Mathematical modelling of the oxyfuel gasification of pulverized coal fuel

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Abstract. In this work, we studied the efficiency of the coal gasification process under oxyfuel conditions. Using mathematical modelling one-dimensional stationary statement, the optimal parameters of coal processing were determined, air and oxyfuel conditions are compared. The calculated dependences of the characteristics of the gasification process on the stoichiometric ratio at different initial temperatures are constructed. The optimal values of oxygen stoichiometric ratio and the maximum values of cold gas efficiency in the selected range of parameters are determined. The contribution of the thermophysical and reactive properties of the gasification agent to the change in the cold gas efficiency is estimated.

1 Introduction

The proposed methods for suppression of carbon dioxide emissions from fossil fuels combustion can be divided into preventive ones, such as increasing the technical efficiency of thermal power plants, reducing the share of combustion by increasing the power of renewable and nuclear energy [1]; and utilizing ones, including CO_2 binding/disposal in compressed/liquefied/mineral form (carbon capture and storage, CCS [2, 3]) and its absorption by biomass (for example, in the form of energy plantations) [4]. Prospects for reducing the costs of CO_2 disposal are opened using new coal technologies, such as integrated gasification combined cycle (IGCC) and combustion in oxygen-enriched atmosphere (oxyfuel) [5, 6]. In this case, the usual combined cycle is realized using gasification products, and CO_2 is extracted in two possible ways. The first is the removal of carbon at the pre-combustion stage, when the generator gas is converted by steam, which allows the use of CO to obtain additional H_2, then the conversion products are separated by absorption or membrane methods, after which the hydrogen-enriched gas is burned in gas turbine, and CO_2 is concentrated and sent for burial. The second method is the removal of carbon from the combustion products, similar to that for traditional CCS systems: the gas mixture is cooled to condensation conditions, after which the CO_2 is separated from other gases and buried. The oxyfuel combustion technology is called to simplify the CO_2 emission in the post-combustion variant, when the oxygen concentration in the blast is usually higher than in air, and the mixture of combustion products (CO_2 and H_2O) plays the role of diluent instead of nitrogen. Combustion in oxyfuel-mode has better environmental characteristics compared to traditional combustion in air: firstly, it is obvious that less nitrogen oxides are formed (mainly fuel oxides);

secondly, the produced flue gases can be purified to high concentrations of CO_2, allowing its simple utilization (disposal). By adjusting the CO_2 supply, the intensity of combustion and heat transfer in combustion/gasification chamber can be controlled [7]. The oxyfuel technology is considered the most technically efficient, but at the same time one of the most expensive CCS systems [8, 9]. Among other problems, the requirements for the purification of flue gases from sulfur and nitrogen oxides can be noted [5]. Combined oxyfuel-combustion technologies are studied, for example, oxyfuel-MILD (decrease in oxygen concentration and increase in blast temperature) [10], the use of CO_2 as a blast or transport agent in pulverized coal gasification [11–13].

Recirculation of combustion products allows controlling the thermal regimes of fuel conversion: for example, in [14] a decrease in the combustion temperature in a fluidized bed by 100 K under oxyfuel conditions is reported; paper [15] describes a pilot plant with staged gasification of coal, where it was possible to increase the degree of carbon conversion (up to 90-95%) by adding CO_2 to the input air. Calculations in work [13] show that an increase in CO_2 concentration can lead to instability of the coal jet flame: under gasification conditions, it is necessary to increase the specific consumption of the oxidizing agent or the oxygen concentration to maintain the stability of the process. One way to increase the stability of the flame may be to heat the blast to the ignition temperature of fuel particles [16–18]. Earlier estimates of high-temperature air-steam coal gasification process efficiency [19, 20] and energy production at coal-fired IGCC using this technology [17, 21, 22] showed that, when choosing suitable conditions, heating the blast allows maintaining high energy efficiency without oxygen enrichment. However, an increase in efficiency due to external heat is only possible with the supply of a suitable gasification agent

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(for example, with significant additions of steam). In this paper, using mathematical modeling, we study the efficiency of the pulverized coal gasification process using high-temperature blasting along with CO_2 recirculation.

2 Mathematical model

In this work, computational tool is used that allows finding values for fuel consumption in different operating modes of the gasifier (described in detail in [23, 24]). By simplifying the model, such calculations can be carried out in a wide range of conditions for the relatively short computational time. The calculation of the process as a whole is carried out as follows. The algorithm is based on the repeated use of a numerical model of the reacting fuel particle in changing thermal field:

$$Uc_p \frac{d(m_p T_p)}{dz} = \alpha S_p (T_g - T_p) + \varepsilon \sigma S_p (T_w^4 - T_p^4) + \sum_j Q_j r_j \quad (1)$$

In eq. (1) z is spatial coordinate (reaction zone length), m; U is particle velocity, m/s; m_p is particle mass, kg; T_p is particle temperature, K; c_p is particle heat capacity, J/kg/K; α is heat transfer coefficient, W/m²/K; S_p is internal surface area of particle, m²; ε is particle surface emissivity; σ is Stefan-Boltzmann constant, W/m²/K⁴; T_g is gas temperature, K; T_w is wall temperature, K; r_j is rate of physicochemical process (drying, pyrolysis, gasification reaction), kg/s; Q_j is heat of a reaction, J/kg.

Equation for drying rate r_{dr} depends on temperature:

$$r_{dr} = \begin{cases} \dfrac{\beta S_p M_{H_2O}}{R_g T} \left(P_{H_2O}^{eq} - P_{H_2O} \right), \ T_p \le T_b \\[4mm] \dfrac{\alpha S_p (T_g - T_p) + \varepsilon \sigma S_p (T_w^4 - T_p^4)}{|Q_{dr}|}, T_p > T_b \end{cases} \quad (2)$$

Here T_b is boiling temperature under given pressure, K; β is mass transfer coefficient, m/s; P_{H2O} – partial pressure of water vapours, Pa; R_g – universal gas constant, J/mol/K.

Pyrolysis rate r_{pyr} depends on temperature as follows:

$$r_{pyr} = k_{pyr} \exp\left(-\frac{E_{pyr}}{R_g T_p} \right) m_V \quad (3)$$

Here k_{pyr} is preexponential factor, 1/s; E_{pyr} is apparent activation energy, J/mol; m_V is volatile fraction of particle, kg.

Heterogeneous reactions proceed according to diffusional kinetics equation [25]:

$$r_g = \frac{S_p C_g}{\dfrac{1}{k_g e^{-\frac{E_g}{R_g T}}} + \dfrac{d_p}{Nu_D D_g}} \quad (4)$$

Here C_g is gasification agent concentration (O_2, CO_2, H_2O), k_g is preexponential factor of heterogeneous reaction, m/s; E_g is activation energy, J/mol; Nu_D is mass transfer Nusselt number; D_g is diffusivity of gaseous oxidizer, m²/s; d_p is averaged particle size, m. Heat of a

reaction Q_j is estimated based on thermochemistry date base [26]. Diffusivities D_g are calculated based on data [27]: diffusivities O_2 and H_2O are binary diffusivities in mixtures with CO_2 or N_2 as diluent; diffusivity of CO_2 in oxyfuel conditions is self-diffusion coefficient. Particle velocity is considered as equal to gas velocity that is determined by continuity equation. Gas composition in every section is considered to be at chemical equilibrium under fixed fuel conversion degree [23, 28]. Numerical solution of a whole equation system is obtained as follows: the kinetics of chemical transformations is calculated using a system of ordinary differential equations (2–4) to change the particle mass for a given temperature distribution; then the stationary problem of heat transfer (1) is solved taking into account heat sources.

Chemical kinetics of reactions in the gas phase is not considered: it is assumed that the substances entering the gas phase quickly reach equilibrium. Thus, chemical transformations are described using a thermodynamic model with macrokinetic constraints on the fuel conversion degree. This approach is applicable for high-temperature processes in which the rate of gas-phase processes is quite high compared to the rate of heterophase ones. In this case, the task of calculating the gas composition is as follows [29]:

Find $\quad \min G(\mathbf{n}^g)$

under constraints:

$$G(\mathbf{n}^g) = \sum_j n_j^g \left(\mu_j^0 + R_g T_g \ln \frac{P n_j^g}{\sum_k n_k^g} \right) \quad (5)$$

$$\mathbf{A} \mathbf{n}^g = \mathbf{b} \quad (6)$$

$$\mathbf{n}^g \ge 0 \quad (7)$$

Here G is Gibbs free energy, J; $\mathbf{n}^g$ is gas phase composition, mol; μ^0 – is a chemical potential, J/mol; P is pressure, atm; $\mathbf{A}$ is matrix of atomic composition; $\mathbf{b}$ is vector containing amounts of atoms, mol. In this case, Gibbs free energy is chosen in order to simplify the calculations: in a stationary state, the local temperature can be considered a constant parameter. The temperature values are iteratively refined when solving equations (1) and (2-4).

The amount of carbon entering the gas phase during gasification is taken into account as an amendment to vector $\mathbf{b}$, which, in addition to the elemental composition of the initial gas phase, includes a change in the elemental composition of solid fuel during pyrolysis and gasification.

3 Results and discussion

Entrained flow gasifier is considered with internal diameter of 3 m and length of 15 m. Fuel is pulverized coal with following characteristics: $W^r = 2\%$; $A^d = 15.38\%$; $V^{daf} = 29.42\%$; $C^{daf} = 85.45\%$; $H^{daf} = 4.86\%$; $N^{daf} = 2\%$; $S^{daf} = 0.67\%$; averaged particle size os 100 μm. Mixtures of O_2/N_2 and O_2/CO_2 are used as gasification agent. An example of calculations based on the model described above is presented at Figs. 1 and 2

for following conditions: initial gasification agent temperature is 573 K, operating pressure is 30 atm, oxygen concentration is 20% vol., fuel consumption is 100 t/h, oxygen stoichiometric ratio α is 0.4. After fuel enters the reaction zone, particles are heated and ignited, the length of the oxygen zone substantially depends on the composition of the gasification agent (0.6 m for air and 1.2 m for O_2/CO_2 mixture). This is due to the deterioration of the conditions for ignition of coal particles ([30, 31]). Under air gasification, fuel conversion takes place along the entire length of the reaction zone; under oxyfuel conditions, the core is much smaller.

All other conditions being equal (such as oxygen concentration, oxygen stoichiometric ratio, initial temperature), the gasification process under oxyfuel conditions occurs at lower temperatures, which is associated with the thermochemical properties of CO_2 [5]: the specific heat capacity of CO_2 is higher than that of N_2; diffusivity of O_2 and H_2O in CO_2 are lower than in N_2; CO_2 enters endothermic reaction with the fuel carbon.

Cold gas efficiency η_{chem} is commonly used as performance criterion of gasification process:

$$\eta_{chem} = \frac{Q_g G_g}{Q_f G_f} 100\%$$

(8)

Here Q_f is heating value of input fuel, J/kg; G_f is coal consumption, kg/h; Q_g is heating value of produced gas, J/Nm^3; G_g is gas production, Nm^3/h. Heating value of produced gas is determined by content of CO, H_2 and CH_4. Gasification process characteristics are compared in Table 1. Under oxyfuel conditions, produced gas contains more CO and less H_2, while heating value of is higher compared to air conditions. Despite temperature decrease, fuel conversion is also higher under oxyfuel conditions, which, apparently, is due to high concentration of gaseous oxidizer. Cold gas efficiency is also higher when using O_2/CO_2 mixtures.

Table 1. Comparison of gasification characteristics.

Process outputs	O_2/N_2	O_2/CO_2
N_2, %	57.75	1.04
CO_2, %	0.68	50.43
H_2O, %	0.43	7.21
CO, %	29.88	34.69
H_2, %	10.12	5.37
CH_4, %	0.36	0.44
Q_g, MJ/Nm^3	5.02	5.52
X_C, %	97.5	98.0
η_{chem}, %	75.20	77.07
T_{max}, K	1837	1322
T_{out}, K	1320	1071

Thus, the addition of CO_2 can improve the efficiency of coal gasification, but significantly reduces the temperature of the process. Thermal stabilization of the gasification process is possible by increasing stoichiometric ratio [13], or by heating the gasification agent. Fuel gasification with high temperature air (HiTAG) was considered earlier in [21]. The possibility of a significant increase in efficiency by using a suitable blast composition (primarily, with the addition of water vapor) was shown. In contrast to this work, the characteristics of the gasification process in regimes with CO_2 recirculation are considered here.
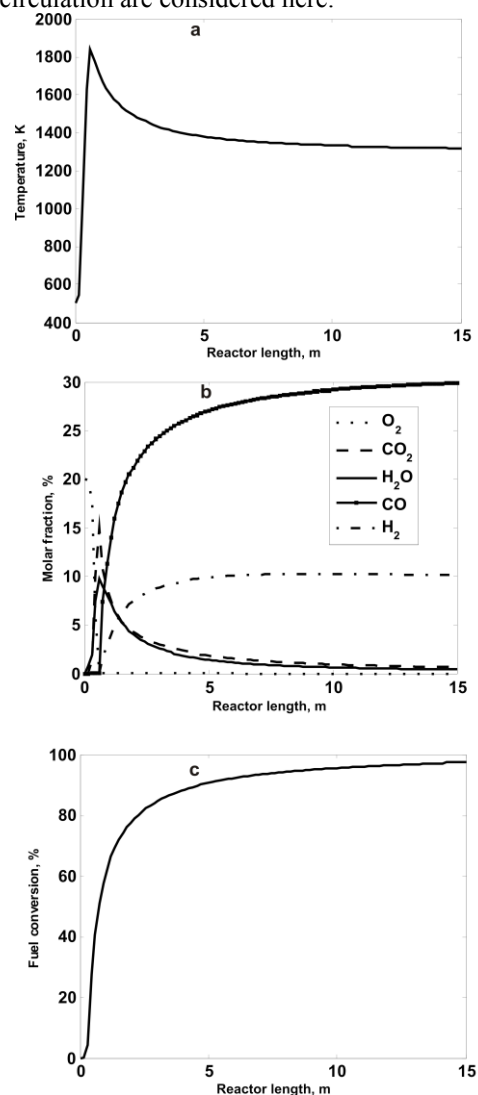
Fig. 1. Distribution of temperature, gas composition and fuel conversion degree along the reaction zone length under air gasification conditions.

Using the mathematical model, it is possible to optimize the gasification process by varying its parameters (first of all, the oxygen stoichiometric ratio). As is known from the thermodynamic analysis of gasification processes (see, for example, Ref. [32]), when gasifying solid fuels using oxygen-containing agent, there is an optimal stoichiometric ratio, which is determined by the conversion of fuel and oxidation of the produced gas. The value of the optimal oxidizer consumption depends on the process conditions: fuel and gasification agent composition, pressure, temperature, size of the reaction zone, etc. Figs. 3 and 4 shows the dependences of the cold gas efficiency of coal gasification process on the composition and oxygen

stoichiometric ratio at oxygen concentration of 20% vol. Coal gasification in O_2/N_2 with a temperature of 573 K allows to achieve a cold gas efficiency of 72.8% at α = 0.4; the use of blast with the composition of O_2/CO_2 makes it possible to obtain an cold gas efficiency of 83.7% at α = 0.3. Such a sharp decrease in optimal stoichiometric ratio value is due to an increase in the concentration of the reacting gases. However, stationary modes of coal gasification in the O_2/CO_2 atmosphere are possible starting from α = 0.25. At α > 0.45, both dependences practically merge, since after complete carbon conversion, the process efficiency is determined by the oxidation of combustible components. An increase in the temperature of the gasification agent to 1173 K practically does not lead to a change in the cold gas efficiency of air gasification, however, the use of O_2/CO_2 mixture can increase the efficiency up to 95.4%. But at the same time, the most efficient modes of oxyfuel gasification are close to the boundary of the thermal stability of the gasification process. Another possibility of thermal stabilization may be increasing the oxygen concentration in the O_2/CO_2 mixtures.
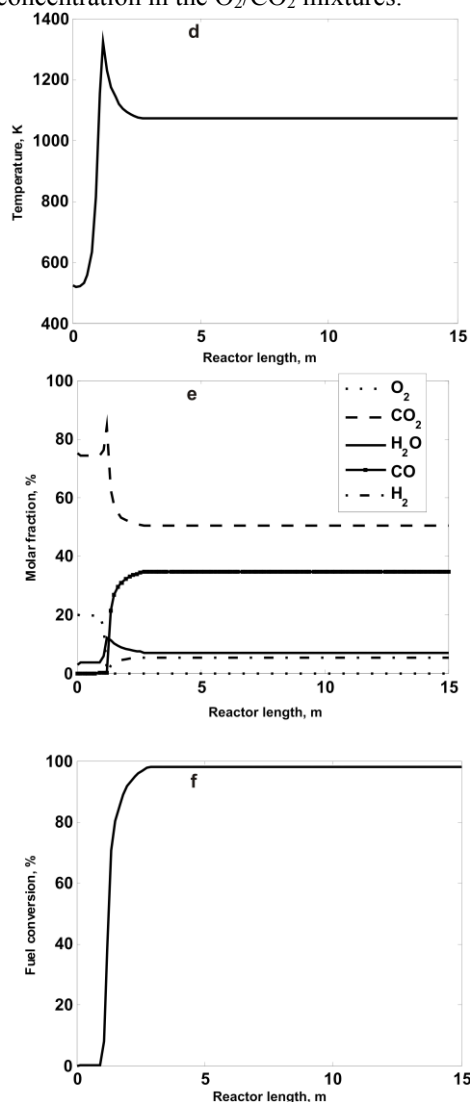

Fig. 2. Distribution of temperature, gas composition and fuel conversion degree along the reaction zone length under oxyfuel gasification conditions.

It should be noted that high-temperature heating of O_2/CO_2 mixtures under high pressure requires the use of special materials: these issues are not considered in this paper, since the main goal is to determine the limiting values of the oxyfuel technology characteristics.
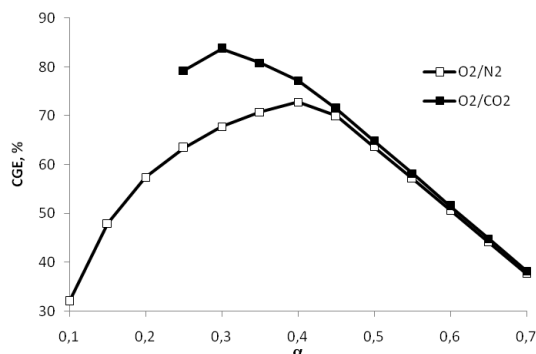
Fig. 3. Dependence of cold gas efficiency on stoichiometric ratio under initial gasification agent temperature 573 K.
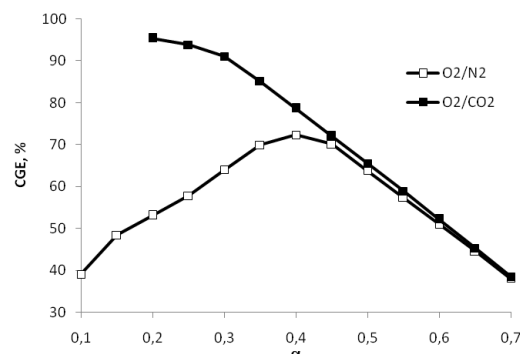
Fig. 4. Dependence of cold gas efficiency on stoichiometric ratio under initial gasification agent temperature 1173 K.

4 Conclusions

Pulverized coal gasification process efficiency in O_2/N_2 (air conditions) and O_2/CO_2 (oxyfuel conditions) is estimated using mathematical modeling. It is shown that under equal stoichiometric ratio, the gasification temperature in O_2/CO_2 mixtures is 200-300 K lower nad cola gas efficiency is 10-20% higher compared to air conditions. Recycling of CO_2 reduces the specific consumption of the oxidizing agent. Gasification agent heating makes it possible to increase the gasification process efficiency by the addition of CO_2.

Oxyfuel gasification in this case is a promising way of high temperature heat recovery. The conversion of fuels using CO_2 can be one of the ways to increase the efficiency of coal-fired thermal plants with the CCS. Of interest is the study of gasification processes with partial replacement of air nitrogen by CO_2.

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