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A numerical and experimental investigation of an axially symmetric RF plasma

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Abstract

The characteristics of an atmospheric pressure, RF discharge in air were determined and compared with a 2D numerical model adapted from that used for a dc glow discharge. For a 15 mm discharge, the RF plasma's electrical and optical characteristics show a close correlation to several equivalent dc plasmas and to the results calculated from the adapted model. For an rms conduction current range of 11–30 mA, the rotational temperature varies between 2800 and 3200 K; the vibrational temperature shows a change of 3500–4000 K with near equilibrium conditions to the rotational state occurring in the central region of the discharge. Spatial measurements and modelling of nitrogen emission spectra identify the changes in the temperatures and dimensions along the vertical z -axis as well as the spatial dependence on the atomic and molecular species generated in the discharge.

(Some figures may appear in colour only in the online journal)

1. Introduction

This paper reports investigations of an atmospheric pressure plasma that has been developed as a prototype sound source. Sound is generated in electrical discharges through atmospheric air when the current is modulated with an audio frequency. The modulated energy of charged particles is transferred through various paths to the surrounding air molecules leading to pressure fluctuations in the gas. A plasma 'loudspeaker' is not a new concept; the effect was discovered and initially investigated at the beginning of the 20th century leading to the first commercially available device in the mid-1950s, variations of which have been available up to the present day [1–4]. There are two mechanisms involved in sound production. The first is momentum transfer from the ions to the neutrals which results in forcing of the air and pressure fluctuations (well known as the 'ionic wind'). The second is a thermal effect, similar to that in a lightning strike, where a pressure fluctuation occurs due to expansion and contraction of the ionized channel as the current through it rises rapidly,

typically in a few microseconds, and afterwards decays over many tens of milliseconds.

Previous comparative studies between models and experiments, have been dominated by use of 'colder' corona and dc glow discharges [5–8]. For the higher temperature 'warmer' plasma used in the present work, thermal mechanisms are anticipated to predominate in the generation of sound. This introduces different kinetic processes and driving conditions from those evaluated previously. With thermal mechanisms, the dominant driver for pressure variation can be probed through measurements of the fluctuations in gas temperature. Generally in molecular gases, gas heating occurs through electron excitation of the vibrational states followed by relaxation to the translational state [9] and in an air plasma this occurs predominantly through nitrogen [10].

Thermal equilibrium in atmospheric pressure, radio-frequency (RF) discharges is not completely achieved, not least because of the continuous periodic energy input from the RF current. It is important therefore to distinguish between electronic, vibrational, rotational and kinetic temperatures

Figure 1. The Tesla coil (left) and the schematic diagram of its driving circuit (right) with the plasma (inset). Approximate component values are $L_p = 24 \mu\text{H}$, $L_s = 28 \text{ mH}$, $C_s = 5 \text{ pF}$. The height of the coil is 600 mm and the electrode separation is 15 mm.

for nitrogen molecules in such plasmas. The vibrational temperature of nitrogen provides a measure of the energy available for a number of chemical processes within the plasma. Note that the fast relaxation time between rotational and translational stages in molecular nitrogen means that the rotational temperature is quite closely coupled to the kinetic gas temperature [10]. Conveniently, the spontaneous optical emission from an atmospheric pressure nitrogen plasma carries detailed information on the molecular thermodynamics. In particular, the so-called nitrogen second positive system (N_2SP) can be used to determine rotational and vibrational temperatures. Comparison of the temperatures in a plasma gives an indication of its state of equilibrium. The spatial structure of a discharge plasma contains regions of varying ionization with strong gradients in temperature and these are expected to have a bearing on the acoustic emission in the present context [7, 11].

The aim of this investigation is to examine the steady state of an unmodulated RF discharge through atmospheric air. This will then be the starting point for further studies of modulated, sound-generating plasmas. First the basic experimental scheme is described, setting out the operational parameter space. Next an outline is given of a 2D fluid model of an atmospheric pressure, low current discharge similar to that studied experimentally. Comparisons are then made between measured and modelled electrical characteristics. Then the spectral analysis of the optical emission from the RF atmospheric pressure plasma is used to define the radial structure of the discharge and to deduce the neutral gas temperature. Once again comparisons are made with the independent numerical model.

2. The RF plasma

2.1. The Tesla coil plasma generator

The plasma under investigation is generated in air at atmospheric pressure using a Tesla coil excited by a simple

square wave oscillator (figure 1). The electrical system is designed to operate in a RF regime where the breakdown voltage of an atmospheric air plasma is at its lowest [9].

The control stage of the drive circuit (figure 1(A)) provides a logic signal that alternately switches MOSFET devices in the power stage, chopping the dc voltage from the power supply unit (B). The inductor, L , protects the dc supply from the back emf of the Tesla coil. The low voltage/high current square wave is then passed to the primary coil (C). The voltage amplitude at this point is equal to the dc input voltage level. The resonant frequency of the coil is determined by the inductances and capacitance of the coils (L_p , L_s , C_s). The circuit resonance was measured from the voltage waveform to be around 325 kHz. This acts as a filter, transforming the fundamental frequency of the square wave to the secondary coil and generating a high voltage between the electrodes (D).

Typically, breakdown can be achieved across a point-to-point gap of several millimetres when the secondary voltage amplitude reaches about 10–13 kV. As a self sustaining discharge develops, the current increases and the electrode voltage drops since the available power is limited by the supply circuit. The electrodes are milled from brass and secured into 150 mm diameter dome structures, also made of brass. The electrodes are configured to provide an axisymmetric, vertical plasma channel. The driving electrode (lower) sits directly on top of the secondary coil. The voltage across the plasma was measured using a Tektronix 6015A high-voltage probe connected between the driving electrode and ground. The upper electrode is connected to ground via a 100Ω 'current sensing' resistor. The inter-electrode separation is adjustable over $50 \text{ mm} \pm 0.1 \text{ mm}$ by means of a screw-thread mounting of the upper electrode. The whole assembly is mounted on a vertical travel stage for adjustment of the axial position that is accurate to within $\pm 0.2 \text{ mm}$.

Upon ignition, the discharge is rooted to the electrodes by visible hot spots. The discharge produces a flame-like

luminous plume some 3–5 mm in diameter. It is stable and silent in ambient air, as shown in the inset to figure 1.

2.2. Numerical model

To complement the experimental platform for investigating RF atmospheric discharges, a numerical model has been used to give an independent characterization of the plasma source.

A detailed 2D numerical model of a short, vertical, low current dc discharge in the open air has been previously developed [12, 13]. This model incorporates the ionization balance of the plasma together with gas dynamics within and surrounding ionization region. The model does not extend to the cathode fall region adjacent to the electrode surface so a boundary condition is set to inject current directly into the channel. Non-equilibrium effects essential in the low discharge current range are accounted for. This includes (i) deviation of the electron energy distribution from a thermalized Maxwellian at the gas temperature (ii) deviation of the vibrational distributions of the molecules from the Boltzmann relationship with the gas temperature and (iii) diffusion of the various plasma species. For the purposes of the present study this model was adapted to the closely related scenario of a RF excited discharge.

The distributions of the plasma parameters along the radial (r) and axial (z) coordinates are governed by the system of equations involving Ohm's law, the mass continuity equation and the axial and radial momentum balance equations, as follows:

$$E(z) = \frac{I}{2\pi \int_0^R \sigma(z, r) r dr}, \quad (1)$$

$$\frac{\partial}{\partial z}(\rho u) + \frac{1}{r} \frac{\partial}{\partial r}(r \rho v) = 0, \quad (2)$$

$$\rho u \frac{\partial u}{\partial z} + \rho v \frac{\partial u}{\partial r} = -\frac{\partial p}{\partial z} - \rho g, \quad (3)$$

$$\rho u \frac{\partial v}{\partial z} + \rho v \frac{\partial v}{\partial r} = 0, \quad (4)$$

where $E(z)$ is the axial component of the electric field at an axial distance z from the lower electrode, I is the conduction current, σ is the electrical conductivity ρ is the gas density, u and v are the axial and radial components of the gas velocity, p is the gas pressure and g is the gravitational acceleration. The model includes also the balance equations for the gas temperature, T and the mean vibrational energy of nitrogen molecules, ε_V , as well as the balance equations for the number densities, n_k , of various plasma components:

$$\rho u C_p^* \frac{\partial T}{\partial z} + \rho v C_p^* \frac{\partial T}{\partial r} = \eta_T \sigma E^2 + Q_{VT} + \frac{1}{r} \frac{\partial}{\partial r} \left(\chi^* r \frac{\partial T}{\partial r} \right), \quad (5)$$

$$n_{N_2} u \frac{\partial \varepsilon_V}{\partial z} + n_{N_2} v \frac{\partial \varepsilon_V}{\partial r} = \eta_V \sigma E^2 - Q_{VT} + \frac{1}{r} \frac{\partial}{\partial r} \left(n_{N_2} D_V r \frac{\partial \varepsilon_V}{\partial r} \right), \quad (6)$$

$$\frac{\partial}{\partial z} (n_k u) + \frac{1}{r} \frac{\partial}{\partial r} (r n_k v) = \frac{1}{r} \frac{\partial}{\partial r} \left(D_k r \frac{\partial n_k}{\partial r} \right) + F_k. \quad (7)$$

Here C_p^* and χ^* are the corrected values of the specific heat and of the thermal conductivity, n_{N_2} is the number density of nitrogen molecules, D_V is the diffusion coefficient of vibrationally excited N_2 molecules, η_T and η_V are the fractions of energy input transferred to gas heating and to the vibrational excitation of N_2 molecules, D_k is the diffusion coefficient for species k (the ambipolar diffusion coefficient is taken for electrons), the source term F_k describes the net rate of generation of species of kind k in the kinetic processes of ionization, dissociation, recombination, etc. The term Q_{VT} describes the vibrational–translational (V–T) relaxation of nitrogen molecules. Note that the mean vibrational energies of other molecular air components, O_2 and NO , are close to their equilibrium values at the gas temperature, due to fast V–T relaxation of these species. The conductivity is $\sigma = en_e \mu_e$ where n_e and μ_e are the number density and mobility of electrons and e is the electron charge; it is assumed that the electrical conductivity does not change significantly over the RF cycle.

Thermal conduction and diffusion in the axial direction is neglected owing to the aspect ratio of the discharge column. No account is taken of heat losses to the electrodes and the radiation losses are also neglected as their role at low currents is small. To accommodate the RF power input the model is operated with rms equivalent values of current, voltage and axial field.

The scheme and rate constants of the kinetic processes (excluding those involving electrons), the diffusion coefficients, the factors η_T and η_V , and the rate of V–T relaxation are taken from [13]. The kinetic scheme includes processes involving N_2 , O_2 , N , O , NO , NO^+ , O^- , O_2^- , O_3^- and electrons. The balance equations are solved for the densities of N , O , NO and electrons. The densities of negative ions are evaluated in a local approximation (such an approach is justified because the times of negative ion destruction in the detachment processes are much less than the times of diffusion), the density of positive ions is obtained from the condition of conservation of charge and the densities of N_2 and O_2 are given by the condition of conservation of N and O nuclei. To evaluate the vibrational temperature of nitrogen excited state $N_2(C)$, the relative number densities of molecules in vibrational levels $v = 0$ and 1 of this state are calculated, in assumption that the rates of excitation of these levels by electron impact from various vibrational levels of nitrogen ground state $N_2(X)$ are proportional to corresponding Franck–Condon factors.

The rate constants of reactions involving electrons are obtained by averaging the corresponding cross sections with the electron energy distribution function (EEDF). Note that for the considered parameters for the RF discharge the relation $\omega \ll \delta \nu$ between the angular wave frequency, ω , and the collision frequency, ν , applies where δ is the mean fraction of electron energy lost in a collision of an electron with a heavy particle. In such conditions, the EEDF follows the periodic change of the electromagnetic field and the averaging of the rate constants of reactions involving electrons over the wave period is required. The dependence of a rate constant, K , on the instantaneous value of the reduced electric field, E/n (here n is the gas number density) may

Figure 2. The voltage (grey) and conduction current (black) waveforms for a rms conduction current of 18 mA. The upper and lower plots show the waveforms for vertical and horizontal electrode orientations, respectively.

be taken in the form $K \exp(-Bn/E)$ which is typical for processes with high energy thresholds (ionization, electronic excitation, dissociation). An approximate expression for the time-averaged rate constant is then obtained from $\langle K \rangle = (2E_{\text{amp}}/\pi Bn)^{1/2} K_{\text{amp}}$ [14] where E_{amp} is the amplitude of electric field and K_{amp} is the corresponding value of the rate constant.

The output from the numerical model will be compared with measurements on the plasma generated by the Tesla coil.

3. Electrical characteristics

With an electrode separation of 15 mm breakdown was observed when the voltage generated by the Tesla coil reached around 25 kV peak to peak. After initiation the discharge was sustained typically by a voltage of between 4.5 and 5 kV peak to peak (1.6–1.8 kV rms). Adjustments to the dc power supplied to the chopping circuit was used to control the sustaining voltage, enabling stable operation over a range of currents. The current in the secondary coil comprises a displacement component associated with the air-filled geometric space between the electrodes and a conduction component associated with the discharge itself. To separate the two components

the electrode gap capacitance was characterized under pre-breakdown conditions. First it was noted that, in the absence of a discharge, the current waveform was sinusoidal and led the secondary voltage waveform by $\pi/2$; it was also noted that the current amplitude was proportional to the voltage amplitude. This current is required to charge the electrodes to a given voltage without driving a discharge. Thus, before breakdown the inter-electrode gap presents an effective capacitance through which displacement current passes. To quantify the displacement current its waveform was recorded when the pre-breakdown gap voltage was 4.5 kV peak to peak; this value was chosen because post-breakdown operating voltages were generally around this level. The pre-breakdown current waveform was sinusoidal and led the voltage by $\pi/2$. Therefore the displacement component for any other condition (pre- or post-breakdown) was determined by scaling this reference level by in proportion to the actual electrode voltage. The conduction current component was then found by subtracting the appropriately scaled displacement waveform from the total current. Stable operation could be achieved with the conduction current in the range 11–30 mA rms.

An example of voltage and conduction current waveforms for $I = 18$ mA rms is given in figure 2(top). The distortion in the peaks of the current waveform indicates a curious limitation on the current flow either at the electrodes, or in the main current channel itself, that is associated with the specific direction of current flow. The distortion arises when the upper electrode is the more positive. The asymmetry between the positive and negative current peaks arises from natural convection around the vertical discharge channel. To confirm this the Tesla coil and electrodes were turned to set the discharge channel horizontally. Figure 2 (bottom) shows the equivalent measurement with the electrodes in this orientation. Some distortion around the current peaks is again present but the conduction current waveform is symmetrical with positive and negative peaks having comparable shapes. In fact in this orientation the channel is bowed upwards by convection into a classic ‘arc’ shape. In view of the presence of distortion around both current peaks in the horizontal discharge it would appear that the effect is linked to the high current density around the electrode spot. The configuration could not be completely inverted owing to the way in which the coil itself is supported. However, given the symmetry of the voltage waveform there should in fact be no electrical or gas dynamic differences between normal and inverted conditions.

One of the variables in numerical model is the axial electric field along the plasma in the conducting channel. A local measurement of this using some invasive form of potential probe is difficult owing to the high kinetic gas temperatures and the highly collisional environment of an atmospheric pressure plasma. Instead a global value can be inferred for the Tesla coil plasma from observations of the additional voltage (ΔV) required to sustain a given rms conduction current when the electrode spacing is increased by a known, small amount ($\Delta z = 1$ mm); several 1 mm increments spanning the 15 mm electrode separation resulted in the same voltage measurement for each increment. The magnitude of the axial electric field

Figure 3. The measured (points) and modelled (line) electric field at $z = 4$ mm (dashed) and 10 mm (solid) as a function of the rms conduction current for a 15 mm electrode separation.

in the channel, $E = \Delta V / \Delta z$, controls the energy input to the electrons that then couple to the various active species through rate constants for thermally activated particle reactions. So changes in the sustaining voltage were made for a succession of short incrementations of the discharge length after adjustments to the input power to restore the conduction current component to its original level. Figure 3 compares measurements of the electric field magnitude with the independently modelled values; the model is capable of calculating spatially resolved results and the electric field for two axial positions, $z = 4$ and 10 mm, are presented. The model neglects the effects of periodic cathode fall regions around the electrodes, but so too does the global measurement. The modelled values lie marginally above the measured values, closely following the same trend over a range of rms conduction currents. This suggests that there are processes not included in the model that weakly enhance the effectiveness of the electric field in sustaining the plasma. Though the overestimation of the modelled values is not severe, it may have further impact on the calculation of other plasma parameters (as the electric field is used to determine the rate constants required in the calculations).

Figure 3 is also representative of the voltage-current characteristics of the discharge and is similar in behaviour to the glow-to-arc transition region seen in a dc glow discharge. Atmospheric pressure dc are generally unstable in this region and subsequently runaway ionization takes the discharge into a fully developed arc in local thermal equilibrium. With RF excitation there is just sufficient ionization retained through the off phase of the cycle for the plasma to be sustained without the full thermalization.

The conductance of the plasma generated by the Tesla coil is comparable to that of a typical atmospheric pressure air plasma [15]. Based on this conductance the electron density for 11 mA rms can be estimated to be approximately $2.4 \times 10^{17} \text{ m}^{-3}$, which is consistent with the average value from the model of $3 \times 10^{17} \text{ m}^{-3}$. The estimated electron density value

is comparable to measurements of a plasma operating under similar driving conditions [16] where the measured electric field and a current density of $0.1\text{--}10 \text{ A cm}^{-2}$ (determined using the plasma dimensions) calculate an estimated electron density of $10^{18}\text{--}10^{19} \text{ m}^{-3}$; for the RF plasma, the current density at 11 mA rms is approximately 1 A cm^{-2} .

The power dissipated by the plasma as a function of the current varies from 18 W at 11 mA rms and up to 34 W at 30 mA rms. The absorbed power does not follow a simple quadratic relationship with rms current owing to the nonlinear current-voltage characteristic. About 65% of the power supplied to the Tesla coil is transferred to the discharge channel.

4. Optical measurements

The visible emissions from the plasma are linked to ground state populations through a rich variety of processes so the wide spectrum light combines a variety of physics. The plasma was imaged using an Andor DH534 iCCD intensified camera fitted with a lensing system designed for close up photography (figure 4). The lensing system consisted of a Tokina 70–200 mm zoom lens combined with a +4 diopter attachment to increase magnification. The lens was stopped down to $f/22$ to compensate for the reduced depth of field when using this combination of lenses. The spatial resolution was determined by focussing on a graticule with 1 mm spacings in the focal region and was calculated to be $15 \mu\text{m/pixel}$.

The model calculations include exchanges between electrons and the rotational and vibrational levels of nitrogen as these can be shown to be dominant processes. Spectroscopic methods were used to probe the gas temperatures in the discharge generated by the Tesla coil. The rotational and vibrational temperatures of an atmospheric pressure discharge can be determined from the spectral emission of the $\Delta v = 2$ band transition of the nitrogen second positive system (N_2SP) between 365 and 385 nm. The intensity of the rotational line structure in a spectrum is related to the rotational temperature while the relative intensities of two separate vibrational band transitions from within the system were used to determine the vibrational temperature.

The plasma was imaged using two different spectrometer systems. The first system was an OceanOptics HR4000 spectrometer fitted with a fibre optic and 5 mm diameter collimating lens to image the region of the plasma onto the entrance slit. The spectral range was 200–850 nm. The grating resolution of this system was $2400 \text{ lines mm}^{-1}$ and the entrance slit width was $6 \mu\text{m}$; for a 40 mm separation between the fibre optic and plasma, the fibre optic acquired light from a cylindrical chord that was approximately 1 mm in diameter. Measurements of the plasma temperatures were made using this system.

The second system used was a Triax HR320 Czerny-Turner type monochromator coupled with an Andor DH534 iCCD camera (figure 5(top)). Light was imaged onto an entrance slit using a 75 mm focal length lens which was optimized for UV transmission. A dual mirror arrangement was positioned between the lens and entrance slit to rotate the image by 90° and thereby capture the intensity in the radial

Figure 4. An optical set-up for measurement of the visible emission (left) and a typical image (top right). The visible discharge radius is taken as the half-width, half-maximum of a intensity profile (bottom right).

Figure 5. The monochromator set-up (top) used for acquiring the nitrogen spectrum (bottom left). The emission integrated over the wavelength direction is used to determine the discharge radius (bottom right).

direction. The light was dispersed with an $1800 \text{ lines mm}^{-1}$ grating and the entrance slit width was 0.15 mm . The monochromator and camera set-up was calibrated using a white light source of known power output between $300\text{--}800 \text{ nm}$ and corrections were applied to the spectral line intensity in order to compensate for the wavelength dependence on the quantum efficiency of the camera CCD; below 300 nm , a nominal correction was applied using the

correction factor applied at 300 nm . Additionally, a flat-field correction and a background correction was applied to the data to correct for variations in pixel sensitivity of the CCD array. Measurements of the UV radial intensity and the on-axis spectral scans were made using this system. The spatial resolution in the radial direction was determined by imaging a source of known dimensions onto the entrance slit and was found to be approximately $13 \pm 0.5 \mu\text{m/pixel}$.

Figure 6. The average optical intensity variation (dashed) and the RF conduction current cycle (solid).

4.1. Visible emission

The time-resolved visible optical intensity is given in figure 6 and is correlated with the conduction current of the vertical electrode configuration presented in figure 2. The peak intensity coincides with both positive and negative peaks of the RF conduction current cycle and corresponds with the near instantaneous timescales for electron excitation ($\tau \approx 10^{-8}$ s). Note that the emission does not go to zero between peaks, but has a mean level of about 75%. This reflects the contribution of metastable states, long radiative times and the overall relaxation processes that include a radiative step. Two sources of visible emission are the upper wavelength range of N_2SP (375–400 nm) along with the nitrogen first positive system (N_2FP) [17–19]. The radiative lifetime of N_2FP is in the low microsecond timescale [20] and corresponds to the emission decay times exhibited by the intensity here. The long-lived metastables sustain emission over longer timescales than that resulting from electron excitation. In nitrogen, the excited metastable $N_2(A)$ causes direct excitation to the $N_2(C)$ state or in a stepwise process through the $N_2(B)$ state. However, in an atmospheric pressure plasma, this process is negligible due to quenching from collisions with other plasma species [21]. It is likely that further prolonged optical emission results from interaction with a long-lived nitric oxide (NO) species as will be discussed later.

4.2. Spectra

Figure 7 presents emission spectra for wavelengths between 200 and 850 nm. Spectra were recorded for two axial positions—at the electrode region ($z = 0$ mm) and in the main body ($z = 6$ mm)—and for two rms conduction currents in the plasma. In addition to the corrections described previously, a high-pass filter was used for spectral emission at wavelengths greater than 450 nm to eliminate the second-order spectrum (mainly from N_2SP emission). A correction was applied for the attenuation of spectral intensity during insertion.

The dominant feature at both axial positions is the N_2SP system between 300 and 400 nm. At the electrode, the nitrogen first negative system (N_2^+FN) and several atomic

Figure 7. The spectral scan (on-axis) over 200–850 nm as a function of two axial positions; $z = 0$ mm (top) and $z = 6$ mm (bottom) and for rms conduction currents of 11 mA and 27 mA.

species such as oxygen, nitrogen and trace elements of argon are also measured. This indicates the electrode region to be a rich source of the high energy electrons needed to instigate the excitation and molecular dissociation required to produce these emissions. Generally, N_2^+FN emission involves an initial ionization of the nitrogen molecule with a threshold of 15.6 eV. This is followed by excitation to the $B^3\Pi_g$ excited state of the formed nitrogen ion, after which optical emission occurs through relaxation [22]. Similarly, the energies needed to produce the atomic species through dissociation must exceed 7 eV for oxygen molecules and 12 eV for nitrogen molecules [22]. The steep potential gradients in the cathode fall region around the electrodes produce the higher energy electrons in sufficient numbers to induce these processes. As previously stated, the visible emission is dominated by the upper wavelength range of N_2SP and the N_2FP . The appearance of the hydroxyl radical (OH), which overlaps the $v = 1-0$ N_2SP transition between 300 and 315 nm, most likely results from the dissociation of water vapour in air. The relative humidity in the laboratory for these particular measurements was 19%; subsequent measurements where the relative humidity was 57% resulted in the appearance of an atomic hydrogen feature at 656 nm. At the electrode, an increase in current results in a near-uniform increase in intensity across the entire spectrum.

The spectrum at $z = 6$ mm can be compared with that around the electrode. At the lower current setting, the spectral intensity followed that of the electrode region albeit being an order of magnitude less in intensity. The absence of the atomic and N_2^+FN emission lines indicates that the average electron energy was significantly lower in the column. The N_2SP system still dominates with clearly defined visible emission from this and N_2FP . As the rms conduction current was increased to 27 mA, however, the relative intensities of the spectrum change. The intensity of the nitric oxide system ($NO\gamma$) around 250 nm was greater, possibly as a consequence of increased recombination rates between N and O atoms being created more readily in the electrode region. The increases in OH and N_2SP emission intensities were more modest in comparison. The most apparent change was in the visible range. The N_2FP emission at low current evolved into a near ‘continuum’ emission at higher current as a result of the increase in the intensity of the rotational line emission of the N_2FP band system or further reactions originating from other emission systems in the plasma to produce visible emission.

4.3. Defining a radius

The spatial dimensions of the plasma were characterized from optical emission measurements using two wavelength regions of the plasma emission: the UV radiation from the $v = 0-0$ vibrational band transition of the N_2SP system (band head origin = 337.14 nm) and the visible region, taken nominally to be between 400 and 750 nm.

For the UV region, the spectral emission was acquired using the Triax monochromator. The ‘UV discharge radius’ was calculated as the half-width, half-maximum (HWHM) of the line of sight integrated intensity (figure 5(bottom right)). For each spectrum, the intensity was integrated along the wavelength axis to give the total intensity emission from which the HWHM provides a ‘visible discharge radius’.

4.4. Comparing optical radii

Figure 8 shows the discharge radius as a function of axial position for three rms conduction currents. The radii from the visible and UV data were determined at several discrete positions along the discharge axis. The equivalent model results for both optical regimes are also presented. At 11 mA rms, the radii determined from the UV and visible emissions are in close agreement and show a measured steady increase with increasing distance up the axis until eventually sharply decreasing towards the upper electrode. The modelled results lie within the uncertainty of the measured data in the mid-region; the boundary conditions described in section 2.2 mean the numerical model does not resolve near-electrode phenomena or include the geometrically restricted upper electrode. The finite diameter of the upper electrode is not taken into account and therefore the model does not account for current constriction and the associated decrease in discharge radius near this electrode. Note, however, that there are no fitting parameters required in the model so the agreement is an indication that the model includes the important physical processes.

When the rms conduction current is increased to 27 mA, the radius derived from visible emission shows a significantly greater increase at all axial positions than the increase in the radius determined from UV emissions. For instance, at $z = 6$ mm, the ‘visible emission radius’ increases from 0.65 to 1.2 mm as the rms conduction current changes from 11 to 27 mA compared with a change in the ‘UV-emission radius’ from 0.75 to 0.8 mm. These different trends are also shown in figure 9, where the discharge radius is plotted as a function of the rms conduction current for two axial positions. The model calculations do not exhibit such marked differences in the UV and visible radii.

The plasma images alongside the axial plots in figure 8 give a good visual indication of the changing (visible) appearance of the plasma. As the conduction current increases, the previously narrow (purple) discharge, which is dominated by emission from the UV/lower visible wavelength range, develops into a source of broader (whiter) emission. The increased current also broadens the plasma as a result of increased axial and radial convection and diffusion within the plasma. This change in the emission reflects the spectra of figure 7; the axial positions at which the spectra were obtained are marked on the plots for the 11 and 27 mA rms settings in figure 8.

In the model, the visible emission which is calculated from the electron excitation accounts for visible emission from the N_2FP emission alone. However, in practice additional species may also contribute to the total visible emission. According to [20], the radiative lifetime of N_2FP is in the ‘low microsecond timescale’ and significantly shorter than the timescales for gas diffusion which, for molecular nitrogen, is on the millisecond timescale. This means that N_2FP emission is limited spatially. An alternative path for N_2FP emission through re-excitation of the $A^3\Sigma_u^+$ to the $B^3\Pi_g$ can also be discounted as (i) higher energy electrons are still required for this process and (ii) they will not lie in sufficient number densities outside of the central ionized channel. The observed discharge radius therefore must be attributed in part to reactions from long-lived metastables or other emission systems in the plasma. The formation of NO_2 fits this criterion and it has been suggested previously as a possible emission source [23]. In an atmospheric pressure plasma under the electrical driving conditions considered here, NO and NO_2 species densities depend mainly from the recombination reactions, $NO + O + M \rightarrow NO_2 + M$ and $NO + O_3 \rightarrow NO_2 + O_2$ [24]. Experimental evidence for this lies in the NO emission systems below 300 nm. If the HWHM radius is determined in a similar fashion to the UV and visible emission radii, it is found to be comparable to the radius derived from the visible emission and may act as the sustaining source of NO_x generation. NO is also a long-lived state and may be active at the radial distances measured here.

5. Gas temperatures

5.1. Data reduction

Rotational and vibrational temperatures for nitrogen molecules in the discharge were determined by fitting a modelled

Figure 8. The axial variation of the discharge radius for three rms conduction current settings; 11 mA (top), 18 mA (middle) and 27 mA (bottom) with corresponding images of the plasma at each setting; measured VIS (squares) and UV (triangles), modelled VIS (dashed) and UV (solid).

spectrum to the measured emission spectrum (figure 10). The modelled spectrum was calculated using a Levenberg–Marquardt (L–M) algorithm to calculate the spectral line intensity for a given temperature. The function calculates the multiplet splitting of the P, Q and R branches in nitrogen

using the rotational terms via the formula from [22] with rotational constants taken from [25]. For the line intensity, the transition probabilities between $C^3\Pi_g-B^3\Pi_u$ electronic states were taken from [26] along with the relevant Honl–London factors, again from [22]. The modelled spectrum

Figure 9. The discharge radius, R , as a function of rms conduction current evaluated at two axial distances, $z = 4$ mm and 10 mm; measured VIS (squares), measured UV (triangles) and modelled VIS (dashed) and UV (solid).

was calculated for the spectral emission lines of the $\Delta v = 2$ transition, with the relative intensities of the 0–2 (380.5 nm) and 1–3 (375.5 nm) band transitions used to determine the vibrational temperature. The fitting procedure gives the vibrational temperature T_v of the state $N_2(C)$, defined by the equation $n_1/n_0 = \exp(-E_c/kT_v)$, where n_0 and n_1 are the number densities of the vibrational levels of $N_2(C)$ state, $v = 0$ and $v = 1$, respectively, and E_c is the corresponding vibrational quantum. Prior to the spectral fitting procedure, corrections were applied for inaccuracies of the wavelength assigned to a data set by the measurement acquisition system. The fitting function uses four free parameters to fit the modelled spectrum: rotational temperature, vibrational temperature, the background emission level and the N_2C excited state population which is used as a measure of the emission intensity. The intensity is independent of the temperatures which are taken as relative measurements. The function optimizes the slit function from an initial value to account for spectral line broadening and improves the fitting to measured data. The calculated spectrum was validated against Specair [27] and against an independently developed fitting algorithm

using the LM method [28], the results of which showed an average difference of around 7% in the calculated rotational temperature when a common test spectrum is analysed. This results from differences in the analysis method such as application of the slit function and the transition probabilities and rotational constants used for calculating the spectral line intensity. The difference has been applied as an uncertainty.

5.2. Rotational and vibrational temperatures

The measured rotational (T_r) and vibrational (T_v) temperatures as a function of axial position are given in figure 11 for a rms conduction current of 11 mA and an electrode separation of 15 mm. As with the discharge radius, the trend on-axis shows two distinct regions. The temperatures in the main body are closely coupled and evidently are in near thermal equilibrium. Around the electrode, T_r and T_v gradually deviate from each other, indicating that here rotational and vibrational energy systems are not in equilibrium. Figure 11 also presents the values for the gas temperature, T , and the vibrational temperatures of $N_2(C)$ state and of the ground state $N_2(X)$ calculated from the model. It is seen that under the conditions considered, the difference between the vibrational temperatures of the ground and excited nitrogen states is small. With growth of the vibrational temperature of $N_2(X)$, the excitation of higher vibrational levels of $N_2(C)$ becomes more effective, resulting in an increase in the vibrational temperature of this state.

T_r ranges from 2300–3000 K and, if taken as being representative of the kinetic gas temperature, indicates that the medium in the channel away from the electrodes can be classified as a warm plasma, but with a lower temperature than expected for the local thermodynamic equilibrium conditions of an arc (where T can be in excess of 10 000 K [20]). The corresponding T_v is consistently a few hundred kelvin or more higher so both temperature ranges lie in the region of that measured for equivalent DC driven plasma [16, 29]. The increase in the T_v around the electrode is due to the molecular nitrogen's dependence on the rate of electron impact excitation of the internal vibrational states. The higher electron density leads to an increased collision frequency, increased excitation and hence higher temperatures. The combination of the steep potential gradients and narrowing discharge radius leads to an increase in the electron number density and provides the necessary conditions for the increased T_v around the electrodes. The dis-equilibrium between T_r and T_v around the electrodes results from the interdependence of the rate of V–T relaxation and the kinetic gas temperature; an increase in gas temperature results in an exponential increase in the V–T relaxation rate [29]. Assuming T_r represents the gas temperature, as it increases so the rate at which energy is transferred the translational state in nitrogen also increases, resulting in faster equilibrium conditions. Hence, an increase in T_r (or T) results in T_v achieving near equilibrium conditions with T_r .

The effects of vertical convective flow, which have been seen in the electrical characteristics and discharge radii, can also be seen in the temperature profiles. The sharper increase in T_v around the upper electrode (compared with a shallower

Figure 10. Spectrum of the $\Delta v = 2$ band transition (top) and the measured (red) and modelled (blue) on-axis nitrogen spectrum (bottom).

Figure 11. The measured T_r (squares) and T_v (triangles) with equivalent modelled temperatures for T (solid) and T_v (dashed—ground state, dots—C-state) as a function of axial position, z for a rms conduction current of 11 mA. The uncertainty in the axial position is ± 0.5 mm.

gradient increase around the lower electrode) coincides with an increase in T_r where a hotter region due to convection has formed. The relative response in the two temperatures again emphasizes the interdependence of V–T relaxation and T . The modelled values share many features with the measured results. For the most direct comparison of T_v to the measured data, the upper C-state temperature has been calculated along with that of the ground state. In the mid-region, the trend in the modelled T_r and T_v values shows close to equilibrium

Figure 12. The comparison of measured T_r (squares), T_v (triangles) and modelled T (solid) and ground state T_v (dashed) as a function of conduction current at an axial position of $z = 7$ mm.

behaviour, albeit around 250–300 K lower than the measured values. While the differences in the central region are modest, the electrode region shows a sharper increase in the modelled values compared with the gradual increase in the measured result. As described in the experimental set-up, the spatial resolution of the fibre optic used to obtain the measured results is approximately ± 0.5 mm which is insufficient to pick out the sharp temperature increases predicted by the model. Figure 12 presents the dependence of T_r on the rms conduction current and shows an increase as expected with an increase in power into the plasma. The modelled results lie marginally outside the measurement uncertainty but follow the trend in the

Figure 13. The axially-dependent vibrational temperature measured for electrode separations of 5 mm (square), 10 mm (circle) and 15 mm (triangle).

measured results. This discrepancy may follow from the subtle differences seen previously in the electric field comparison which then influence subsequent calculations of the plasma parameters.

Comparative measurements at three electrode separations show that the T_v profile remains constant around the electrode regions (figure 13). It is seen that in the middle of the gap, sufficiently far from the electrodes, the T_v values at various electrode separations are nearly the same and are close to the modelled T_v values (figure 11); note that the modelled data at the given distance from the bottom electrode, obtained without account of electrode effects, are independent of the electrode separation. A direct relationship can be established between the electric field, E , and the electron collision frequency via the electron energy distribution. It may be possible for the T_v profile to be used as a non-invasive method to determine the electric field variation within the plasma and the feasibility of this is being investigated further.

6. Discussion

This investigation was undertaken to understand a steady RF atmospheric pressure discharge as part of wider investigations into how this plasma produces sound when it is modulated at audio frequencies. It is anticipated that periodic spatial and temporal variation of the plasma would be associated with thermal expansion of the air in the plasma plume. However, unlike the fluid/structural attributes of a mechanical loudspeaker, it is difficult to define a ‘boundary’ between the plasma and air where the pressure wave originates. The results for the discharge radius complicate this further with two notional boundaries determined for the plasma depending on the optical region used for analysis. Correlation between the measured and modelled vibrational temperature gives confidence that V–T relaxation is the dominant energy transfer mechanism that leads to gas heating and instigates thermal expansion. Typically, vibrational excitation in the nitrogen

molecule is optimal when the electron energies are between 1 and 3 eV which in effect coincides with the discharge volume where visible emission occurs. Therefore, it is feasible that the discharge radius based on visible emission also defines the zone within which the pressure wave originates. The asymmetry of the plasma radius may lead to a more complex acoustic field in the near-field region, though this is of lesser importance to a listener as the experimental results indicate that the emission in the far-field is close to a cylindrical source and behaves as a isotropic radiator. The axial variation of the vibrational temperature and specifically the axial dependence in the rate of V–T relaxation may lead to spatial phase differences in the response of the plasma to a time-varying electrical signal (and in the subsequent acoustic signal). However, V–T relaxation times calculated from the temperature results at the electrode and the main body, show a modest difference in the time constants associated with relaxation, which are on the microsecond timescale [30]. This is significantly shorter relative to the period of audio modulation and implies that, while a small phase difference would occur along the plasma length, the response of the human ear would not be sensitive enough to detect it.

Other practical considerations when using a plasma in a domestic environment can also be addressed. Ozone generation is acknowledged to be a problem for corona-type plasmas with prolonged exposure in a confined area being hazardous to health. Ozone synthesis in air relies on a three body reaction between oxygen, diatomic oxygen and a third body. This process is significantly affected by gas temperature and humidity within the environment. The effects of temperature are twofold; firstly, NO_x production is increased at elevated gas temperatures where faster reactions between O and NO_x (NO and NO_2) use up the oxygen atoms that would otherwise contribute to ozone synthesis. Secondly, ozone is naturally unstable at elevated temperatures. As an example a DBD discharge operated at a gas temperature of 800 K produces a negligible level of ozone [24]. If the rotational temperature is assumed to represent the translational gas temperature in the Tesla coil plasma then ozone levels would be negligible. Measurement using an ozone detector shows this to be the case with ozone levels remaining below 0.01 ppm and within workspace exposure limits [31]. The short lived nature of ozone under these conditions leads instead to increased levels of other hazardous species such as the nitrogen oxides (NO_x) identified in the spectral scan; the health implications of NO_x are well documented and further investigation is required to determine the level of generation while the plasma is operational.

7. Summary

The characteristics of an RF atmospheric pressure discharge in air have been determined from a number of non-invasive techniques. Instantaneous current and voltage measurements have been used with spectrally and spatially resolved optical emission. The rotational temperature of nitrogen molecules has been shown to vary between 2800 and 3400 K for a corresponding rms conduction current of

11–30 mA and is significantly above that expected for the cooler, corona-type discharges. The corresponding vibrational temperatures are in the range 3500–4000 K. A numerical model adapted from dc to rms RF currents has been used to provide an independent expectation of molecular temperatures (vibrational and rotational), electron number density and spatial structure. The temperatures calculated using the model are comparable to those measured and give confidence that the gas heating is dominated by both joule heating and vibrational–translational relaxation of nitrogen molecules. A reasonably close agreement is seen between the modelled and observed discharge radius as a function of axial position. The effects of vertical convective flow can be seen in all results. In addition, spatial variations can be seen in the discharge dimensions and the species temperatures are identified. The spatial variation in the plasma parameters has been measured and provide an interesting combination of conditions which could impact on the acoustic emission when the discharge is modulated. Broadly, two regions are evident; around the electrode and in the main column. These regions are reflected in the plasma characteristics with regions of thermal and non-thermal equilibrium as well as the relative intensity changes in the spectral emission. Atomic species (O, N and Ar) and radical species (OH, NO) have been identified though some are limited spatially to the electrode region.

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The uncertainties expressed in this paper are calculated from statistical variation of the measurements and from systematic evaluations of the measurement system. The type B (systematic) uncertainty was derived from a combination of manufacturer's specifications, evaluation of the instrumentation and the methods used for data analysis. Unless stated, a rectangular probability distribution has been assumed with the combined, expanded uncertainty calculated to a 95% confidence level [32].

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