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Design of the experimental rig for retrieving kinetic data of char particles



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ABSTRACT

A new concept for evaluating chemical kinetics data of combustion of coal char particle is proposed. The experimental rig based on the measurement of mass loss of the char particle during the combustion process has been designed and built. The char particles are dropped into a laminar carrier gas stream flowing in a narrow horizontal channel. Temperature and gas composition inside the channel are fully controlled and correspond to those encountered in industrial furnaces and boilers. The idea of the measuring method is to trace the falling particle in the carrier gas, retrieve the mass reduction rate from the curvature of the recorded trajectory of the particle, and measure its diameter. An original technique of determining the diameter using a medium resolution camera has been developed. The kinetic constants are evaluated using an original inverse technique.

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1. Introduction

Coal-fired power plants currently fuel 40% of global electricity production and, in some countries e.g. in Poland the portion of energy produced from coal exceeds 90% [1]. Highly efficient and clean coal combustion process requires proper design and operation of the combustion chamber of the boilers. To meet the present and future efficiency and pollution limits, new combustion chambers are optimized while the existing chambers undergo appropriate modifications. The tool used for both processes is the Computational Fluid Dynamics (CFD). To produce reliable results, CFD at its present state of development, requires reliable experimental data. In the case of coal combustion, critical information concerns chemical kinetics. Due to the complex chemical composition of coal, these data cannot be obtained from molecular simulations, thus experiments remain the only way to retrieve kinetic models. The process is costly and time consuming. Examples of such CFD simulations for both fluidized and pulverized coal combustion are available in the literature. Some representatives examples of such an approach can be found in literature [2–6].

Combustion of a coal particle is composed of three elementary phenomena: drying, devolatilization and char combustion. Depending on the particle size, these three processes can occur

* Corresponding author. E-mail address: wojciech.adamczyk@polsl.pl (W. Adamczyk). simultaneously (large particle) or sequentially (small particle). The work deals with the longest stage of these elementary processes, namely the char combustion, which can take up to few seconds. Char oxidation is controlled by two phenomena, diffusion and kinetics. At high temperatures, diffusion control is limiting, whereas at low temperatures it is kinetics that controls the rate of char oxidation. The picture of the process is even more complex, as the chemical reactions can take place on the particle surface or/and within the particle pores.

The primary parameter to be determined in experiments whose aim is to retrieve the kinetics of coal combustion is the temporal variation of the burnout rate (mass loss) of the particle. However, to find the necessary data entering the kinetic equation, the changes in time of several other parameters should be known. This set of data comprises particle shape and temperature as well as the composition and temperature of the surrounding gas. The final kinetics equation is given by formulae containing several coefficients [7,8], whose values are evaluated by fitting the experimental data to measured values. To determine the kinetic data of a given type of coal, several parameters must be known, including particle dimension, shape, rate of the particle mass loss and particle surface temperature. Mentioned physical parameters are often used as the input parameters for numerical modeling of industrial Pulverized Coal (PC) and Circulating Fluidized Bed (CFB) boilers using Computational Fluid Dynamics (CFD) methods. To accurately predict combustion process of coal, the kinetics data plays crucial role and has to be precisely defined. In computational codes combustion is modeled

by employing mathematical models with number of coefficients which are obtained by fitting mathematical model to experimental data. The most popular methods to acquire experimental data are Drop Tube Furnaces (DTF) [9–12] and Thermogravimetric analysis (TGA) [13–16].

Over the years, the DTF achieved status of the experimental technique which can be applied for determining both combustion rates of volatile matter and char in various operating conditions. The heating rates, which achieve up to 10,000 K/s are similar to those expected within the real pulverized boilers. The measurement strategy requires that the coal particles are injected to the reactor at the upper zone which next are carried by the gases. To ensure isothermal conditions the external walls of the DTF are covered by electrical heaters. Depending on the position of the collector the residence time of the particles can change which ensure different combustion rates of the particles. The composition of the oxidizing atmosphere is controlled by a set of flow meters. Collected material at the bottom of the DTF in the next stage is analyzed using TGA to determine the remaining fraction of combustible matter in particles. This method provides accurate data, nevertheless the running of experiments is very expensive and required experienced operating stuff. Some application of the DTF for investigation pyrolysis process of coal and wood biomass can be found in [17], whereas in work [18] the useability of DTF for retrieving kinetic correlation for coal under high temperature.

The TGA method allows tracking the changes of the sample weight as the function of time and temperature [19,20]. It can be applied also for other materials like biomass, wastes, and swage sludge rather then only for coal. The sample of coal (typically 1 mg to 30 mg) is placed on the load cell in an enclosure with controlled oxidation atmosphere. The TGA can also be used for carrying out proximate analysis of coal which is proven by the American Society for Testing and Materials (ASTM) [21]. Huge advantages of the TGA over the DTF is that the measurement process is relatively simple and cheap. However, huge disadvantages of the TGE is the low values of heating rate (1 K/s). In real pulverized coal combustion process the particles heating rates are much faster than those achieved in the TGA. The very rapid heating of coal particles may affect many physical processes and in consequence influences on predicted kinetics. That is the main reason why the kinetic data received from TGA experiments cannot be directly used while simulating pulverized coal boilers. Nevertheless, the kinetic data retrieved in chemically controlled regime [22] using TGA can be farther scaled to kinetic/diffusion or diffusion range [23]. Nevertheless, to use this concept the particle temperature, and oxygen fraction in vicinity to the particle has to be known, which in nature is difficult.

To mitigate the difficulties and restrictions of both mentioned methods, a new and unique experimental test-rig that can be used to measure the particle mass lost during combustion under different oxidizing atmospheres is proposed in this study. To combine advantages of both mentioned measurement methods (DTF and TGA), a new and unique experimental rig and particle tracking software have been proposed in this study. Experimental rig provides possibility to observe particle ignition, exact combustion time, instantaneous two-dimensional particle sphericity, and changes of particle diameter during combustion process. The particle tracking system collects particle trajectories which are then used by inverse analysis to determine the rate of burned char. Combining the tracking application with inverse code gives possibility to retrieve char kinetic data almost online which is extremely fast in contrary to other methods. Kinetic data in the next step can be relatively easily used as the input to sub-model for heterogeneous reactions of solid particles in a CFD code.

2. Designing of the experimental rig

The first step in the designing of the experimental rig was the formulation of the measurement methodology and concept of the test-rig. In the following stage Computational Fluid Dynamics (CFD) has been used to aid the detailed design of the experimental rig before its construction and assembly. Finally, the development of the numerical model was performed to retrieve the kinetic data out of the recorded particles trajectories.

2.1. Methodology

The main idea of the measuring procedure is illustrated in Fig. 1. The char particle is dropped vertically into a laminar gas stream flowing in a narrow horizontal channel. The composition of the gas is typically a controlled mixture of O_2/CO_2 or O_2/N_2 , other options can readily be used. For a given composition and temperature of the gas, the velocity of free fall of the particle can be determined. Similarly, the horizontal velocity of the carrier gas can be measured or evaluated. The vertical component of the particle velocity and the horizontal component of the gas velocity component, should have similar magnitudes. The trajectory of the particle in an inert atmosphere (no combustion) would then be a straight line of an inclination close to 45° if the particle falling velocity was equal to that of the carrier gas velocity. Appropriate gas velocity for given coal type and flow condition (gas temperature and composition) is adjusted before measurement are running. In practice it is difficult to find the exact velocity in which the inclination angle will be equal to 45°. However, the developed algorithm for retrieving coal particle mass lost rate is non-sensitive on the particle inclination angle which means that the gas velocity is taken into account by the mathematical formulation of the inverse algorithm. For combusting particles, the chemical reaction consumes carbon causing mass loss. This in turn reduces the free fall velocity leading to a deviation of



Fig. 1. Concept of the measurement procedure.



Fig. 2. Expected particle trajectories depending on particle diameter for two different oxygen fraction in oxidation gases.

the trajectory from straight line. Thus, the curvature of the trajectory depends strongly on the rate of mass loss of the particle. In the same atmosphere particle trajectory will bend differently depending on the type of char and the size of the particle. Namely, the trajectories of more reactive particles will depart from the straight line more pronounced than their less reactive counterparts. The particle motions are recorded online through transparent window using a high speed camera. The acquired particles trajectories are then treated with the mathematical model to retrieve the kinetic data by applying the inverse approach [24].

2.2. Prediction of the particle trajectories

Prior to building the experimental rig, a sequence of CFD simulations have been carried out in order to assess the main features of the rig. The geometry of the numerical model encompassed the measuring channel of the rectangular cross section of the dimensions 15×250 mm, and length limited to 500 mm. At the inlet to the channel constant gas velocity equal to 0.3 m/s and temperature equal to 800 °C were assumed. The set of simulations was carried out for different composition of the inlet gas in order to check how particles behave within the narrow channel. The walls of the channel were kept at constant temperature equal to 800 °C. Constant ambient pressure was assumed at the channel exit. The flow was treated as laminar, as the Reynolds number was low (Re<500). Radiation of the gas has been neglected due to very short radiation path length and isothermal walls. Since the particle size was very small (around 100 μ m) its temperature reaches very quickly that of the gas. Thus the irradiation of the particle by the walls can be ignored. Fig. 2 shows the expected particle trajectories, assuming that the particles diameter was equal to 100, 106, 125, 140 μ m. Two gas compositions were considered: 6% and 12% of oxygen. Since the numerical tests provide only gualitative information about the particle trajectory, it was arbitrarily assumed that char particle contains 80% of carbon and 20% of ash. The kinetic-diffusion limited char combustion model was used [7,8] with ANSYS Fluent default coefficient for low volatile coal.

The particle mass and temperature changes are tracked by solving the mass and energy balance equations for individual particle. The particle mass changes during combustion process can be described as

$$\frac{\mathrm{d}m_p}{\mathrm{d}t} = \frac{\mathrm{d}m_{\mathrm{char}}}{\mathrm{d}t} \tag{1}$$

where m_{char} are the mass of the char in combustible particle. The convective heat transfer between surrounding gases and the particle is taken into account by solving particle energy balance equation

$$m_p c_p \frac{dT_p}{dt} = A_{\text{ext}} h \left(T_f - T_p \right) + Q_c \tag{2}$$

where c_p and T_p stands for the particle heat capacity and temperature respectively, T_f is the temperature of gases inside a cell where the particle is located, h denotes the heat transfer coefficient, and A_{ext} represents the particle surface area, Q_c represents the heat released in the surface combustion processes. The char combustion process was modeled using one heterogeneous reaction.

$$C(s) + O_2 \to CO_2 \tag{3}$$

The trajectories presented in Fig. 2 show two regimes of particle motion: char combustion and ash free fall. The curved part of trajectory represents char combustion while the straight line trajectory corresponds to ash free fall. As expected, small particles burn



Fig. 3. Gas horizontal velocity profiles positioned on horizontal planes at different distances from top of the measuring channel.



Fig. 4. Predicted gas and particles velocities at short distance from feeding port.

faster than the bigger do. Similar tendency can be observed for two levels of oxygen concentrations. As expected for 12% of oxygen in the carrier gas, the burning process is faster than for 6% of oxygen. The differences are large enough to be seen later in the physical experiment. The vertical spread of trajectories also gives rise to the definition of the observation area in inspection window and placement of char particle injection ports.

Fig. 3 shows the gas horizontal velocity profiles corresponding to horizontal cross sections at 5, 10, 50 and 100 mm from top of the measuring channel.

Low Reynolds number flow generates parabolic velocity profiles. At 5 and 10 mm these profiles are different, but at 50 and 100 mm they are the same meaning that the velocity profile is fully developed. This can also be seen in Fig. 4, where the horizontal velocity profile (see line with x crosses) of the carrier gas is plotted on vertical plane positioned in the center of the channel.

Fig. 3 shows also particle spread area in the horizontal plane. The feeding system supply particles through a drill shape feeder. As a result, the velocity of some particles entering the channel are not vertical. The tilt angle was selected randomly between 0 and 30°. Such assumption produced particles horizontal spread not exceeding 1.6 mm.

Fig. 4 shows the predicted gas and particles velocities close to the feeding port. Low Reynolds number caused the development of flow boundary layer that is visible on the graph. Horizontal gas velocity component increased from zero at the wall to stable value of 0.53 m/s at around 2.0 cm from the top wall. Two carrier gas velocity profiles are shown: first at the channel center and second shifted by 0.8 mm of the center plane. This is the distance bounding the spread of the particles injected to the channel.

The horizonal component of the particle velocity is plotted for two sizes of the particle 100 μ m and 120 μ m. The 100 μ m particles reached gas velocity after less then 2 cm of the vertical fall from injection point whereas 120 μ m particles moved with gas velocity after slightly more then 3 cm of the vertical fall. Afterwards, particle horizontal velocity component is equal to the horizontal gas velocity.

2.3. Experimental setup

The experimental rig is illustrated in Fig. 5. Electric heaters are used to heat up and maintain a constant temperature of the oxidizer flowing in the channel. The total length of the heating section is 2.05 m. The maximum heating temperature of each module cannot exceed 1100° C. The cross section of the gaseous slot is set to 15×240 mm. The heating temperature of each heater is controlled by external heating controllers. The heating module with observation window is



Fig. 5. Experimental rig.

equipped with fuel feeding rail cooled by an external water circuit, which prevents uncontrolled particle ignition in the coal injection channel. The feeding rail consist of 30 ports, which are evenly distributed in 25 mm distance. In current rig design the position of the coal feeder can be manually changed. To ensure an appropriate particle feeding rate and particle dispersion, a custom coal feeder has been designed. It consists of funnel, step motor, regulated screw, system for controlling step motor velocity and motor step division to ensure smooth particle feeding rate. The gas flow through the rig during experiments, was controlled by gas mixing system. The system gives possibility to control flows of two gases mixture O_2-N_2 or O_2-CO_2 using *Bronkhorst* flow controllers. They give the possibility to accurately regulate the flow in the range from 0 Nm³/h to 2 Nm³/h.

In order to record particles trajectories, one of the module is equipped with observation windows on both sides of the channel. One window is used for backlight illumination and on the other side a CMOS camera is used for recording the trajectories. The last channel segment is only insulated. The outlet of the channel is entirely open to the atmosphere. The heating modules located, are equipped with fuel feeding ports, used to deliver char particle to the rig. To prevent heat losses through the observation window, additional quartz barriers were placed in observation gap. To record the particle trajectories, a Vieworks VC-4MC-180 camera link equipped with a CMOS sensor and a maximum resolution of 2048×2048 was used. Nevertheless, the interesting field of view (FoV) should be very small to capture particles, a special optical system had to be used. In the presented application a ZEISS lens Apo Sonnar with a high focal length (135 mm) and small lens distortion (0.3%) was used. To ensure a good sharpness quality of the images, the camera was placed at 800 mm from the rig. In consequences the FoV observed by the camera is equal to 25×25 mm. The camera frame rate in this configuration reaches 183 Hz. The brightness of burned particles is enough to saturate the camera sensor, but additional treatments were required for tracking black particles (inert or ash) carried by the inert gas (N₂). In presented application a LED backlight installed at the opposite side of the observation module (see Fig. 5) was used to ensure appropriate illumination during the recording of the inert particle trajectories over a short exposure time. Important aspect during in-plane measurement is the depth of field, which for the used camera-lens configuration is 4 mm. This parameter is used by the particle tracking algorithm described in work [25]. To ensure smooth and accurate control of the camera against the observation window it was installed at the positioning system. The camera processes (recording, controlling), camera positioning and the gas mixing system were fully integrated through the in-house control application written in LabVIEW software.

An in-house procedure whose aim was both to track the particle position and the particle diameter has been used in this study. The detailed information about this code are included in work [25], here only a brief description is provided. This software is capable of tracking a number of individual particles in subsequent snapshots, using time steps that are lower than in standard Particle Image Velocimetry. The mathematical model used for calculating combustion rates uses both the position of the particle and its diameter. While the first data can be obtained by analyzing the recorded frames in subsequent time steps, the measurement of the diameter requires a more sophisticated approach. The original algorithm developed to deal with this question counts the pixels of the image of the particle recorded by the monochrome CMOS camera. The dimensions of the particles are proportional to the number of pixels covered by particle, as it is shown in Fig. 6. The dimension defined by horizontal and vertical distances H and V (see Fig. 6) is far enough from the particle. Close to the border of the detected particles, the intensities are lower than the saturation intensity. The reason for this is that, the limits of the pixels do not exactly correspond to the border of the particle.



Fig. 6. Graphical illustration of the procedure used for detecting particle size and area.

As a result, external pixels are illuminated partially by the burning particle and partially by the background. Similar phenomenon referred to as *Halo effect* has been described in [26] where dust particles were traced. Due to their small size and high resolution of the camera, in the experiments conducted in Igathinathane et al.[26] no additional treatment of the results were required. To determine the size of the particle, the partial illumination of the external pixels has to be accounted for. The surface area of the particle is obtained by summing up all pixels covering the particle. When the information about particle area and perimeter are known the particle sphericity can be calculated.

2.4. Evaluation of the mass loss of the char particle

The trajectories of the particles during experiments are recorded using the CMOS camera. At every time instance the camera takes a snapshot of traveling particle and an in-house application retrieves the particles positions in time. The simulated trajectory is then fit to the measured positions of the particles. An inverse procedure is applied to determine the kinetics data for which the simulated particle trajectory matches best the measured one. The inverse problem is an iterative optimization loop within which, the search for parameters (decision variables) is modified, so as to produce minimum discrepancy between the model and the measurements. The model equations, also known as the direct problem are solved at each step of the iterative loop.

2.4.1. The direct problem

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In order to obtain the particle trajectories, i.e. the functions $x_p(t)$ and $y_p(t)$ a force balance is formulated, where t is time and x_p and y_p are the horizontal and vertical positions of the particle, respectively. The functions $x_p(t)$ and $y_p(t)$ can be calculated by solving

$$\frac{\mathrm{d}x_p}{\mathrm{d}t} = w_{\mathrm{x},p} \tag{4}$$

$$\frac{\mathrm{d}y_p}{\mathrm{d}t} = w_{y,p} \tag{5}$$

The velocities $w_{x,p}$ and $w_{y,p}$ can be obtained from the force balances in *x* and *y* direction, which are written as follows

$$\frac{\mathrm{d}w_{x,p}}{\mathrm{d}t} = \frac{18\mu}{\rho_p d_p^2} C_d \frac{\mathrm{Re}_p}{24} (w_x - w_{x,p}) \tag{6}$$

$$\frac{\mathrm{d}w_{y,p}}{\mathrm{d}t} = \frac{18\mu}{\rho_p d_p^2} C_d \frac{\mathrm{Re}_p}{24} (w_y - w_{y,p}) + \frac{\mathrm{g}(\rho_p - \rho)}{\rho_p} \tag{7}$$

where ρ and μ are the density and dynamic viscosity of the carrier gas, ρ_p and d_p are particle density and diameter, C_d is the drag

coefficient, w_x and w_y are gas velocities in x and y direction, g is the gravitational acceleration and Re_p is particle's Reynolds number defined as

$$\operatorname{Re}_{p} = \frac{\rho d_{p} w_{p}}{\mu} \tag{8}$$

where w_p is the relative velocity of the fluid with respect to particle velocity (either $||w_x - w_{x,p}||$ or $||w_y - w_{y,p}||$). It is assumed that the particles are spherical and the drag coefficient is determined using the correlation by Morsi and Alexander [27]. The vertical gas velocity w_x is assumed to be 0 at all times. The horizontal gas velocity profile w_y is determined from a CFD simulation of the flow in the reactor performed for the same conditions as in the experiment. Furthermore, at the current stage of the work, it is assumed that the rate of change of particle density due to reaction is given by

$$\frac{\mathrm{d}\rho_p}{\mathrm{d}t} = -\frac{\mathrm{6}R_c}{d_p} \tag{9}$$

where R_c is the reaction rate constant to be determined. Set of ordinary differential Eqs. (4)–(9) is solved using an explicit Runge-Kutta (4,5) method, the Dormand-Prince pair, implemented in MAT-LAB [28] (ode45 solver) with the following initial conditions $x_p(0) = 0$, $y_p(0) = 0$, $w_{x,p}(0) = 0$, $w_{y,p}(0) = 0$, and $\rho_p(0) = \rho_{p,0}$.

2.4.2. Formulation of the inverse problem

The reaction rate constant R_c entering Eq. (9) can be determined by solving the following minimization problem

$$\min_{R_c} F = \min_{R_c} \sum_{i=1}^{N} w_i \left[(x_{p,i}(R_c) - \bar{x}_{p,i})^2 + (y_{p,i}(R_c) - \bar{y}_{p,i})^2 \right]$$
(10)

where the $\bar{x}_{p,i}$ and $\bar{y}_{p,i}$ are the experimentally determined mean particle positions at *N* points and w_i is the weight calculated as a ratio of the number of trajectories used to determine the mean position at point *i* to the total number of recorded trajectories. The minimization is accomplished using the Levenberg-Marquardt method [29,30] implemented in MATLAB [28].

3. Sample results

The methodology presented in this work was tested using bituminous coal where the coal composition is provided in Table 1. Before the exact measurement produce could be run the coal has to be appropriately prepared in order to ensure that only char combustion process will be observed. The raw coal particles primarily consist of water, ash, volatile matter, and char. Presence of water influences on the particle ignition delay, where water evaporation process changes the gas temperature in vicinity to the particles. Presence of volatile matter within particles injected to the rig influences on particle swelling which as a consequence leads to particle breakage into a number of smaller particles. Small particles could not be effectively tracked by the tracking application [25] because the number of pixels covered by them is too small to evaluate their size and they will be rejected by the tracking procedure. Moreover, the gases released form particle causes fumigation of the observation window which is an undesirable effect. The sample preparation was divided into few steps, firstly the moisture associated with coal was evaporated, secondly the volatile matters were volatilized from the particles. This process was run in constant temperature (1000°C) and in the presence of inert gas. In the last stage the char particles are intensively milled to produce char particles and sieving them to ensure a narrow range of the particle size injected in the experimental rig.

Table 1

Raw ultimate and proximate coal composition used in experiments.

Prox., %	Ulti., %
Ash 12.3	C 58.21
Water 11.0	H 4.00
Vol 30.43	S 1.52
Char 45.9	N 0.91
	0 12.06

Measurements of the particle trajectories for four O_2/CO_2 mixtures ($O_2 = [9,12,15,18]$ vol.%) at gas temperature $T = 1000^{\circ}C$ have been taken. Analysis of the data using the inverse procedure described above produced the reaction rate constant R_c for each case. In Fig. 7 a fitted particle trajectory to the mean experimental data collected for gas mixture $18\%O_2/82\%CO_2$ is presented. The recorded combustible particles trajectories are marked by red dots. It can be seen that after a certain time, the predicted trajectory becomes a straight line which corresponds to a point where all carbon content has been consumed and the particle becomes non-reacting (ash) as it can be seen in Fig. 7.

The retrieved reaction rate constants R_c for the analyzed oxidizer compositions are equal to 1.6×10^{-2} , 1.75×10^{-2} , 2.21×10^{-2} , and 3.5×10^{-2} kg/m²s, respectively. As expected the reaction rate constant increases with increasing oxygen concentration and the increase is non-linear. More experimental tests, as well as their detailed analysis will be carried out in the research program to obtain results for other conditions and solid fuel chars.

4. Summary

The experimental test rig for evaluating the diameter and mass loss of burning pulverized coal char particles has been developed. The char particles motion is driven by gravitation and drag of the carrier gas, which corresponds to combustion conditions similar to that of industrial boilers. The proposed method relies on the analysis of the particle trajectories in carrier gas of controlled composition. The falling particle trajectory is acquired by a high speed monochromatic camera. The curvature of the trajectory results from particle mass loss due to combustion. From the shape of that trajectory, the mass loss is retrieved. Employing an inverse analysis, the reaction rate constants can be determined. To achieve the post-treatment, an



Fig. 7. Trajectories of combustible and inert particles tracked by algorithm with mean value and fitted function.

original technique of determining the diameter of the particle from their low resolution images has been developed.

The design process of the rig has been supported by a series of CFD simulations for different configurations and physical parameters. The final shape of the developed installation is shown. Preliminary results reproduce quantitatively the reaction rate constants of burning char particle.

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