Numerical investigation of transient, low-power metal vapour discharges occurring in near limit ignitions of flammable gas

Rajiv Shekhar¹², Sergey Gortschakow³, Holger Grosshans¹, Udo Gerlach¹ and Dirk Uhrlandt³

¹ Physikalisch-Technische Bundesanstalt (PTB), Bundesallee 100, 38116 Braunschweig, Germany
² School of ITEE, The University of Queensland, Qld, Australia
³ Leibniz Institute for Plasma Science and Technology (INP), 17489 Greifswald, Germany

E-mail: rajiv.shekhar@uqconnect.edu.au

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Abstract

This article presents an investigation of a transient (30 μs–5 ms) electrical discharge in metal vapour with low voltage (≤30 V) and current (≤200 mA), drawn between two separating electrodes. Discharges of this type are rarely studied, but are important in electrical explosion safety, as they can ignite flammable gases. An empirical model is developed based on transient recordings of discharge voltages and currents and high speed broadband image data. The model is used for predicting the electrical waveforms and spatial power distribution of the discharge. The predicted electrical waveforms show good accuracy under various scenarios. To further investigate the underlying physics, the model is then incorporated into a simplified 3-D gas dynamics simulation of the discharge occurring in a flammable atmosphere. This simulation includes chemical reactions, molecular diffusion, heat transfer and evaporation of metal from the electrode surface. The local thermodynamic equilibrium (LTE) assumption is next used to calculate electrical conductivity from the field outputs of the simulation, which in turn is integrated to produce electrical resistance over time. This resistance is then compared to that implied by the voltage and current waveforms predicted by the empirical model. The two methods produce differing estimates, demonstrating that the studied discharge very likely deviates from LTE. Finally, assuming that electrical conductivity is approximately independent of the heavy particle temperature, the empirical model is used together with the calculated electrical conductivities to estimate the electron temperatures present in the discharge.

Keywords: combustion, metal vapour plasma, cathode phenomena, OpenFOAM

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

1. Introduction

Electrical discharges have been studied extensively as a means of igniting flammable gasses in the field of combustion science. Previous works have typically focused on high voltage spark discharges, possessing substantially different characteristics to the type of discharge considered here. Although scientific literature on the subject is limited, these low power discharges, referred to variously as break arcs, break sparks, or contact arcs, are well known in electrical explosion safety. Industry standards prescribe methods for managing the explosion risk they pose [1, 2], but require experiment based methods for establishing safety limits which have been proven to be unreliable [3]. There is thus an increasing demand for a reliable alternative based on sound science to be developed, which requires a fundamental understanding of the relevant phenomena.
The specific scenario being investigated is described in figure 1. A tungsten wire anode is positioned with its tip in contact with the surface of a cadmium block cathode. The wire moves along the surface until it reaches the edge of the block, where contact is broken. As the two electrodes separate, the discharge is drawn between them, reaching a typical maximum length of around 300 µm. The electrode materials and geometry are based on a reference scenario for standardised explosion safety testing [1]. Additionally, the electrodes are energised by an electronic circuit which adjusts output voltage so that a programmed constant current flow is maintained.

A previous investigation applied a combination of optical diagnostic experiments and reactive computational fluid dynamics simulation to the problem, focusing on the combustion resulting from the discharge [4]. Here, the discharge was represented by a resistive heating term derived from an empirical model of the discharge. The empirical model was based on a quasi-static correlation between voltage, current and length of the discharge. This work presents an expanded derivation of this empirical model, including several improvements and the incorporation of new experimental data. A partial validation is also presented utilising electrical measurement data. A key challenge of the empirical approach is quantifying the spatial distribution of resistive heating. This is currently done using high speed broadband image data, which is difficult to analyse. Additionally, this aspect of the model is not easy to validate.

The direct imposition of a particular spatial distribution for the ohmic heating term could be avoided if electrical conductivity within the discharge region were known. This calculation would be significantly simplified by the assumption of local thermodynamic equilibrium (LTE), i.e. that all species present at any point in the plasma have kinetic energies described by a Maxwell–Boltzmann distribution for a given single temperature. This has the important implication that the parameters of the plasma such as ionisation fraction and electrical conductivity are defined uniquely by this temperature. Previous studies of high voltage sparks conducted simulations under the LTE assumption, where tabulated data for plasma parameters as functions of temperature were used to couple the equations of gas dynamics to those of electromagnetics [5, 6].

Applying a similar approach to modelling the low power discharge in question would require some indication of whether the LTE assumption is valid. In models of discharges in switching electrodes at high currents, the bulk of the plasma is often considered to be in LTE [7]. Quasi-static empirical models are also used for equilibrium arcs, and have shown some success in this application [8]. The radiation emitted by the discharge was also confirmed to comprise predominantly of atomic lines of cadmium [9]. Characterising this discharge as an equilibrium arc would, however, be questionable, considering the extremely small energy and time scales involved. This work therefore aims to provide insight into this matter. Given the substantial difficulties involved in modelling a non-equilibrium plasma, it is desirable to know beforehand whether doing so is necessary.

For this purpose, a similar approach to [4] is taken using the empirical model as a resistive heat source term in a gas dynamics simulation. This approach does not make assumptions as to the underlying physics of the discharge, reflecting only its measured characteristics. Plasma properties of the discharge are estimated from postprocessing of the simulation output and the calculations based on the LTE assumption. The article is presented as follows: section 2 describes the development of the empirical discharge model, section 3 describes the simulated solid, gas phase physics, and section 4 presents the post-processing and analysis of the simulation and empirical model results. Conclusions from the work and avenues for future development are presented in sections 5 and 6.

2. Empirical discharge model

The empirical part of the model defines the total power input into the discharge over its duration, as well as how that power is distributed over the discharge region. This section outlines the derivation of these two relationships. A similar approach to [4, 8] is taken, applying it to the recent dataset of [10] and adding a more detailed consideration of varying arc geometry. In this previous work, discharges in a mixture of air with 21% hydrogen were considered, this being a standard mixture used to investigate ignitions by the discharges [1]. A considerable evaporation of the Cd electrode also contributed to the gas composition in the discharge area. The corresponding dataset consists of voltage and current waveforms, together with corresponding high speed image recordings from which length variation of the discharge over time is measured. The image data were recorded with high speed camera and image intensifier, providing a 3 dB spectral range of 200 nm–700 nm. The dataset is thus a series of
voltage/current/length triples—one for each recorded time instant of each discharge.

An example of this data for one discharge event is shown in figure 2. As explained in section 1, the current remains roughly constant over the duration of the discharge as regulated by the electronic source circuit, while the voltage and length of the arc increase continuously, the former up to a maximum of 30 V. Data for four different current values (70, 100, 150 and 250 mA) are used in the proceeding analyses. Images are recorded every 10 μs, of which five are shown. A voltage and current value is assigned to each image as shown.

2.1. Voltage and current

The quasi-static relationship between voltage (v), current (i_d) and length (l) in μm of the discharges takes the form of [8]

\[ v(i, l) = v_{\text{fall}} + \alpha l \left( 1 + \frac{\beta}{l^0} \right) ; \quad t_0 < t < t_{\text{end}} \]

\[ l(t) = u_{\text{arc}} t. \]  

The length is modelled as increasing linearly over the duration of the discharge (t_0 to t_{end}, as shown in figure 2 with velocity u_{\text{arc}}). The quantity v_{\text{fall}} is known as the fall voltage, and is the minimum voltage for which the discharge can exist. Fall voltage is a property of the electrode materials, and is given a constant value of 10 V based on experimental observation. As this value compares favourably to cathode fall voltage values for cadmium given in the literature [11, 12], it is assumed that v_{\text{fall}} consists entirely of a cathode fall and does not arise from anode effects. The remaining parameters α, β and n are fitted from the data.

Discharge lengths are determined using simple image analysis techniques. Here, and for the subsequent analyses of section 2.2, it is assumed for simplification that the light intensity of the discharge recorded over the spectral sensitivity of the high-speed camera (mainly the optical range) scales in good approximation with the current density of the discharge, and therefore indicates the active discharge area. Details like the change of atomic and ionic radiation intensities with species temperatures in the plasma are not considered. This assumption is utilised because the aim of the approach applied here is not the detailed determination of the power density distribution of the discharge. Rather, a rough estimation of the length and the thickness of the current path is to be obtained. Spectroscopic measurements have shown that the visible part of the discharge light emission is dominated by Cd line radiation. It is known, e.g. from welding arcs, that metal vapour regions can show lower temperatures and conductivities in comparison with the surrounding gas. In gas metal arc welding processes within argon shielding gas, the current path does not correspond to the region where metal vapour is present [13]. In molecular shielding gases, however, this effect disappears and the current path well coincides with the light emission in the visible range (see e.g. [14]). The situation of a low-power metal vapour discharge in an air-hydrogen mixture studied here is assumed to behave similarly to the latter case.

In the first step of the analysis, the greyscale image of the discharge is converted to monochrome, setting to white all parts of the image with intensity greater than 20% of the maximum value. Next, small white areas of the image are removed using a
non-linear filtering operation known as morphological opening [15]. The largest contiguous white region of the image is then selected, and presumed to be the discharge area. The centroid and major and minor axes of this region are then calculated, with the major axis taken as the estimate of discharge length. An example of this analysis procedure is shown in figure 3. Here, the ellipse corresponding to the identified major/minor axes is shown, superimposed on the original greyscale image.

The empirically determined parameters of (1) and a systematic length error \( \theta_4 = l_{\text{err}} \) comprise a vector \( \theta \), whose value can then be estimated by non-linear least squares minimisation of the function

\[
\hat{\theta} = \arg\min_\theta f(\theta) = \hat{\mathbf{V}}(\theta_1, \theta_2, \theta_3, \mathbf{I}, \mathbf{L}, \theta_4) - \mathbf{V}
\]

where \( \mathbf{V}, \mathbf{I} \) and \( \mathbf{L} \) are vectors of all measured voltage data, and their corresponding currents and lengths. \( \mathbf{V} \) is the voltage predicted by the model (1), and is therefore a function of the fit parameters \( \theta_1-\theta_4 \) as well as measured current and length data. Here, the systematic length error \( (\theta_4 = l_{\text{err}}) \) is added to the measured length. The measured data together with the fitted voltage/length relationship is shown in figure 4 for current values of 100 and 250 mA.

By linearising \( f(\theta) \) about its least squares minimiser \( \hat{\theta} \), asymptotic estimates of the standard error for each parameter \( \theta_i \) can be calculated as [16]

\[
\text{ASE}_{\theta_i} = \frac{||f(\hat{\theta})||}{\sqrt{N-P}} \sqrt{\text{diag}(J_f(\hat{\theta})J_f(\hat{\theta})^T)}
\]

where \( N \) and \( P \) are the sizes of \( \mathbf{V} \) and \( \theta \) respectively. The fitted parameter values with asymptotic estimates of the associated standard error are given in table 1. The corresponding data and fitted length/voltage lines for two of the current values are shown in figure 4. The relatively large errors are a reflection of the difficulties involved in the experiment and the image based evaluation methods, as discussed in [10].

### 2.2. Spatial resistive heating distribution

As noted in section 2.1, the discharge light intensity is used to approximate current density. Under a further assumption of uniform electric field within the conducting region, the light intensity also provides the relative spatial distribution of resistive heating. A rotationally symmetric and axially homogeneous structure of the current path is assumed to provide an estimate for this distribution. Unfortunately, the image data are not appropriate to deduce the axial variation of the structure. However, this is of lower importance because of the electric current conservation. Note that this description of resistive heating is later incorporated into the computational model, which includes convection and conduction to calculate the discharge power distribution (see section 3.1). The distribution of resistive heating is given by another empirical term, namely

**Table 1.** Fitted parameter values for (1) with corresponding asymptotic standard error estimates (ASE).

| Parameter/unit | Value | ASE (±%) |
|----------------|-------|----------|
| \( \alpha/\text{V} \mu\text{m}^{-1} \) | \( 3.35 \times 10^{-2} \) | 11 |
| \( \beta/\text{A}^2 \) | \( 3.16 \times 10^{-1} \) | 35 |
| \( n/- \) | \( 7.09 \times 10^{-1} \) | 9 |

![Figure 4](image1.png)

**Figure 4.** Voltage/Length data with corresponding fitted lines from (1). Data for two of the four current \( (I_d) \) values are shown.

![Figure 5](image2.png)

**Figure 5.** Example of intensity line profile (above) and curve fitting (below) to determine \( \sigma \) value.
\[ q_{ac}(r, x) = \frac{(v - v_{fall})i_d}{\sigma^2 2\pi} \exp \left( -\frac{r^2}{2\sigma^2} \right) \]

for \( t_0 < t < t_{end}, \ 0 < x < l \).

This term describes the discharge as cylindrically symmetric, for the axial coordinate \( x \) and the radial coordinate \( r \). The distribution of power is thus Gaussian in the radial direction and a uniform in the axial direction. This Gaussian approximation has been used in classical models of electric arcs [17].

The parameter \( \sigma \) of (5) is derived from line profiles of intensity taken from the images (figure 5). The Gaussian function is fitted to this profile data, providing an estimate of \( \sigma \). This analysis is carried out across the image dataset to determine the relationship between \( \sigma \) and the length and current of the discharge. In the absence of a physical model, the dependence of \( \sigma \) on both quantities is presumed to be linear, with an expression of the form

\[ \sigma = \sigma_0 + a_\sigma l + b_\sigma i_d \]  

for length coefficient \( a_\sigma \) and current coefficient \( b_\sigma \). This expression is fitted to the data using a conventional linear least squares method, and standard errors of the parameters calculated in the usual manner [16]. The data and fit are shown in figure 6, and parameters with corresponding standard errors of regression are shown in table 2.

### 2.3. Model validation

The validity of the electrical part of the empirical model can be assessed by examining voltage and current predictions under various circumstances. In section 1, it was noted that the electrodes were connected to an electronic source circuit designed to maintain constant current. By replacing this special circuit with a simple resistive or inductive source, a discharge with time varying current is produced, and its electrical waveforms recorded. These circuits, together with a component described...
The source terms $-\sigma \cdot (\rho u) = \frac{1}{\tau}u(\nabla \cdot p)$ are calculated by solving \( \nabla \cdot \sigma = p - \nabla \cdot \mathbf{t} \). These are derived from the Newton–Euler equations

\[
\frac{\partial}{\partial t} \cdot (\rho u) + \nabla \cdot (\rho uu) + \nabla p = \nabla \cdot \mathbf{t} + \rho \mathbf{g}
\]

for pressure $p$ and gravity vector $\mathbf{g}$. The viscous stress vector $\mathbf{t}$ is calculated from Newton’s law

\[
\mathbf{t} = -\mu(\nabla u + (\nabla u)^T) - \frac{2}{3}(\nabla \cdot u)\mathbf{I}
\]

for dynamic viscosity $\mu$ and identity matrix $\mathbf{I}$. Conservation of enthalpy $(h)$ is given by

\[
\frac{\partial}{\partial t} \rho(h + e_k) + \nabla \cdot (\rho(h + e_k) + \mathbf{j}_k) = \frac{\partial}{\partial t} q_{\text{src}} + q_e
\]

for heat flux $\mathbf{j}_\mathbf{q}$, specific kinetic energy $e_k = \frac{1}{2}u \cdot \mathbf{u}$, chemical heat release term $q_e$, and $q_{\text{src}}$ as per (5). Finally, conservation of the $i$th chemical species is given by

\[
\frac{\partial}{\partial t} \rho Y_i + \nabla \cdot (\rho u Y_i + \mathbf{j}_i) = \Omega_i
\]

for species mass fraction $Y_i$, production rate $\Omega_i$, and diffusive flux $\mathbf{j}_i$. The source terms $q_e$ and $\Omega_i$ are calculated by solving a 19-reaction mechanism for the oxidation of hydrogen [19]. The simulated gas model thus includes a total of 10 species, namely those of the original mixture (H$_2$, O$_2$, N$_2$), products of reaction and dissociation (H$_2$O, H$_2$O$_2$, HO$_2$, OH, O, H), as well as evaporated Cd.

In the above equations, pressure is related to density by the ideal gas equation $p = \rho RT$ for specific gas constant $R$ and temperature $T$. Temperature is derived from enthalpy by numerically inverting the equation $h = \int_0^T c_p dT$, where $c_p$ is the temperature dependent mixture specific heat, obtained from a thermodynamic database [19]. The diffusive fluxes are given by

\[
\mathbf{j}_i = -\rho D_i \nabla Y_i - \frac{D_T}{T} \nabla T
\]

and

\[
\mathbf{j}_k = -k \nabla T + \sum h_i \mathbf{j}_i
\]

for mixture averaged diffusion coefficients $D_i$, thermodiffusion coefficients $D_T$, and thermal conductivity $k$. These transport parameters, together with viscosity $\mu$, are determined as a function of temperature using the first Chapman–Enskog approximation [20] for each of the above-mentioned pure species. The required collision integrals are based on the Lennard-Jones potential and interpolated from the tables of [21]. The mixture transport properties are then calculated using mixing rules [22].

The use of the empirical term $q_{\text{src}}$ in equation (10) replaces the explicit consideration of ionic cadmium and free electrons as species in the gas mixture, as well as the explicit calculation

3. Conservation equations

The conservation equations of the model are given in (7)–(11). Conservation of mass is stated by

\[
\frac{\partial}{\partial t} \rho + \nabla \cdot (\rho u) = 0
\]

for density $\rho$ and velocity $u$. Additionally, conservation of momentum is given by the Navier–Stokes equations

\[
\frac{\partial}{\partial t} \rho(uu) + \nabla \cdot (\rho(uu) + \nabla p) = \nabla \cdot \mathbf{t} + \rho \mathbf{g}
\]

for pressure $p$ and gravity vector $\mathbf{g}$. The viscous stress vector $\mathbf{t}$ is calculated from Newton’s law

\[
\mathbf{t} = -\mu(\nabla u + (\nabla u)^T) - \frac{2}{3}(\nabla \cdot u)\mathbf{I}
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\]

for mixture averaged diffusion coefficients $D_i$, thermodiffusion coefficients $D_T$, and thermal conductivity $k$. These transport parameters, together with viscosity $\mu$, are determined as a function of temperature using the first Chapman–Enskog approximation [20] for each of the above-mentioned pure species. The required collision integrals are based on the Lennard-Jones potential and interpolated from the tables of [21]. The mixture transport properties are then calculated using mixing rules [22].

The use of the empirical term $q_{\text{src}}$ in equation (10) replaces the explicit consideration of ionic cadmium and free electrons as species in the gas mixture, as well as the explicit calculation

3. Physics

The resistive heating term of (5) is simulated in a gas dynamics model together with a simplified consideration of heat conduction and evaporation in the solid cadmium electrode via boundary conditions. The same gas mixture of 21% hydrogen in air is used for the simulations.

The geometry and dimensions of the computational domain are described in figure 9. This is intended to represent the scenario of figure 1, excluding the moving tungsten wire electrode. The relevant conservation equations and boundary conditions are discussed in this section, together with details of the calculation of electrical conductivity based on an equilibrium plasma composition.

Figure 9. The computational domain of the simulation. The region is bounded by a plane of symmetry (at $z = 0$), two surfaces of the block electrode, and ‘open’ boundaries (marked in blue).
of electric and magnetic fields, current density, and electrical conductivity within the simulation. This is in contrast to a full magnetohydrodynamic model. The assumptions implicit in the formulation of $q_{acc}$ have been noted in sections 2.1 and 2.2. The approach presented here also assumes that magnetic field and Lorentz forces can be neglected (justifiable by the low current), and that radiation losses are low (justifiable by the dominance of Cd lines in the radiation spectrum, and their relatively low intensity compared to other metal vapours). An additional assumption is that the presence of free electrons and cadmium ions do not appreciably affect the specific heat $c_p$, or the transport coefficients of equations (9), (12) and (13). The implications of these assumptions will be further discussed in section 5.

3.2. Boundary conditions

The open boundaries of the computational domain are simulated using a Dirichlet boundary condition for pressure $p = 1$ atm and applying Neumann boundary conditions to all other variables.

Heat conduction at the electrode surface is approximated by solving the 1-D diffusion equation in the perpendicular direction for each point on the surface

$$\rho_s c_{ps} \frac{\partial T}{\partial t} = \frac{\partial q}{\partial x_s}$$

$$q = k_s \frac{\partial T}{\partial x_s} \quad \text{for} \quad x_s > 0$$

where $x_s$ is the depth. Here $\rho_s$, $c_{ps}$, and $k_s$ are the density, specific heat and heat conductivity of cadmium, assumed constant for the simulation. The equation is solved up to a depth of 100 $\mu$m, corresponding to a 2 ms characteristic time of heat conduction. As suggested by [23], heat flux and constant temperature boundary conditions are used for the solid (14) and gas (10), respectively, with the value for each derived from the other’s solution.

The heat flux at the surface $q_{x_s=0}$ comprises of three components, namely

$$q_{x_s=0} = q_{cond} + q_{fall} - q_{vap}$$

The first of these is the heat conducted from the gas domain.

$$q_{cond} = -k_g \frac{\partial T_{gas}}{\partial x_s}$$

The second term represents the heating produced in the cathode fall region, which is assumed to possess the same radially symmetric spatial distribution as the power within the discharge—as described in (5)

$$q_{fall} = \frac{v_{fall} i_d}{\sigma^2 2\pi} \exp \left( -\frac{r_s^2}{\sigma^2} \right)$$

where $r_s$ represents distance along the surface from the origin of the discharge. It is also assumed here that this energy is completely transferred towards the cathode surface and not to the plasma. The last component represents heat lost due to evaporation

$$q_{vap} = (L_v + c_{ps} T_{x_s=0}) J_m$$

where $L_v$ is the specific heat of vaporisation. $J_m$ is the surface normal mass flux due to evaporation. This is calculated with

$$J_m = -\rho T_{Cd} \frac{\partial Y_{Cd}}{\partial x_s} + \alpha_e \frac{p_v}{\sqrt{2\pi R_{Cd} T_{x_s=0}}}$$

Table 3. Overview of simulated cases. Parameter values are as described in figure 2 and (1).

| Case | Current/mA | Duration/µs | $u_{acc}$/m s$^{-1}$ |
|------|------------|-------------|---------------------|
| 1    | 70         | 200         | 0.96                |
| 2    | 100        | 200         | 1.14                |
| 3    | 70         | 1000        | 0.19                |
| 4    | 100        | 1000        | 0.23                |

Figure 10. Example isosurface plots for subset of field data for temperature (left) and cadmium partial pressure (right). Data is from case 2 at $t = 150$ µs.

Table 3. Overview of simulated cases. Parameter values are as described in figure 2 and (1).

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| 1    | 70         | 200         | 0.96                |
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The heat flux at the surface $q_{x_s=0}$ comprises of three components, namely

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The first of these is the heat conducted from the gas domain.

$$q_{cond} = -k_g \frac{\partial T_{gas}}{\partial x_s}$$

The second term represents the heating produced in the cathode fall region, which is assumed to possess the same radially symmetric spatial distribution as the power within the discharge—as described in (5)

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where $r_s$ represents distance along the surface from the origin of the discharge. It is also assumed here that this energy is completely transferred towards the cathode surface and not to the plasma. The last component represents heat lost due to evaporation

$$q_{vap} = (L_v + c_{ps} T_{x_s=0}) J_m$$

where $L_v$ is the specific heat of vaporisation. $J_m$ is the surface normal mass flux due to evaporation. This is calculated with

$$J_m = -\rho T_{Cd} \frac{\partial Y_{Cd}}{\partial x_s} + \alpha_e \frac{p_v}{\sqrt{2\pi R_{Cd} T_{x_s=0}}}$$
where the first term represents diffusion flux and the second bulk evaporation according to the Langmuir relation [24]. Here, $D_{Cd}$ and $Y_{Cd}$ represent the diffusion coefficient and mass fraction of cadmium at the surface. $R_{Cd}$ is the specific gas constant for cadmium. The equilibrium vapour pressure $p_v$ is estimated as a function of temperature from the August equation coefficients given in [25]. An accommodation coefficient $\alpha_e$ is calculated from the ratio of $p_v$ to the pressure in the gas domain at the surface ($p$), according to the equations of [24]. Here, $\alpha_e = 0$ for $p_v/p \leq 1$ and asymptotically approaches a value of approximately 0.82 as $p_v$ increases beyond $p$. The two terms of equation (20) thus account for the evaporation mechanisms dominant when vapour pressure is below and above the surrounding gas pressure respectively.

To account for the evaporation process, Dirichlet boundary conditions on the surface are also imposed for velocity in the surface normal direction ($|u| = J_m/\rho$) and cadmium mass fraction ($Y_{Cd} = pM_{Cd}/(pM)$), where $M_{Cd}$ and $M$ are the molar mass of cadmium and and the mixture average molar mass, respectively.

4. Simulation results and analysis

The equations of section 3 are implemented in the OpenFOAM finite volume method solver [26]. The pressure implicit with splitting of operator (PISO) method is used to solve for pressure and velocity [27]. Transport parameters in the conservation equations are calculated using the Cantera software [28].

Simulations are conducted for the discharge parameter values given in table 3, these being representative of the data collected in [9] and of the application in general. Here, the simulated discharges are specified in terms of current ($i_d$) and duration ($t_{end}$), from which velocity $u_{arc}$ is calculated using (1). For each simulation, the time varying fields of mass fraction for cadmium are used to calculate the corresponding partial pressure. Example fields for temperature and cadmium partial pressure are shown in figure 10. A notable characteristic of figure 10 is the elongated distribution of temperature and cadmium partial pressure. Note that the discharge length at this point is around 170 $\mu$m, however the high temperature region extends well beyond this length. This is due to a jet of evaporated material produced by the cathode fall region heating, which can be seen in the velocity distribution of figure 11.

4.1. LTE electrical conductivity

The cadmium partial pressure and temperature fields are next used to calculate an electrical conductivity field, under the assumption of LTE. For the calculation of electrical conductivity as a function of these two quantities, a pure cadmium plasma at a pressure equal to the partial pressure of cadmium in the gas mixture has been assumed. This result provides a good indication that the presence
of other neutral species would not appreciably affect electrical conductivity.

Determination of the transport properties of this cadmium plasma has been performed using the procedure presented in [29]. First, the plasma composition is calculated. The temperature dependent densities of plasma species, namely, electrons, atoms and ions can be predicted using the method of the minimisation of generalised Gibbs free energy [29]. The following components have been taken into account: electrons $e$, atoms Cd, ions Cd$^+$, Cd$^{2+}$, Cd$^{3+}$ and Cd$^{4+}$. The densities have been calculated for various vapour pressures. When the densities are known, transport parameters for the corresponding pressures and temperatures can be calculated.

Similarly to the calculation of gas transport parameters (section 3.1), the Chapman–Enskog theory is utilised for obtaining transport properties of the thermal plasma in equilibrium state [20]. In the specific case of a monatomic gas, transport parameters can be obtained using the approach developed by Devoto [30]. The fourth order of approximation was used for determination of the transport coefficients. The starting point for the determination of transport coefficients is the calculation of kinetic integrals which requires the knowledge of the scattering cross sections as input data. Kinetic integrals for Cd–Cd collisions have been calculated using the Lennard-Jones model potential with parameters from [31]. For the determination of the data for the collisions between electrons and Cd atoms scattering cross sections from [32, 33] have been adopted. In order to describe the ion-atom interaction, polarization interaction and charge transfer collisions have been taken into account. The polarisability of cadmium has been taken from [34]. Cross section data for the resonant charge transfer have been calculated using the relations from [35]. Finally, interaction between charged particles has been characterised by the Coulomb cross-section [36]. Figure 12 shows the resulting temperature dependence of electrical conductivity for different vapour pressures.

This data can then be used to calculate electrical conductivity fields, an example of which is shown in figure 13.
4.2. Discharge resistance

The electrical conductivity field is sampled to produce an averaged distribution in 2-D cylindrical coordinates. An example of the axial and average radial profile of electrical conductivity is shown in figure 14.

The sampled axisymmetric distribution of electrical conductivity can be numerically integrated to estimate overall resistance $Z$,

$$Z = \int_{L_0}^{L_1} \int_0^{R_1} dl \int_0^{2\pi r \sigma_e} dr.$$  \hspace{1cm} (21)

The radial limit of integration $R_1$ can simply be made large enough to include the conductive region (200 $\mu$m is selected here). The axial limits $L_1$ and $L_0$ require consideration. As the axial profile of figure 14 shows, conductivity is negligible near the ends of the discharge, particularly on the cathode (left) side. Since the electrode layer effects are not modelled, including the areas near the electrodes in the integration would not be meaningful. The analysis is thus conducted by integrating only over the region where conductance is no less than 20% of its maximum value. Under these conditions, the evaluation of (21) will provide a lower bound on possible resistance.

The resistance at each time step of the simulated cases was calculated both (21) and the empirical relationships of (1), as $Z = v(l,i)/i_d$. A comparison of the results from both calculation methods can be seen in figures 15 and 16, for the simulated discharges of 200 and 1000 $\mu$s, respectively. Here, the expected values are those predicted by (1) as described above. The comparison provides an indication of consistency of the model under the LTE assumption. The results show a difference of at least an order of magnitude, which is particularly stark at the beginning of the discharge period.

4.3. Electron temperature

As the resistance calculated from the simulated gas temperatures are significantly higher than the purely empirical estimates, it can be concluded that a departure from thermodynamic equilibrium is present. This would mean that electron temperatures are significantly higher than those of the heavier species in the gas, and that electrical conductivity is determined primarily by the former. Here, an assumption can be made that the relationship between electrical conductivity and electron temperature is approximately independent of heavy particle temperature, where electron temperatures are low. This was previously demonstrated for a non-metallic plasma [37]. Based on this assumption, together with that of uniform electric field (as noted in section 2.2), the data of figure 12 can be used together with equation (5) to estimate the distribution of electron temperatures in the discharge. This is accomplished by noting that

$$\sigma_e = \frac{q_{ac}}{E} = \frac{i_d}{(v - v_{fall})\sigma_e^2} \exp \left( -\frac{r^2}{2\sigma_e^2} \right)$$  \hspace{1cm} (22)

for electric field $E = (v - v_{fall})/l$, and then interpolating the corresponding temperature values from figure 12 (neglecting variations in cadmium partial pressure). The results of this analysis are summarised in figures 17 and 18. It can be seen here that a final electron temperature around 35% higher than the heavy particle temperature would account for the observed resistances at the end of the discharge.

5. Discussion

The results of section 4.2 show that the predictions of an LTE model diverge significantly from the empirically observed properties of the electrical discharge. Even under the favourable assumptions made (i.e. no radiation losses, near electrode regions assumed perfectly conducting), the discrepancy between electrical resistances predicted by simulation and those determined empirically is not less than one order of magnitude. In addition to the difference in magnitude of the electrical resistance, the way in which this resistance varies over the duration of the discharge is also noteworthy. Under the LTE assumption, the resistance would be expected to
decrease over time. This is because the average temperature increases over time, and the electrical resistance decreases near-exponentially with temperature (and increases only linearly with length). The data and empirical correlations of section 2, however, show that voltage is linearly proportional to length. This implies a constant electrical resistance per unit length over the duration of the discharge.

The further analysis of section 4.3 predicts that a relatively modest difference in the electron and heavy particle temperatures would be sufficient to account for the observed discrepancy, at least in the later stages of the discharge. This is due to the strongly non-linear relationship between temperature and electrical conductivity at low temperatures. Based on this predicted difference in temperatures, the plasma could potentially be described as a type of glow discharge.

A question then arises as to how the empirical model of section 2 is able to describe the electrical characteristics of the discharge with reasonable accuracy. This is surprising, given that the hyperbolic relationship between voltage and current is based on LTE, specifically, the idea that higher currents produce a broader and hotter (therefore more conductive) discharge column. It is possible that the apparent applicability of the model is coincidental. One theory of stable glow discharges suggests that a constant current density at the cathode is maintained, with increases in current only increasing the size of the electron emitting area (or cathode spot) [38]. It is conceivable that this would, in turn, also increase the width of the discharge column, leading to a qualitatively similar voltage/current relationship as in the case of a stable arc. These ideas would require confirmation through experiment and/or more detailed simulations.

5.1. Assumptions and uncertainties

The various assumptions made in this work inevitably introduce uncertainties into the results. Although a rigorous quantification of the uncertainties is currently not possible, some qualitative arguments will be presented here.

The use of neutral gas CFD simulation together with the pure Cd based calculation for electrical conductivity implies the assumption that:

- Electrical conductivity is negligibly affected by the non-metal gas components, and
- Thermodynamic and transport properties (excluding electrical conductivity) are negligibly affected by excitation and ionisation processes

Significant errors would not be expected due to the former of these, because, as noted in section 2.2, previous work shows that current flows predominantly through the metal vapour when the surrounding gas is molecular. To gain some insight into the implications of the second assumption, viscosities and thermal conductivities for pure cadmium are calculated both with a neutral gas (as per section 3.1) and LTE plasma (as per section 4.1) approach. The results of this comparison, shown in figures 19 and 20 indicate that the errors in the neutral gas based calculation increase with temperature, reaching around 25% for both viscosity and thermal conductivity of a temperature of $6000$ K. This could reasonably be considered an upper bound, as the calculation is conducted in pure cadmium, whereas other neutral gases are present in the majority of the simulated domain. The implications of underpredicting these parameters is likely to be a slight over-prediction of velocities and heavy particle temperatures.

The numerical uncertainty of the calculated composition and transport properties is determined by the numerical methods used, and is estimated to be as low as 1 ppm. However, the main source of possible deviations in transport parameters are the cross sections for various collision processes. Thus, for the temperature region of interest ($4000–6000$ K) the most relevant collisions are between electrons and Cd atoms, as well as the charge transfer collisions. There are only few sources with corresponding data available. Therefore, it is not possible to estimate the accuracy of available data. The values of electrical conductivities has been calculated using the fourth order approximation [30] which guarantee accuracy of at least a few percent. On the other hand, the electrical conductivity is strongly temperature dependent in the above mentioned temperature region. Thus, when the temperature rises...
over 50%, electrical conductivity changes over more than one order of magnitude. Therefore, even when the value of electrical conductivity deviates by a factor of two (upper level for data uncertainty), the error in temperature determination will be less than 10%.

6. Conclusions and future work

The results have significant implications for the simulation of combustion initiated by the electrical discharges in question. Thus far, the empirical modelling approach has shown some success in predicting electrical properties of the discharge, and has previously been applied to the modelling of the combustion process [4]. If a more rigorous approach is desired, however, this work shows that non-equilibrium thermodynamics must be considered. As a first step, emission spectroscopy could be used to determine excitation temperatures in the plasma, which can be compared to the rough estimates of electron temperature presented in this work. Ultimately though, a two temperature approach is required for a physical simulation of the plasma. This in turn would necessitate a more rigorous treatment of the electrodes, including thermo-field electron emission. Additionally, the treatment of radiation and a more sophisticated experimental comparison would be enabled by a collisional-radiative model. These extensions to the work pose significant challenges due to the uncommon material involved (cadmium), for which limited published data exists.

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ORCID IDs

Rajiv Shekhar https://orcid.org/0000-0002-1692-9848
Holger Grosshans https://orcid.org/0000-0001-6441-7225
Dirk Uhrlandt https://orcid.org/0000-0001-6534-147X

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