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Theoretical Modeling of Chemically Reacting Flows

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Abstract: ***This paper describes a modeling investigation of a three-dimensional fluid flow and heat transfer inside a high temperature optical cell. The convective fluid flows are driven by the hot tungsten filament located centrally in the spherical cell which contains nitrogen gas doped with halogen-containing gas, added as part of a filament regenerative cycle. The fluid flows within the cell play a vital role in governing the chemical and physical processes operating within the system. The model equations of steady continuity, Navier-Stokes and energy are numerically solved with a finite volume method. The chemical reactions at thermodynamic equilibrium are solved using local Gibbs energy minimization method. The numerical results of the thermal and dynamic study inside shows the convective flow within the cell and the effect of concentration dependence of different chemical species present in the cell in operation with the filament temperature. Amongst several important outcomes of the work is the observation of the effect of thermal diffusion on the relative concentration of Brome atoms in the high temperature, intra-coil region.***

Keywords: Heat transfer, fluid flow, CFD, Gibbs energy, halogen light source.

1. Introduction

Many modern day processes of scientific and industrial importance involve fluid flow and heat transfer [1-2], the understanding of which has drastically improved in recent decades. In addition to making experimental measurements, many of these complex processes can be modeled through the use of numerical simulation techniques. The different light source types make up a wide range of systems where high temperature reactive gas flows can be found [3-5]; others include propulsion and heat generation systems [6-7].

The purpose of this study is to investigate the complex and hostile environment found within a simulated halogen light source and develop a representative theoretical model. The spatially precise knowledge of both physical conditions, and multispecies chemical composition, over a wide range of operating conditions of the source creates an important opportunity to understand such complex systems through the use of computer based models. The model is simulated numerically on the basis of the computational fluid dynamics (CFD) method which implements local Gibbs energy minimization [8-9]. This work was carried out using the general purpose fluid dynamic computer code, Fluent [10]. Fluent is a state of the art CFD computer package for modeling fluid flow and heat transfer problems in complex geometries.

2. Problem Definition

The present model consists of a spherical optical cell of diameter 22mm, containing 95% of molecular nitrogen and 5% of hydrogen bromide HBr. Nitrogen gas filling is required because it has favorable thermal conductivity properties and has little tendency to arc. The HBr dose is added into the lamp as part of a filament regenerative cycle. A filling pressure of 0.79 atm is used to maintain significant buoyancy driven gas flows in the cell. The coil tungsten filament is represented by rings enabling the simulation of the flow inside and outside of the filament. The filament temperature can be as high as 3500K with a corresponding temperature of the wall of the envelope of 600K. These are experimental values. Physical constants relevant to the case under study include the operating pressure, density, heat capacity and transport properties of the fluid. The transport properties (viscosity and thermal conductivity) and the heat capacities are specified as temperature dependent polynomials. In the present investigation, the data of Svehla [11] is used to generate the best fit polynomials.

3. Numerical Procedure

The flow and heat transfer phenomena to be investigated here are basically described by the equations of continuity, Navier-Stokes and energy. The fluid is regarded as Newtonian and incompressible; the flow is laminar in steady regime. Radiation heat transfer is negligible. Equation of type "poisson" for the correction of the pressure, derived from the equation of continuity and the equations of momentum linearized, will be solved to obtain the corrections of the fields pressure and velocity necessary to satisfy continuity.

Continuity equation:

$$\frac{\partial \rho}{\partial t} + div(\rho \mathrm{u}) = 0 \qquad (1)$$

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} = div(\mathrm{u})$$

Momentum conservation

x-momentum

$$\frac{\partial(\rho u)}{\partial t} + div(\rho u \mathrm{u}) = -\frac{\partial p}{\partial x} + div(\mu grad u) + S_{Mx}$$

y-momentum

$$\frac{\partial(\rho v)}{\partial t} + div(\rho v \mathrm{u}) = -\frac{\partial p}{\partial y} + div(\mu grad v) + S_{My}$$

z-momentum

$$\frac{\partial(\rho w)}{\partial t} + div(\rho w \mathrm{u}) = -\frac{\partial p}{\partial z} + div(\mu grad w) + S_{Mz}$$

(2)

$S_{Mx} = -\rho g$, $S_{My} = 0$ et $S_{Mz} = 0$.

S_{Mx}, S_{My}, S_{Mz} : source terms

Energie equation:

$$\frac{\partial(\rho i)}{\partial t} + div(\rho i \mathrm{u}) = -p div \mathrm{u} + div(\mu grad T) + \phi + S_i \quad (3)$$

$div(\mathrm{u}) = 0$ For an incompressible fluid.

The general equation of transport is written as follow:

$$\frac{\partial(\rho\phi)}{\partial t} + div(\rho\phi \mathrm{u}) = div(\Gamma grad\phi) + S_\phi$$

ρ is volumic mass, i is intern energy and is the exchange diffusion coefficient of

To provide a link between Fluent and a sub-model to calculate the thermodynamic chemical distribution the species user defined function set was written. The model takes an initial set of conditions and starting concentrations and allows equilibrium thermodynamic chemical distribution to be calculated as the fluid flow field is solved. The state of equilibrium of a chemical system can be determined by the minimization of the Gibbs energy (Equation 4) with constant volume and constant pressure:

$$G = \sum_{j=1}^{N} n_j . \mu_j \quad (4)$$

n_j is the number of mole and μ_i is the chemical potential of the species j

The governing equations were discretized using a finite-volume method. The pressure-velocity calculations have been performed using the SIMPLE pressure correction algorithm. In this study, the solution is taken as converged when the mass continuity residual and velocities is in the order of 10^{-8}An exception to this general value is that taken for the enthalpy residual "energy equation", for which a value of 10^{-6} is more appropriate.

The symmetry of the problem (2 symmetry planes xy plane and yz plane) reduces the computational domain to ¼ of the total volume of the cell. The mesh chosen for this application was a triangular mesh consisting of 144469 grid cells. This computational grid is illustrated in Figure1.

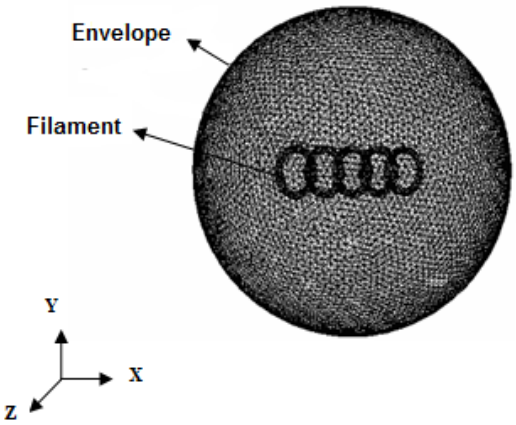

Figure 1: 3D computational grid

4. Results and Discussion

4.1 Thermal Fluxes and Fluid Flow

The importance of the fill gas is on reducing the tungsten vapor pressure at high filament operating temperatures, thus allowing the filament to be run at an elevated temperature. The fill gas temperature profiles are predicted for a filament temperature of 3500K. The temperature gradients through the fill gas caused by the difference in temperature between the walls and the filament can be extremely high; Figure 2 shows gradients as large as 1500K/mm.

The contour maps, shown in Figure 2, are taken through the xy plane and the yz plane of the cell. It can be seen that the temperature drops very quickly in the region immediately around the filament in the Langmuir sheath region [12] and that a large proportion of the cell is relatively cold.

The fluid flows within the optical cell play a vital role in governing the chemical and physical processes operating within the system. The observed fluid flows and other changes in fluid properties are a direct result of thermal energy transfer from the filament. The high filament temperature acts to increase the fill gas molecules internal energy and therefore large changes in the gas pressure and density can be observed. The changes in density are local to the different regions of the cell and it is these density gradients which, when combined with gravity, lead to the observed convective flows.

Figure 3 shows a good example of buoyancy driven flow. The hot gas forms a central plume above the filament which due to its lower density rises to the cooler top region of the cell. As the gas cools its density increases and it flows to the lower regions of the cell where it can encounter the hot filament region. The yz plane clearly shows fast flows occurring between the filament coils and more stagnant regions where the coils block this flow. In these regions the flow is at its fastest with velocities up to $12 cms^{-1}$.

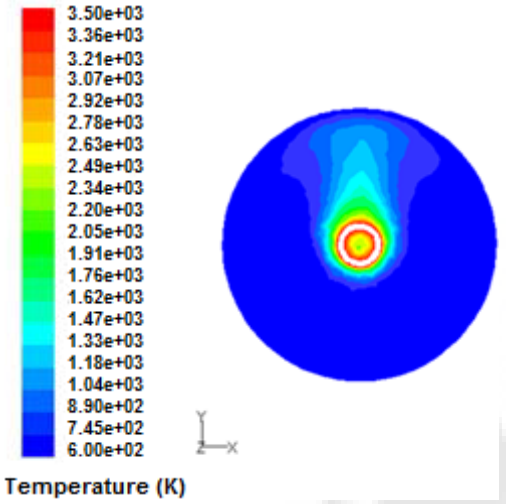

Figure 2(a): Temperature distribution in the xy planes with a filament temperature of 3500K

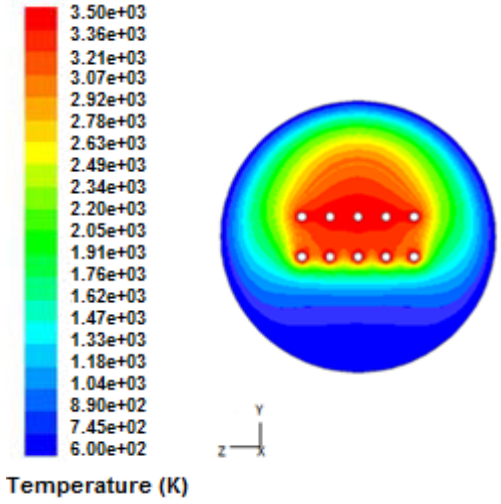

Figure 2(b): Temperature distribution in the yz planes with a filament temperature of 3500K

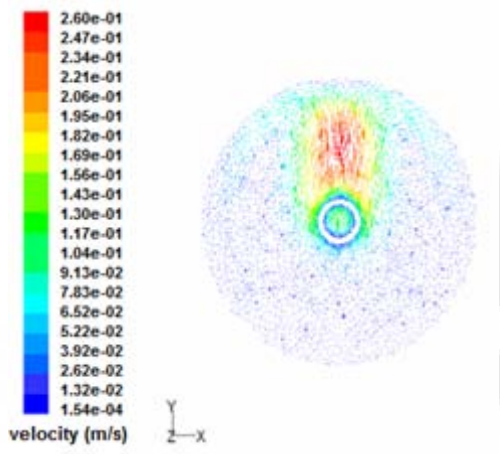

Figure3 (a): Velocity vector representing the fluid flows in the xy planes with a filament temperature of 3500K

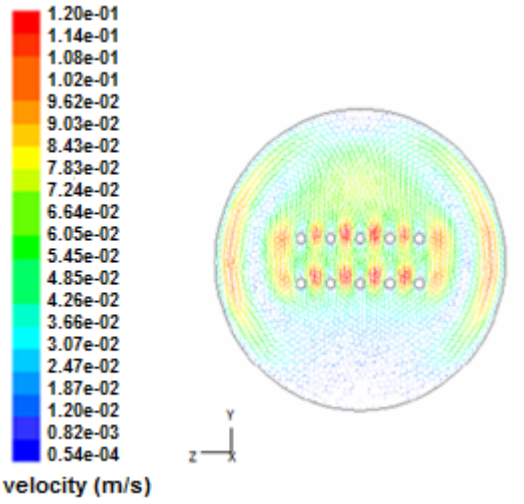

Figure3 (b): Velocity vector representing the fluid flows in the yz planes with a filament temperature of 3500K

4.2 Distribution of Chemical Species

At even modest operating temperatures the tungsten filament will gradually evaporate from the filament surface and condense on the cool wall regions causing wall blackening. The halogen undergoes reaction with the tungsten vapour, which previously would have been deposited on the walls of the lamp causing blackening, transferring the tungsten vapour back to the filament. This is known as the regenerative cycle [13].

HBr is a gas at room temperature. Within the cell this gas mixes and flows along with the fill gas; nitrogen. As the temperature of the HBr gas increases it starts to dissociate into more stable compounds through a series of well-studied mechanisms shown below. These compounds are Br, Br_2, HBr, H and H_2. It is the production of bromine atoms that is necessary for the halogen cycle.

$$H_2 + N_2 \leftrightarrow 2H + N_2$$

$$Br_2 + N_2 \leftrightarrow 2Br + N_2$$

$$HBr + H \leftrightarrow H_2 + Br$$

$$HBr + Br \leftrightarrow Br_2 + H$$

$$HBr + N_2 \leftrightarrow Br + H + N_2$$

The principle point of interest in any tungsten halogen system is the filament region. It is this region where most of the active chemistry and physical phenomena are found. A detailed investigation into how the filament temperature effects the species distributions is shown in Figure 4.

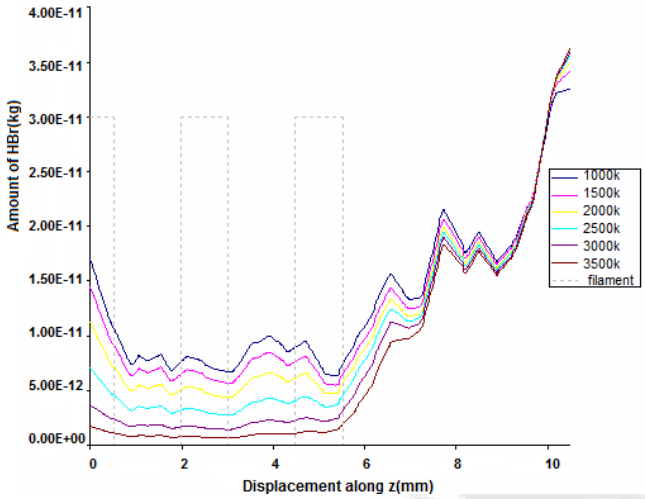

Figure 4 (a): Species distributions as a function of displacement in the z direction at different filament temperatures (HBr)

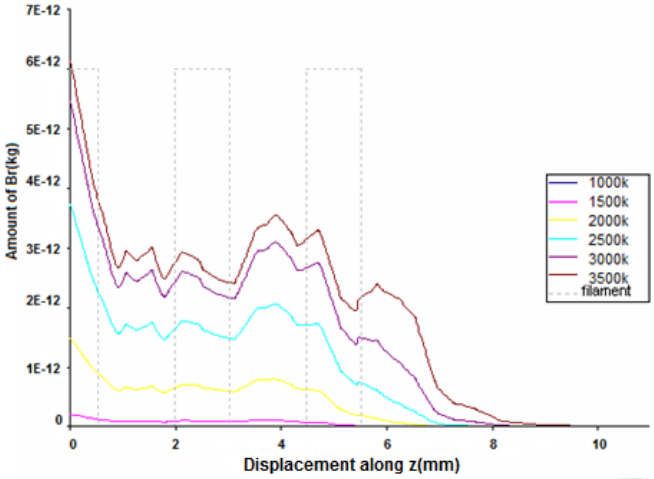

Figure 4 (b): Species distributions as a function of displacement in the z direction at different filament temperatures (Br)

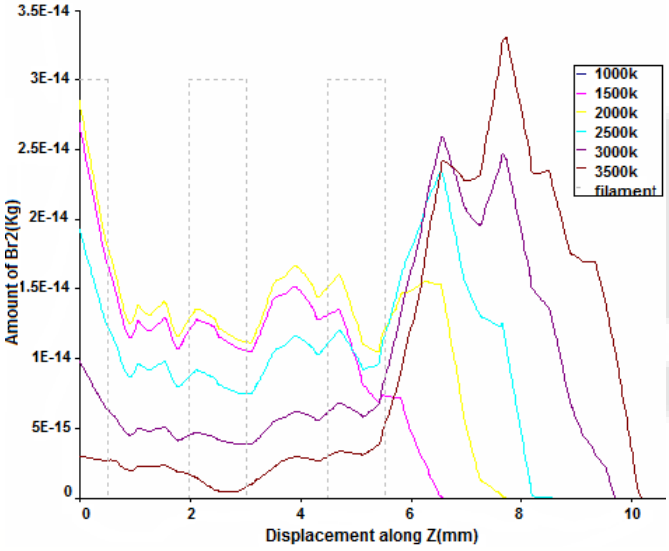

Figure 4 (c): Species distributions as a function of displacement in the z direction at different filament temperatures (Br2)

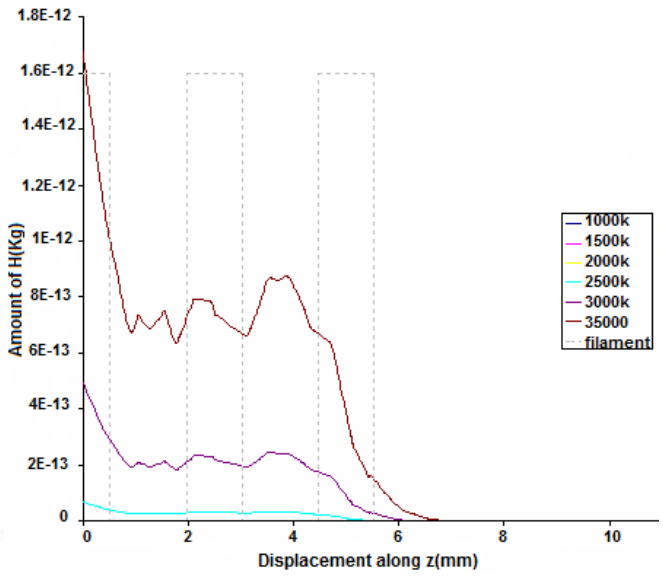

Figure 4 (d): Species distributions as a function of displacement in the z direction at different filament temperatures (H)

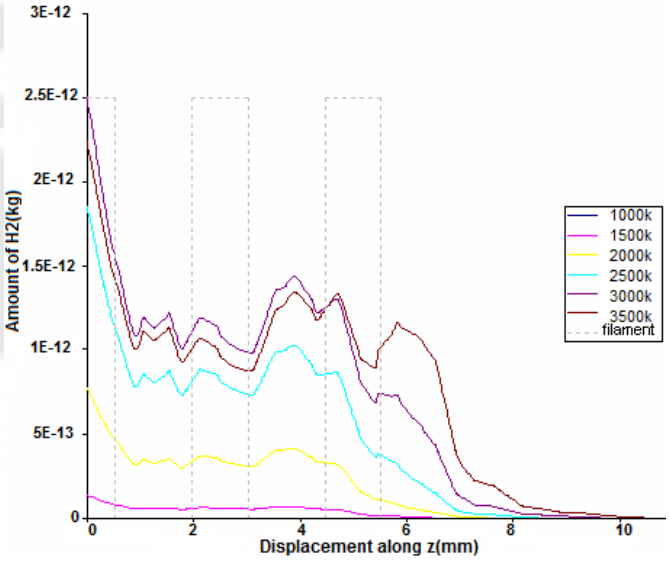

Figure 4 (e): Species distributions as a function of displacement in the z direction at different filament temperatures (H2)

Hydrogen bromide is the starting point for the halogen cycle. HBr dissociates easily at elevated temperatures. Figure 4 shows how HBr is found in the cooler regions of the cell envelope. These regions can act as a reservoir of bromine for the regenerative cycle. HBr also acts as to prevent excess Br atom being present at lower filament operating temperatures. At lower filament temperatures, in the region of 1800K, bromine atom will cause erosion to the tungsten filament.

The amount of bromine atom depends on the temperature; the hotter the filament temperature, the higher the bromine atom concentration. Figure 4 shows the largest amounts of bromine atoms can be found in the hottest parts of the cell. The hottest gas temperatures are found within the coils and within the convection plumes from the filament surface. The major bromine atom production occurs directly around the filament, or when compared to the temperature profile occurs

within the Langmuir sheath around the hot filament. This is exactly what is required by the halogen cycle; the greatest abundance of bromine atoms is where the greatest abundance of tungsten vapor would be found.

There is a small degree of competition between bromine as a molecule and bromine as an atom in the lower temperature region, 900 to 3200K. The most favorable temperature for Br_2 production is about 1800K, bromine molecule is the predominant form of bromine in regions of the cell with temperatures between 980K and 1040K. Figure 4 shows that the bromine molecule is being formed at the cooler boundaries of the Langmuir sheath where the temperature is low enough not to dissociate the molecule into its constituent atoms.

To get hydrogen atoms produced by the thermal dissociation of HBr the temperature has to be very high. Hydrogen atom does not start being produced until the temperature is in excess of 2000K. At lower temperatures the preferred state for hydrogen is the hydrogen molecule which is stable at much lower temperatures. Figure 4 shows that the hydrogen atom production occurs very close to the filament.

The hydrogen molecule gets produced as HBr starts dissociating at the lower temperatures. The hydrogen molecule acts as a store for the excess hydrogen which is created as bromine is produced. As the temperature increases, so do the temperature gradients and also the flows, the effect of which is to isolate a region within the coils where a low temperature region is confined by the hot filament. In this region the effects of mixing by the gas flows becomes reduced allowing thermal diffusion to become more apparent. The lighter hydrogen species will migrate to the hot regions immediately around the filament and the heavier bromine species will migrate to the cooler region in between the coils.

5. Conclusions

The numerical results of the thermal and dynamic study inside a cell simulating halogen source, shows the convective flow within the enclosure and the effect of concentration dependence of different chemical species present in the cell in operation with the filament temperature. Increasing the filament temperature, results in an increase of the gas temperature near the filament, and the creation of large temperature gradients in the volume of the cell. An important observation is the effect of thermal diffusion on the concentration of the Bromine atoms at high temperature. The results presented are showing that chemical species distributions can be solved at the same time as the temperature and flow fields in a CFD analysis of a high temperature optical cell. This is the basis for a powerful and complex chemical model. As the scalar transport takes care of the individual elements the final chemical distribution is calculated on an individual cell level then multiple phases can be included. Overall this approximates condensation and evaporation effects.

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Author Profile

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