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RESEARCH ARTICLE - MECHANICAL ENGINEERING

CFD Modeling of Toxic Element Evolved During Coal Combustion

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Abstract The combustion of coal is a complex series of reactions, dominated by the transport mechanism. An aspect that is poorly understood is the influence that the evolution of trace elements has on the environment and the pollution level. To explore this impact, a mathematical model of arsenic was incorporated into a computational fluid dynamics code to predict the behavior of its evolution during the combustion of pulverized coal inside a Drop Tube Furnace. Coal particles are treated as non-interacting spheres with full coupling of mass, momentum and energy with the gaseous phase. The eddy dissipation model is coupled with Arrhenius-type expressions for devolatilization, char combustion, and CO_x production. The simulation employs the $k - \varepsilon$ turbulence model, the eddy dissipation model for the gas mixture phase, and the discrete transfer model for particle radiation. The char remaining after devolatilization is considered to be pure carbon, and its reaction is governed by external diffusion of oxygen to the particle surface. The simulation outcome showed a fair prediction of the flame progress, as the flame base was found to be close to the fuel feed source. Flow recirculation was shown to be very lean, resulting in no coal and/or ash particulates near the furnace walls, hence keeping the by-product gas streams flow smoothly toward the furnace exit. The simulation results have also proved that complete combustion in the combustion zone was obtained for the prescribed coal/air ratio. Hence, less NO_x gasses are emitted. Arsenic oxide, the product of oxidization of the evolved arsenic, is observed to concentrate at high temperature spots, while the trioxide arsenic spreads out through the combustion zones. The conclusion obtained from the simulation could be used as a benchmark for comparison with available experimental data for agreement.

Keywords Coal · combustion · Trace element · Arsenic · CFD · DTF

1 Introduction

The combustion of fossil fuels has been and still is the backbone of the power, manufacturing and transportation industries. However, the evolution of undesired inorganic elements during the burning process continues to be a challenge, as such elements cause severe problems in the heat exchanging systems [1]. Trace elements, on the other hand, have a significant influence on the environment, as they could either be a component of fly ash or of pure elements in the bottom ash, thus expanding their risk on human health.

Due to scarce research on the hazardous effects of trace elements retrieved from coal, considerable attention has thus been oriented toward the behavior of certain trace elements during the course of combustion. Among the many elements associated with coal particles, trace metals such as mercury, arsenic, sodium, calcium, and selenium are of particular concern [2].

In pulverized coal, the host of trace elements (the mineral grains) can either be included in coal particles or excluded from coal particles [3]. Some alkali and alkaline elements may also exist in their organically associated form in the coal particles [4].

Throughout the process of combustion, many of these elements vaporize and may react with other elements at varying degrees. The reaction depends upon the characteristics of the element, the thermofluid conditions of combustion, and the existence of other constituents in the gas stream.

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Arsenic (As) is chosen in the current investigation as a sample of a trace element released during coal combustion. Arsenic emission to the atmosphere ranges between 0.1 and 80 mg per kg of coal [5]. The majority ends up in fly ash, causing severe environmental hazard, thereby affecting the community at large.

Arsenic fate does not only depend on the temperature and thermal condition of the combustion process, but it also depends on the condensation–vaporization processes, the presence of calcium, gaseous sulfur, and chlorine compounds in flue gases. Temperature is the main factor that governs the phase of arsenic in the fly ash. Once the temperature passes 600 °C, a substantial amount of Arsenic converts into a gaseous phase [6]. The elemental arsenic (As) and its oxide forms (e.g., As_2O_3) are considered the most probable arsenic species in the flue gas during the process of coal combustion. However, (As) is much more volatile as As_2O_3 than its elemental form (As); hence, researchers have concluded that As could only be present in the flue gas in the form of an oxide [7].

Seams et al. [8] have experimentally proved that the arsenic evolved from coal combustion, could follow, in order, the following processes: oxidization (in gas phase), condensation during cooling, and then be concentrated in the sub-micrometer ash size fractions or may remain in the vapor phase. The existence of calcium and other alkaline elements accelerate such reaction to form a refinery substance, resulting in a dramatic increment of the thermal resistance of the boiler [9].

Formulating a model of the evolution, reaction, and transformation of the trace elements, as well as their impact on the combustion performance, is still a challenge for researchers [7]. As such, further studies are currently demanded to describe the physical and chemical behavior of trace elements during the course of combustion.

Modeling the vaporization of arsenic during the combustion of individual coal and pyrite particles was mathematically developed [10]. Since arsenic exists in pyrites, as included in coal particles and/or excluded from coal particles, both types of associations were separately formulated [11]. The model predicts the amount of elements vaporized as a function of coal particle size, mineral grain size, mineral grain association, flame temperature, and residence time.

The current study attempts to incorporate the arsenic vaporization models into CFD code to understand the morphology and behavior of included and excluded arsenic during coal combustion. A 3D Drop Tube Furnace, built in the Department of Chemical Engineering at the University of North Dakota, is numerically simulated to illustrate the shape of the flame temperature and predict the by-product concentration, including the arsenic fate during combustion.

2 Modeling Overview

Combustion of pulverized coal laden with pyrite in a Drop Tub Furnace (DTF) is coded using FLUENT, a commercial CFD software that utilizes a pressure-based segregated finite volume method solver. Fluent software contains physical models for a wide range of applications and possesses capabilities needed to model flow including: turbulence, heat transfer, reacting flows, chemical mixing, combustion, and multiphase flows. It provides physical models on unstructured meshes, manifesting the virtues of easier problem setup and greater accuracy using solution-adaptation of the mesh.

The CFD software has capability to link the eddy dissipation concept with the finite rate chemistry model when calculating the near flame zone field and the far-field regions of the flame. The main target is to minimize the interaction of the solids inside the combustion chamber with the chamber walls by optimizing the flow pattern and trajectories of the particles. Such optimization requires uniformity of temperature inside the combustion zone, uniformity of mixing patterns of the combusted solids, uniformity of trajectory patterns of the injected particles inside the combustor, and sufficient residence time for the particles to release moisture and to devolatilize.

The model, which is based on a finite volume discretization technique, solves the mass, momentum and energy equations using the Semi-Implicit Method for the Pressure-Linked Equations (SIMPLE) algorithm [12]. Initially proposed by Patankar and Spalding [12], the algorithm employs boundary conditions to compute the gradients of velocities and pressure by solving the discretized continuity and Navier–Stoke equations. An approximation of the velocity field is obtained by solving the momentum equation. The pressure gradient term is calculated using the pressure distribution from the previous iteration or an initial guess. The pressure equation is formulated and solved in order to obtain the new pressure distribution. Velocities are corrected and new sets of conservative fluxes are calculated.

The furnace model is based on the numerical solution of the three-dimensional differential equations for conservation of mass, momentum and energy. The simulation uses comprehensive representation of the features of gas phase fluid dynamics, combustion of coal particles, evolution of trace elements, heat transfer including radiation and chemical reactions to investigate the velocity profile, flame temperature, and product emissions at varying locations in the combustion zone.

Coal combustion is calculated by combining a particle transport model of the coal particles with an eddy dissipation model for the combustion of volatiles gases in the gas phase. Coal particles are treated as non-interacting spheres with the full coupling of mass, momentum and energy with the gaseous phase.

The combustion sub-models treat particle devolatilization, char oxidation, and additional gas phase reactions. The model uses the Lagrangian approach for tracking the particulate phase, the Eulerian approach for formulating the gas phase time-averaged conservation equations, the $k-\varepsilon$ turbulence model, the eddy dissipation model for the gas mixture phase, particle to turbulence interaction, particle dispersion by turbulence and the discrete transfer model for particle radiation. Coal devolatilization is described by a first-order 'single-reaction' model [13], while the char remaining after devolatilization is considered to be pure carbon and its reaction is governed by external diffusion of oxygen to the particle surface [11].

The particle tracking includes laws for inert heating, drying (wet combustion model), devolatilization, and char burnout. Chemical reactions were modeled using the Eddy-Break-Up model. The reaction process was based on a two-step mechanism with carbon monoxide as an intermediate combustion species.

2.1 Devolatilization

The devolatilization model is based on the single-reaction mechanism using generic Arrhenius reactions. The coal particle has the initial mass C_0 of raw coal, and the mass of residual char becomes purely carbon after the devolatilization process. The rate constant k_v of the single reaction determines the rate of conversion of raw coal:

$$\frac{\mathrm{d}C_0}{\mathrm{d}t} = -k_v C_0 \tag{1}$$

The rate of volatiles production is given by:

$$\frac{\mathrm{d}V}{\mathrm{d}t} = -Yk_v C_0 \tag{2}$$

where V is the mass of the volatile, and Y is the volatile's yield. The rate constant k_v is expressed in Arrhenius form as:

$$k_v = A_v \exp\left(-\frac{E_v}{RT_\mathrm{p}}\right) \tag{3}$$

where T_p is the temperature of coal particle (assumed uniform), A_v and E_v are constants, evaluated experimentally for the particular coal. The standard experimental Arrhenius constants of devolatilization reaction may give good estimates for most problems and are used in current simulation [14].

3 Furnace Layout

Figure 1 depicts the compact arrangement of the DTF. The burning zone dimensions are: 6″ ID (15.25 cm), 8″ OD (20.3 cm), and 52″ long (132 cm) for the combustion zone and about 188″ length (477.5 cm) of the post-combustion zone. The primary air–coal mixture enters the furnace via 1/2″ (1.25 cm) water cooled pipe, while the secondary air is fed perpendicular 2″ (5.1 cm) from the top. The preheated secondary air undergoes a 3″ (7.6 cm) thick flow straightener to absorb vortices before mixing with the coal/primary air. However, the mixing of the coal/primary air with the secondary air occurs at ∼7″ (17.8 cm) down the top of the furnace. The temperature of the primary coal/air mixture entering the furnace is 60 C while the secondary air enters at a temperature of 450 C.

3.1 Coal Properties

The model requires direct data input obtained from the laboratory ultimate/proximate coal analysis to compute the properties and reactions. Table 1 illustrates the ultimate/proximate analysis of coal used in the simulation.

The higher heating value (HHV) of the coal utilized in the analysis is 21.31 MJ/kg. The average values for coal particle density and specific heat capacity, namely 1560 kg/m^3 and 1450 J/kg K, were used in this simulation. The particles' diameters are between 5 and 250 μm, randomly distributed and equal in distribution by number for total particles.

3.2 Boundary Conditions

The coal mass flow rate, the primary and secondary air velocities, and the temperatures inlet to the furnace are presented in Table 2. The wall temperature of the combustion zones has been treated as thermally equilibrium with external enclosed air since the wall is surrounded by air with heating elements that work to keep the wall temperature within 1400 C. Radiation interaction between particles and the furnace environment was also enabled in all simulation's runs using the Discrete Ordinates Radiation model.

Particles are set up with nine different particle sizes. Diameters between 5 and 250 μm corresponding to CCSEM analysis have been used. The size distribution has been applied based on the typical coal size distribution analysis for PC burners. Constant temperature boundary conditions were considered for combustion zones walls, whereas adiabatic insulated conditions were used for the rest of the walls.

4 Geometry and Meshing Scheme

In order to accurately simulate the combustion process, the computational grid must be capable of handling the complex flow features, particularly near the flame zone. Since the number of cells and their art-distribution play a great role in numerical calculations and time consumption, a grid

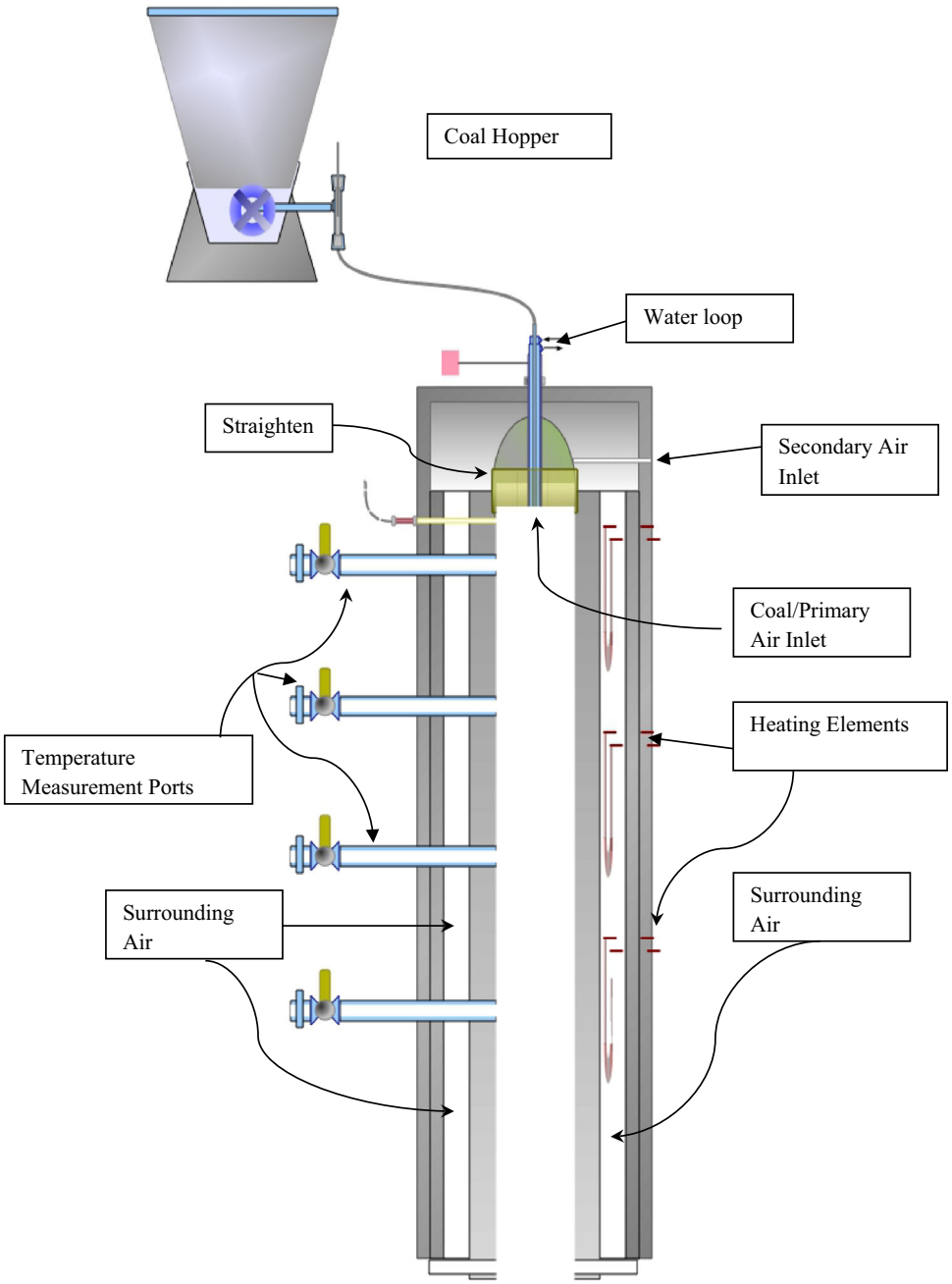


Fig. 1 Layout of the Drop Tube Furnace built in Chemical Engineering Department at University of North Dakota (UND)

Table 1 Proximate/ultimate analysis of coal

Proximate analysis (%)				Ultimate analysis (%)					HHV (MJ/kg)
Ash fixed	Carbon	Moisture fraction	Volatiles	C	H	O	N	S	
7.06	40.79	22.5	29.57	54.49	6.26	30.82	0.82	0.49	21.31

independence testing study at different grid resolutions was conducted. If the grid is too coarse in some areas, the product of the elemental chemical reactions, the temperature contours, and the coal/air thermal interaction will not be resolved properly. Hence, the flow features can potentially be lost or misplaced in the flow. The parameters considered for the grid independence study were the flow characteristics inside and around the flame.

Table 2 Inlet boundary conditions for the furnace

Inlet type	Velocity (m/s)	Coal mass flow rate (kg/s)	Temperature (K)
Primary air	1.7	0.0014	333
Secondary air	1.7	0	723

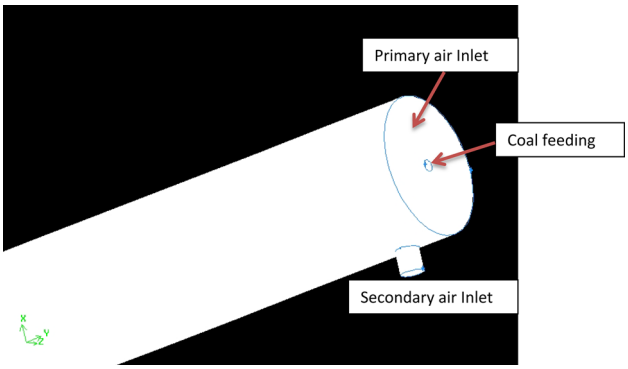


Fig. 2 3-D CFD geometry of DTF

Figure 2 shows the outline of the combustion zones, as well as the inlet ports for the computed cases. Meshes with approximately 42,780 tetrahedral cells (Fig. 3) have been found to give good grid resolution for the modeled cases. The computational domain is filled with an unstructured tetrahedral and hexahedral mesh. A refined mesh was employed around the inlet ports to allow the cell size to grow from the areas with the high strain rates to the rest of the domain. With this approach, it was possible to resolve the recirculation pattern of the air flow and the possible recirculation patterns of the solid particles. The mesh was further refined during the calculations in the areas with higher rates of devolatilization of solids.

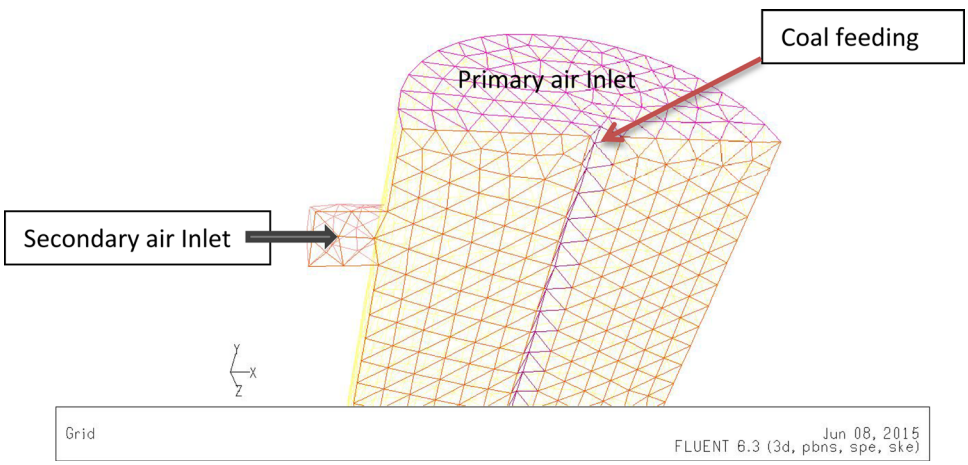
The comparison of the flow structure begins to show the disparity between the different computational grids. Cell size affects the flame shape and temperature magnitude for a given upstream condition because large cells cannot fully demonstrate the flow features in their correct locations. Hence, sufficient cell resolution should be maintained in the flame region where the inception of combustion occurs.

Three different sizes of cells at the flame zone are selected for the investigation of the grid resolution. Namely, cells with an average of 10 mm (adopted size), 20 mm (coarse grids), and 1 mm (fine grids) element sizes were used. Figure 4 illustrates the average predicted static temperature along the axis of symmetry passed through the core of the flame for the three cases. The wiggly and unsystematic trend of the temperature for the coarse case implies that grid resolution has to be performed. The comparison result of the coarse pattern with the recommended grid size (i.e. 10 mm) concludes that the coarse cell pattern has a substantial difference in predicting the flame shape. However, such discrepancy nearly vanishes when the grid size is shrunk to 1 mm. Hence grids with 10 mm average size at the flame were adopted, while larger sizes of cells were chosen at the rest of the domain for obtaining better resolution and grid optimization per minimum CFD running time.

5 Results and Discussion

Fluent uses a coupled solver, in which all the hydrodynamic equations for velocity components (u, v, w), pressure (p), energy equation for temperature (T), turbulent equations for ε and k, and equations of species are solved as a single system. The set of governing differential equations together with the boundary conditions are solved numerically using an iterative procedure on each of the control volume to obtain a value for each parameter of interest.

Fig. 3 Axisymmetric meshing scheme of DTF



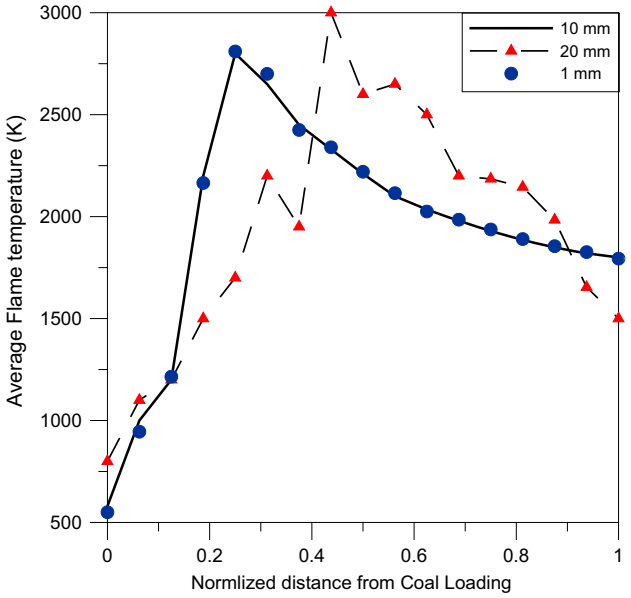


Fig. 4 Average flame static temperature at different mesh sizes

5.1 Particle Trajectory, Flame, and Devolatilization

The trace of particle path during the process of combustion is very important in the design of DTF, as the particle trajectories exhibit less interaction between the solids with the combustion chamber walls. Otherwise, the particles would deposit on the furnace walls.

The CFD design approach illustrated in Fig. 5a–c is shows the trajectories of 5, 50 and 75 μm PC particles colored by particle mass. It is observed that coal is denser in the core of the furnace and burns systematically while falling down along the furnace. No significant amount of coal has been predicted near the wall region, meaning that the trajectories of the pulverized coal particles do not interact with the walls. Hence, a high devolatilization is anticipated inside the combustion chamber. Such behavior guarantees uniformity in the flow pattern of the particles, making the system approaches the optimum design criteria.

5.2 Flame Shape

Stability of the flame during coal combustion is of primary interest to examine during the design process of DTF, as it impacts the performance of the burner (including NO_x production), the flame shape and length, and the char burnout.

As illustrated in Fig. 6, no strong recirculation pattern is found in the combustion zone. This could be considered as an advantage because the recirculation could maintain the coal particles and fly ash particulates in the combustion zone, or bring them back to the coal/air inlets. The mass fraction of coal, presented in Fig. 7a, b, gives an impression that the degree of devolatilization is reasonable since the pattern is uniform.

5.3 Temperature, Velocity and Species Contours

Regions of very high temperatures near the core of the furnace confirm that no coal has been shuttered toward the wall. The flame's mean temperature predicted by simulation is found to be ∼2800 K, as demonstrated in Fig. 8. The flame length is shown to be approximately 0.75–1 m long. The phenomena of flame liftoff may also be seen from the 3D contours of temperature in Fig. 9. The flame root position is an important parameter in the design of low NO_x combustors. When the flame root is closer to the burner it could be a sign for less NO_x production compared to the flame root away from the burner. This may attribute to the fact that when the flame is detached from the burner then the concentration of oxygen near the flame-envelope increases the temperature locally-thereby increasing thermal NO_x.

The velocity vector and contour plotted in Figs. 6 and 9, respectively, demonstrate clearly the smoothness of the flow, which prevents the plumes from pushing toward the wall. The flow vectors show that a uniform plume penetrates throughout the system despite the vortex on the top of the furnace. There is a miniscule shifting of the flow stream to the left at the mixing spot near the coal/primary air inlet as a result of the secondary air vortex. Such minor shifting causes no alteration to the flame stability. Nevertheless, the experiment apparatus has a straightener just below the secondary air inlet chamber to keep the stream at the mixing position uniform and stable.

As a general rule, a certain fraction of O_2 should be the benchmark of the combustion nature. Contours of O_2 mass fraction, shown in Fig. 10, portrays that a considerable amount of oxygen remains at the outlet of the combustion zone, thereby confirming that excess air has been fed to the system. The zero oxygen concentration in and around the flame indicates that complete combustion has occurred at such region.

As a result of the volatile released during the devolatilization process, CO peak emerges near the flame core. Figure 11 demonstrates the contours of mass fraction of CO inside the combustion zone. It is clearly shown that no CO is predicted at the furnace exit.

The presence of CO_2 in the flue gases at furnace outlet also gives an impression that complete combustion has occurred, as seen in Fig. 12. The high concentration of carbon dioxide at furnace outlet proves that almost all the carbon was converted to CO_2, despite the existence of carbon monoxide as an intermediate combustion species.

A similar conclusion is obtained in regards to the hydrogen/water vapor converting process, from the mass fraction contours of the H_2O vapor, as illustrated in Fig. 13.

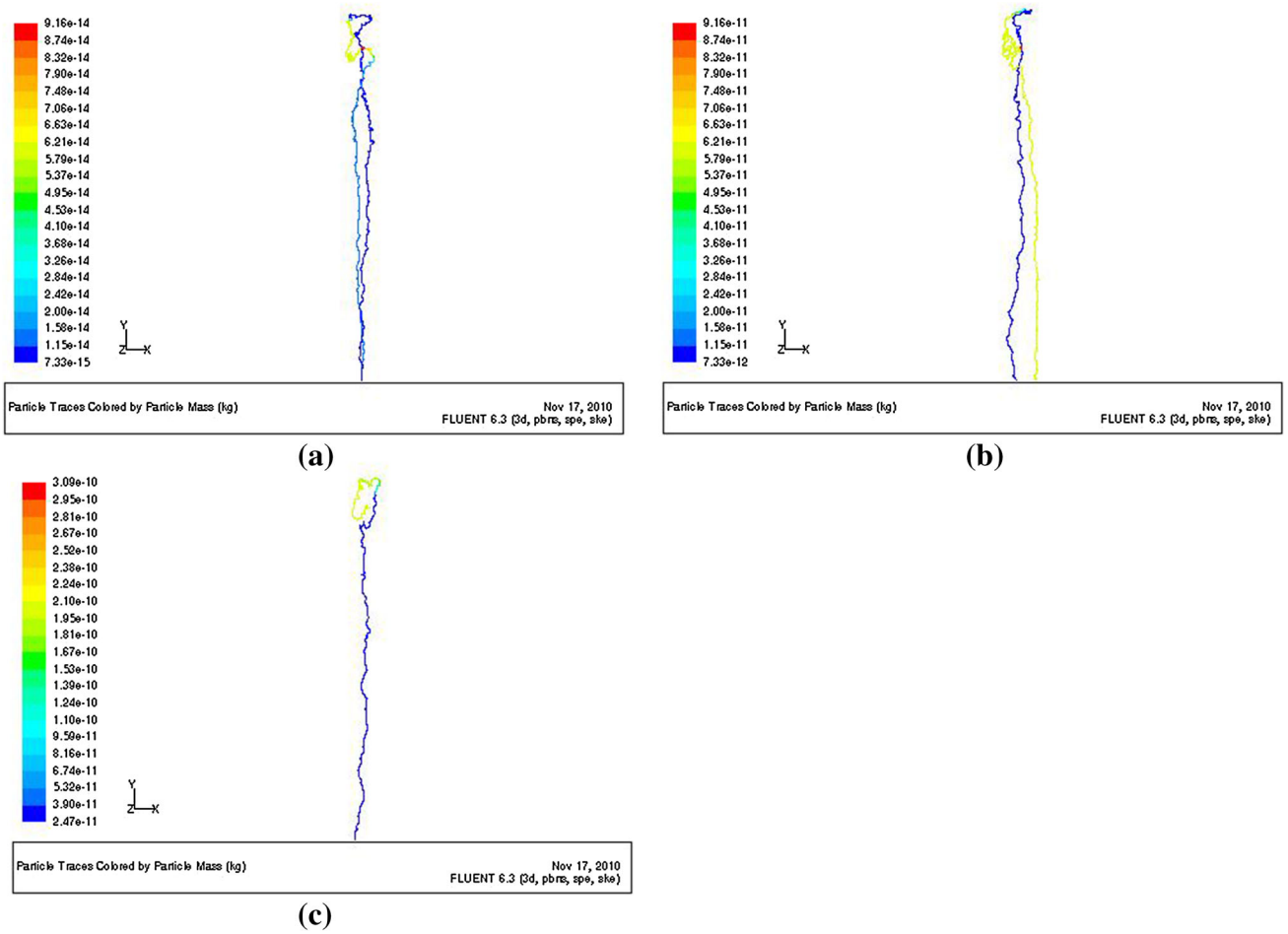


Fig. 5 Particle trajectory colored by particle mass for three different sizes

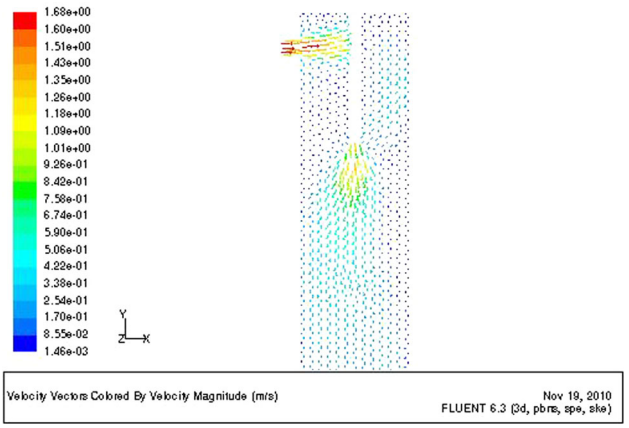


Fig. 6 Flow stream pattern near inlets

6 Trace Element Evolution

It is likely that one of the most important factors that influence the behavior of arsenic during combustion is the form of occurrence of arsenic in the coal. The conversion of arsenic forms in coal to arsenic forms in ash as a result of combustion depends upon many factors. The release of arsenic vapor, as a result of devolatilization, and the presence of Oxygen, accelerate the oxidization process and convert the released arsenic to vaporous arsenic oxides. In turn, arsenic oxides will react with alkaline-earth minerals to from stable compounds, such as calcium orthoarsenate. The latter condensates and acts as a refractory substance that sticks on the heat transfer surfaces, causing reduction in the heat transfer efficiency.

Since the objective of the study is to show how the trace element behaves during combustion, we incorporated the mathematical model of the arsenic evolution into the CFD simulation. The assumption we made is that arsenic reacts chemically with oxygen to form an arsenic trioxide base on the following expression:

$$\mathrm{As} + \frac{3}{4}\mathrm{O_2} \rightarrow \frac{1}{2}\mathrm{As_2O_3} \tag{4}$$

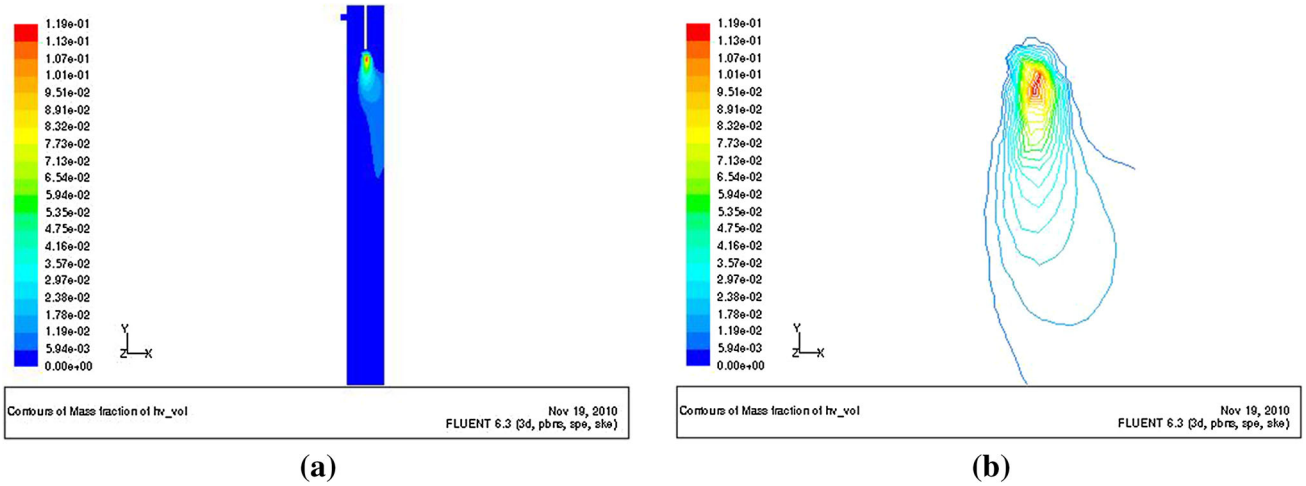


Fig. 7 Mass fraction profile of volatile matter. **a** Combustion zones. **b** Enlargement contours

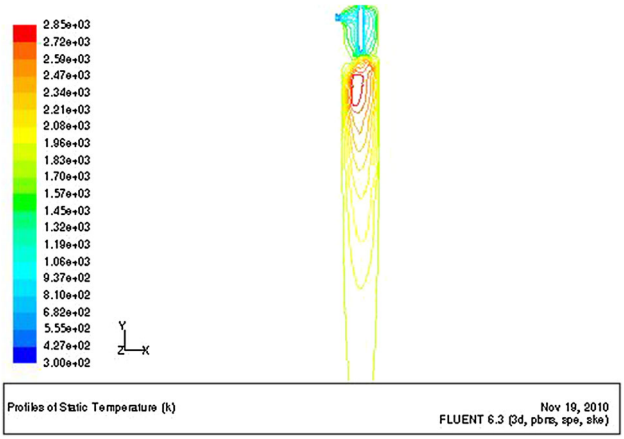


Fig. 8 Flame shape at the center axis of the furnace

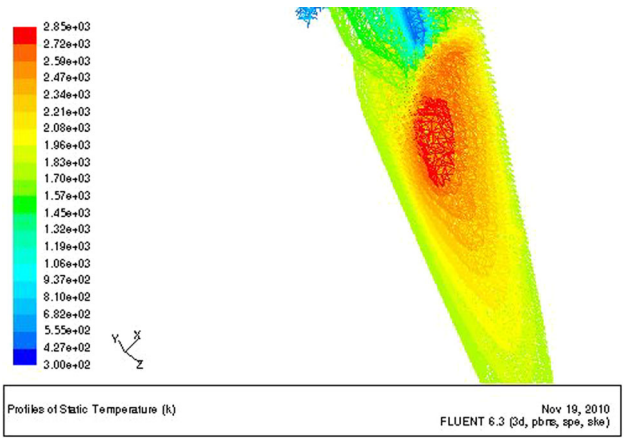


Fig. 9 3-D temperature contours near the flame

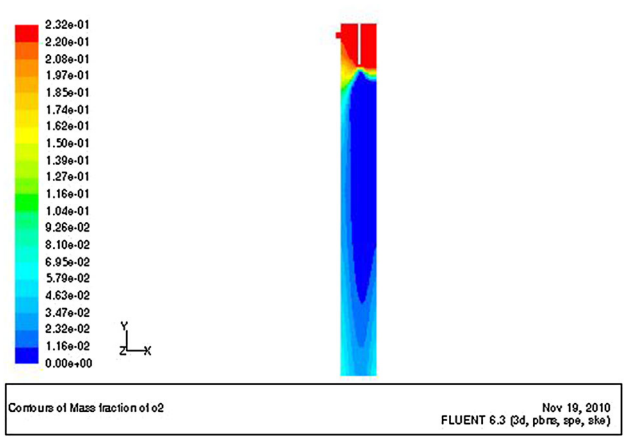


Fig. 10 Concentration profile of oxygen in the combustion zone

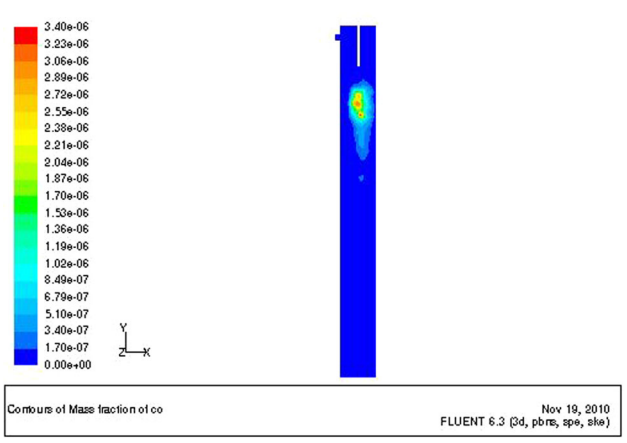


Fig. 11 Mass fraction of CO

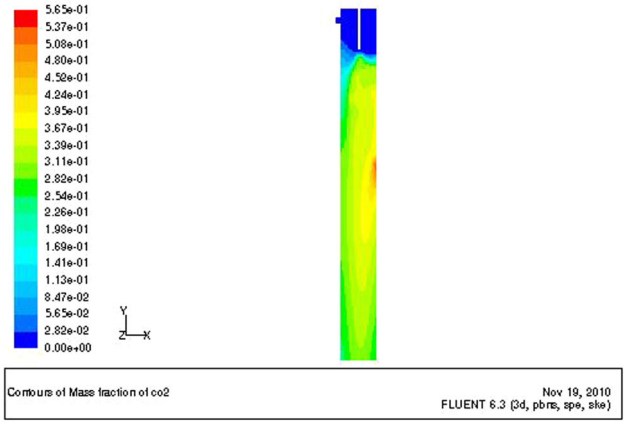


Fig. 12 Mass fraction of CO_2

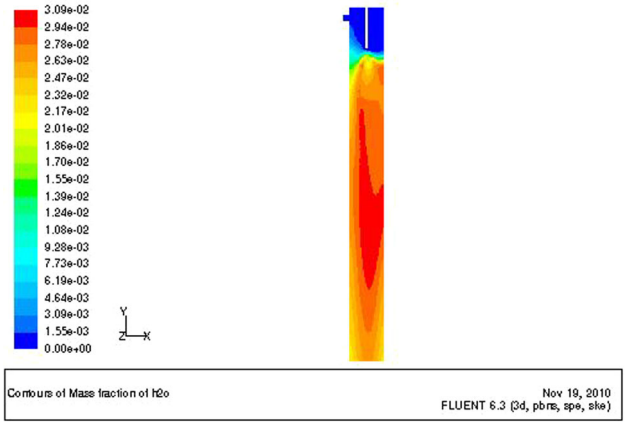


Fig. 13 Mass fraction of H_2O

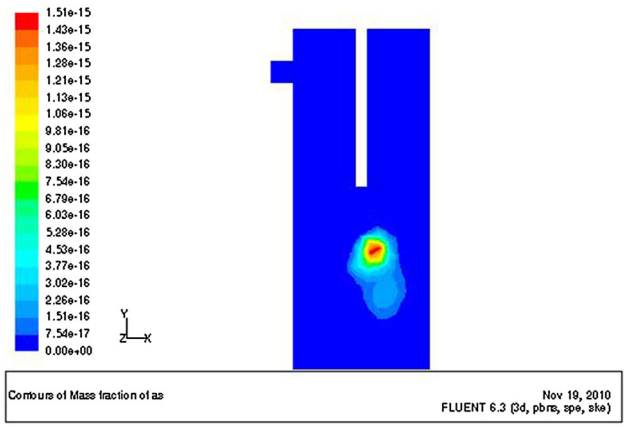


Fig. 14 Mass fraction of As

Contours of arsenic shown in Fig. 14 give an insight of the spot of arsenic vapor released, which is mostly done in the high temperature zone. In turn, trioxide arsenic spreads out through the combustion zones (as seen in Fig. 15), leading to greater chances of forming refractory substances in substantial amount throughout the furnace. Such substances would significantly drop the boiler efficiency, as well as increase the hazard to the environment and community, when they are carried by the fly ash.

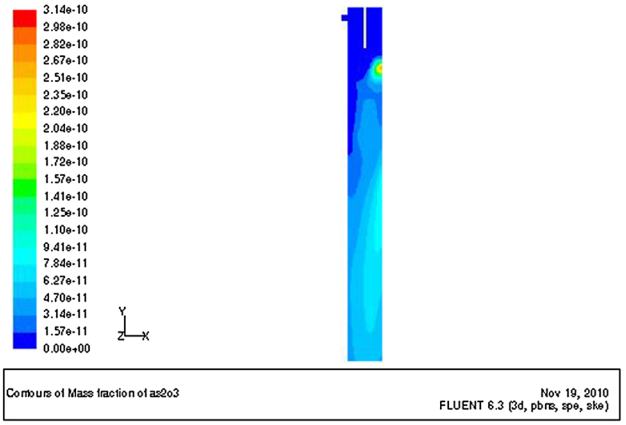


Fig. 15 Mass fraction of As_2O_3

7 Conclusion

The evolution of arsenic (As) released during coal combustion is modeled using the criteria of three consecutive transportation processes. Formulated expression of arsenic evolution is incorporated into CFD simulation code in order to predict the behavior of the trace element that releases during coal combustion. In-situ Drop Tube Furnace was numerically simulated and real experiment data was used to describe the transformation processes that occur during the course of combustion. Profiles of temperature, velocity, by-product gases, and trace element vaporization and reaction have been presented.

The simulation predicts good design in term of flame progress, as the flame base is shown to be close to the fuel source. Flow recirculation is very lean, resulting in no coal and/or ash particulates near the furnace walls. The results of the simulation also show that complete combustion occurs in the combustion zone and less NO_x gasses are emitted.

Arsenic Oxide, which is the product of the oxidization of evolved arsenic, is observed to evolve at higher temperature spots, while trioxide arsenic is spread out through the combustion zones. The outcome obtained from the current simulation could be a useful resource for comparison with experimental data seeking verification and agreement.

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