

NUMERICAL TRACKING OF SORBENT PARTICLES AND DISTRIBUTION DURING GAS DESULFURIZATION IN PULVERIZED COAL-FIRED FURNACE

by

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Original scientific paper

<https://doi.org/10.2298/TSCI160212196T>

Furnace sorbent injection for sulfur removal from flue gas presents a challenge, as the proper process optimization is of crucial importance in order to obtain both high sulfur removal rates and good sorbent utilization. In the simulations a two-phase gas-particle flow is considered. Pulverized coal and calcium-based sorbent particles motion is simulated inside of the boiler furnace. It is important to determine trajectories of particles in the furnace, in order to monitor the particles heat and concentration history. A two-way coupling of the phases is considered – influence of the gas phase on the particles, and vice versa. Particle-to-particle collisions are neglected. Mutual influence of gas and dispersed phase is modeled by corresponding terms in the transport equations for gas phase and the equations describing the particles turbulent dispersion. Gas phase is modeled in Eulerian field, while the particles are tracked in Lagrangian field. Turbulence is modeled by the standard k - ϵ model, with additional terms for turbulence modulation. Distribution, dispersion and residence time of sorbent particles in the furnace have a considerable influence on the desulfurization process. It was shown that, by proper organization of process, significant improvement considering emission reduction can be achieved.

Key words: *model, two-phase reactive flow, turbulence, sorbent particles, furnace*

Introduction

Coal combustion in power plants on global scales is one of major contributors to pollution of air, and consequently the reason for appearance of photochemical smog and acid rains [1]. As the use of fossil fuels increases, and fuels of lower quality are combusted, an effort must be made to treat combustion products in order to remove or reduce the amount of pollutant gasses released into the atmosphere. This problem is globally recognized, and multiple legal regulations are passed emphasizing the importance of gas treatment in new power plant boilers and also setting the emission limits within a demand for retrofit of old power plants in order to extend the boiler live expectancy.

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In this paper a numerical simulation is used to examine possible sulfur oxides removal from flue gas exiting furnace of Kostolac B power plant boiler. More detailed description of furnace can be found in [2]. The furnace is part of a 350 MW_e pulverized lignite-fired boiler with eight tangentially arranged burners divided in four tiers. Air and coal mixture and secondary air are injected at each burner tier. Calcium based sorbent – limestone, is injected in furnace through dedicated openings above the burner tiers, or premixed with air and coal mixture. Calcium is injected in the form of CaCO₃, and, when in furnace, at high temperatures it is converted in CaO, which reacts with sulfur oxides and reduces their amount in furnace.

Furnace sorbent injection [3] might not be effective as flue gas scrubbing [4, 5], but it is still an attractive solution due to considerably lower costs, and when combined with wet scrubbing, dry sorbent injection is becoming especially important in low water technologies [6], where the water resource is limited, and should be preserved as much as possible. Injecting particles in furnace leads to development of their internal surface, and its sintering. Particles exposed to low temperatures tend to have undeveloped reactive surface, and thus they do not react well with SO₂ during the time they remain inside of the boiler furnace. On the other hand, particles exposed to excessively high temperatures rapidly develop surface, but at same time suffer great surface loss due to sintering [7, 8].

Efficiency of this desulphurization method is highly dependent on mixing between combustion products and sorbent in the furnace regions with the so called temperature window of about 950-1250°C. However, to determine proper sorbent injection configuration, knowledge of optimal temperature window for sorbent injection is not sufficient by itself. An insight in boiler flame temperatures, particle thermal history and dispersion are important when determining proper injection configuration. Multiple models of particle sulfation with accompanying calcination and sintering models are available in literature for single sorbent particle only [7-13]. Few models with complete furnace modeling by commercially available software can be found.

Into previously developed in-house CFD code [2, 14], particle calcination, sintering and sulfation models are incorporated together with sorbent particle tracking [15], making complex tool for analysis of processes that occur in furnace during combustion of Serbian lignite combined with injection of pulverized sorbent.

Mathematical model

Comprehensive 3-D differential mathematical model is used for numerical prediction of processes in two-phase turbulent reactive flow. Large-scale coal-fired boiler furnace is simulated under stationary conditions [2].

Euler-Lagrangian approach is employed for treatment of two-phase reactive gas-particle flow [14]. Time-averaged Eulerian partial differential equations for mass, momentum, energy, gas mixture component concentrations, turbulence kinetic energy and dissipation are used to describe gaseous phase, given for general variable Φ in index notation (1):

$$\frac{\partial}{\partial x_j} (\rho U_j \Phi) = \frac{\partial}{\partial x_j} \left(\Gamma_\Phi \frac{\partial \Phi}{\partial x_j} \right) + S_\Phi + S_\Phi^p \quad (1)$$

Closure of the gas phase momentum conservation equations is done by k - ϵ turbulence model. Radiative heat exchange in furnace is simulated by six flux model.

Interaction between fluid and particles is modeled using Particle Source In Cell (PSI Cell) method. Fluid motion influences the motion of immersed particles. This influence de-

pends on the drag coefficient which depends on particle size. Particle influence fluid flow via additional source terms in equations that describe fluid flow. The two-way coupling is used to describe particle interaction with fluid and vice versa, neglecting particle collisions with other particles. Particle number density has such value to allow for two-way coupling to be used in simulations.

Dispersed phase was described using Basset equation in Lagrangian field [14], with less important effects neglected (2):

$$m_p \frac{d\vec{u}_p}{d\tau} = \frac{1}{2} C_D \rho A_p |\vec{u} - \vec{u}_p| (\vec{u} - \vec{u}_p) + \frac{d_p^3 \pi}{6} (\rho_p - \rho) \vec{g} \quad (2)$$

The problem with modeling influence of turbulent fluctuations components in flow on particle velocity can be overcome by splitting velocity in convective \vec{U}_{pc} and diffusion \vec{U}_{pd} part. Convective velocity (\vec{U}_{pc}) can be obtained by solving particle motion equation (3), and this equation is solved when determining particle convective velocity:

$$m_p \frac{d\vec{U}_{pc}}{d\tau} = \frac{1}{2} C_D \rho A_p |\vec{U} - \vec{U}_{pc}| (\vec{U} - \vec{U}_{pc}) + \frac{d_p^3 \pi}{6} (\rho_p - \rho) \vec{g} \quad (3)$$

The drag coefficient C_D for particle Reynolds numbers having value equal to or less than 1000 is calculated using the equation $C_D = (24/Re_p)(1 + 0.15 Re_p^{0.67})$ [16], and for values above that C_D has fixed value $C_D = 0.44$. Particle Reynolds number can be determined by using $Re_p = d_p |\vec{u} - \vec{u}_p| / \nu$. The particle inertia effect may be estimated in every individual case, influencing the difference between the particle response time and the characteristic turbulence time scale.

Diffusion velocity (\vec{U}_{pd}) is obtained by using the concentration of particles (particle number density) N_p in given control volume: $\vec{U}_{pd} = -(1/N_p) \Gamma_p \nabla N_p$, where $\Gamma_p = v_p^t / \sigma_p$ represent the coefficient of turbulent diffusion of particles. Equation used to solve diffusion velocity is empirical. Total particle velocity is obtained by adding convective and diffusion velocity, and can be noted as eq. (4) [14]:

$$\vec{U}_p = \vec{U}_{pc} + \vec{U}_{pd} \quad (4)$$

Modeled equations for turbulent kinetic energy (5) and its dissipation (6) with terms for turbulence modulation are used in following form:

$$\frac{\partial}{\partial x_j} (\rho U_j k) = \frac{\partial}{\partial x_j} \left[\left(\mu + \frac{\mu_t}{\sigma_k} \right) \frac{\partial k}{\partial x_j} \right] + \mu_t \left(\frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i} \right) \frac{\partial U_i}{\partial x_j} - \rho \varepsilon + 2k S_p^m + S_p^k \quad (5)$$

$$\frac{\partial}{\partial x_j} (\rho U_j \varepsilon) = \frac{\partial}{\partial x_j} \left[\left(\mu + \frac{\mu_t}{\sigma_\varepsilon} \right) \frac{\partial \varepsilon}{\partial x_j} \right] + C_{\varepsilon 1} \frac{\mu_t}{k} \varepsilon \left(\frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i} \right) \frac{\partial U_i}{\partial x_j} - C_{\varepsilon 2} \rho \frac{\varepsilon^2}{k} + 2\varepsilon S_p^m + S_p^\varepsilon \quad (6)$$

In these equations source of turbulent kinetic energy due to fluctuations of drag force (relative motion), with an assumption that turbulence is isotropic is modeled according to eq. (7).

The concentration of particles N_p can be obtained from the transport equation in the form of eq. (1) [14, 17].

Source in turbulent kinetic energy dissipation and term that describe influence of change in particle mass are modeled and included as described in eqs. (8) and (9) [14]:

$$S_p^k = \sum_i \sum_j 2\pi\mu d_{p,ij} k \left[\tau_i^{0,5} - (\tau_i + \tau_{p,ij})^{0,5} / (\tau_i + \tau_{p,ij})^{0,5} \right] N_p \Delta\tau \quad (7)$$

$$S_p^\varepsilon = 2\nu \frac{\partial u'_i}{\partial x_l} \frac{\partial s'^{ij}_p}{\partial x_l} = C_{\varepsilon 3} \frac{\varepsilon}{k} S_p^k \quad (8)$$

$$S_p^m = \Delta\dot{m}_p = \frac{1}{V} \frac{\pi}{6} \rho_p \sum_i \sum_j \left[(d_{p,ij}^3)_{in} - (d_{p,ij}^3)_{out} \right] \dot{N}_{ij} \quad (9)$$

Variable \dot{N}_{ij} represents the flow of number of particles. Relation between flow of number of particles and concentration of particles in the considered control volume can be given as $N_p = \sum_i \sum_j \dot{N}_{ij} / A_{inl} U_{p,ij}$.

Turbulence modulation effects were previously investigated in [18] under coal particle combustion conditions, and it was noticed that, as expected, relatively small particles (with respect to ratio of particles diameter and turbulence length scale), had dampening effects on turbulence. The particle interactions with walls are considered as fully elastic collisions.

Sorbent particle reaction rates are calculated using Partially Sintered Spheres Model (PSS Model) presented by Alvfors [8] and Lindner [9]. Model consist of two parts: calcination and sintering reactions, from which we obtain information about particle surface development during its exposure to high temperatures, modeled according to eq. (10), and sulfation reaction, representing the capture of sulfur oxides in particle, in form of CaSO_4 , according to eq. (11). Both equations and their detailed explanation can be found in papers by Alvfors [8], Lindner [9], and Kocaefe [10].

$$S_{\text{CaO}}^n = \sum_{k=1}^n S_{0,\text{CaO}} \left(\{1 - [K_s(T_k)t_n]^{1/\gamma(T_k)}\} [X_c(t_k) - X_c(t_{k-1})] \right) \quad (10)$$

$$-\bar{r} = \frac{-r_A}{k_{cA0}S_0} = \frac{C}{S_0/S_2 + \text{Da}LS_0/l_0S_{\text{avg}}} \quad (11)$$

This reaction rate is closely linked with reactant diffusion through the particle, representing interaction between local chemical kinetics and pore diffusion (12):

$$\frac{1}{z^2} \frac{d}{dz} \left(z^2 \delta \frac{dC}{dz} \right) = -\Phi_{\text{th}}^2 \frac{C}{S_0/S_2 + \text{Da}LS_0/l_0S_{\text{avg}}} \quad (12)$$

During the particle tracking, source terms from particles are obtained. Sulfur dioxide production term is obtained from lignite combustion reactions, while the term for depletion is obtained from sorbent particle sulfation reaction. These terms influence sulfur dioxide concentration field, and the way they are included can be seen in eq. (13):

$$\frac{\partial}{\partial x_j} (\rho U_j X_{\text{SO}_2}) = \frac{\partial}{\partial x_j} \left(\Gamma_{\text{SO}_2} \frac{\partial X_{\text{SO}_2}}{\partial x_j} \right) + S_{\text{p,SO}_2}^m + S_{\text{p,SO}_2}^{\text{CaO}} \quad (13)$$

Model implementation was verified by numerical simulations performed on drop down tube reactors, by simulating two-dimensional circular pipe, and verifying obtained data with available experimental data [19].

Numerical model and simulation set-up

Previously developed numerical model of Kostolac B1 furnace is used in simulations. Numerical mesh consists of 591,500 nodes. Stability and convergence of model were previously tested and it was shown that this grid was sufficient to obtain grid independent numerical results [18], and further grid refinement would have not brought improvement.

Pulverized coal is injected through eight burners with four vertical tiers each, and depending on combustion configuration one or two burners are always out of operation, with six burners maintaining the constant boiler load. Simulated sorbent injection is done by injecting sorbent through dedicated sorbent ports above the burners, or together with coal through selected burner tiers. The amount of injected sorbent is calculated based on the total lignite mass flow, and lignite sulfur content, maintaining constant average (Ca/S) ratio. Sorbent is than distributed among injection ports, and burners, depending on the process configuration.

In all simulations coal and air flows are kept at constant rates, and the sorbent parameters are being varied.

Nominal boiler operation regime set-up was selected as a starting base for simulations of furnace sorbent injection. In this regime six burners are in operation, while the other two are standing by as reserve if needed. The pulverized coal ultimate analysis used for simulations is C = 36.52%, H = 3.45%, S_{sag} = 1.00%, O = 12.50%, N = 1.52%, W = 8.83%, A = 36.18%. Same fuel is used in all simulated test cases, with same type of sorbent, in order to make test cases comparable with each other. The sorbent is considered to be high purity limestone, consisting only of CaCO₃ part. In the test cases sorbent distribution was varied. The pulverized coal is distributed as 15%, 15%, 25.5%, and 44.5% going from the top burner tier to the bottom one. Part of secondary air that is injected through the two bottom – main burner tiers is 70.3%.

Average sorbent particle size used in simulations is 10 μm. Sorbent particles are modeled as monodisperse powder, containing 10 μm particles as representative diameter for all fractions. An assumption that particles rapidly heat up to the gas temperature is used, thus the change in their temperature is nearly instantaneous. This assumption is based on experimental measurements from Fan [13].

Test cases for sorbent injection are conceptualized in such a manner to allow for analysis of direct impact of furnace internal conditions on sorbent behavior. Sorbent is injected above the burner tiers in one case, and through the burners in various configurations in the other ones. In all test cases sorbent to sulfur ratio (Ca/S) is equal to 1.0.

Results and discussion

Results obtained by numerical simulations are given in tab. 1. The table presents data obtained by numerical simulation results for given test cases. In the test case 1 reduction is lowest due to short residence time combined with low gas temperature and the fact that sorbent is not well penetrating to the central zone, where the majority of SO₂ produced by combustion of coal is concentrated.

It can be noticed that by lowering the injection point towards the flame, and improving the sorbent penetration towards the furnace center the efficiency of reduction increases.

However, it must be also noted that, during the injection in the test case 4, which gives good reduction of SO₂, the sorbent particles are exposed to extreme temperatures, which lead towards rapid deactivation of sorbent particles. Even though this lead to high initial reduction, those sorbent particles lost their internal surface and as such, are not suitable for

further SO₂ emission reduction processes, which is expected to continue to a certain level in the boiler convective pass after the furnace itself.

On fig. 1 the trajectories of the sorbent particles in selected test cases are given, colored by particle axial velocity. It can be seen that overall distribution is better when sorbent particles are injected through burner tiers, compared to case when they are injected through dedicated sorbent ports placed above the burner tiers. By examining fig. 2 – temperature fields that correspond to test cases from fig. 1, it can be seen that different injection of sorbent transport air (portion of secondary air carrying the sorbent particles) affects also the combustion process, as well as the flame position within the furnace. In this case, also, high initial

Table 1. The SO₂ emission reduction - numerical simulation results

Test case	Sorbent injection configuration	Furnace exit gas temperature [°C]	SO ₂ content, at furnace exit [mgNm ⁻³]	SO ₂ reduction, at furnace exit [%]
1	Injection above burner tiers	1026	5946	12.8
2	Upper-stage burner (upper section)	1025	5848	14.2
3	Upper-stage burner(lower section)	1039	5232	23.2
4	Bottom main burner	1025	4308	36.8
5	Three upper burner tiers	1031	4962	27.2
6	Through all burner tiers	1022	4143	39.2

*Reduction is calculated compared to the reference test case with same combustion configuration and without sorbent injection (the reference test case: Furnace exit gas temperature = 1029 °C, SO₂ emission = 6816 mgNm⁻³)

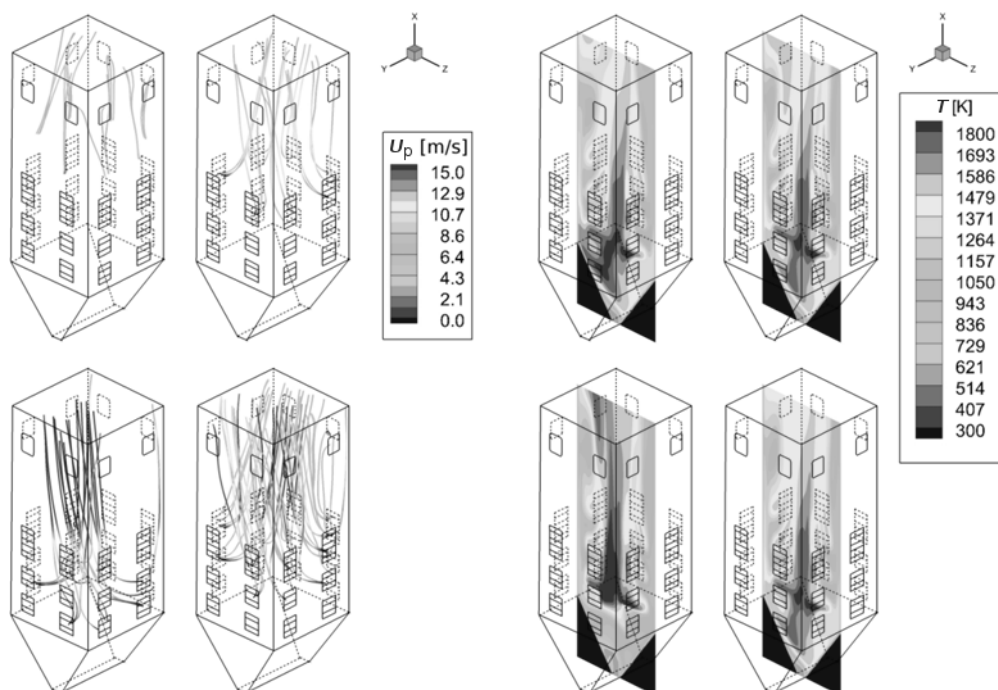


Figure 1. Sorbent particle trajectories for test cases 1, 3, 4, and 5, colored by particle axial velocity
(for color image see journal website)

Figure 2. Temperature field for test cases 1, 3, 4, and 5
(for color image see journal website)

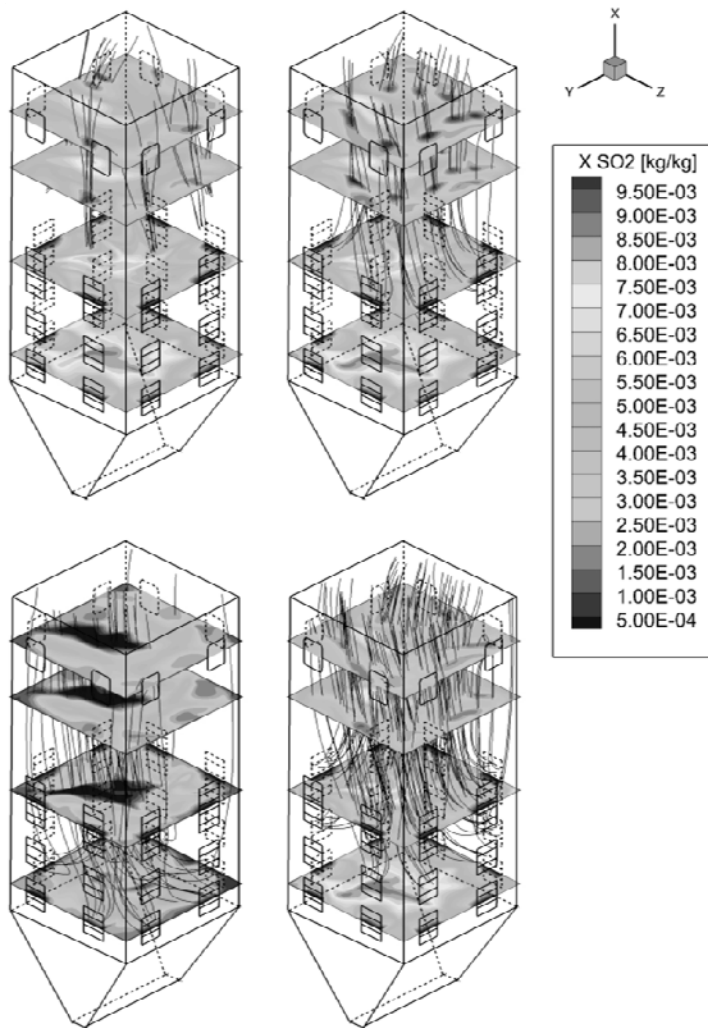


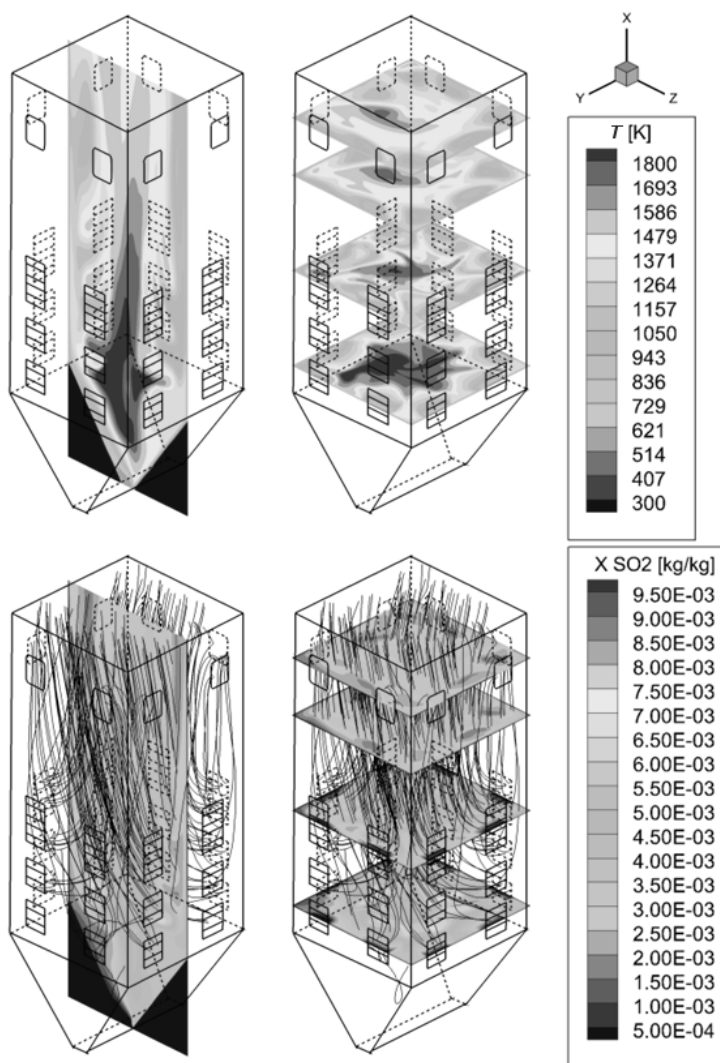
Figure 3.
The SO₂ concentration field
for test cases 1, 3, 4, and 5,
with sorbent trajectories
(for color image see
journal website)

SO₂ capture was noticed, but at a cost of sorbent usability in further processes (rapid loss of sorbent internal surface).

Influence of sorbent particle distribution on SO₂ concentration is given in fig. 3. In flow regions following trajectories of sorbent the reduction of SO₂ is the highest one. In test case 1 reduction occurs only in near wall zones, due to the fact that most of particle trajectories are in these zones, or are being sucked in by recirculation openings. Injection through upper tier burners in test case 3 leads to widening of SO₂ capture zones, as the trajectories cover larger cross section area of the furnace. In both test cases 1 and 3 it can be noticed that high SO₂ concentration zone near the furnace center remains almost intact.

Injection of sorbent through the bottom main burners in test case 4 lead to directing all of the sorbent in central flame region, with most of the sorbent reactions occurring in burner zone of furnace. It can be seen that concentration of SO₂ in flame center was greatly reduced, but SO₂ remains in zones around the flame center.

Figure 4.
Temperature and SO₂
concentration field with
sorbent trajectories, for
test case 6
(for color image see
journal website)



In test case 5 better reduction is obtained compared to previous test cases, except for test case 4. By injecting sorbent through all burner tiers (equally distributed among the burner tiers) – fig. 4, the best reduction was obtained, with only one quarter of sorbent being injected into the furnace fireball. Three quarter of sorbent is not affected by high temperature central flame zone, and its surface remains well developed and suitable for further use.

Conclusions

In this paper it was shown that by proper sorbent distribution over furnace it was possible to achieve high SO₂ reduction with proper sorbent distribution. The initial boiler operation regime influences the way the sorbent particles will travel when they are injected in furnace. All simulations were performed for single (Ca/S) ratio of 1.0, and by using higher (Ca/S) ratios higher initial removal could be achieved.

The influential factors that have impact on SO₂ reduction can be summarized as: (Ca/S) ratio, SO₂ concentration, mixing of SO₂ and sorbent particles, sorbent thermal history and residence time. (Ca/S) ratio influences the SO₂ reduction in such way that higher ratio gives better reduction up to a certain point. But this factor alone does not guarantee that overall reduction will be high, due to the fact that uncontrolled injection in furnace can lead to poor distribution of sorbent, thus leading to low utilization. Local concentration of SO₂ is important factor, and getting the sorbent particles in zones with high SO₂ concentration is one of main goals considering increase in process efficiency. However, particle thermal history and residence times also must be monitored, as the particles in high temperature zones tend to lose available internal surface (due to sintering) if exposed to high temperatures for prolonged periods of time.

With all this in mind, an optimal group of solutions to the problem would be the one in which a good distribution over furnace cross section is achieved, aiming for high SO₂ zones with optimal injection temperatures for particle internal surface development, and long particle residence times.

Numerical CFD code presents valuable tool for analysis of complex processes that occur during combustion and SO₂ capture processes in furnace, as they can give insight in parts of process that are difficult to measure directly, and if used together with experimental measurements they can help in efficient, reliable and environmental friendly operation of boiler unit.

Acknowledgement

This work was supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia (project: *Increase in energy and ecology efficiency of processes in pulverized coal-fired furnace and optimization of utility steam boiler air pre-heater by using in-house developed software tools*, No. TR-33018).

Nomenclature

A_{inl}	– particles inlet surface area, [m ²]	N_p	– particle concentration, [m ⁻³]
A_p	– particle surface projection area, [m ²]	Re_p	– particle Reynolds number (= $d_p \vec{u} - \vec{u}_p / \nu$), [-]
C	– dimensionless concentration, [-]	\bar{r}	– local reaction rate, [-]
C_D	– drag coefficient, [-]	r_A	– reaction rate per unit vol., [kmol m ⁻³]
$C_{\varepsilon 1}$	– constant in ε equation, [-]	S_0	– initial particle specific surface, [m ² m ⁻³]
$C_{\varepsilon 2}$	– constant in ε equation, [-]	S_1	– product layer specific surface, [m ² m ⁻³]
$C_{\varepsilon 3}$	– constant in ε equation, [-]	S_2	– unreacted specific surface, [m ² m ⁻³]
c_{A0}	– SO ₂ concentration, [kmol m ⁻³]	S_{avg}	– average spec. surface area, [m ² m ⁻³]
Da	– Damkohler number (= kl_0/D_s), [-]	$S_{0,CaO}$	– nascent calcium surface area, [m ² g ⁻¹]
$D_{eff,0}$	– effective diffusion coefficient in the particle, [m ² s ⁻¹]	S_{CaO}^s	– sintered CaO area, [m ² g ⁻¹]
D_s	– diffusion coefficient of product layer, [m ² s ⁻¹]	S_p^k	– turbulent kinetic energy source due to particles, [kg m ⁻¹ s ⁻³]
$d_p, d_{p,ij}$	– particle diameter, [m]	S_p^m	– source term due to mass exchange between phases, [kg m ⁻³ s ⁻¹]
\vec{g}	– gravitational constant, [m s ⁻²]	S_p^e	– turbulent kinetic energy dissipation source due to particles, [kg m ⁻¹ s ⁻⁴]
K_s	– sintering rate factor, [-]	S_{p,SO_2}^m	– source term due to mass exchange (SO ₂ released from coal), [kg m ⁻³ s ⁻¹]
k	– turbulent kinetic energy, [m ² s ⁻²]		
L	– thickness of product layer, [m]		
l_0	– characteristic distance, [m]		
m_p	– particle mass, [kg]		
\dot{N}_{ij}	– particle number flow, [s ⁻¹]		

S_{p,SO_2}^{CaO}	– source term due to mass exchange (SO ₂ captured by sorbent), [kg m ⁻³ s ⁻¹]	Γ_Φ	– transport coefficient, general variable
S_Φ	– source term for general variable Φ	γ	– sintering rate factor, [–]
S_Φ^p	– source term for general variable Φ due to reactions between phases	δ	– relationship between effective diffusivity at the beginning and at the current time step, [–]
T_k	– temperature, [K]	ε	– turbulent kinetic energy dissipation, [m ² s ⁻³]
t_n	– time at nth time step, [s]	μ	– dynamic viscosity, [kg m ⁻¹ s ⁻¹]
U_j	– time avg. velocity component, [m s ⁻¹]	μ_t	– turbulent dynamic viscosity, [kg m ⁻¹ s ⁻¹]
\vec{U}_p	– time avg. particle velocity, [m s ⁻¹]	ν	– kinematic viscosity, [m ² s ⁻¹]
\vec{U}_{pc}	– convective velocity, [m s ⁻¹]	ν_t	– turbulent kinematic viscosity, [m ² s ⁻¹]
\vec{U}_{pd}	– diffusion velocity [m s ⁻¹]	ν_p^t	– turbulent kinematic viscosity due to particles, [m ² s ⁻¹]
\vec{u}	– current velocity, [m s ⁻¹]	ρ	– fluid density, [kg m ⁻³]
\vec{u}_p	– particle velocity, [m s ⁻¹]	ρ_p	– particle density, [kg m ⁻³]
X_c	– extent of calcination, [–]	τ	– time [s]
X_{SO_2}	– SO ₂ concentration, [kg kg ⁻¹]	τ_t	– Kolmogorov time scale [s]
x_j	– coordinate, [m]	$\tau_{p,ij}$	– particle time scale [s]
z	– position inside of the particle, [m]	Φ	– general variable
Greek symbols		Φ_{th}	– Thiele modulus [= 0.5d _p (kS ₀ /D _{eff,0}) ^{0.5}], [–]
Γ_p	– coefficient of particles diffusion, [m ² s ⁻¹]		
Γ_{SO_2}	– transport coefficient for SO ₂		

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