A method of retrieving char oxidation kinetic data from reacting particle trajectories in a novel test facility under oxy combustion conditions

W. Adamczyk[a], R.A. Białecki[a], M. Ditaranto[b], N.E.L Haugen[b], A. Katelbach-Woźniak[a], A. Klimanek[a]*, S. Sładek[a], A. Szlęk[a], G. Węcel[a]

[a]Institute of Thermal Technology, Silesian University of Technology
Konarskiego 22, 44-100 Gliwice, Poland
[b]SINTEF Energi A.S., Sem Saelands vei 11, 7034 Trondheim, Norway

*Corresponding author:
Adam Klimanek, adam.klimanek@polsl.pl, tel.: +48 32 237 2974

ABSTRACT

In this paper, a new method of retrieving char combustion rates is presented. The method is based on an observation that trajectory of a freely falling and reacting char particle in a horizontal laminar flow changes due to change of its mass. Non-burning particles follow straight lines whilst burning particles bend due to mass loss. Since the deviation from the non-reactive particle trajectory depends on the rate of change of mass, it was postulated that recorded trajectories can be used to determine the rate constants of mass loss. An experimental facility was designed and built at the Institute of Thermal Technology, which allows recording reacting particles' trajectories and determining oxidation kinetics. In this procedure a model of a freely falling particle in a laminar horizontal flow was developed. The model comprises a set of ordinary differential equations that predict a particle trajectory for given rate constants. The trajectory predicted by the model is then fitted to the measured trajectories by changing the rate constants. The best fit corresponds to the mean rate constants. The inverse procedure was developed in the MATLAB software. The experimental facility and measurement procedure are briefly described. This is followed by a description of the applied inverse procedure. Measurement results for various O_2/CO_2 mixtures for two polish coal chars are then presented.

1. INTRODUCTION

Improvement of existing combustion chambers and development of new combustion techniques requires understanding of the elementary processes occurring during fuel conversion. The understanding should be accompanied with reliable data that can be used to predict the system behaviour at the various conversion stages. In recent years, numerical modelling of combustion processes by means of computational fluid dynamics (CFD) tools became an integral part of the design and optimization process of existing combustion systems. The models used in the simulations, require closure approximations, which are based on experimental data. In order to improve the predicted system behaviour, the quality of the data should be high and the closure models themselves should be fast, robust and accurate. Kinetic parameters of drying, devolatilization and char gasification/combustion, together with ignition characteristics, are

crucial elements of simulations of combustion processes. It should be stressed that the experimental data, from which the kinetic parameters are obtained, should be determined at relevant process conditions. Specifically the temperature, the heating rate, the thermal history, the pressure and the atmosphere are of importance [1,2]. Drop tube furnaces (DTF) are already standard devices used to retrieve such data. In recent years, many studies were devoted to characterization of solid fuels. These include ignition characteristics [3-7], kinetic parameters of devolatilization [1,2,8,9], combustion [10-13] and gasification [14-17]. The conditions achievable in drop tube reactors are close or the same as in the real pulverized coal systems. Thermogravimetric analysis (TGA) is also frequently used to obtain kinetic data. It is however restricted to lower temperatures (below 1000 °C) due to the inability of observing the real diffusion restrictions at high temperatures. At lower temperatures combustion and gasification rates are controlled by chemical reactions at the solid fuel particle surface. Therefore, TGA is frequently used in combination with a DTF to determine fuel characteristics at low and high temperature limits [15,16]. The investment and operating costs of a DTF are relatively high and the measurement procedures are time consuming. In this paper a new concept experimental facility, that could reduce the overall cost and measurement time, while retaining the important characteristics of DTF, is presented. Preliminary results obtained for the combustion of two polish coal chars in O2/CO2 mixtures are presented.

2. THE EXPERIMENTAL FACILITY

The experimental facility is based on an observation that the trajectory of a freely falling particle in a flow changes due to changes of the particle's mass. This obvious behaviour can be visualized by comparing trajectories of burning and non-burning char particles introduced to a laminar horizontal flow, as shown in Fig. 1.

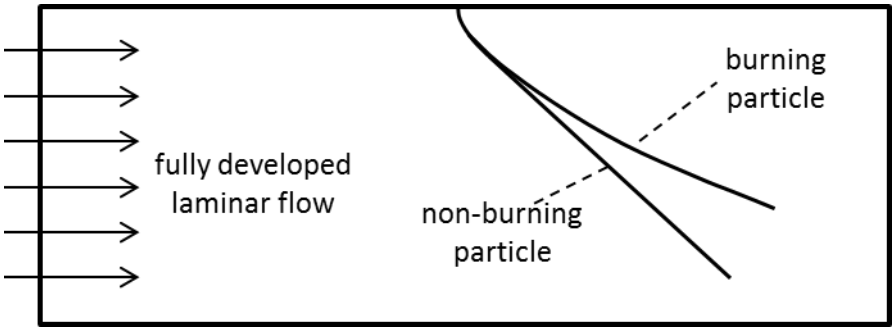

Fig. 1. Behaviour of non-reactive and reactive particle falling in laminar flow field

The trajectory of a burning particle deviates from the trajectory of constant-mass particle due to its mass loss. It is anticipated that the change of particle trajectory can be used to determine the mass loss of the reacting particle. This can be achieved by recording reacting particles' trajectories by means of a high-speed camera and retrieving their mass changes in an inverse analysis. These postulates are being verified at the Institute of Thermal Technology in Gliwice,

Poland, where an experimental laboratory scale facility was designed and built. The furnace is a rectangular duct in which hot gas flows horizontally and the particles are introduced through fuel injection ports at the top wall of the duct. This idea is presented in Fig. 2, where the dimensions of the duct are shown as well.

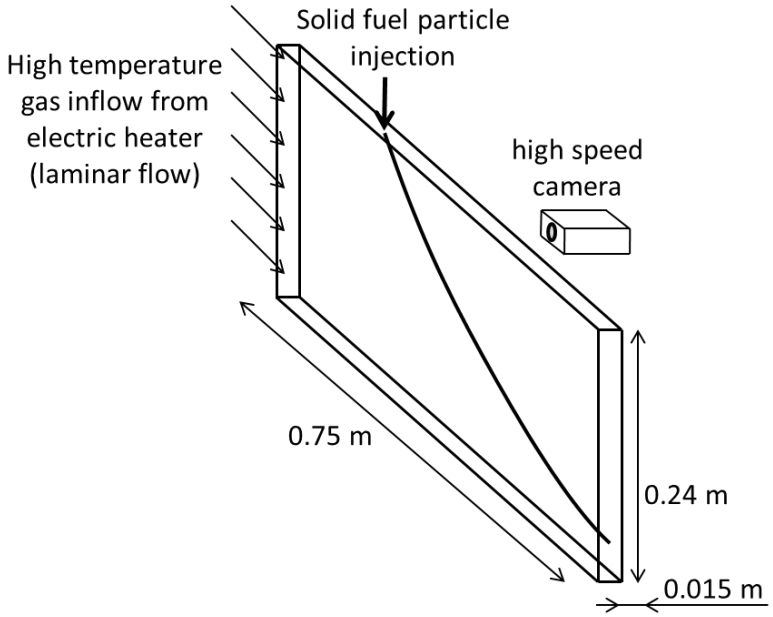

Fig. 2. Schematic diagram of the experimental facility

The walls of the furnace are built out of quartz slabs and are separated by a 1.5 cm gap. The quartz slabs are surrounded by electric heaters and insulated, allowing a constant temperature to be maintained in the furnace. Particles are visualized by a CMOS camera situated in front of a vertical observation window, as visualized in Fig. 3. During the experiment, the fuel feeding system can be moved and the pulverized char particles can be supplied from various ports. At the same time the

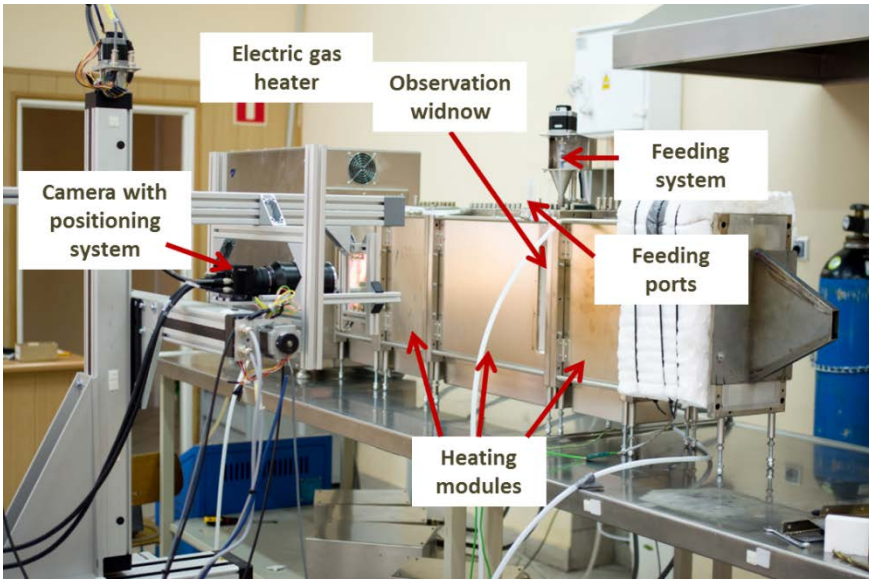

Fig. 3. The experimental facility

camera can move in the vertical direction so that a large portion of the furnace space can be observed. The gas flow is controlled by two mass gas flow meters for preparing oxidizers of desired compositions. The gas can be heated up to 1400 °C. The length of the duct is selected such that a fully developed laminar flow is obtained at the char injection region.

3. RETRIEVING THE COMBUSTION RATES

During the experiment, the particles' trajectories are recorded using the camera and an in-house application retrieves the particles positions in time. At each camera position, a number of particles are recorded. Although the char particles are carefully prepared and sieved to obtain a narrow size distribution, their shapes as well as their composition varies slightly, influencing their trajectories. Since the combustion rate should represent a mean reaction rate representative of the whole sample, a mean trajectory representing the whole sample is also desired. This mean trajectory is obtained by averaging the recorded trajectories released from a single port. Therefore, the final data representing the mean trajectory comprises a set of a few discrete points. The data can then be fitted by a model. The model comprises a set of ordinary differential equations. In order to obtain the particle trajectories, i.e. the functions $x_p(t)$ and $y_p(t)$ (the horizontal and vertical positions of the particle respectively as a function of time), a force balance is formulated. The force balance accompanied with the definitions of particle velocities form the following set from which $x_p(t)$ and $y_p(t)$ can be calculated

$$\frac{dx_p}{dt} = u_p \tag{1}$$

$$\frac{dy_p}{dt} = v_p \tag{2}$$

$$\frac{du_p}{dt} = \frac{18\mu}{\rho_p d_p^2} C_d \frac{Re_p}{24}(u - u_p) \tag{3}$$

$$\frac{dv_p}{dt} = \frac{18\mu}{\rho_p d_p^2} C_d \frac{Re_p}{24}(v - v_p) + \frac{g(\rho_p - \rho)}{\rho_p} \tag{4}$$

where u_p and v_p are the horizontal and vertical particle velocities respectively, μ is dynamic viscosity of the gas, g is the gravitational acceleration, ρ_p and ρ are particle and gas densities respectively, d_p is particles diameter, C_d is the drag coefficient, and Re_p is the particle Reynolds number defined as

$$Re_p = \frac{\rho d_p u_s}{\mu} \tag{5}$$

where u_s is the relative velocity of the fluid with respect to particle velocity. The drag coefficient is determined using the correlations by Morsi et al. [18]. The vertical gas velocity *v* is assumed to be 0 at all times. The horizontal gas velocity profile $u(y)$ is determined from a CFD simulation of the flow in the reactor performed for the same conditions as in the experiment. At the current stage of the work, it is assumed that the rate of change of particle density due to the reactions is

$$\frac{d\rho_p}{dt} = -\frac{6R_c}{d_p} \tag{6}$$

where R_c is the reaction rate constant to be determined. If R_c is known, solving the above set of equations allows the particle trajectory to be determined, i.e. $x_p(t)$ and $y_p(t)$. The discrete mean particle trajectory known from the experiment (i.e., $\bar{x}_{p,i}$ and $\bar{y}_{p,i}$ as a function of time) can now be used to determine R_c by solving the following minimization problem

$$\min_{R_c} F = \min_{R_c} \sum_{i=1}^{N} \omega_i \left[\left(x_{p,i} - \bar{x}_{p,i}\right)^2 + \left(y_{p,i} - \bar{y}_{p,i}\right)^2 \right] \tag{7}$$

where N is the number of discrete mean trajectory positions and ω_i are weights calculated as the ratio of number of measured particle trajectories contributing to the computation of the mean at point i to the total number of trajectories. Solution of the set of ordinary differential equation (1)-(6) is accomplished using the explicit Runge-Kutta (4,5) and the minimization is realized using the Levenberg-Marquardt method implemented in MATLAB.

4. RESULTS

Two coal chars were examined using the new facility; hard coal derived char from Janina coal mine and lignite derived char from Turów coal mine. Both chars were prepared at 1000 °C and sieved. The particles retained at sieve opening of 100 μm and passing through 106 μm were used. The coal chars were then introduced to the furnace of the rig and oxidized in various O2/CO2 mixtures with O2 = {9,12,15,18} % (vol.). The oxidizer was heated to 1000 °C and the temperature in the furnace was kept constant. During the experiments, the trajectories were recorded and analysed. In the analysis the mean positions were determined, which were then used in the inverse problem to retrieve the reaction rate constant R_c using the approach described in the previous section. The result obtained for the hard coal Janina at 9 % O2 (vol.) is presented in Fig. 4, where the recorded trajectories, their mean values in each injection port and the fitted trajectory predicted by the model are presented. The position (0,0) corresponds to the particle injection point. The resulting reaction rate constant for the data presented in Fig. 4 is $R_c = 0.039$ kg/m^{2}s. As can be seen, the trajectory predicted by the model fits well with the averaged positions of the recorded trajectories. It can also be seen from the figure that at the beginning the particle falls vertically and then starts to follow the gas flowing from left to right. The reason for this behaviour is the presence of the laminar boundary layer of the gas near the top wall. Furthermore, it is is apparent from the figure that after a certain position (close to 0.22 m) the modelled trajectory becomes a straight line, which corresponds to the flow of the inert ash. Although the analysed char particles were selected from a narrow range of diameters, the inhomogeneity of their shape and composition (ash content in particular) are the reason for the large observed scatter of the recorded trajectories. Since the trajectory is fitted in the weighted least square sense the contribution of each mean position to the fit is different. In Fig. 5 the reaction rate constants for all the analyzed cases and both coal chars are presented. A growth of the reaction rate constant is observed with increasing oxygen content. The growth is however larger for the lignite derived char. The rate constant for the lignite char is larger than that for hard coal.

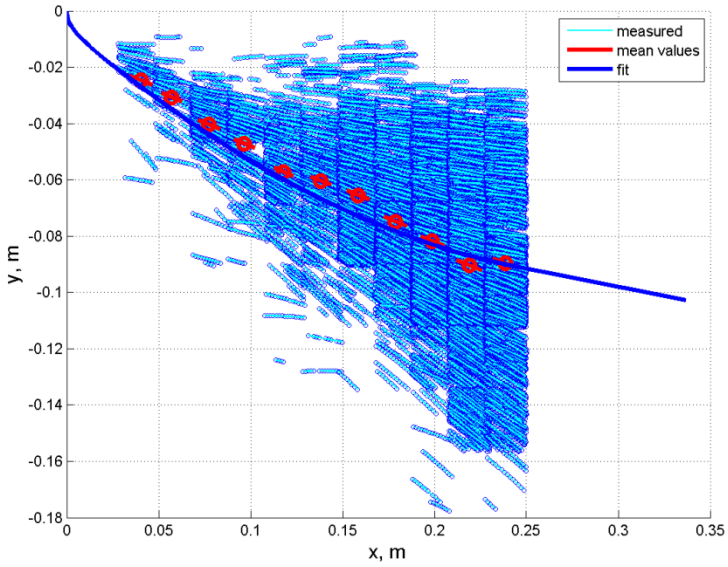

Fig. 4. Recorded trajectories (measured), their mean values in each injection port and the fitted trajectory predicted by the model

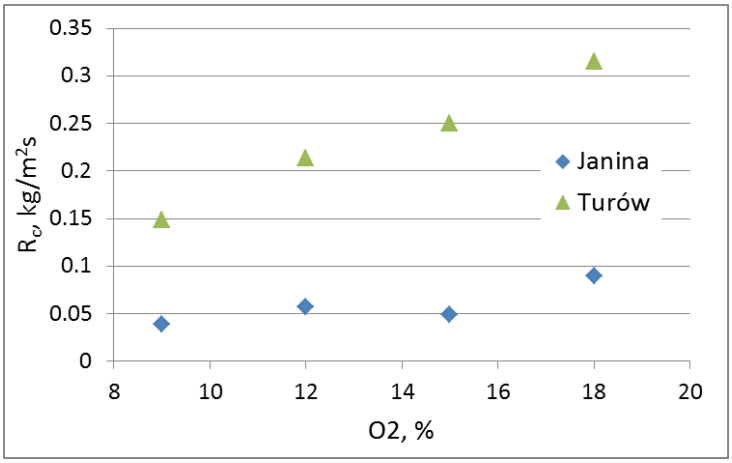

Fig. 5. Char combustion rate constants at various O2/CO2 mixtures for the Janina hard coal and Turów lignite

5. CONCLUSIONS

The new experimental facility designed and built at the Silesian University of Technology was briefly described in the paper. The aim of building the facility was to verify the concept of retrieving coal char burning rates from recorded reactive char particle trajectories. The methodology required developing a model for predicting particle trajectory of reactive particle together with an inverse procedure that allows fitting the modelled trajectory to the experimentally determined mean particle positions. The code was then used to determine burning rates of a hard coal and a lignite coal char in various O2/CO2 mixtures at 1000 °C. The reaction

rate of the lignite coal char was observed to be approximately 3 times larger than for the char from the hard coal. A linear increase in reaction rate with increasing oxygen concentration was observed for both chars. The results are therefore intuitively coherent, however a more detailed comparison with other experimental methods is required. Further work will be devoted to a sensitivity study of the model and inclusion of a more detailed rate expression.

ACKNOWLEDGEMENT

The research leading to these results has received funding from the Polish-Norwegian Research Programme operated by the National Centre for Research and Development under the Norwegian Financial Mechanism 2009-2014 in the frame of Project Contract No Pol-Nor/232738/101/2014.

REFERENCES

[1] Biagini E., Tognotti L., Fuel Processing Technology, 126 513 (2014)
[2] Authier O., Thunin E., Plion P., Schönnenbeck C., Leyssens G., Brilhac J.-F. and Porcheron L., Fuel, 122 254 (2014)
[3] Zhang D.-K. and Wall T.F., Fuel, 73:7 1114 (1994)
[4] Wall T., Gupta R., Gururajan V., Zhang D.-K., Fuel, 70:9 1011 (1991)
[5] Essenhigh R.H., Misra M.K. and Shaw D.W., Combust. Flame, 77 3 (1989)
[6] Khatami R., Stivers C. and Levendis Y.A., Combust. Flame, 159 3554 (2012)
[7] Shaddix C.R. and Molina A., Proc. Combust. Inst., 32:2 2091 (2009)
[8] Septien S., Valin S., Dupont C., Peyrot M. and Salvador S., Fuel, 97 202 (2012)
[9] Jones J.M., Pattersona P.M., Pourkashanian M., Williams, Arenillas A., Rubiera F. and Pis J.J., Fuel, 78 1171 (1999)
[10] Wang G., Zander R., Costa M., Fuel, 115 452 (2014)
[11] Bejarano P.A. and Levendis Y.A., Combust. Flame., 153 270 (2008)
[12] Rathnam R.K., Elliott L.K., Wall T.F., Liu Y. and Moghtaderi B., Fuel Processing Technology 90:6 797 (2009)
[13] Zhang L., Binner E., Qiao Y. and Li C.-Z., Fuel, 89 2703 (2010)
[14] Wang Y. and Bell D.A., Fuel, 140 616 (2015)
[15] Umemoto S., Kajitani S. and Hara S., Fuel, 103 14 (2013)
[16] Kajitani S., Hara S. and Matsuda H., Fuel, 81:5 539 (2002)
[17] Harris D.J., Roberts D.G. and Henderson D.G., Fuel, 85:2 134 (2006)
[18] Morsi S., Alexander A., An investigation of particle trajectories in twophase flow systems, J. Fluid Mech. 55:2 193 (1972)