Abstract

Introduction: More than 80% of the world's energy comes from fossil fuels. Coal is one of the main sources of fossil fuel energy, whereas coal combustion is a key way of energy conversion. Unfortunately, this ongoing process tremendously impacts the natural environment, climate, and human health. As a result, specific measures are taken to promote and develop efficient technologies which can mitigate the negative impact on our planet. Coal, despite its gradually decreasing consumption in many countries, will continue to be a meaningful energy source for many years to come. This results in continuous research in environmentally benign coal-based technologies. Entrained flow coal gasification is one of the promising coal-technologies with high efficiency and low environmental impact. The gasification main product – syngas, can be utilized in many branches of industry and electricity production (e.g., in internal combustion engines, fuel cells, and gas turbines). It also can be converted into other chemical products of commercial interest (diesel, ethanol fuels). However, the process is still not fully understood on a fundamental level and many models that describe specific gasification sub-phenomena still lack high accuracy. Many aspects in gasification differ from conventional pulverized coal combustion and many issues have not yet been thoroughly investigated.

Objectives: This work aims to investigate thoroughly the most commonly applied modeling techniques of devolatilization, gas phase, and char conversion which are the key sub-processes of entrained flow coal gasification, and to propose optimization techniques and most optimum modeling approaches for these sub-processes that improve the accuracy of the CFD modeling of entrained flow coal gasification. These optimized models will be validated for a wide range of operating conditions.

Research methodology: The extensive literature review allowed me to gather the most important information regarding the formulas of the utilized models and applied kinetic parameters, their benefits, and drawbacks, made assumptions and simplifications.

As regards devolatilization, the emphasis was laid on two global models (single-step first-order approach, competing two-step reaction approach). The literature review indicated that the majority of researchers incorporated literature kinetic parameters for these models. Considering the fact that kinetic parameters are only valid for the conditions for which they were determined, such a simplification can cause huge inaccuracies. Therefore, I have focused on incorporating an optimization technique that would consider unique kinetic parameters for the specific conditions.

As regards the gas phase, the literature review indicated that the majority of authors utilized global combustion mechanisms and turbulence-chemistry interaction approaches the same as in coal combustion studies. However, these processes differ substantially. On top of that, no study was found that would thoroughly investigate and compare the gas phase models strictly in gasification conditions which would help to assess their accuracy in such conditions. On this basis, I have decided to study these aspects thoroughly.

In the char conversion process the main focus, similarly to devolatilization, was laid on

the optimization techniques of the global empirical model (kinetic/diffusion model) based on the advanced carbon burnout kinetic (CBK) approach [1–4]. This procedure will also allow me to obtain optimized kinetic parameters corresponding to the investigated coal and operating conditions.

Main results: Devolatilization - The optimized kinetic parameters depend on operating conditions (heating rate) and fuel properties (proximate and ultimate analysis). Such an outcome was possible thanks to the complex devolatilization models which require heating rate as input and data from proximate and ultimate analyses. Direct utilization of complex network approaches within CFD can substantially raise the computational effort, therefore such an optimization procedure is an optimum choice of raising the accuracy without increasing the computational burden. Moreover, the use of experimental techniques could be very challenging, or even inviable, especially in the case of large-scale reactors. The literature review indicated that similar approaches of gaining kinetic parameters have already been undertaken, but only for a constant heating rate. The novelty of this dissertation consists in employing the instantaneous heating rate as input data. This extended analysis was performed for both entrained flow coal gasification and for coal devolatilization in an inert atmosphere (coal pyrolysis). Moreover, the research considered the comparison of two detailed devolatilization models (CPD [5–7] and FG-DVC [8]) with respect to the volatile yield prediction, providing clear drawbacks of the latter. The entire optimization procedure has an iterative character and its utilization allowed to increase the accuracy of simulations with regard to the experimental data for both coal gasification and coal pyrolysis on the basis of the error analysis.

Gas phase - The investigation considered two ideal reactors (plug flow reactor and a perfectly stirred reactor) and three actual gasifiers, as part of the CFD analysis. I laid emphasis on the temperature distribution and main syngas components distribution in each reactor. The reference data that I compared the numerical results with were the experimental results and two detailed mechanisms – GRI-Mech [9] and CRECK [10]. These detailed mechanisms allowed me to obtain accurate results for a wide range of operating conditions. However, the two reactors that I examined considered pressures higher than the ones for which GRI-Mech was validated. On this basis, I have also decided to carry out additional simulations to see what would be the performance of GRI-Mech outside of its validation/optimization range. The results confirmed the utility of GRI-Mech for higher pressures. The analysis of the plug-flow reactor and the perfectly stirred reactor allowed me to determine the most accurate global reaction mechanism with regard to GRI-Mech and CRECK in gasification conditions. A substantial influence of the turbulence-chemistry interaction on the gas composition was pronounced. Specific attention was also paid to the impact of the water-gas shift reaction which was found to be the key gas-phase reaction.

Char conversion – the optimized kinetic parameters for the kinetic/diffusion global model are obtained based on the advanced carbon burnout kinetic model for oxidation (CBK/E) [11] and gasification (CBK/G) [12], which determines the oxidation and gasification surface reaction rates taking into account such aspects as intrinsic surface area evolution, pore diffusion, thermal annealing, and ash inhibition. Thanks to the procedure, the optimized parameters accurately matched the prediction of the advanced approach. A CFD analysis was performed in order to estimate, whether the optimized global model provided more accurate results than the model with literature kinetic parameters, based on the experimental measurements. It turned out that, similarly to devolatilization, the optimization method for char conversion managed to increase the agreement of the syngas composition and char conversion degree with experimental data. Additionally, it was emphasized that the applied kinetic parameters had a very strong impact on the gas formation in the reforming zone and the flame zone. After the comparison of each of the gasification main sub-processes, I determined that the char conversion phase and accompanying its surface reactions have the greatest impact on the gasification process. Nevertheless, each of the subprocesses is of crucial importance, when one considers effective and credible simulations of entrained flow coal gasification.

Work layout: The work consists of 7 chapters. Introduction aims to present and justify the scientific problem which I decided to investigate. Chapter 2 considers the review of literature on devolatilization modeling, gas-phase modeling, and char conversion modeling which are the key phases in entrained flow coal gasification. In sub-chapter 2.1 the main objectives and scope of the thesis are summarized. Chapter 3 regards the CFD model description that was used to model the entire entrained flow coal gasification processes for various cases. Chapter 4 discusses the optimization procedure of devolatilization that was applied to improve the accuracy of CFD simulations of entrained flow coal gasification. Chapter 5 presents the utilized gas-phase modeling techniques with the aim of assessing which are most suitable for reproducing the entrained flow gasification process. Chapter 6 discusses the optimization procedure of char conversion which was employed to improve the accuracy of CFD simulation of entrained flow coal gasification. Chapter 7 considers conclusions.

[1] R. Hurt, J.-K. Sun, M. Lunden, A Kinetic Model of Carbon Burnout in Pulverized Coal Combustion, Combust. Flame. 113 (1998) 181–197. doi:10.1016/S0010-2180(97)00240-X.

[2] H. Hurt R, M. Lunden M, G. Brehob E, J. Maloney D, Statistical kinetics for pulverized coal combustion, Symp. Combust. 26 (1996) 3169–3177.

[3] R.H. Hurt, J.M. Calo, Semi-global intrinsic kinetics for char combustion modeling††Entry 2 has also been referred to as "Langmuir kinetics." The present paper adopts common chemical engineering usage, in which the designation "Langmuir" is applied to the equilibrium adsorption, Combust. Flame. 125 (2001) 1138–1149. doi:10.1016/S0010-2180(01)00234-6.

[4] T. Lang, R.H. Hurt, Char Combustion Reactivities for a Suite of Diverse Solid Fuels and Char-Forming Organic Model Compounds, Proc. Combust. Inst. 29 (2002) 423–431. doi:10.1016/S1540-7489(02)80056-6.

[5] D.M. Grant, R.J. Pugmire, T.H. Fletcher, A.R. Kerstein, Chemical Model of Coal Devolatilization Using Percolation Lattice Statistics, Energy and Fuels. 3 (1989) 175–186. doi:10.1021/ef00014a011.

[6] T.H. Fletcher, A.R. Kerstein, R.J. Pugmire, M.S. Solum, D.M. Grant, Chemical Percolation Model for Devolatilization. 3. Direct Use of 13C NMR Data To Predict Effects of Coal Type, Energy and Fuels. 6 (1992) 414–431. doi:10.1021/ef00034a011.

[7] T.H. Fletcher, A.R. Kerstein, R.J. Pugmire, D.M. Grant, Chemical percolation model for devolatilization. 2. Temperature and heating rate effects on product yields, Energy & Fuels. 4 (1990) 54–60. doi:10.1021/ef00019a010.

[8] P.R. Solomon, D.G. Hamblen, R.M. Carangelo, M.A. Serio, G. V. Deshpande, General Model of Coal Devolatilization., ACS Div. Fuel Chem. Prepr. 32 (1987) 83–98. doi:10.1021/ef00010a006.

[9] G. Smith, D. Golden, M. Frenklach, N. Moriarty, B. Eiteneer, M. Goldenberg, C. Bowman, R. Hanson, S. Song, W. Gardiner, V. Lissianski, Z. Qin, GRI-Mech 3.0, (2000). http://combustion.berkeley.edu/gri-mech/version30/text30.html (accessed October 14, 2018).

[10] E. Ranzi, C. Cavallotti, A. Cuoci, A. Frassoldati, M. Pelucchi, T. Faravelli, New reaction classes in the kinetic modeling of low temperature oxidation of n-alkanes, Combust. Flame. 162 (2015) 1679–1691. doi:10.1016/j.combustflame.2014.11.030.

[11] S. Niksa, G.S. Liu, R.H. Hurt, Coal conversion submodels for design applications at elevated pressures. Part I. Devolatilization and char oxidation, Prog. Energy Combust. Sci. 29 (2003) 425–477. doi:10.1016/S0360-1285(03)00033-9.

[12] S. Niksa, G.S. Liu, Coal conversion submodels for design applications at elevated pressures. Part II. Char gasification, Prog. Energy Combust. Sci. 30 (2004) 679–717. doi:10.1016/j.pecs.2004.08.001.

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