section details of the plasma reactor is given in section 2.1. Details of the Langmuir probe system utilized is
82 given in section 2.2. Section 2.3 describes a I-V probe which is used to corroborate Langmuir probe measurements.

83

84 *2.1 Reactive ion etcher - Plasmalab system 100*

85 Figure 1 show a schematic of the Oxford Instruments Plasmalab System 100 [36] rf excited (13.56 MHz) parallel
86 plate capacitively-coupled plasma (CCP) source used in this work. It consists of lower powered/driven electrode and
87 grounded electrode of diameters 205 mm and 295 mm respectively with a 50 mm separation gap. Power was applied
88 to the lower electrode through an auto-match unit. The feedstock gases from the mass flow controller (MFC) unit
89 were fed to the chamber through the shower-head provided on the upper electrode. The reactor walls and electrodes
90 are made of aluminium alloy (6082). An automatic variable throttle valve was located beneath the powered electrode
91 to control the pressure. An Alcatel (ATP400HPC) turbo-molecular pump, with a pumping speed of 400 l/s, was used
92 to evacuate the chamber and backed by an Alcatel rotary pump with a pumping speed of 6 l/s. The MKS (627B-
93 15968) capacitance manometer and Edwards cold cathode ionization (Edwards AIM-S-NW25) pressure gauges

94 were used with this chamber to measure the process pressure and the base pressure (4.5×10^{-5} to 5×10^{-6} Torr)
 95 respectively.
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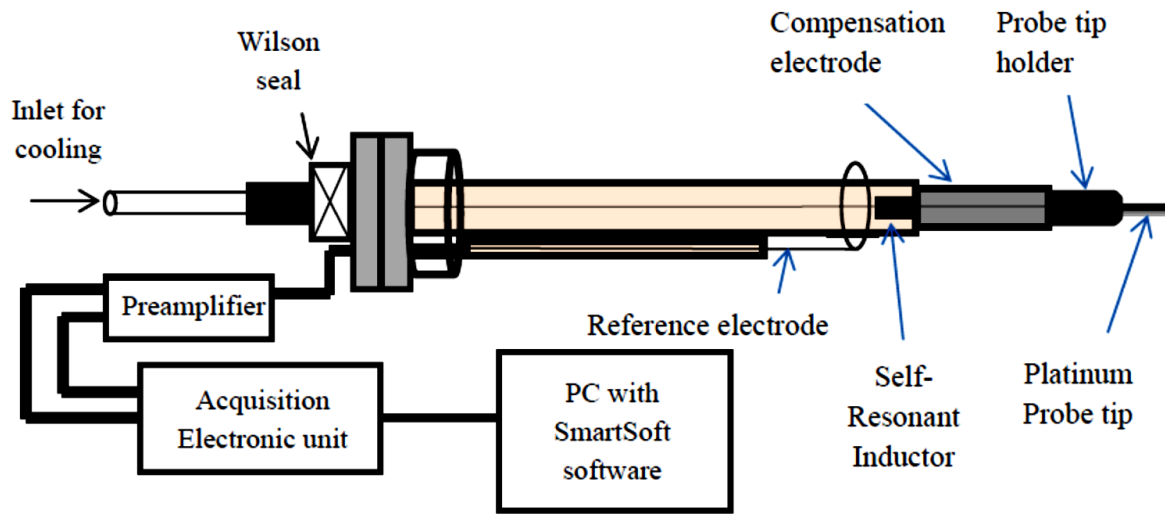


98
 99 Figure 1 Schematic of Oxford instruments Plasmalab system 100

100 *2.2 Langmuir probe-The SmartProbe*

101 The SmartProbe, which is a commercial Langmuir probe system used in this work, is shown schematically in
 102 Figure 2. Developed by Scientific Systems [37], the SmartProbe consists of a Langmuir probe, reference probe with
 103 preamplifier, acquisition electronic unit (AEU) and SmartSoft software. The probe tip, which constitutes the active
 104 part of the Langmuir Probe, is made of platinum wire with a 0.15 mm diameter and 11.0 ± 0.5 mm in length. The
 105 probe is inserted into the chamber from the side and positioned at radial center (2.5 cm above the powered electrode)
 106 for most measurements. The advantage of using a platinum wire is to minimize the contamination level preventing
 107 distortion of the I-V characteristic. The probe tip is connected to the circuitry through a coaxial cable covered using
 108 ceramic shaft as to prevent it making contact with the plasma. The I-V characteristic was obtained by measuring the
 109 current drawn by probe at varying probe voltages. The plasma parameters calculated using I-V characteristics
 110 assumes that voltage between the probe and the plasma contains only direct current (DC) component and no
 111 significant rf component. If there is a contribution from rf component, this would produce distorted I-V data and the
 112 plasma parameters measured from this would be incorrect. In order to eliminate the rf component, a compensation
 113 electrode along with a set of inductors forms the part of the probe circuitry. The function of this so-called rf
 114 compensation is to keep the probe-ground impedance higher in comparison to impedance between the probe and the
 115 plasma. To achieve this, a set of self resonant inductors, located close to probe, are tuned to 13.56 MHz and related

116 harmonics to increase the probe impedance. The inductors were cooled using air through a 4 mm diameter teflon air
 117 inlet tube. The SmartProbe also consist of reference electrode (reference probe) which tracks and corrects for any
 118 low-frequency or DC shifts in the plasma potential when the probe tip is biased. Low frequency noise, which
 119 originates within the discharge due to instability in applied rf voltage, can cause distortion in the probe
 120 characteristics [9] as this instability in applied rf voltage can result in the probe voltage noise and drift. The
 121 reference probe is made of stainless steel and forms a wire loop around the main shaft of the Langmuir probe. The
 122 major part of the reference probe in the plasma was covered with a ceramic shaft for insulation. The other end of the
 123 reference probe is connected to probe circuitry through an electrical feed-through. In this work the probe voltage
 124 was varied from -95.0 to 95.0 V and the number of samples per point was chosen between 100-200. Moreover, I-V
 125 characteristics measured in this work involved an average of 10-20 sweeps. The probe tip was cleaned before each
 126 sweep, by biasing the tip at high positive voltage (electron bombardment) for $\approx 2-3$ s based on plasma density, to
 127 ensure it was free from contamination and changed regularly to minimize the effect of etching by the oxygen
 128 discharge.



129
 130

131 Figure 2 Schematic of the Smart-Probe – commercial Langmuir probe system used in this work.

132

133 The Druyvesteyn method [38] was used in this work to determine n_e , T_{eff} and EEDF to allow for non-Maxwellian
 134 distributions. Alternative methods often assume a Maxwellian EEDF which can lead to a high degree of uncertainty
 135 in the measured plasma parameters in low-pressure discharges [39]. Moreover, this method was successfully
 136 employed for previous studies of this discharge [31] and elsewhere [9] for EEDF measurements. The EEDF, $f(\varepsilon)$,
 137 obtained using this method is given by,

$$f(\varepsilon) = \left(\frac{d^2 I}{dV^2} \right) \frac{2m_e}{e^2 A} \left(\frac{2e\varepsilon}{m_e} \right)^{\frac{1}{2}} \quad (1)$$

138 where ε is the electron energy, I the electron current, V the probe bias, m_e the mass of electron, e the charge of an
 139 electron and A is the surface area of probe. The electron density and effective electron temperature can be
 140 determined using,

$$n_e = \int_0^\infty f(\varepsilon) d\varepsilon \quad (2)$$

$$T_{eff} = \frac{2}{3n_e} \int_0^\infty \varepsilon f(\varepsilon) d\varepsilon \quad (3)$$

141 The electron distribution is often illustrated as an electron energy probability function, $f_e(\varepsilon)$, (EEDF), where,

$$f_e(\varepsilon) = \frac{f(\varepsilon)}{\sqrt{\varepsilon}} \quad (4)$$

142 The ion current was removed from total current by the procedure described by Steinbruchel [40] to obtain
 143 electron current which is required by the Druyvesteyn method to determine EEDF.
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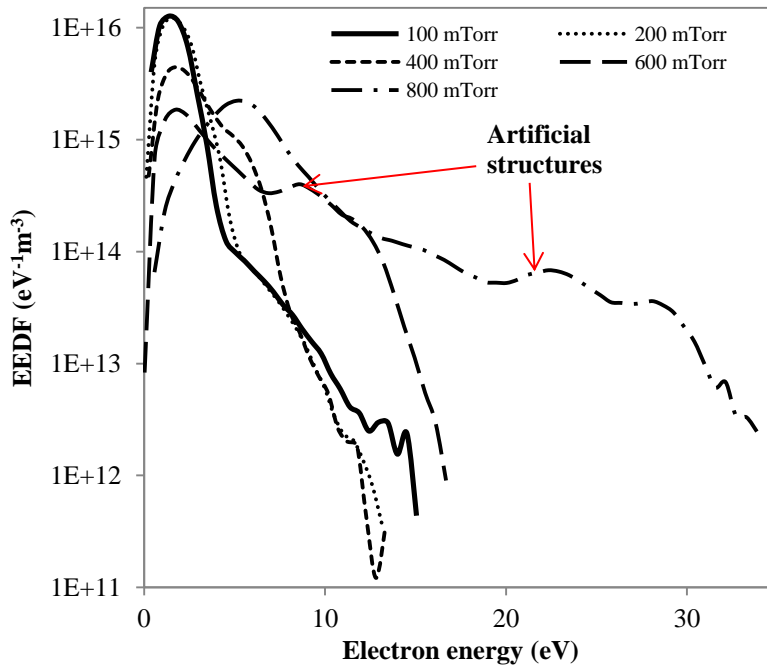
145 *2.3 Octiv VI probe*

146 In this work, the phase shift angle between the plasma current and voltage was measured to corroborate
 147 Langmuir probe predictions of changes in electron heating mode. An Octiv VI probe developed by Impedance Ltd
 148 [41] is employed here. The inline probe was installed the between powered electrode and automatic matching unit.
 149 The probe incorporates capacitive sensing for voltage acquisition with inductive current pickups in combination
 150 with analogue-to-digital conversion (ADC) circuitry for signal digitisation. The fast Fourier transform is utilised for
 151 frequency domain analysis and extraction of phase data via dedicated logic performing noise reduction.
 152

153 **3 Results and discussion**

154
 155 In this section, we present evolution of EEDF in oxygen plasma for a wide range of pressures measured using
 156 Langmuir probe to understand the electron heating mechanism. In section 3.1 inline arrangement of a main probe &
 157 reference probe/electrode is considered to understand the structures in measured EEDF. In section 3.1.1,
 158 investigation of structures in measured EEDF was extended and correlated to positions of reference probe and
 159 compensation electrode with respective to main probe tip. Finally in section 3.2 the pressure sensitivity of EEDF's
 160 using modified arrangement is presented with analysis of the heating behaviour.
 161

162 *3.1 Investigation of “artificial” structures in measured EEDF*



163
 164 Figure 3 “Artificial” structure(s) observed on EEDFs measured (using standard Langmuir probe setup shown in
 165 Figure 4 (a)) as a function of O₂ pressure at 200 W rf power in capacitively-coupled plasma.
 166

167 In figure 3 we see the evolution of EEDF with increase in oxygen pressure at constant applied rf power of 200
168 W. As pressure increases a transition to a different electron heating regime is apparent here. At pressures less than
169 200 mTorr, EEDFs were found to be bi-Maxwellian (see figure 3). Similar bi-Maxwellian characterized EEDFs
170 have been observed previously in capacitive oxygen discharge for pressures less than 100 mTorr [25]. In this
171 scenario the discharge was predominately controlled by stochastic heating, where electrons predominately gain
172 energy from the oscillating rf sheaths at plasma-sheath boundaries [9, 42]. It has been well established in both
173 electropositive discharges of argon [9], helium [9] and electronegative chlorine discharge [43], that ohmic heating
174 dominates with increasing pressure and the EEDF evolves from a bi-Maxwellian to a Druyvesteyn type distribution
175 [8, 49]. In this investigation for pressures greater than 200 mTorr the shapes of EEDFs were found to deviate from
176 bi-Maxwellian indicating that the discharge is clearly in a transition mode. At higher pressures (as the collision
177 frequency increases shown in Figure 9) the plasma bulk tends to become more collisional/resistive. This allows the
178 rf electric field to penetrate into the plasma bulk, where it can heat low energy electrons contained by the ambipolar
179 field. Eventually, with further increase in gas pressure, discharge enters collisional heating regime where ohmic
180 heating is the main power dissipation process and low-electron energy group in plasma bulk are heated by the rf
181 electric field [42].

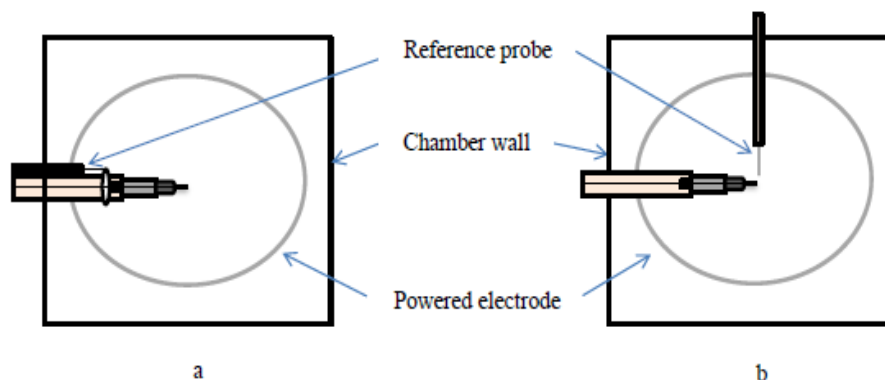
182 Here, at oxygen pressures greater than 400 mTorr, “artificial” structures (or dips) were observed in measured
183 EEDFs. Indeed at the highest pressure investigated here (800 mTorr) the high energy EEDF tail is found to increase
184 significantly (as shown in figure 3) which is a behavior not commonly observed in oxygen discharge [25, 30]. It is
185 important to note that this structure or dip in EEPF is different to that usually reported in nitrogen plasma in the
186 energy range of 2-4 eV which corresponds to loss of electrons due to inelastic collisions involving vibrational
187 excitation of the nitrogen molecules [44]. Capitelli *et al.*, [45] reported that super-elastic collisions between
188 electrons and molecules in a metastable state can cause additional peaks or structures in an electron energy
189 distribution. Indeed in capacitive oxygen discharge, metastable molecule $O_2(a^1\Delta_g)$ plays a significant role in the
190 governing heating mechanism [46]. However, no evidence of additional peaks or structure on EEPFs tails in oxygen
191 has been reported to date. This includes computational studies such as the 1-D object-oriented PIC/MCC code
192 ‘oopd1’, which incorporated the metastable molecule $O_2(a^1\Delta_g)$ along with numerous other species in the reaction set
193 [30, 47]. Appearance of structures and/or high energy tails in EEDF can be possible if there is any distortion in
194 obtained I-V characteristics [9]. This is, therefore investigated extensively in the next section.

195

196 3.1.1 Investigating the position of compensation electrode and reference probe

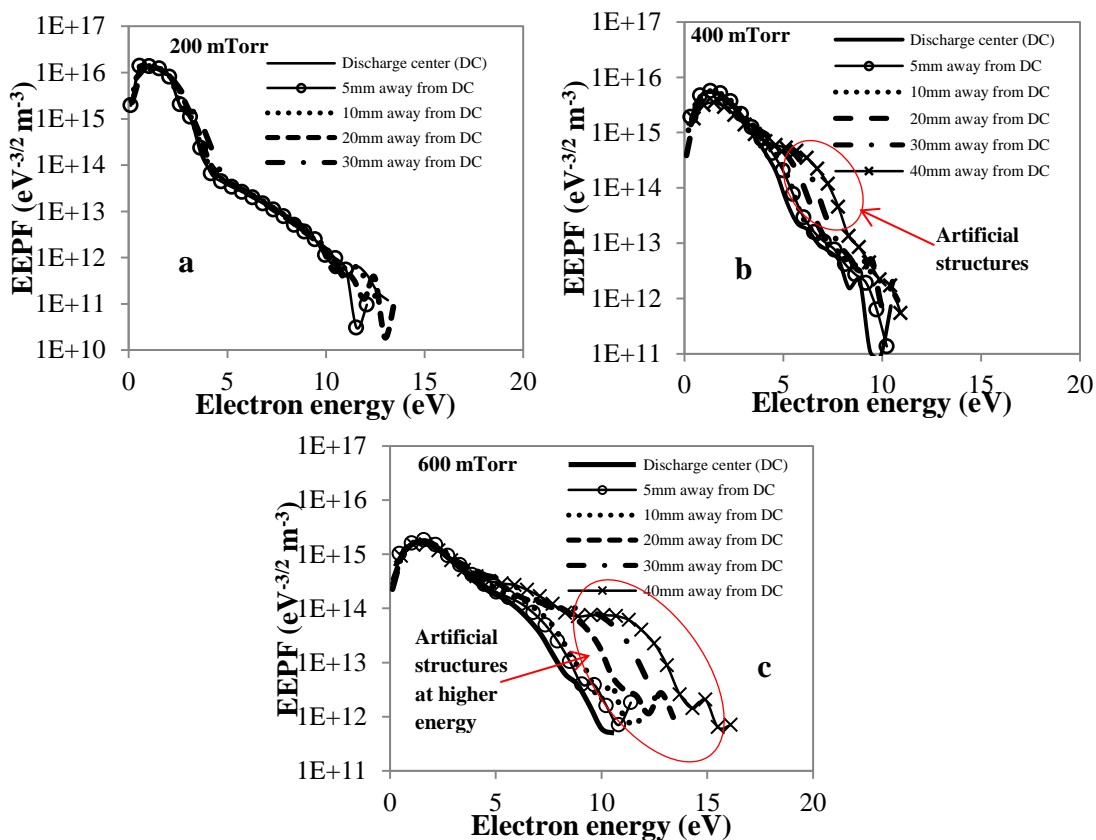
197 Structures on an EEDF can be the result of distorted probe I-V characteristic and can be caused by such factors
198 as probe contamination, inadequate rf compensation and poor compensation of any low frequency plasma-potential
199 noise and drift [9, 48] due to instability in the rf applied voltage. Distortion of probe I-V data would be possible if
200 any part of probe assembly- the probe tip, probe holder, reference electrode/probe and compensation electrode - is
201 not in the plasma. Here the Langmuir probe arrangement was modified as shown in figure 4 (b) from the standard
202 setup shown in figure 4 (a) to ensure the reference probe always held in plasma bulk so as to avoid any shift in
203 plasma potential due to plasma sheath resistance. In the standard Langmuir probe arrangement shown in figure 4 (a),
204 the wire loop of reference probe is 20 mm in length and is positioned at a distance of ≈ 31 mm from probe tip. At
205 higher oxygen pressures, part of the reference probe in standard Langmuir probe arrangement was visually found to
206 be out of the plasma bulk due to discharge confinement and eventually, fails to accurately account for any changes
207 to plasma potential when probe tip is biased. To verify any distortion to I-V data, the position of reference probe was
208 fixed at the radial center in the plasma bulk, as shown in figure 4 (b) and the main probe (excluding reference probe
209 here) was moved along the radial direction. Structures were observed on measured EEPFs especially at 400 and 600
210 mTorr for the probe to be furthest away from discharge center shown in figure 5 (b) & (c). With an increase in
211 oxygen pressure, discharge confinement was visually evident. Moreover, asymmetric capacitive oxygen discharges,
212 which are of interest here, are found to become increasingly symmetric with increase in pressure as evidenced by a
213 drop in the dc self-bias voltage [49]. Particularly at 600 mTorr pressure, as the main probe was moved away from
214 discharge center, additional structure began to appear due to the compensation electrode (≈ 15 mm in length and \approx
215 24 mm away from probe tip) no longer being in the plasma bulk. When the probe tip was positioned ≈ 30 -40 mm

216 from discharge center, most part of compensation electrode was out of the plasma bulk (which is readily visually
 217 observed). It can be concluded that the compensation probe not being in the plasma bulk distorts the measured I-V
 218 trace which resulted in additional “artificial” structures at higher energy on measured EEPFs shown in figure 5 (b) &
 219 (c).



220
 221 Figure 4 Schematic of Langmuir probe arrangement (a) standard setup used to obtain EEPFs shown in figure 3 ; (b)
 222 modified setup with reference probe held close to main probe.
 223

224 This inadequate referencing to the main probe produced additional structures and/or enhanced tail on EEDF
 225 shown in figure 3 due to distorted probe I-V characteristic. Therefore, to improve probe referencing especially at
 226 higher pressures, modified Langmuir probe was used for all subsequent data presented here.



227
 228 Figure 5 Measured EEPFs as a function of distance from discharge center (DC) in radial direction at (a) 200, (b) 400
 229 & (c) 600 mTorr oxygen pressures operated at 200 W with Langmuir probe set up as in figure 4 (b). Additional

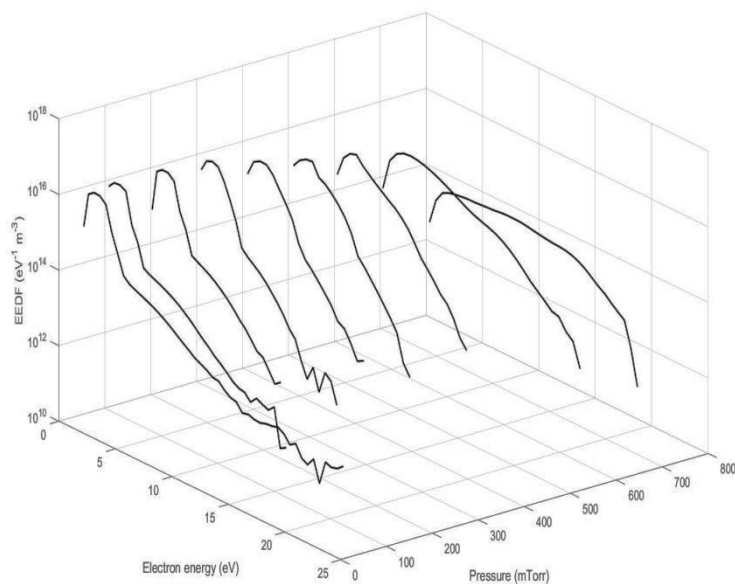
230 structures and enhanced high energy tails in EEDFs (in 400 & 600 mTorr) caused due to distorted I-V
231 characteristics.

232
233 *3.2 Investigation of EEDF evolution as a function of O₂ pressure with the modified Langmuir probe arrangement*

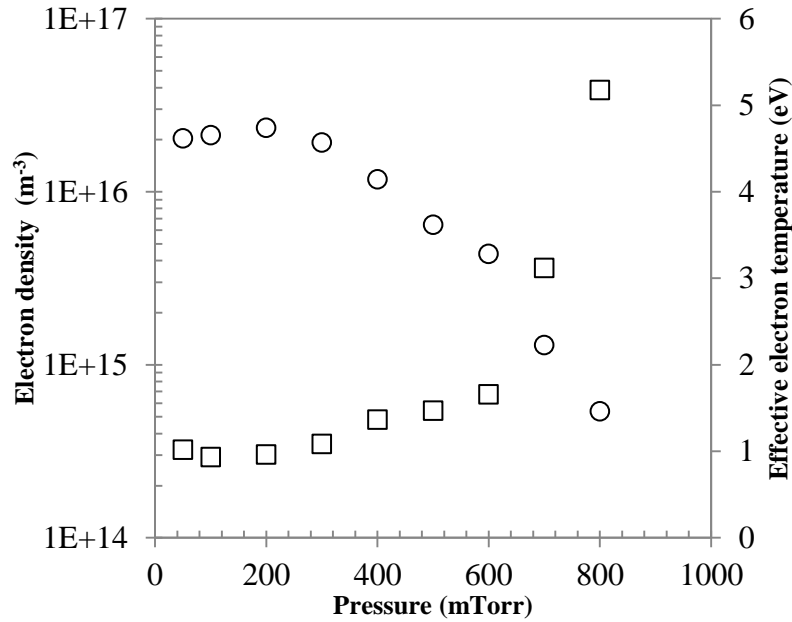
234 In this section we investigate EEDFs measured as function of O₂ gas pressures (50-800 mTorr) with the modified
235 Langmuir probe arrangement (as shown in figure 4(b)) for constant applied rf power of 200 W are shown in figure 6
236 with the corresponding plasma parameters such as electron density and effective electron temperature calculated
237 from the EEDF are shown in figure 7.

238 *3.2.1 Pressures 50-300 mTorr*

239 For pressures in the range 50-300 mTorr, the EEDFs exhibited a bi-Maxwellian shape with low bulk electron
240 temperature (≈ 0.4 - 0.5 eV) and high tail energy temperature (≈ 1 - 2 eV). A similar bi-Maxwellian characteristic was
241 reported by Lee *et al.*, [25] in a capacitive oxygen discharge between pressures 20-100 mTorr and is well known for
242 a capacitively coupled argon discharges at lower pressures [9, 10]. Such EEDF characteristics can result from
243 stochastic dominant electron heating, where electrons are heated by oscillating rf sheaths, which is typical in low-
244 pressure rf discharges [9]. In this heating regime, low energy electrons, with low electron-neutral collision
245 frequency, oscillate without any collisions are unable to gain energy either from weak rf electric field or from the
246 oscillating rf sheaths to overcome the ambipolar potential barrier and are confined in the plasma bulk, whereas the
247 high-energy electron group can overcome ambipolar potential and reach oscillating sheath boundary. Thereby, high-
248 energy electron group gain energy from the collision with oscillating rf sheath boundaries [9, 42].



249
250 Figure 6 Pressure evolutions of EEDFs measured using Langmuir probe with modified arrangement as in figure 4
251 (b) in O₂ discharge operated at 200 W applied rf power.



252

253 Figure 7 Variations of electron density n_e (\circ) and effective electron temperature T_{eff} (\square) measured using Langmuir
 254 probe with modified arrangement as in figure 4 (b) for different gas pressures in O_2 discharge operated at 200 W.
 255 Errors due to statistical uncertainties on these measurements arising from uncertainties in the I-V measurements of
 256 the Langmuir probe are insignificant and error bars are too small to indicate.

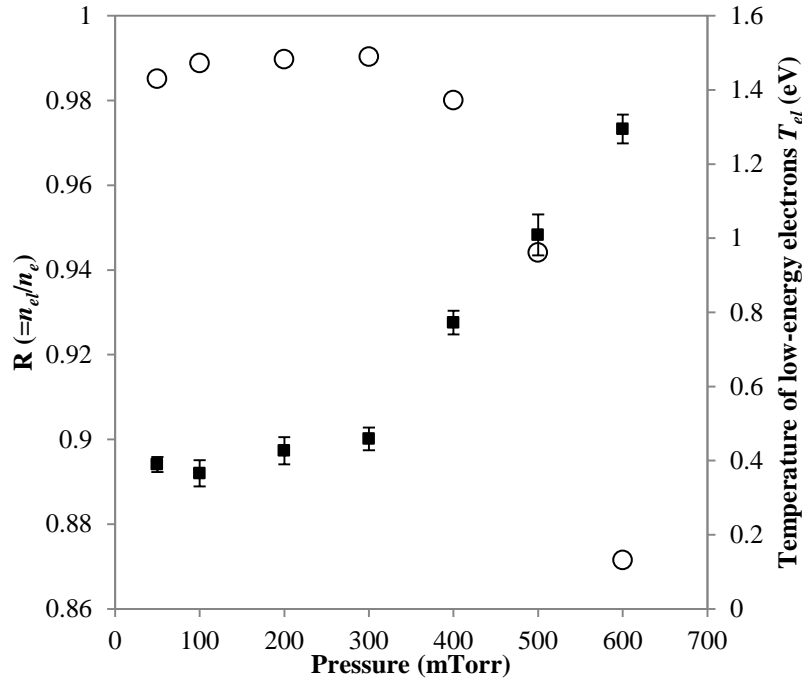
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3.2.2 Pressures 400-600 mTorr

258

An increase in gas pressure results in an increase in electron-neutral collision frequency and therefore,
 259 population of high energy electron group decreases, by losing their energy through inelastic collisions, with
 260 increasing gas pressures. This is clearly evident in EEDFs for pressures in the range 400-600 mTorr in figure 6.
 261 Moreover, the temperature of low-energy electrons, which constitutes to majority ($\approx 90\%$) of electrons in the plasma
 262 bulk, found to increase by a factor of approximately 3 as the pressure increased from 200 to 600 mTorr (shown in
 263 figure 8). As a result, the shape of EEDFs changed from bi-Maxwellian to Maxwellian distribution for pressures in
 264 the range 400-600 mTorr as shown in figure 6 and similar EEDF evolution was reported in capacitively coupled
 265 argon discharge [25]. Enhanced electron-electron collisions can also thermalize the electron population towards a
 266 Maxwellian distribution in the plasma bulk. This is mostly occurring at large electron densities and lower electron
 267 temperatures as electron-electron collision frequency proportional to $n_e T_{eff}^{-3/2}$ allows the EEDF to evolve to a
 268 Maxwellian. However, as seen in figure 7, the electron density is found to decrease and T_{eff} increases for pressures in
 269 the range 400- 600 mTorr and therefore, evolution to Maxwellian distribution here is less likely due to electron-
 270 electron collisions. Another important aspect of EEDF evolution to Maxwellian distribution as gas pressure
 271 increases can be collisional ohmic heating of low-energy electrons becoming significant and thereby, low-energy
 272 electrons gain sufficient energy to overcome the ambipolar potential field and reach the oscillating sheath to
 273 participate in collisionless heating. In contradiction, reports in the literature suggest that collisional heating is
 274 inefficient at higher pressures in a capacitively coupled oxygen discharge due to the weak electric field strength [25,
 275 30] and also the reported decrease in the temperature of low-energy electrons with increase in oxygen pressure [25].
 276 However, it has been found here that for pressures above 300 mTorr, the discharge begins to depart from a
 277 stochastic heating regime as the temperature of low-energy electrons, determined by the negative inverse slope of
 278 linear fit to the data of $\ln f_e(\epsilon)$ vs ϵ in the electron energy in the range 0.1 to 4 eV, is found to increase significantly
 279 (figure 8), due to enhancement of collisional heating. In addition, Lee M.H *et al.* [25] reported a decrease in the
 280 density ratio of low-energy electrons to total electrons with an increase in oxygen pressure. Here, ratio of low-

281 energy electron density, n_{el} , (determined by integrating EEDF in the electron energy range 0.1 to 4 eV) to electron
 282 density denoted by $R (= n_{el}/n_e)$ is found to increase only slightly (figure 8), with pressure until 300 mTorr. This small
 283 increment is due to contributions from inelastic collisions, however for pressures above 300 mTorr R was found to
 284 decrease significantly (shown in figure 8) as low-energy electrons, confined in the plasma bulk, gain sufficient
 285 energy to depart from ambipolar potential field due to enhancement of collisional heating. Moreover, an increase in
 286 low-energy electron temperature with gas pressure can be due to additional collisional heating. In contrast to this
 287 experimental result, simulated results in an oxygen capacitively coupled discharge by Gudmundsson *et al.*, [30]
 288 reported the opposite trend in EEDF behavior with increase in pressure (10- 500 mTorr) i.e., EEDF changes from
 289 Druyvesteyn-like (convex) at low pressures to bi-Maxwellian (concave) at higher pressures. Inconsistency between
 290 these two results may be to the fact that experimental measurements reported here were investigated by varying
 291 pressure at constant (or fixed) applied power whereas the simulations reported in [30] vary pressure at constant
 292 voltage (222 V) which shows more than an order increase in the bulk electron density. This increase in bulk electron
 293 density at higher pressures can give an impression that bulk plasma becomes more conductive and easier to screen
 294 the rf electric field. Apparently, low energy electrons in their simulations would have not gained enough energy to
 295 overcome the ambipolar potential barrier even as pressure increases and therefore, behavior of EEDF transition in
 296 their simulations differ to that reported here.



297
 298 Figure 8 Variations of density ratio of low-energy electron, n_{el} , to total electrons, n_e , R (○) and temperature of low-
 299 energy electrons T_{el} (■) with gas pressures in O_2 discharge operated at 200 W measured using Langmuir probe with
 300 modified arrangement as in figure 4 (b). Errors bar shown here are calculated using standard error on the slope of a
 301 linear fit to the data of $\ln f_{el}(\epsilon)$ vs ϵ in the electron energy range with 95% confidence interval.

302
 303 **3.2.3 Pressures above 600 mTorr**

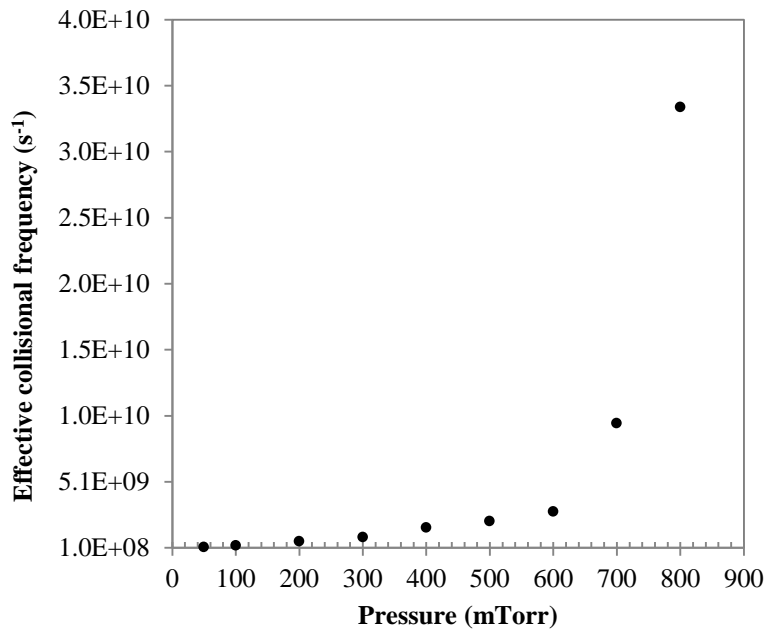
304 At pressures above 600 mTorr the discharge exists in collisional dominated heating regime where ohmic heating
 305 is the main rf power dissipation process. Moreover, the effective electron-neutral collision frequency, ν_{en} , as shown
 306 in figure 9, calculated using,

$$\nu_{en} = n_g \int_0^{\infty} \sigma_{en}(\epsilon) f_n(\epsilon) v(\epsilon) d\epsilon \quad (5)$$

307 is found here to saturate at lower pressures as collisional (ohmic) heating becomes less important. Here v_{en} obtained
 308 using normalized EEDF, f_n , momentum transfer collision cross-section, σ_{en} , for oxygen [50] and electron velocity,
 309 v . Here, at higher pressures (> 600 mTorr), $v_{en} \gg \omega$ ($= 2\pi \times 13.56$ MHz) for most electrons, therefore the power
 310 absorbed by a single electron in the rf electric field of amplitude E is

$$P_{abs} = \frac{e^2}{m_e v_{en}} \times \left(\frac{E}{\sqrt{2}} \right)^2, \quad (6)$$

311 is equivalent to the power absorption in the dc field. For low-energy electrons, the electron-neutral collision
 312 frequency will be small due to smaller electron velocity. Therefore, low-energy electrons are more effectively heated
 313 up as the energy gain by the low-energy electrons will be higher than high-energy electrons. Thus, the EEDF
 314 evolved close to Druyvesteyn for pressures above 600 mTorr as shown in figure 6 with abrupt raise in T_{eff} .

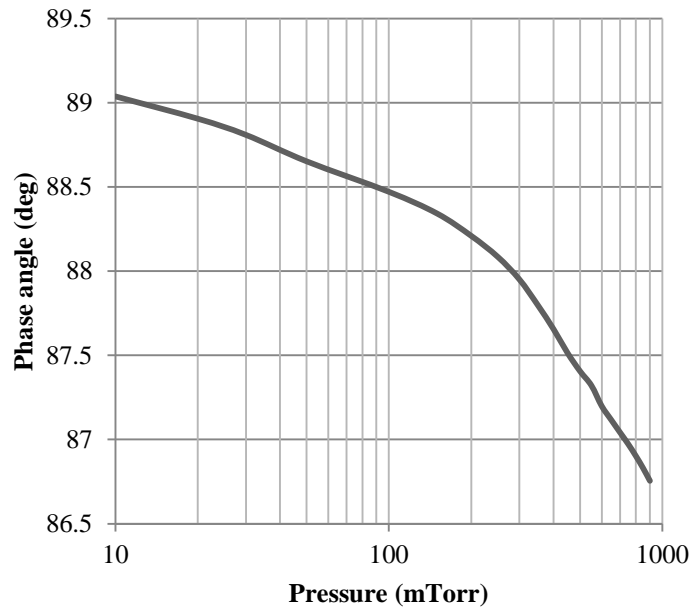


315
 316 Figure 9 Effective collision frequency calculated as a function of pressure in O₂ discharge operated at 200 W applied
 317 rf power.

318 3.2.4 Phase-shift angle measurements

319 Additional corroboration of this collisionless to collisional heating mode transition is given by the investigation
 320 of the phase shift angle measured using Octiv VI probe shown in figure 10. Two distinct slopes are evident here. A
 321 smaller rate of phase change at lower pressures (less than 300 mTorr) indicates a departure from collisional heating.
 322 With increase in pressure (>300 mTorr), the phase shift decrease indicates $v_{en} \gg \omega$, synonymous with greater rf
 323 power dissipation. Lisovskiy *et al* [32] found similar change in phase shift angle which found to decrease with
 324 oxygen pressure until it reached the end of α - γ transition in the capacitive oxygen discharge. A more straightforward
 325 comparison of phase-shift angle measurements with that of [32] is not be possible here due to facts that gas pressure
 326 was varied at constant rf power and discharge limitations. However, it can be derived that at a particular rf voltage,
 327 phase shift angle was found to decrease with increasing pressure, as in figure 10, reaching a minimum, where
 328 discharge is sustained through ohmic heating mechanism, which can also be referred as collisional or α -mode
 329 regime.

330



331

332 Figure 10 Phase angle measurements using Octiv VI probe for different gas pressures in O₂ discharge operated at
 333 200 W applied rf power. Errors due to statistical uncertainties (standard error on the mean) is of order 10⁻³.

334 **4 Conclusion**

335 The heating mode transition in a capacitively coupled oxygen discharge using a commercial Langmuir probe has
 336 been investigated at greater pressures than have been reported which has been made possible by modification the
 337 geometry of the reference probe. The electron heating mode in an oxygen discharge has been found to transit from
 338 collisionless to collisional dominant regime with increase in gas pressure and is reported for the first time in a
 339 capacitive oxygen discharge. Electron heating transition was marked with bi-Maxwellian shape EEDFs in
 340 collisionless regime (50-300 mTorr), Maxwellian EEDFs in the pressure range 400-600 mTorr and Druyvesteyn-like
 341 EEDFs in collisional dominant regime (greater than 600 mTorr). Such transition is due to electron behaviors, which
 342 at lower pressures gain energy by colliding with oscillating sheath whereas at higher pressures, low energy electrons
 343 are heated by rf electric field in the plasma bulk. Moreover, as the discharge begin to deviate from collisionless
 344 regime, the temperature of low-energy electrons was found to increase with pressure and the calculated effective
 345 electron-neutral collision frequency found to increase by an order magnitude when the discharge exists in collisional
 346 dominated regime. Additional corroboration from phase shift angle, measured using a VI probe, which has been
 347 found to decrease at faster rate for oxygen pressures ≥ 400 mTorr indicating the discharge transition from
 348 collisionless to collisional dominated regime.

349

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353

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