# Sensitivity analysis of reacting two-phase flow in nuclear heat-based gasification process

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## Abstract

Current work investigates influence of operating parameters on chemical reactions occuring within two-phase reacting flow. This particular flow analysed, corresponds to processes in coal gasifier unit supplied in heat by a high temperature gas cooled nuclear reactor (HTGR).

Due to the fact that gasification is a complex process, in which multiphase mixture undergoes chemical reactions, it crucial to answer questions about sensitivity to parameters changes.Performed analysis was dedicated to answer question about the optimal flow parameters. Controll of flow patern, namely the swirl of coal-oxygen mixture traversing the gasifier domain, allowed creating efficiency curve, relating gas composition with non-axial component of the velocity vector.

Using numerical model of the process, numbers of simulations were run in order to determine operation point yielding the highest efficiency, defined as a ratio of higher heating values of a syngas product of gasification process and coal feed into the unit. Obtained results concerning the most favorable operating parameters can be valuable information of evaluation of such gasification system from the economical point of view. Created tool can be used to study the system performance for various types of coal-fed.

*Keywords:* Gasification, two-phase model, reacting mixture, CFD

## 1. Introduction

Among many industrial processes involved in energy generation, gasification has recently become an interesting measure for reduction of energy generation-related environmental burdens. As proposed in early 70s and 80s, gasification can be coupled with high temperature nuclear reactors, using the fission as a source of heat for endothermic chemical reactions involved.

In the last decades gasification-based systems have been widely analyzed. Various models investigating variety of phenomena were proposed. Vast numbers of analysis performed were focused on the interference between thermal and chemical phenomena involved. Among developed models, those focused on two-phase flow with chemical reaction are of a high attention.

Presented work is dedicated to entrained flow gasifier, in which coal is delivered into the unit with approximately the same velocity as the fed gas. This approximation means that velocity of solid- and

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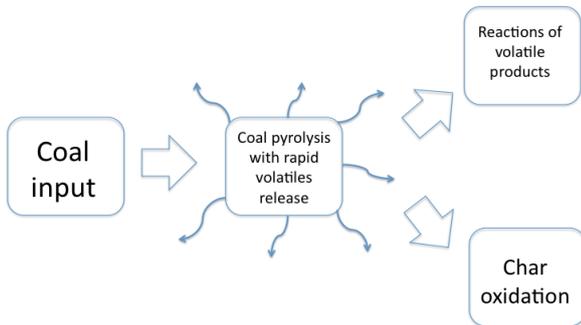
Figure 1: Stages of gasification process

gas-phase are equal [1].

Regardless type of gasifier and particular application, process comprises always the same four stages, subject to next section.

Composition of generated gas highly depend on the process temperature and pressure, type of coal and its physical structure, as well as characteristics of the flow, and eventually on the geometry of the gasifier. Among available literature, recognized paper by Work of Chen et al. [2] presents results of investigation of carbon monoxide generation during gasifier operation. Since gasification technology is dedicated to reduction of energy-related $CO_2$, wide discussion on the gas production was crucial for understanding the formation mechanism on carbon monoxide production. At the same time investigations of $CO_2$ generation in the gasification systems have been undertaken [3, 4]. As reported, back in the beginning of 1990s formation of carbon dioxide varies substantially with temperature of reaction and cannot be explained easily. Influence of pressure of the gasification process was analyzed by Ranish and Walker [5] at operating pressures up to 70 bars. Operation of a gasifier at low pressurewas reported to be favorable [6, 7], therefore presented simulations were run at pressure of 1 bar.

## 2. Chemistry of Gasification

Gasification is a multistage and multiphase process involving number of phenomena and interactions between them. In order to implement physics

behind investigated system in a correct way, an appropriate numerical approach must be carefully selected. Model of chemical reactions involved in coal gasification must account for processes occurring in two phases. The entire process of gasification combines particle transport, mass and heat exchange, phase change, and chemical reactions that strongly depend on the flow character. Four stages of gasification can be distinguished, which can be seen in Fig. 1.

### 2.1. Gas-phase reactions

Since the process was mainly analyzed from the gas-phase reactions point of view, this stage will be the only one discussed in details.

Chemical reactions of volatile products, undergo in a single gas-phase, transport equation in fluid is solved separately for each of materials released from coal particles. Capability of ANSYS CFX 12.1 allows modeling volatile product either as a group of materials, mixture with fixed composition, or a pure substance. For purpose of this modeling, volatiles are modeled as a fixed-composition mixture. Reactions considered in the gasification process are listed in the Table 1. Where $A$ and $N$ are the pre-exponential and temperature coefficients of Arrhenius equation (1), respectively. $E_a$ is the activation energy.

$$k = A \left( \frac{T}{T_o} \right)^N exp \left( -\frac{E_a}{RT} \right) \qquad (1)$$

Since last two reactions can proceed both ways, i.e. forward and backward with different reaction rates, and activation energies, those have been considered separately and indicated with $a$ for reaction proceeding forward, and $b$ for reaction backward. Kinetic parameters werebased on data found in the literature [8–11].

### 2.2. Chemical reaction modeling

Modeling of gasification incorporates investigation of turbulent reacting flow with chemical reactions. In case of such flows, mass balance equations must be solved separately for each of species. Generally saying, mass balance includes convection of species with the flow, generation and/or destruction

Table 1: Summary of reactions included in the analysis

| Nr | Reaction | N | A | $E_a$, J/mol |
|---|---|---|---|---|
| 1 | $CH_4+0.5O_2 \rightarrow CO+2H_2$ | 3.00E+08 | -1 | 1.26E+11 |
| 2 | $H_2+0.5O_2 \rightarrow H_2O$ | 6.80E+15 | 0 | 1.68E+11 |
| 3 | $CO+0.5O_2 \rightarrow CO_2$ | 2.20E+12 | 0 | 1.67E+11 |
| 4a | $CH_4+H_2O \rightarrow CO+3H_2$ | 4.40E+11 | 0 | 1.68E+11 |
| 4b | $CO+3H_2 \rightarrow CH_4+H_2O$ | 5.12E-14 | 0 | 2.73E+07 |
| 5a | $CO+H_2O \rightarrow CO_2+H_2$ | 2.75E+10 | 0 | 8.38E+10 |
| 5b | $CO_2+H_2 \rightarrow CO+H_2O$ | 2.65E-02 | 0 | 3.96E+06 |

of species by chemical reactions, and turbulent diffusion. In order to simplify computation and reduce numerical expense, two models have been proposed and are widely used, namely Finite Rate Chemistry and Eddy Dissipation Model.

Finite Rate Chemistry (FRC) Model The main assumption under this model states that the rate of progress of any elementary reaction can be reversible only if backward reaction is defined. With this assumption, rate of reaction progress $R_k$ can be determined with formula:

$$R_k = \left( F_k \prod_{I=A,B...}^{N_c} [I]^{r'_{kI}} - B_k \prod_{I=A,B...}^{N_c} [I]^{r''_{kI}} \right) \quad (2)$$

where:

*[I]* molar concentration of component

$F_k$, $B_k$ forward and backward reaction rate constants, respectively

*r* reaction order of component *I* for the elementary reaction *k*.

For elementary reaction the stoichiometric coefficient is equal to the reaction order. Reaction rate constants are temperature dependent, and this dependence is given by Arrhenius formula, and can be written separately distinguishing reaction forward and backward:

$$F_k = A_{kF} T^{\beta_{kF}} exp \left( -\frac{E_{kF}}{RT} \right) \quad (3)$$

and

$$B_k = A_{kB} T^{\beta_{kB}} exp \left( -\frac{E_{kB}}{RT} \right) \quad (4)$$

where, in respect to new notation:

$A_{kF}$, $A_{kB}$ pre-exponential factors

$\beta_{kF}$, $\beta_{kB}$ dimensionless temperature exponents

$E_{kF}$, $E_{kB}$ activation energies

$T$, $R$ absolute temperature and gas constant.

In particular cases $R_k$ may be directly specified, without implementation of presented correlation, however this requires sufficient knowledge. Direct specification of $R_k$ usually involves experimental determination of this parameter.

### 2.3. Eddy Dissipation Model (EDM)

In particular cases of turbulent flows, transport process can become relatively slower than chemical reaction rate. For such cases Eddy Dissipation Model has been introduced.

It is based on the assumption that a reaction time scale can be related to the dissipation of turbulent eddies occurring within the domain filled with reactants and products. There are two main differences between mixing of reactants in a gas phase and a liquid phase, which are of importance when reacting flows are to be modeled. The first difference is that the coefficient of molecular diffusion is much higher in gases than in liquids, meaning that the Schmidt number in the gas phase is much smaller ($Sc \sim 1$) than in the liquid phase ($Sc \gg 1$). The second difference results from the density variation of the gas phase and the resulting sensitivity of the gas phase density to pressure and temperature variations. Then reaction is not being controlled by the kinetic any more, therefore different approach is needed.

Moreover, prediction of behavior, where chemical kinetics limits reaction rates, is poor. With EDM, reaction rate is calculated as based on the turbulent mixing rate of the flow [12], while FCR reaction rate calculations are based on the chemical kinetics. General concept assumes that reactants mixing at the molecular scale leads to products formation. Reac-

tion rate corresponding to products formation can be related to the time required to mix reactants at mentioned molecular scale. While investigating turbulent flow, eddy properties will influence time required for mixing, and rate will be in this case proportional to turbulence model parameters, i.e. fraction with turbulent kinetic energy $k$ placed in the numerator and the dissipation $\varepsilon$ in the denominator.

Implementation of Eddy Dissipation Model in CFX incorporates two additional parameters – limiters.

It can be assumed that each of reactions occurring in the gas phase can be written in the following form:

Unit mass of **A** + x-mass of **B** → (unit mass of **A** + x-mass of **B**) **C**
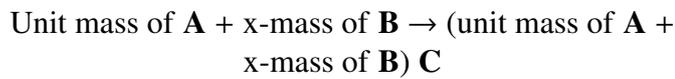
For such generic reaction the following transport equation can be written:

$$\frac{\partial}{\partial t}(\rho Y_A) + \frac{\partial}{\partial x_i}(\rho u_i Y_A) = \frac{\partial}{\partial x_i}\left[\frac{\mu_T}{S_{cT}} \cdot \frac{\partial Y_A}{\partial x_i}\right] + R_A \quad (5)$$

$$\frac{\partial}{\partial t}(\rho f) + \frac{\partial}{\partial x_i}(\rho u_i f) = \frac{\partial}{\partial x_i}\left[\frac{\mu_T}{S_{cT}} \cdot \frac{\partial f}{\partial x_i}\right] \quad (6)$$

$$\mu_T = \frac{C_D \rho k^2}{\varepsilon}$$

$$f = \frac{\beta - \beta_\infty}{\beta_o - \beta_\infty}$$

where $\mu_T$ is the turbulent viscosity with drag coefficient $C_D = 0.09$, $f$ is the mixture fraction, where $\beta$ is based on turbulent dissipation rates of reactants: $\beta = Y_A - Y_B/x$. Subscripts 0 and $\infty$ refer to value $x$ of $\beta$ at $A$-rich inlet, and to $\beta$ at $B$-rich inlet, respectively. The number that relates viscous diffusion rate and mass diffusion rate as their ratio can be written as:

$$S_{cT} = \frac{\mu}{\rho D} \quad (7)$$

Validity range of equations (5) and (6) is based on turbulent Schmidt number $S_{cT}$ of each ofinvolved species. Generally, equations are valid when Schmidt number is equal for all ofthem. This approach is fairly good for turbulent flows.

According to generic reaction presented earlier, turbulent dissipation rates of species $A$, $B$, and $C$ can be written as:

$$Y_A, \frac{Y_B}{x}, B\frac{Y_C}{1+x} \quad (8)$$

Based on them, transport equation for themass fraction of species $A$ is solved, where reaction rate of $A$ is taken as a minimum, which can be written:

$$R_A = -A\rho\frac{\varepsilon}{k}min\left(Y_A, \frac{Y_B}{x}, B\frac{Y_C}{1+x}\right) \quad (9)$$

In should be emphasized that $R_A$ is a mean reaction rate, i.e. time mean. Coefficients $A$ and $B$ are reactants and products limiters with values of 4 and 0.5, respectively, found in literature available for gaseous reactions [12]. However approach is well know, it is sometimes advised to set $B$ to negative value in order not to have product limiter, as it has been done in the current work.

Default values of $A$ and $B$ coefficients are 4 and -1, which in case of the second means that the product limiter does not apply. Such setting is an ANSYS CFX recommendation for multistep reaction schemes present within gasification process [13]. In this case the reaction rate is determined by reactants concentrations and turbulence.

In current analysis both models were used simultaneously. Such approach is justified, since each of reactions involved had different driving mechanism. The effective reaction rate was either computed as the minimum of rates computed using EDM and FRC models or only one model was associated with each of reactions, as proposed by Marklund et al. [14].

## 3. Multiphase flow with dispersed solid phase modeling

According to the operation principle of any type of gasifier, coal particles are introduced to the unit, hence from the very beginning the domain is filled with more than one phase. Solution of governing equations requires proper approach to each of the present phases.

Two models are generally considered for modeling multiphase flow, when one phase is solid, while

the other is of a fluid type (including both gas and liquid), namely Eulerian-Eulerian and Lagrangian Particle Tracking. Simultaneous application of two models allowed overcoming downsides of both. Volume of dispersed solid-phase was in order of 1–3% of the entire mixture volume, where less than 10% is a condition for application of Lagrangian Particle Tracking.

### 3.1. Forces acting on coal particles

Considering a single particle traveling in a continuous medium, number of forces influence particle acceleration and hence its velocity, and especially the difference between particle and fluid velocity. Momentum equation for such particle, in rotating reference frame was derived by Basset, Boussinesq and Ossen in the following form:

$$m_p \frac{dU_p}{dt} = F_D + F_B + F_R + F_{VM} + F_P + F_{BA} \quad (10)$$

where

$F_D$ drag force acting on the particle

$F_B$ buoyancy force

$F_R$ force due to domain rotation (centripetal and Coriolis forces)

$F_{VM}$ virtual mass force

$F_P$ pressure gradient force

$F_{BA}$ Basset force accounting for the deviation in the flow pattern from a steady state.

In current analysis $F_R$, $F_{VM}$ and $F_{BA}$ were not taken into account.

### 3.2. Swirl

In order to enhance mixing and provide better driving mechanism for turbulence-controlled reactions, swirl parameter was introduced. In principle swirl number has been defined [15] as the ratio of axial fluxes of angular and linear momentum, including the radius of the injector section, what can be written:

$$S = \frac{G}{G_x r_i} \quad (11)$$

where:

$$G = 2\pi \int \rho u \omega r^2 dr \quad (12)$$

and

$$G_x = 2\pi \int \rho \omega^2 r dr + 2\pi \int \rho r dr \quad (13)$$

This basically can be considered as a non-dimensional measure of the tangential momentum of a flow considered.

Since linear momentum depends on the velocity profile, it is difficult to determine it through measurements. Therefore, Kerr and Fraser [16] introduced modification to the swirl number definition. Under assumption that density variation and the pressure-integral contribution to the axial momentum flux is negligible, swirl number can be written as:

$$S = \frac{\int u \omega r^2 dr}{\int \omega^2 r dr \, r_i} \quad (14)$$

## 4. Results

Usingcreated CFX modelnumbers of simulations were runfor different gasifier inlet flow patterns. Summary of the most important model parameters, beside those concerning chemical reactions discussed earlier, can be seen in Table 2.

While using cylindrical coordinate system with radial component equal to zero, the azimuth became arbitrary andwas simply user-specified. This was a unique case of cylindrical reference frame very beneficial for presented work.

Simulations were run with discrete values of the angle in range 3–33°, as given in Table 2. According to technical specification of various swirl generators the angle can be as low as 2–50° up to as high as 45°, therefore particular selection was justified. With temperature ranging from 773 K up to 1173 K, simulations were run, and results for CO, $CO_2$, $H_2O$, and $H_2$ can be seen in Fig. 2 and 3.

As discussed earlier, chemical reactions involved in the process where controlled either by the residence time or by the turbulence of the flow.

### 4.1. Errors

Error estimation is a complementary part of any numerical analysis performed. In presented work, data corresponding to gas yield for different components were extracted from Lagranian Particle Tracking calculations. Due to high computation expense,

Table 2: Summary of model parameters

| | |
|---|---|
| Type of analysis | Steady-state |
| Model applied for coal | Lagrangian Particle Tracking |
| Model applied for gas | Eulerian-Eulerian |
| Turbulence model | $k$-$\varepsilon$ |
| Mass relaxation | 0.75 |
| Energy relaxation | 0.75 |
| Particle diameter | user-specified discrete diameter distribution |
| Coal particle size distribution [$\mu m$] | 12; 38; 62; 88 |
| Coal particle mass fractions [%] | 0.18; 0.25; 0.21; 0.36 |
| Coal particle number fractions | 0.25; 0.25; 0.25; 0.25 |
| Turbulence intensity | 10% |
| Transport equation solved for | $CH_4$, $H_2$, CO, $CO_2$, $H_2O$, $O_2$, and $N_2$ |
| Heterogeneous reaction | between fixed carbon and $CO_2$, $O_2$, $H_2O$ |
| Particles inlet temperature | 343 K |
| Gas inlet temperature | 673 K |
| Gasifier operating pressure | 1 bar (up to 4 bar in certain cases) |
| Swirl angle | 0–33° |
| Physical time scale of analysis [s] | 0.005 |
| Gas-coal fluid pair drag force | Schiller-Naumann model |
| Gas-coal fluid pair heat transfer | Ranz-Marshall model |
| Gas-coal fluid pair emissivity | Modeled as black bodies, emissivity 1 |
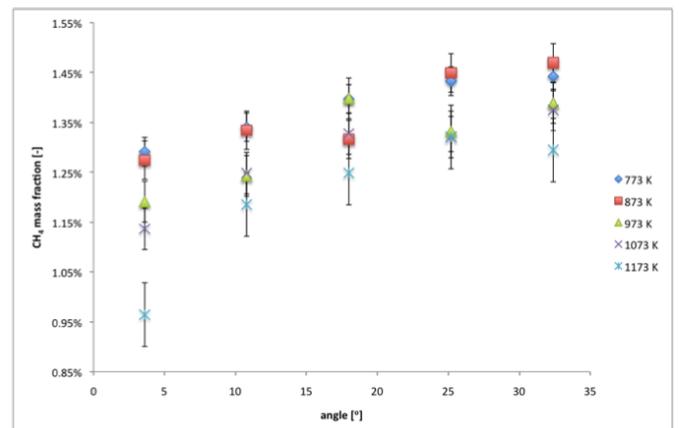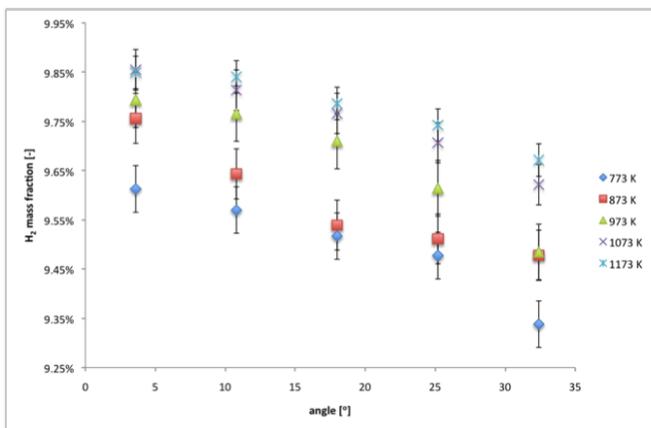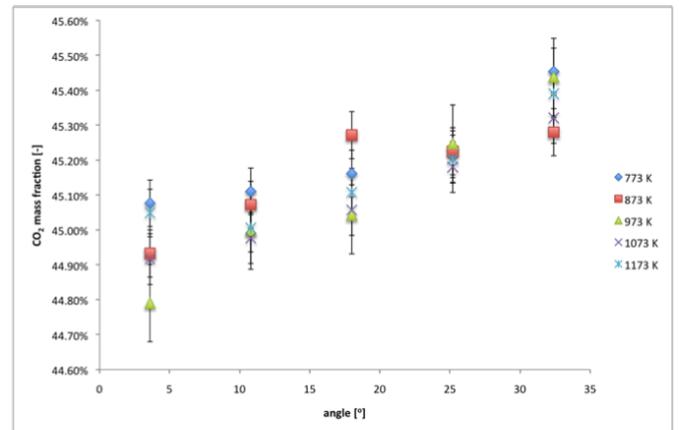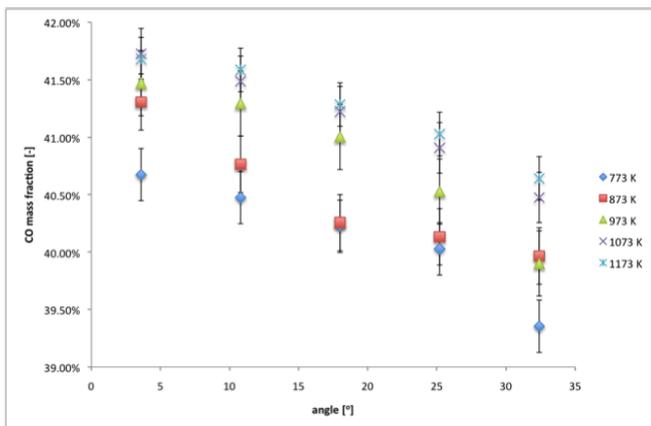
Figure 2: CO and $H_2$ yields for various swirl angle



Figure 3: $CO_2$ and $CH_4$ yields for various swirl angle
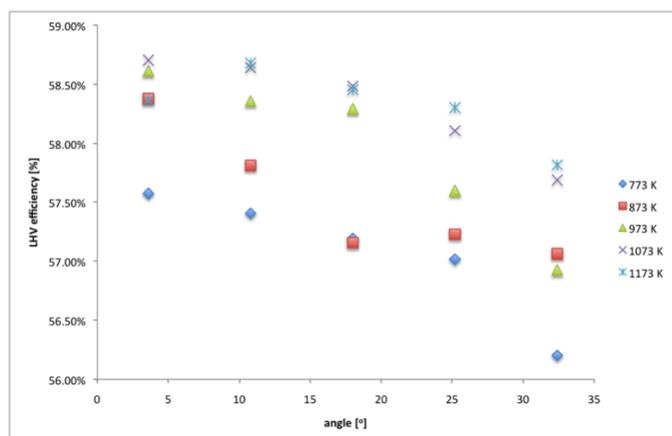
Figure 4: Efficiency of gasification process

there were only 25 particles traced, hence statistical uncertainty might had been an issue. In order to account for that fact, corresponding standard deviation is presented next to gas yields.

*4.2. Efficiency*

As proposed by Watanabe and Otaka [17] evaluation of gasification system performance can be done on the efficiency basis. Commonly used approach employs determination of the chemical heat of the produced gas in order to compare it with the coal feed. Efficiency distribution determined through calculations performed can be seen in Fig. 4.

## 5. Conclusions

Created numerical model reflected physics of proposed synthetic fuel generation system. Performed calculations provided information about system sensitivity to swirl angle, temperature of the gasification process. Parameters of chemical reactions found in the literature were based on experiments and are believed to be correct. Obtained efficiency was in agreement with literature available, and small angle of swirl corresponded to the highest efficiency of the process. Lack of commercially operating units limited possibility of validation with real-world unit, however experimental data were sufficient to do so.

Composition of generated gas corresponded to various experiments and similar simulations. High amount of carbonmonoxide is characteristic for proposed configuration with oxygen blown system. Carbon dioxide generation was promoted by intensive

mixing corresponding to high swirl numbers. Slight increase of $CO_2$ gas was accompanied by reduction of CO yield. Amount of hydrogen in order of 9% was roughly twice smaller than usually occurring in gasifier operating at comparable temperature level [17, 18].

It was possible to define the most favorable operating point of investigated gasification system, therefore the objective of current work was reached. Created model allowed evaluation of the system under various operating conditions. It can be further used to determine the conversion efficiency for various types of coal.

## Acknowledgements

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