Environment Protection Engineering

Vol. 37 2011 No. 2

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NUMERICAL SIMULATION OF NO PRODUCTION IN A PULVERIZED COAL FIRED FURNACE

Behaviour of air–coal mixture has been described using the Navier–Stokes equations for the mixture of air and coal particles, accompanied by the turbulence model. The undergoing chemical reactions are described by the Arrhenius kinetics (reaction rate proportional to exp($-E/RT$)). Heat transfer via conduction and radiation has also been considered. The system of partial difference equations is discretized using the finite volume method and the advection upstream splitting method as the Riemann solver. The resulting ordinary differential equations are solved using the 4th order Runge–Kutta method. Results of simulation for typical power production level are presented together with the air staging impact on NO production.

1. INTRODUCTION

The main motivation of the examination of the combustion model is its potential inclusion in the current model of a steam generator [1], to use this model for development of control systems for the industrial installation. Another application for the combustion model is the optimization of the production of nitrogen oxide which strongly depends on the temperature and unburned char distribution, and thus it can be controlled by intelligent distribution of fuel and oxygen into the burners. Because the experiments on a real device are prohibitively cumbersome and expensive, in extreme cases even hazardous, the only way to test the behaviour of the furnace is mathematical modelling.

The mathematical model of combustion is based on the Navier–Stokes equations for a mixture of multiple components where coal particles are treated as one of the phases. We use this approach, as it simplifies the model especially when dealing with

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turbulence, and also removes several empirical relations and constants, moreover, it shortens the computational time of the simulation.

2. MATHEMATICAL MODEL

The mathematical model of combustion is based on the Navier–Stokes equations for a mixture of multiple components where the coal particles are treated as one of the phases. Unlike e.g. in [2], where the gas particles are treated separately and use separate equations of momentum, we chose to use this approach, as it simplifies the model especially when dealing with turbulence, and also removes several empirical relations and constants.

Currently, the following components of the mixture are considered:

- chemical compounds engaged in major thermal and fuel NO reactions (nitrogen (N_2), oxygen (O_2), nitric oxide (NO), hydrogen cyanide (HCN), ammonia (NH_3), carbon dioxide (CO_2) and water (H_2O)),
- char and volatile part of the coal particle.

The gas phase is described by equations given below. The mass balance is described by equations of mass balance of each sub-component (the Einstein summation is used)

$$\frac{\partial}{\partial t}(\rho Y_i)+\frac{\partial}{\partial x_j}(\rho Y_i u_j)=\nabla\vec{J}_i+R_i, \tag{1}$$

where ρ is the flue gas mass density, Y_i – mass fraction of the i-th component, and u_i are the gas velocity components. The right hand side terms describe the laminar and turbulent diffusion of the components and either production or consumption due to chemical reactions within the R_i term. The equations of mass balance of components are supplemented by the equation of total mass balance:

$$\frac{\partial\rho}{\partial t}+\frac{\partial(\rho u_j)}{\partial x_j}=0. \tag{2}$$

Equations of momentum conservation are as follows

$$\frac{\partial}{\partial t}(\rho u_i)+\frac{\partial(\rho u_i u_j)}{\partial x_j}=-\frac{\partial p}{\partial x_i}+\frac{\partial}{\partial x_j}\left[\mu_{eff}\left(\frac{\partial u_i}{\partial x_j}+\frac{\partial u_j}{\partial x_i}-\frac{2}{3}\delta_{ij}\frac{\partial u_l}{\partial x_l}\right)\right]+g_i, \tag{3}$$

where $\vec{g}=[g_1,g_2,g_3]$ is the external force acting on the fluid, in our case it is the gravity force. The effective friction coefficient μ_{eff} is calculated from the turbulence model as

$$\mu_{\text{eff}} = \mu + \mu_t = \mu + \rho\, C_\mu \frac{k^2}{\varepsilon},$$

where μ is the laminar viscosity, k – the turbulent kinetic energy, and ε – the turbulent energy dissipation rate. Constant C_μ, like additional constants introduced later in the description of the turbulence model, has to be chosen empirically for a particular problem, in our case we use $C_\mu = 0.09$, which appears to give satisfactory results.

The last equation describes the conservation of energy

$$\frac{\partial}{\partial t}(\rho h) + \frac{\partial(\rho u_j h)}{\partial x_j} = -n_{coal} \frac{dm_{\text{coal}}}{dt} h_{\text{comb}} + q_r + q_c + q_s, \tag{4}$$

where the right hand side terms are the heat of combustion, heat transfer by radiation, heat transfer by conduction, and heat source or sink. The heat transfer terms are computed as follows

$$q_c = -\nabla \cdot (\lambda \nabla T),$$

for the transfer by conduction, which is described by the Fourier law of heat conduction, and

$$q_r = -\nabla \cdot (cT^3 \nabla T),$$

for the transfer by radiation. The radiation heat transfer is fully described by an integral-differential equation of radiation, which is very computationally expensive to solve. However, as the flue gas can be considered an optically thick matter, the above approximation of the radiation flux called the Rosseland radiation model can be applied.

The heat sink term is nonzero only in the edge computation cells and describes the energy exchange with the walls of the furnace via conduction and radiation

$$q_s = A(T_{\text{gas}} - T_{\text{wall}}) - B(T_{\text{gas}}^4 - T_{\text{wall}}^4),$$

where A and B are constants dependent on the properties of the interface between the modelled region and its surroundings.

The particle mass change rate is currently described by the one-step Arrhenius kinetics, which is used separately for the char and volatile coal components – combustion of the volatiles is more rapid than combustion of the char

$$\frac{dm_p}{dt} = -A_v m_p^\alpha [\mathrm{O}_2]^\beta \exp\left(-\frac{E_v}{RT_p}\right),$$

where m_p is the particle combustible mass, A_v, E_v are empirical constants, $[O_2]$ – oxygen concentration, T_p is the particle temperature and R is the universal gas constant.

As usual, these equations are supplemented by the equation of state

$$p = (\kappa - 1)\rho\left(e_{\text{gas}} - \frac{1}{2}v_{\text{gas}}^2\right),$$

where κ is the Poisson constant and e_{gas} is the gas energy per unit mass.

For the turbulence modelling, we use the standard k–ε model, which describes the evolution of turbulence using two equations, former for the turbulent kinetic energy

$$\frac{\partial}{\partial t}(\rho k) + \frac{\partial(\rho u_j k)}{\partial x_j} = \frac{\partial}{\partial x_j}\left[\left(\mu + \frac{\mu_t}{\sigma_k}\right)\frac{\partial k}{\partial x_j}\right] + G_k - \rho\varepsilon, \tag{5}$$

and the other one for the turbulent kinetic energy dissipation rate

$$\frac{\partial}{\partial t}(\rho\varepsilon) + \frac{\partial(\rho u_j \varepsilon)}{\partial x_j} = \frac{\partial}{\partial x_j}\left[\left(\mu + \frac{\mu_t}{\sigma_\varepsilon}\right)\frac{\partial \varepsilon}{\partial x_j}\right] + C_{1\varepsilon}\frac{\varepsilon}{k}G_k - \rho C_{2\varepsilon}\frac{\varepsilon^2}{k}. \tag{6}$$

Constants in this model have once more to be determined empirically, in our case they are: $C_{1\varepsilon} = 1.44$, $C_{2\varepsilon} = 1.92$, $\sigma_k = 1.0$, $\sigma_\varepsilon = 1.3$.

Left hand sides of the equations describe passive advection of the respective quantities by the advection velocity $\vec{u}$. Right hand sides describe their spatial diffusion, production and dissipation.

The term G_k describing the production of turbulence, can be derived from the Reynolds averaging process and written in terms of the fluctuating part of the velocity as

$$G_k = \tau_{jl}\frac{\partial u_j}{\partial x_l} = -\rho\overline{u_j' u_l'}\frac{\partial u_j}{\partial x_l} = \mu_t S^2 \qquad \text{with} \qquad S_{ij} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right),$$

where τ_{jl} is the Reynolds stress tensor and S_{ij} is a mean strain rate. However during practical computation, fluctuations u_j' and u_l' are unknown, we use the Boussinesq hypothesis that the Reynolds stress is proportional to the mean strain rate.

Diffusion of the species consists of two processes, laminar and turbulent ones, and the diffusion term in Eq. (1) can be written in the form

$$\vec{J}_i = -\left(\rho D_{i,m} + \frac{\mu_t}{Sc_t}\right)\nabla Y_i.$$

The former term corresponds to linear laminar diffusion, the latter one to turbulent diffusion. Given the fact that the turbulent diffusion generally predominate the laminar one, and the term $D_{i,m}$ is difficult to determine, the laminar diffusion can be usually ignored. The coefficient Sc_t is the turbulent Schmidt number and we have $Sc_t = 0.7$.

3. SIMPLIFIED MODEL OF NO CHEMISTRY

This model has been developed to approximately describe the amounts of NO emissions leaving a coal combustion furnace. The real mechanism of coal flue gas production seems to be very complicated, so that just the most important phenomena and reaction paths were considered to provide maximum possibility of using this model in real-time control and operation systems.

Two major processes contribute to the total NO chemistry. The former is known as thermal NO or Zeldovich NO and simply consists of oxidation of atmospheric nitrogen at high temperature. The latter is called fuel NO and describes creation of NO from nitrogen, which is chemically bounded in coal fuel. Fuel NO is usually the major source of NO emissions, when the thermal way is suppressed. These are the only mechanisms involved, although a few more could be considered (such as prompt NO (Fenimore) or nitrous oxide (N_2O) intermediate mechanisms).

3.1. THERMAL NO

Thermal NO generation mechanism is applicable only at high temperature and is represented by a set of three equations, introduced by Zeldovich [2] and extended by Bowman [3]

$$\mathrm{O} + \mathrm{N}_2 \xleftrightarrow{k_1} \mathrm{N} + \mathrm{NO},$$

$$\mathrm{N} + \mathrm{O}_2 \xleftrightarrow{k_2} \mathrm{O} + \mathrm{NO},$$

$$\mathrm{N} + \mathrm{OH} \xleftrightarrow{k_3} \mathrm{H} + \mathrm{NO}.$$

All these reactions are considered to be reversible. Rate constants were taken from [4]. In order to compute the NO concentration, concentrations of nitrogen radical [N], oxygen radical [O] and hydroxyl radical [OH] must be known. It is useful to assume that N is in a quasi-steady state according to its nearly immediate conservation after creation. In fact, this N radical formation is the rate limiting factor for thermal NO production, due to extremely high activation energy of nitrogen molecule, which is caused by a triple bond between two nitrogen atoms. Hence, the NO formation rate can be stated as

$$\frac{d[\mathrm{NO}]}{dt} = 2k_1^+[\mathrm{O}][\mathrm{N}_2]\frac{1 - \dfrac{k_1^- k_2^-[\mathrm{NO}]^2}{k_1^+ k_2^+[\mathrm{N}_2][\mathrm{O}_2]}}{1 + \dfrac{k_1^-[\mathrm{NO}]}{k_2^+[\mathrm{O}_2] + k_3^+[\mathrm{OH}]}}.$$

Concentrations of O and OH can be profitably described by following partial equilibrium approach [5, 6]

$$[\mathrm{O}] = K_1[\mathrm{O}_2]^{1/2}T^{1/2}, \qquad [\mathrm{OH}] = K_2[\mathrm{O}]^{1/2}[\mathrm{H}_2\mathrm{O}]^{1/2}T^{-0.57}.$$

Equilibrium constants are as follows

$$K_1 = 36.64\exp\left(-\frac{27123}{T}\right), \qquad K_2 = 212.9\exp\left(-\frac{4595}{T}\right).$$

3.2. FUEL NO

The composition analysis shows that nitrogen based species are more or less present in coal, usually in amounts of tenths to units of percent by weight. When coal is heated, these species are transformed into certain intermediates and then into NO. Fuel itself is therefore a significant source of NO pollutants. When a coal particle is heated, it is presumed that nitrogen compounds are distributed into volatiles and char. The parameter α is introduced to describe the distribution of the coal-bounded nitrogen between the volatiles and char part of the coal particle.

$$m_{\mathrm{vol}}^{\mathrm{N}} = \alpha m_{\mathrm{tot}}^{\mathrm{N}}, \qquad m_{\mathrm{char}}^{\mathrm{N}} = (1-\alpha)m_{\mathrm{tot}}^{\mathrm{N}},$$

where $\alpha \in \langle 0, 1\rangle$, $m_{\mathrm{tot}}^{\mathrm{N}}$ is the total mass of nitrogen per 1 kg of the coal used in boiler, $m_{\mathrm{vol}}^{\mathrm{N}}$ is the mass of bounded nitrogen in volatiles and $m_{\mathrm{char}}^{\mathrm{N}}$ is the mass of bounded nitrogen in char. As mentioned above, nitrogen transforms to pollutants via intermediates which usually are ammonia NH_3, and hydrogen cyanide HCN. To proceed further, we must define four parameters to describe complex partitioning of the fuel bound nitrogen:

- β is a part of volatile bounded nitrogen which converts to HCN,
- δ_1 is the distribution of char bounded nitrogen which converts to HCN,
- δ_2 is the distribution of char bounded nitrogen which converts to NH_3,
- δ_3 is the distribution of char bounded nitrogen which converts to NO,

$$\beta \in \langle 0, 1\rangle, \qquad \delta_1 + \delta_2 + \delta_3 = 1.$$

Different parametric studies should be carried out to find the best values of these parameters suitable for a specific type of coal. Five overall reactions of either NO formation or depletion were incorporated in the combustion part of the numerical code.

NO, HCN, NH_3 reactions. According to [7], we have following reactions and their formation rates

$$\mathrm{HCN} + \mathrm{O}_2 \xrightarrow{R_1} \mathrm{NO} + \mathrm{Z}, \qquad R_1 = 1.0\times 10^{10} X_{\mathrm{HCN}} X_{\mathrm{O}_2}^{a} \exp\left(-\frac{33732.5}{T}\right),$$

$$\mathrm{NH_3 + O_2 \xrightarrow{R_2} NO + Z}, \qquad R_2 = 4.0\times10^{16} X_{\mathrm{NH_3}} X_{\mathrm{O_2}}^{a} \exp\left(-\frac{16111}{T}\right),$$

$$\mathrm{HCN + NO \xrightarrow{R_3} N_2 + Z}, \qquad R_3 = -3.0\cdot10^{12} X_{\mathrm{HCN}} X_{\mathrm{NO}} \exp\left(-\frac{30208.2}{T}\right),$$

$$\mathrm{NH_3 + NO \xrightarrow{R_4} N_2 + Z}, \qquad R_4 = -1.8\cdot10^{8} X_{\mathrm{NH_3}} X_{\mathrm{NO}} \exp\left(-\frac{13593.7}{T}\right),$$

where X is the mole fraction, a is the oxygen reaction order and Z are other products we simply neglect.

Heterogeneous reduction of NO. Present char allows following adsorption process to occur

$$\mathrm{Char + NO \xrightarrow{R_5} N_2 + Z}.$$

Levy [8] uses surface area of pores (BET) to define NO source term

$$S_{\mathrm{ads}}^{\mathrm{NO}} = R_5 c_s \mathrm{A_{BET}} M_{\mathrm{NO}} p_{\mathrm{NO}},$$

where $R_5 = 2.27\times10^{-3}\exp(-17168.33/\mathrm{T})$ is the rate constant, $S_{\mathrm{ads}}^{\mathrm{NO}}$ is the NO source term, c_s is the concentration of particles, A_{BET} is the pore surface area and P_{NO} is the partial pressure of NO.

In order to evaluate overall NO source term, single source terms have to be summarized. This overall source term can be further used in transport equations. As for HCN and NH_3 source terms, it is possible to determine them from coal burnout rate. It is assumed that nitrogen from both char and volatiles transforms to intermediate species quickly and totally.

For numerical solution of the equations, the finite volume method is used. For left and right hand sides in Eqs. (1)–(6), advection upstream splitting method (cf. [9]) is used to approximate fluxes in the FVM formulation, and edge dual volume approximation is used to approximate the second order derivatives, respectively. For detailed description of the solution procedure see [1].

4. RESULTS

In Table 1, the values of various parameters used in simulation are given as well as summarized numerical values of air and fuel distribution among the burners, excess air coefficients for the burners and outlet concentration of NO, CO_2 and O_2 taken from the outlet area of the boiler. In Figure 1, profiles of mass fractions of NO are shown spatial-averaged over the boiler cross section area.

Table 1

Simulation parameters and fuel/air proportions for studied cases

Parameter	Symbol	Value
Gas heat capacity [J·kg^{-1}·K^{-1}]	c_V	1037.0
Molar weight of gas [kg·mol^{-1}]	M	0.02896
Raw coal density [kg·m^{-3}]	ρ	1345.0
Coal particle radius [m]	r_{coal}	0.2×10^{-3}
Coal heat capacity [J·kg^{-1}·K^{-1}]	c_{coal}	2048
Boiler cross-section [m^2]	S	49
Coal feeding rate [kg·s^{-1}]	$\dot{m}_{coal}$	5.63
Initial pressure [Pa]	p_{ini}	1×10^5
Initial temperature [°C]	T_{ini}	800
Mixture temperature at the inlet [°C]	T_{mix}	350
Pressure at the inlet [Pa]	p_{in}	1×10^5
Pressure at the outlet [Pa]	p_{out}	$p_{in} - 100$
Excess air coefficient	α	1.3
Temperature of the wall [°C]	T_{wall}	300
Width of the side wall [m]	a, b	7.0
First burners row position [m]	B_1	2.74
Second burners row positions [m]	B_2	3.84
Third burners row position [m]	B_3	4.93
Fourth burners row position [m]	B_4	6.03
Number of burners in a row	N_B	4
Burner diameter [m]	D_B	0.8
Gas–wall heat transfer coefficient due to conduction [W·m^{-2}·K^{-1}]		30.0
Gas–wall heat transfer coefficient due to radiation [W·m^{-2}·K^{-4}]		0.6×10^{-8}
Results of coal analysis [%]		
Ash	A	9.5
Water	W	30
Carbon	C	45.72
Hydrogen	H	3.69
Sulfur	S	1.16
Oxygen	O	9.42
Nitrogen	N	0.5

Case	Air distribution [%]				Fuel distribution [%]				Excess air coefficient				NO [ppm]	CO_2 [%]	O_2 [%]
	B_1	B_2	B_3	B_4	B_1	B_2	B_3	B_4	B_1	B_2	B_3	B_4			
1	25	25	25	25	25	25	25	25	1.3	1.3	1.3	1.3	357	20	1.9
2	50	20	20	10	25	25	25	25	2.6	1.04	1.04	0.52	177	20	1.9
3	10	20	20	50	25	25	25	25	0.52	1.04	1.04	2.6	134	20	2
4	25	25	25	25	50	20	20	10	0.65	1.63	1.63	3.25	182	18	4.5
5	25	25	25	25	10	20	20	50	3.25	1.63	1.63	0.65	233	20	2
6	10	20	20	50	10	20	20	50	1.3	1.3	1.3	1.3	355	20	2
7	50	20	20	10	50	20	20	10	1.3	1.3	1.3	1.3	388	20	1.9
8	50	20	20	10	10	20	20	50	6.5	1.3	1.3	0.26	173	20	1.9
9	10	20	20	50	50	20	20	10	0.26	1.3	1.3	6.5	124	20	2

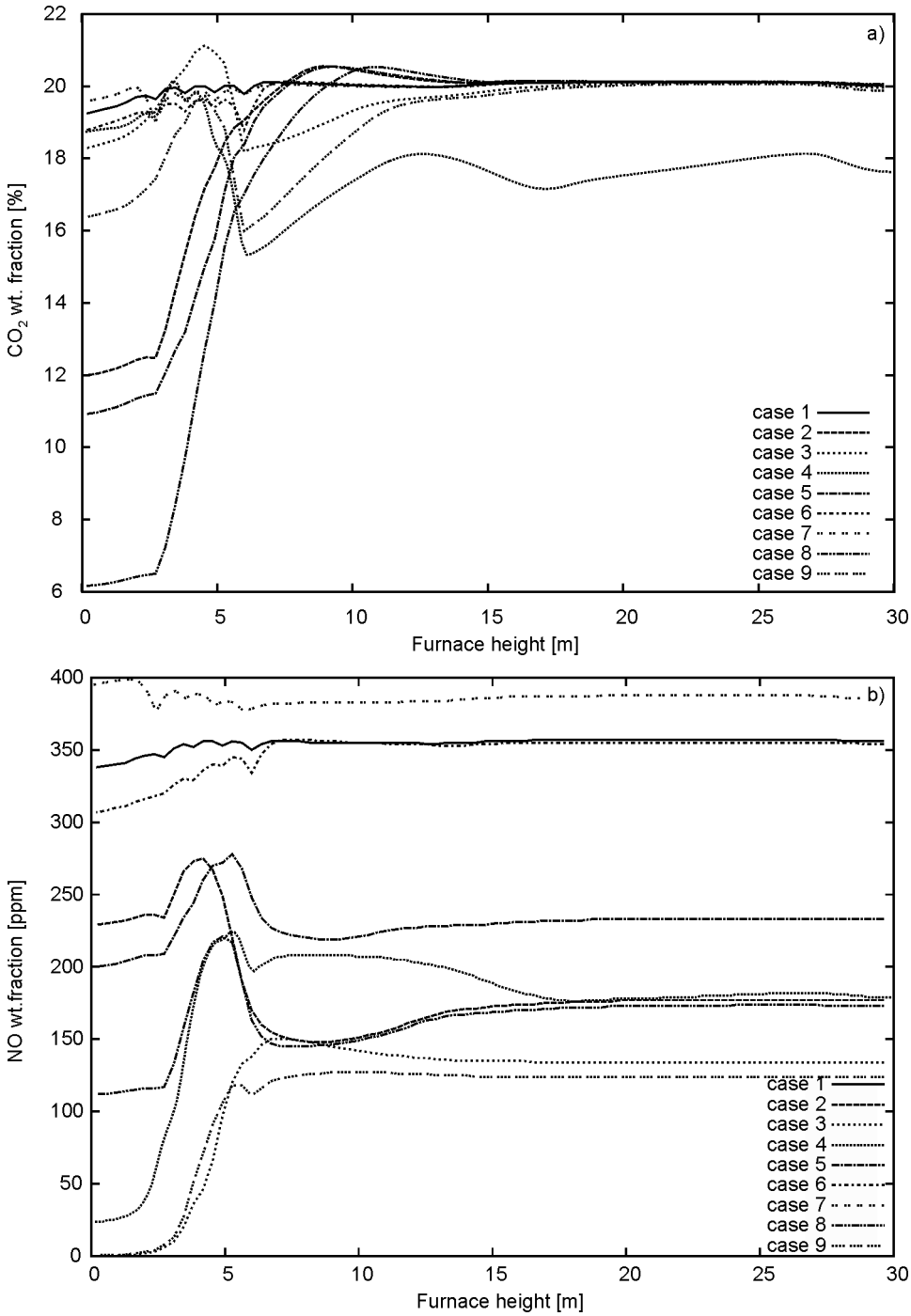

Fig. 1. Profiles of CO_2 (a) and NO (b) weight fractions for various arrangements of air and fuel

Averaged values are computed after 35 s of boiler operation. One can see that the lowest NO concentrations are obtained for the configuration 3 and 9. This is consistent with creating fuel-rich, oxygen-lean zone in the lower part of the burners, where the temperature is low and thermal NO production is inefficient, fuel NO is low because of low oxygen concentration and NO is also reduced on the char surface which is greater because of high concentration of fuel.

5. CONCLUSION

A numerical model of pulverized coal fired boiler has been presented. It has been developed taking into consideration the balance between computation complexity and the precision of simulation. This was also the reason for using a simplified reaction kinetics instead of full reaction set for the production of NO and simple kinetic approach of the coal particle combustion. At current state, the model can be used as air-stage designing tool for coal fired boilers. Presented results with emphasis on the NO production are adequate when compared with real life devices, as well as model behaviour for different cases. Still there are places where enhance of the model is needed, mainly the radiation heat transfer modelling and more accurate coal particle combustion model.

ACKNOWLEDGMENTS

This work has been partly supported by the project A*pplied Mathematics in Physical and Technical Sciences* MSM 6840770010 and *Jindřich Nečas Center for Mathematical Modelling* of the Czech Ministry of Education, Youth and Sport.

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