

<http://dx.medra.org/10.7424/jsm130303>

Received: 2013.04.08 | Accepted: 2013.09.24 | Available online: 2013.10.28

CASE STUDIES INVESTIGATING SINGLE COAL PARTICLE IGNITION AND COMBUSTION

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Abstract

Studies focused on single fuel particles are designed to provide direct and unbiased information regarding the combustion process. The resulting data is primarily used to create and/or validate mathematical theories and models of the combustion process. The use of a single coal particle as a research object was first initiated over 40 years ago and nowadays is still one of the most important stages in a number of fundamental coal research techniques. Such experiments are especially important in the context of modern concepts that are now under development for new, sustainable and environmentally neutral coal processing technologies.

Article summarizes a broad spectrum of research methodologies, which were created in the recent history of single coal particle studies and motivated by the need to develop knowledge for new, clean coal technologies. The purpose of the experiments presented herein was to find the most comprehensive examination of the processes, where coal particles undergo changes at high temperatures. This objective in the case of coal combustion technology generally boils down to the characterization of particle ignition phenomenon and sub-stages of particle combustion. However, recent data presented by different research groups is still not always in agreement even when describing the same investigated issue. These differences often result from the shortcomings of the study methodology itself, which our article also attempts to highlight and analyze.

Keywords

single particle, clean coal technology, fundamental research, combustion

1. INTRODUCTION

Investigation of coal dust ignition began in the 19th century, when coal had already been fairly widely used for power generation purposes. The main motivation for undertaking such research concerned safety issues – those associated with the usage of coal dust, mainly due to its high explosiveness. The first results, obtained experimentally, determined the so-called relative ignition temperature, the values of which had been reported in a very wide spectrum, ranging from 500 to 1000°C. Hence, scientific research had been mistrusted by power engineers, because results of experiments greatly contradicted the principle arising from industrial experience – that the temperature of air leaving coal mills cannot exceed 70°C – due to the hazard of coal dust explosion. The questioned research data, mainly its disparity in relation to the reality was later justified by scientists, who attributed it to the scale effect of laboratory experiments, when compared to boiler conditions (size and construction of the reactor, coal dust stream, type and power of the igniting agent, measurement uncertainty, etc.)

An explicit definition of the measured parameter constituted the most important issue for researchers. The relative ignition temperature appeared to be an unclear parameter,

dependent mainly on the conditions of the conducted experiments and the interpretation of the researcher. A proper explanation of the ignition theory also required the development and verification of a theoretical approach, including mathematical models, which were not created until the 1930s. On the one hand it was the result of insufficient knowledge regarding the fuel itself, while on the other experienced researchers considered the theoretical investigation of ignition irrelevant in real measurement conditions (Essenhig, Misra, Shaw 1989).

A theoretical bases of coal ignition began to form on par with the creation of radical reactions and thermal equilibrium concepts in the 1930s. One of the consequences of the proposed combustion theories was the change of perspective regarding the method of conducting experimental research. The necessity to verify these theories implied a need to reduce the complexity of experiments, what was limited by the fact of placing a single coal particle as an object of research. The first studies on coal particles examined independently of each other and burnt in a tube reactor were introduced by Cassel and Liebmann in 1959 (Cassel, Liebman 1959). The idea of investigating single fuel particles was then developed by Essenhig and co-workers (Chen, Fan, Essenhig 1985;

Brooks, Essenhigh 1988; Seixas, Essenhigh 1986). Until the late 1990s many other concepts of studies regarding single coal particles were developed, of which the experimental results have turned out to be substantially compliant with the findings anticipated in theoretical calculations [1–2, 4, 6–8, 14, 17, 19, 22, 24, 26, 36, 40, 44]. This in turn has encouraged other researchers to continue studies on single coal particles, also in view of issues not related to the phenomenon of coal ignition. Currently, these experiments are among some of the most fundamental research studies on coal. If a need arises for the development of a new technology regarding coal utilization (the currently applied co-firing or oxy-fuel combustion), one of the key experiments, expanding the scope of available knowledge on coal, is the study on single coal particles (Chen, Yong, Ghoniem 2012). This paper attempts to compile the latest achievements in this type of research, the results of which extend the knowledge needed for the development of clean coal technologies.

2. COAL PARTICLE IGNITION AND COMBUSTION THEORY

One of the first theories developed within the field of studies on coal, was related to the ignition of coal particles and the process of combustion. It has been observed that, depending on temperature conditions, partial pressure of the oxidant, the type of coal and the size of the particle – when considering the mechanism of ignition, the ignition of a single coal particle can occur:

- homogeneously
- heterogeneously
- simultaneously hetero-and homogeneously

The first of the considered mechanisms – homogeneous ignition – occurs as a result of pre-ignition pyrolysis of the particle and then the ignition of released volatile matter (Faraday mechanism). This case applies to considerably large particles – with a diameter over 100 μm , which are heated sufficiently slowly, i.e. particle heating rate is less than 100°C/s (Juntgen, Van Heek 1979). Homogeneous ignition mechanism is observed usually during the combustion of hard coals with a high content of volatile matter. Such ignition takes place, if the temperature of the mixture consisting of released volatile matter and oxygen exceeds the temperature of self-ignition. The jet of gaseous products released from the particle transfers the reaction zone beyond the particle surface, thus protecting it against the direct attack of oxygen (Tang et al. 2010).

Heterogeneous mechanism occurs as a result of a direct attack of oxygen on the coal particle. This type of reaction can occur in the case of smaller coal particles (< 100 μm), very rapidly heated to the temperature of ignition or in the case of coke/char particles, deprived of volatile matter.

Hetero-and homogeneous ignition boils down to the simultaneous and competing occurrence of both the mentioned mechanisms and can appear in the case of a very wide spectrum of particle sizes and heating rates (Golec 1989).

Heterogeneous, as well as mixed type: hetero- and homogeneous ignitions are characteristic of the combustion process of hard coking coals and brown coals. The oxidation of the solid surface of coal can (but not necessarily) be accompanied by the disintegration of particles into smaller fragments

(Katalabmula et al. 1997; Mühlen, Sowa 1995; Wendt et al. 2002).

A graphic presentation of the three above ignition mechanisms depending on particle diameter and heating rate is shown in Fig. 1.

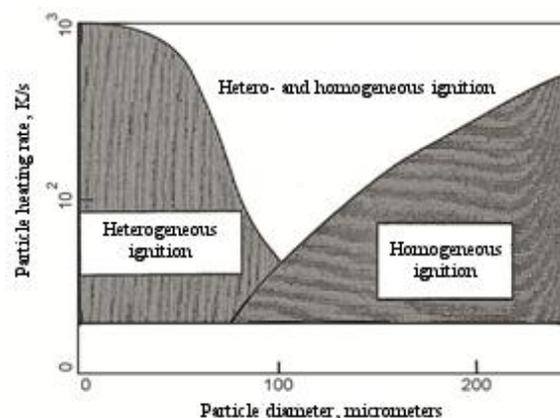


Fig. 1. Ignition mechanism as a function of particle heating rate and particle diameter (Juntgen, Van Heek 1979)

3. RESEARCH METHODS OF SINGLE COAL PARTICLE COMBUSTION PROCESSES

The use of a single particle in studies on coal combustion was supposed to provide experimental data to prove and/or to verify the correctness of theoretical concepts of the combustion process. Nowadays, this data is also utilized for the numerical modeling of coal dust combustion processes. From the point of view of a designer of a combustion chamber, it is vital to know the time of total burn-out of coal particles and the amount of heat emitted during combustion (Golec 1989). As it was suggested by Tang and others (Tang et al. 2010), a full understanding of the thermal processes taking place in a coal particle is also very important in the construction of gassifiers. Besides, this knowledge is still very desirable for reasons of safety and contributes to the prevention of uncontrolled coal dust explosions.

The chief advantage arising from the study of isolated particles is the elimination of the influence of other particles from the considered research setup. In a cloud of burning coal dust the particles compete for oxygen. Due to that, the combustion mechanism can be different in the case of particles inside the cloud, where the oxidant concentration is negligible than in the case of particles in the outer layer of the dust cloud, where the effect of struggle for oxygen is the weakest. Additionally, the temperature of the burning group of particles is higher than of a single particle, resulting in an increase in the devolatilization rate and the following oxidation reaction rates.

Coal particle combustion experiments are intended to determine one or more of the below parameters:

- coal particle ignition temperature
- change of particle internal temperature during the combustion process
- change of particle surface temperature during the combustion process
- determine the combustion mechanism in the given experimental conditions (homogeneous, heterogeneous, hetero-homogeneous combustion)

- particle burning time as the function of particle size against changing temperature conditions and/or varying gas composition
- the moment of particle ignition (defined as the time delay from the start of the experiment to the appearance of the flame or from the start of the experiment to the moment when particle temperature exceeds the ambient temperature)
- the time of particular stages of particle combustion – time of volatile matter combustion, time of carbon residue combustion
- change of particle size during combustion
- particle mass loss during combustion
- to determine kinetic constants for the combustion reactions in given measurement conditions

Most studies are dedicated to the determination of particle temperature during combustion, either on the surface as well as inside the particle. The reasons are stated by Joutsenoja (Joutsenoja et al. 1999):

1. The kinetics of carbon residue oxidation is strongly dependent on particle temperature, so the measurement of this parameter constitutes the key element for the determination of combustion reaction rates.
2. Around 2/3 of the heat generated in the process of combustion is emitted during the reaction of CO oxidation, whereas the measurement of particle temperature is required to determine the CO/CO₂ ratio.
3. Particle temperature is the most important parameter in the calculation of heat flow to/from the particle, both by convection, as well as by radiation.
4. Particle temperature can also be an important indicator in the identification of problems appearing during coal utilization, such as slagging and fly ash formation, and its deposition on the walls of the boiler.

It has been proved that the temperature of a burning particle is correlated with other examined parameters, such as particle diameter, mass loss, time of combustion, etc. Experimental determination of one of the mentioned parameters can be of use for the estimation of further ones (primarily particle temperature, if it was not measured directly) and ultimately for the mathematical presentation of the process of coal combustion.

The techniques used to conduct experiments with single coal particles differ to a great extent. These differences are mainly related to: the construction of the test stand, the temperature in the reactor, the method of heat transfer to the particle, the composition of the gas containing the oxidant, the method of measuring the examined parameter and the size of the single particle used. This review presents most of the latest methods of single coal particle studies and the most significant results of recent experiments. They have been grouped based on the methodology of the experiment and the construction of the reactor used.

3.1. Heated-grid reactor

The analysis of single coal particle combustion can be done with the application of a heated-grid reactor – HGA (heated-grid apparatus). The first laboratory stands of this type were created in the 1980s. Tests in heated-grid reactors are also still conducted today (Qiao et al. 2010).

In the HGA reactor, a single coal particle is placed on a metal grid. During the experiment, high voltage is applied to the grid, which results in its very fast heating – and at the same time, simultaneous heating of the coal particle.

The obtained heating rates reach 1200 K/s (Mühlen, Sowa 1995). This method makes the assumption that the temperature of the particle is equal to the temperature of the heated grid. As a direct implication, the definitions of ignition temperature and the moment of ignition, are related to the temperature of the heating grid and the moment, in which the flame or glow appears on the examined particle.

Studies on the combustion of coal particles using heated-grid reactors were undertaken by Mühlen and Sowa (Institute for Cokemaking and Fuel Technology, Essen) (Mühlen, Sowa 1995). The scheme of the reactor is shown in Fig. 2. The rig can be operated at elevated pressure, with the use of gases such as H₂, CO, N₂ and water vapour. Coal particles, size 0.5–0.8 mm were placed on the heated grid. Grid heating rate was set by a computer. The experiment was recorded with a high-speed camera.

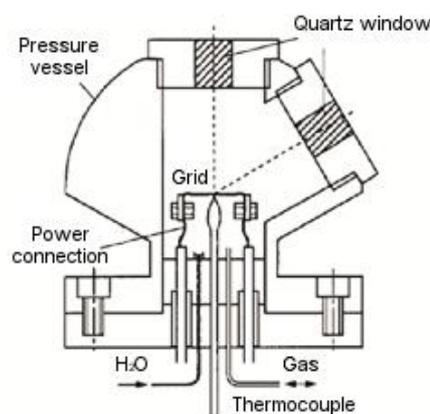


Fig. 2. Scheme of heated-grid reactor (HTG) (Mühlen, Sowa 1995)

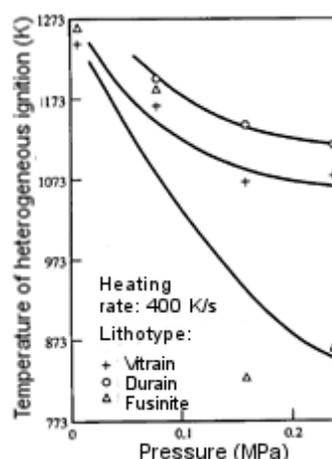


Fig. 3. Influence of gas pressure on ignition temperature for different coal ranks (Mühlen, Sowa 1995)

The main results obtained by Mühlen and Sowa pertain to research on the influence of oxygen partial pressure on the process of combustion – Fig. 3. The rise of the partial pressure of oxygen increases the burning rate and decreases the ignition temperature for every type of tested fuel. On the other hand, higher total pressure has an impact on the degassing of coal, obstructing the free outflow of volatile matter

from inside the particle. Thus, only heterogeneous ignition is observed at high pressure.

The main disadvantage of experiments with the application of a heated grid is the assumption that particle temperature is the same as the temperature of the grid. This might lead to considerable errors in the case of big particles. Besides, as in every optical method, the finding of the initial moment of ignition is limited to the observation of a particular plane of the particle surface, thereby the determination of ignition is possible only when the flame or glow becomes visible to the eye or the optical instrument being used. The influence of the grid on the course and initiation of combustion cannot be excluded. At relatively low heating rate, abrupt ignition of volatile matter can result in reversing the direction of heat flow. Reversed heat transfer from the particle to the grid retards the further progress of combustion, substantially influencing the process. Qiao and others (Qiao et al. 2010) in some way attempted to avoid this effect in their research on coke particles. They applied the flow of oxidizing gas perpendicular to the grid, in order to ensure the uniform distribution of temperature in the system.

Method with heated-grid reactor (HG) allows users to obtain very high particle heating rates, which constitutes this technique's greatest advantage. It is possible to conduct research with heating rates reached in commercial power plants. Furthermore, this parameter can be set very precisely in a very wide range, allowing researchers to perform comparative combustion studies on particles of specific sizes at different heating rates.

3.2. Pulsation tests of ignition

The idea of the pulsation ignition experiment was developed by researchers from Newcastle University in the 1980s (Wall et al. 1991). The test stand is an electrically heated tube furnace and some additional equipment. The experiment boils down to an injection of a small sample of fuel into the furnace (0.2–0.5 mg) and the observation of the appearance of a flash, interpreted as coal ignition. For every tested temperature of furnace, a series of repetitions of dust injections is carried out, further the share of successful experiments, i.e. those concluded with a flash, in the whole series of dust injections is calculated. The presentation of results is shown in Fig. 4. The fraction 63–90 μm of two fuels: coal with a high content of volatile matter and quick coke made out of it were used for the experiments. The diagram in Fig. 4 presents the range of temperatures, in which the frequency of light flashes increases from 0 to 100%.

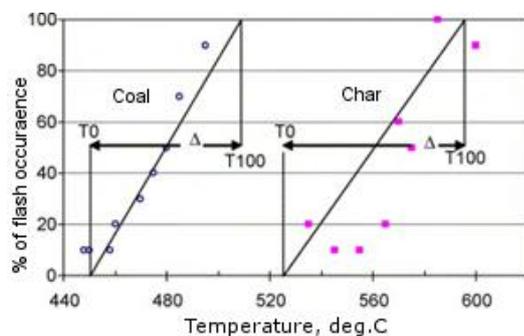


Fig. 4. Results from pulsation experiments carried out for highly volatile coal and char prepared from this coal (Gupta 2005)

The objective of the research, as stated by the authors - Wall and others (1991) and Gupta (2005), was the determination of the minimum ambient temperature required for the ignition of a single particle. However, during the experiment actually the ignition of the dust cloud was observed, hence such an explanation of the results might be questioned. Further to that, for the tested dusts the ignition appearance frequency curve in a certain temperature range is obtained – and, as admitted by the researchers themselves (Wall et al. 1991) – the interpretation of which temperature on the diagram is the ignition temperature can be arbitrary.

The indisputable advantage of the pulsation method of ignition testing is the uncomplicated construction of the test stand and the simple method of performing the experiment. The obtained results are difficult to interpret unambiguously in view of determining particle ignition temperature. The presented method might be useful, above all, for the qualitative comparison of different fuels ignition points. Nowadays, the pulsation method for ignition testing is not applied in studies on single coal particles.

3.3. Research at microgravity conditions

One of the recently identified problems manifesting itself during coal particle combustion is the so called “buoyancy effect” (Tang et al. 2010). Thanks to the force of gravity on the burning particle, the heavier and colder gasses shift towards the bottom of the particle, whereas the burning, hot and light volatile matter floats, forming a flame over the particle. The majority of the combustion models, created so far, neglect this “buoyancy effect”, assuming that the flame of burning gasses perfectly envelopes the particle. Nonetheless, as mentioned by Wendt and others (Wendt et al. 2002), the action of gravity, and as a result of it – the influence of natural convection on the combustion process – disturbs the transport of heat and mass around and within the particle, what in extreme conditions (e.g. in the course of burning of suitably small particles) can lead to blowing off the volatile matter cloud – away from the particle surface. Data provided by the experiments conducted so far, does not meet all the requirements of theoretical models, which leads to inestimable errors. Hence, in 2001 Ronney (2001) suggested the necessity for single particle combustion experiments at microgravity. As stated by Wendt (Wendt et al. 2002), the conditions of tests carried out this way are more idealized, thus closer to the assumptions of mathematical models. The ignition process then depends only on the temperature, pressure and composition of the gaseous atmosphere, shape, size and composition of the particle and on the radiation.

Studies aimed at comparing the ignition temperature of coal particles under normal gravity conditions (1-g) and at microgravity ($\mu\text{-g}$) were performed by Tang and others (2010). Tests have been done for hard coal and brown coal particles with a diameter of 1.5 and 2 mm. Fig. 5 shows a diagram of the construction of a test stand. The system consists of a tube furnace electrically heated to 1123 K, a device for introducing the particle, a CCD camera and a stand operation control. In the research conducted by Tang and others, the coal particle was glued on top of a thin quartz needle and could have been easily introduced/removed from the furnace by the use of a mechanical guide. The entire system was placed in a microgravity generating capsule – pro-

ducing microgravity up to 10^{-2} – 10^{-3} g. Videos recorded during the tests, through the use of visual analysis of the RGB colorimetric method, were used to determine the moment of ignition and the particle surface temperature.

The observations made at μ -g conditions allowed researchers to notice strong explosions of volatile matter, which were not observed in the series of tests carried out at normal gravity (1-g). The flame surrounding the particle at μ -g was more spherical, thicker and darker when compared to the flame produced at the same time from the moment of ignition in the 1-g experiment (Fig. 6). Apart from that, the analyzed visual data revealed that at μ -g conditions, the ignition of the particle happens at a temperature lower than at 1g conditions by ca. 50–80 K μ -g.

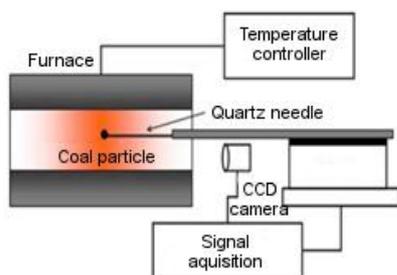


Fig. 5. Scheme of research rig with the possibility of microgravity generation (Tang et al. 2010)

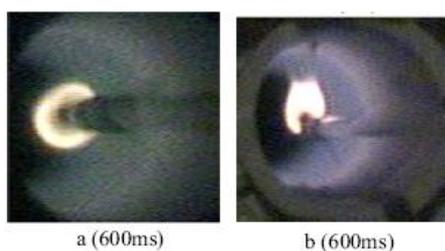


Fig. 6. Picture of burning particle in: a) μ -g, b) 1-g – conditions (Tang et al. 2010)

The characteristic feature of studies at microgravity is the possibility of minimizing the influence of the buoyancy effect on the particle combustion process. An attempt to eliminate the effect, which always accompanies heat exchange processes, seems to be purely scientific and useful only for currently created models. It should also be noted that these studies require complicated apparatuses, in particular, the μ -g generating capsule. A simpler and cheaper solution could be the development of models in such a way, that they would also take the buoyancy effect into account. Then, studies carried out at normal gravity conditions would be sufficient.

3.4. Research on single coal particle in a fluidized bed boiler

In a fluidized bed boiler, the single fuel particles are comparatively convenient research objects, provided that appropriate measurement methodology is ensured. In a fluidized bed boiler, fuel particles are separated from each other by the bed material and can be treated as single particles.

Due to the high temperature in the system, coal particles are seen as the brightest objects, contrasting with the darker background of the bed material. In studies carried out on single particles, the selection of the appropriate measurement

methodology, as well as ensuring proper optical access for the necessary instrumentation could be a problematic issue.

Joutsenoja and others (Joutsenoja et al. 1999) performed experiments in a laboratory scale reactor with a fluidized bed, located at the Institute for Cokemaking and Fuel Technology in Essen (Fig. 7). For the purpose of particle temperature measurement, the authors used a pyrometric method with optical fibre for the collection of measuring signals. The key advantage of the application of a pyrometer with optical fibres is the lack of interference of the measuring system with the combustion process, which – as suggested by the authors – can take place while using contact sensors. Besides, a pyrometer is capable of measuring the temperature of smaller particles, than i.e. a thermocouple. Similar merits can be attributed to the measurement technique implementing a camcorder/camera, yet in this case only the objects ‘visible’ in the foreground (top of the bed) are measured. On the other hand the usage of a thermocouple sensor allows researchers to investigate the whole process of particle combustion, starting from its heating, whereas in the pyrometric technique the particle becomes ‘visible’ only after reaching the minimum temperature of the pyrometer calibration range. The thermocouple also does not require a suitably big difference of temperature between the background (reactor) and the particle, which is essential in the case of measuring the radiation signal, emitted by the examined objects.

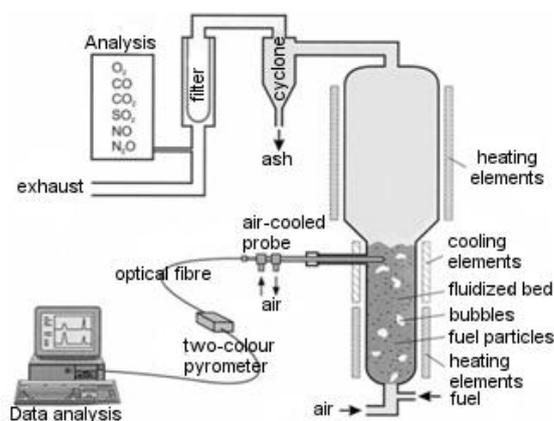


Fig. 7. Scheme of research rig with application of pyrometer and fiber optics for particle temperature measurements in fluidized bed reactor (Joutsenoja et al. 1999)

The objective of the studies, carried out by Joutsenoje and others was to find the correlation of pyrometric measurements of particle temperature with its size (diameter), for particles smaller than 1.2 mm. During the experiments, the temperature in the boiler as well as oxygen concentration was controlled.

The Statistical Pyrometric Sizing of Particles (SPPS) – numerical method, proposed by (Heino, Hernberg, Stenberg 1997), has been applied for the simultaneous measurement of temperature and the diameter of particles. It involves the determination of the so-called parameter X, connected directly to both of these values. Parameter X itself depends on the difference of temperatures between particles and the bed, ‘visibility’ coefficient and the tested emissivity, divided by the optical fibre diameter.

The results obtained by Joutsenoje and others (Joutsenoja et al. 1999) indicate that increased oxygen content in the

measuring system results in the observation of a greater number of particles with temperature considerably higher than bed temperature (200 K difference). In the same way – though to a smaller extent – the temperature of particles is affected by a rise in the bed temperature.

The results prove that the hottest particles (temperature higher than the temperature of reactor by 300 K) have a diameter less than 0.4 mm, whereas particles with lower temperature have wider particle size distribution. The occurrence of very hot particles has also been discovered (temperature difference 400–600 K), but their number was not statistically significant.

Studies on single particles in fluidized bed reactors are relevant mainly for coal utilization technology. For example, experiments conducted at the Czestochowa University of Technology focused on the study on phenomena characteristic for fluidized combustion, such as the erosion of single particles in contact with bed material and the resulting change of their internal structure (Pełka 2009). Some of the developing measurement techniques implemented for the first time in fluidized bed reactors, like the pyrometric method for the determination of particle temperature and diameter, introduced by Joutsenoje and described herein, are increasingly being adapted at other test stands, also in the case of pulverized coal combustion experiments for coal fractions in the range of 50–80 μm (Khatami, Levendis 2011).

3.5. Research in batch type reactor

A batch type reactor was elaborated at the Royal Institute of Technology, KTH in Stockholm.

The results of studies on single coal particles have been presented by Ponzio and others (Ponzio et al. 2008).

Actually, the authors have used a pellet produced from pulverized and pressed coal dust, assuming it as a single coal particle. This pellet was of a cylindrical shape, with dimensions of 35 \times 15 mm and weight of ca. 6.85 g. Prior to the experiment, the prepared sample was attached to a piston by means of a thin metal wire (Fig. 8). When the reactor was properly heated, the piston was lowered manually, resulting in the sample introduction to the furnace (the pellet was suspended in the reactor on a wire). At any time of the experiment, the pellet could have been drawn out and weighed, which provided data on mass loss during the combustion of coal. Ignition temperature was also measured, it was evaluated at the moment of flame appearance on the pellet surface, as the difference of temperatures between the two thermocouples located at the pellet's two ends. The progress of the experiment was recorded by a camera.

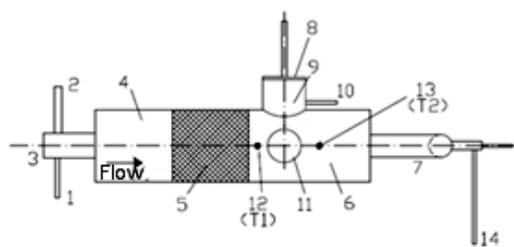


Fig. 8. Batch type reactor in KTH (Ponzio et al. 2008)

(1 – propane inlet, 2 – air/O₂ inlet, 3 – burner, 4 – combustion chamber, 5 – ceramic filler, 6 – reactor chamber, 7 – gases outlet, 8 – lid with a piston, 9 – cooling chamber, 10 – quenching gas inlet, 11 – observation window, 12, 13 – thermocouples, 14 – gas sampling)

The authors have carried out research for varying oxygen content in the range from 5 to 100%, for different reactor temperatures: 500, 800 and 1000°C. Owing to that, it has been observed that the ignition temperature of the tested pellet varies on par with the changing temperature and concentration of the oxidant: the ignition temperature drops if the oxygen content in the mixture increases or if the gas temperature rises (Fig. 9). Thanks to recorded films, the authors have also defined the ignition mechanism under different measurement conditions. The samples, which were ignited with a visible flame at a high reactor temperature and a low concentration of oxidant, have been identified as undergoing the homogenous ignition mechanism. At a high concentration of oxidant and the high temperature at which the experiments were conducted, ignition with many visible sparks was observed, which was interpreted as the heterogeneous ignition mechanism. The same mechanism was assumed in the case of experiments at low reactor temperature and low oxygen content, when ignition was only observed as a glowing of the sample.

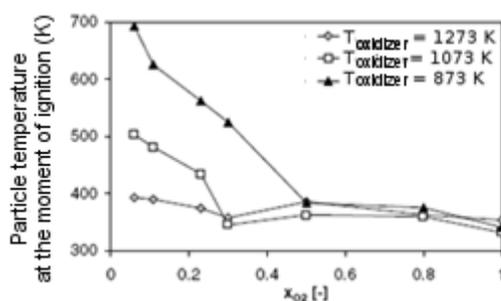


Fig. 9. Particle temperature at the moment of ignition as function of the oxygen content at varying experimental temperatures (Ponzio et al. 2008)

The studies presented by Ponzio and others (Ponzio et al. 2008) are based on a risky assumptions concerning equating the ignition and combustion of a single particle with the ignition of a coal pellet. The application of thermocouples next to the sample, was aimed at avoiding the interference of sensors with the combustion process, but it can be also contributed to the increasing uncertainty of particle temperature evaluation. However, this method seems to be considerably simple and reproducible (big samples of coal). The acquired results made it possible for the authors to present the mathematical model depicting the correlation between ignition time of the pellet and the concentration and temperature of the oxidant.

3.6. Research on single coal particle in entrained flow reactor

An exemplary, relatively complicated test stand with entrained flow of coal particles is located at Sandia National Laboratories in the USA. Studies carried out within this stand are focused on the investigation of the size and temperature of burning coal particles and on measuring the degree of burn-out of particles after different residence times in the reactor. While applying oxygen-enriched atmosphere it is possible to define the combustion kinetics of coal particles (Khatami, Levendis 2011; Murphy, Shaddix 2006; Liu 2011).

The diagram of the Sandia flow reactor is shown in Fig. 10. Experiments can be performed at temperatures even as high as 2000 K. Gas mixtures used in the reactor are prepared

by means of a Hencken burner. Thanks to that, the hot gas atmosphere instantly heats the coal particles introduced into the reactor, simultaneously acting as an oxidizing medium in the conducted experiments. Composition of the gas mixture can be freely composed, for oxygen content up to 50%.

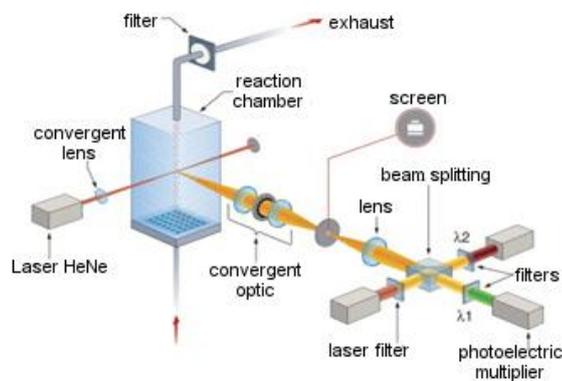


Fig. 10. Entrained flow reactor Sandia (Khatami, Levendis 2011; Murphy, Shaddix 2006)

Coal is introduced into the reactor from the bottom. Transport is provided with the aid of unheated nitrogen, which reaches the furnace temperature at the height of ca. 5 cm from the chamber inlet. Apart from heating the gaseous medium, this passage is also assumed to be sufficient for degassing and burning of volatile matter. The very small stream of fuel in the experiments – 1 g/h, is used to ensure the isolation of particles and as a result it can be assumed that the introduced particles burn independently from each other.

There are points of optical access for pyrometer measurements located along the reactor. The pyrometer, employed in the studies, is used for the simultaneous measurement of speed, the diameter and temperature of single particles at a selected height of the reactor. Measurement is triggered by means of a laser light.

Experiments described by Murphy and others (Murphy, Shaddix 2006) aimed at finding the comparison of coal particles' combustion in atmospheres with varying oxygen content. Gases with the following O_2 concentration were used – 6, 12, 24 and 36% in mixtures with nitrogen and water vapour – 14% and CO_2 – 4%.

The 106–125 μm fractions, obtained from two hard coals were tested. From recordings of the experiments, the authors reproduced their progress in a visual way by superimposing the photographs of burning coal particles – Fig. 11. The juxtaposition of images obtained during measurements – for different concentrations of oxygen proves that an increase of oxidant's content accelerates the degassing of particles. Besides, the more oxygen in the gaseous atmosphere, the light emitted by burning particles of carbon residue is observed in the lower part of the reactor. This implies a rise of particle temperature and increases the rate of particle combustion. These conclusions have been confirmed by temperature measurements taken simultaneously with the use of a pyrometer: in atmospheres with a low concentration of oxygen – 6% O_2 – the measured particle temperature amounted to ca. 1800 K, whereas the temperature of a particle burning in a mixture with 36% O_2 amounted to 2400 K. Further to that, data acquired as a result of optical measurements were utilized for the assessment of combustion kinetics of chars.

According to the authors, the results proved the kinetic controlled regime of combustion in oxidant-enriched atmospheres, in spite of the fact that from recorded particle temperatures it can be implied that the limitation of the combustion process can as well be attributed to the diffusion phenomenon. Summarizing, the works of Murphy et al. demonstrated in two ways that an oxygen-enriched atmosphere causes a considerable rise of temperature and the reduction of combustion time of coal particles.

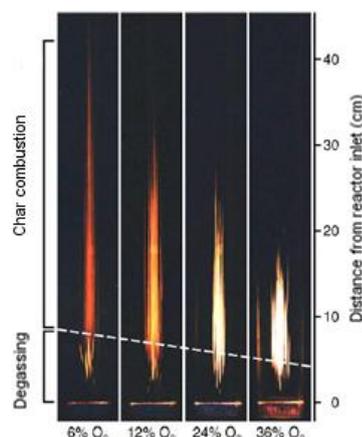


Fig. 11. Lapsed pictures from coal combustion experiments in Sandia entrained flow reactor. In each photograph, the oxygen concentration in each gas mixture is indicated. The dashed line suggests points of devolatilization end and the beginning of char combustion. The length scale in relation to the reactor inlet is placed to the right of the pictures (Murphy, Shaddix 2006).

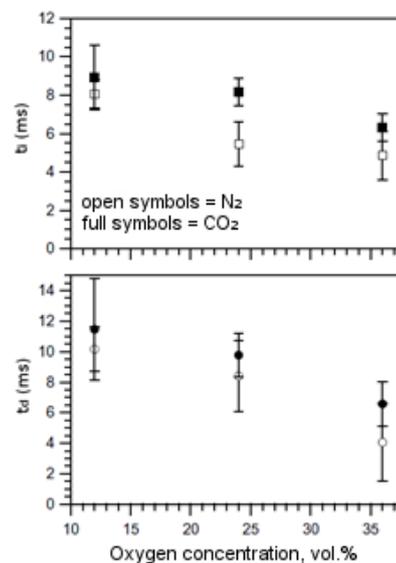


Fig. 12. Moment of ignition (top) and duration of volatile matter combustion (down) for bituminous coal in varying O_2 concentration (Shaddix, Molina 2009)

Similar experiments were conducted in the same reactor by Shaddix and Molina (2009). These were targeted at comparing the combustion of coal particles in an atmosphere containing nitrogen and CO_2 . In both types of mixtures the concentration of O_2 was altered in the range from 12 to 36%. Again, two hard coals were tested, fraction 75–105 μm . Experiments were carried out at the reactor temperature of 1700 K.

Results of this series of experiments in the flow reactor indicate, that the presence of CO₂ delays the ignition of coal particles and slightly extends the combustion time of volatile matter. The increased concentration of oxidant in the gaseous atmosphere accelerates the ignition of particle – in the mixture with N₂, as well as with CO₂ (Fig. 12). Changes were observed in the flame formed around the particle, which appears in the moment of volatile matter ignition. Ignition of hard coal particles with high content of volatile matter was accompanied by the generation of a hot cloud of soot particles, which was not noticed in the case of the other tested coal. The size and temperature of this cloud are strongly related to the concentration of oxygen and the use of N₂ instead of CO₂. It has been deduced that CO₂ retards the diffusion of released volatile matter, this results in a more difficult formation of a combustible gaseous cloud around the particle, and finally a smaller diameter of an observed flame and not particularly bright flame luminosity as it was observed for the flame formed in the N₂ atmosphere.

Studies undertaken at Sandia laboratories have great scientific potential and have provided quite a bit of valuable information concerning the combustion process of coal particles in modified atmospheres. However, it must be kept in mind that the presented flow reactor is well equipped with additional instrumentation, often technically sophisticated. The application of a pyrometer or camera (Shaddix, Molina 2009) in studies on very fine coal fractions implies the use of very sensitive devices, which in turn leads to obtaining noisy and often unclear experimental data. Analysis of these kinds of results requires that researchers use their many years of experience and knowledge of measurement methodology, including the processing of visual data acquired through experiments.

3.7. Particle ignition with free fall

The idea of creating a test stand, in which a free falling and burning coal particle would be the object of research, resulted from the lack of sufficient data concerning heat exchange between the particle and its close surroundings. Huang and others (Huang, Vastola, Scaroni 1998) constructed a stand, in which during experiments the falling particle went past several thermocouples, and thanks to that temperatures in the whole space of the measuring system were registered. The idea of the conducted experiments is shown in Fig. 13.

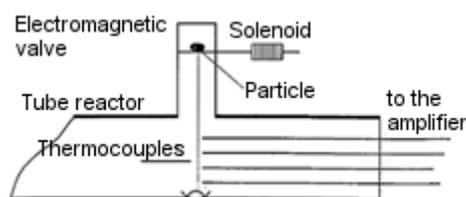


Fig. 13. Scheme of experiments carried out with coal particle freely falling on (Huang, Vastola, Scaroni 1998)

In Huang's and others' studies, coal and quick coke particles, with ca. 1 mm diameter, constituted the objects of research. In order not to disturb the path of particle fall, after the stand was heated to the suitable temperature, gas flow was turned off for the time of performing the experiment. The possibility of introducing nitrogen or air into the reactor,

enabled them to follow the temperature of changes during particle pyrolysis or in the course of its combustion.

Coal and char particles were used as tested objects in Huang et al. experiments. In order to not disturb the particle free fall and after the reactor was properly heated up, the gaseous flow was shut down at the moment of the experiment. The possibility of filling the reactor with N₂ or air allowed researchers to follow temperature changes during the conducted experiment of particle pyrolysis or combustion. Examples of results of the latter process are presented in Fig. 14. In the initial phase of the experiment, the particle heats up just after it is inserted into the reactor. At the same time, as a result of coal particle introduction, the temperature in the reactor in the surroundings of the particle drops – within the distance of 1.5 mm from the particle (point A). Next, the temperature measured at the particle surface sharply rises, exceeding the temperature in the reactor (point B). The researchers explain this rise as particle ignition and the formation of a flame enveloping the particle. As a proof, the findings of the tests with volatile-free char particles are quoted, where – after an initial drop in temperature, no rapid rise was noted, as between points A and B on Fig. 14.

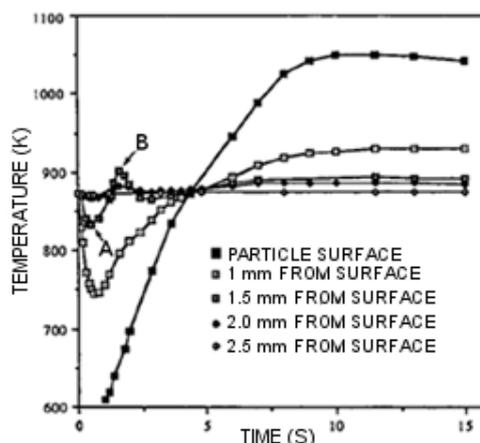


Fig. 14. History of temperature changes measured in the burning coal particle environment (Huang, Vastola, Scaroni 1998)

The reactor presented by the investigators from Pennsylvania makes it possible and easy to follow the history of combustion or degassing processes. However, it should be noted that even thin thermocouples have relatively large time-lag (signal inertia) in relation to the speed of the occurrence of such fast processes, like the combustion of coal. In connection with that, the experiment should guarantee the adequately long burning of particles and that the amount of heat released during this process will be sufficient enough to raise the temperature in the reaction space. This means that the measurement of free falling particles can be performed only in the case of considerably big particles [as in (Huang, Vastola, Scaroni 1998) about 1 mm].

3.8. Studies on falling particle in drop tube furnaces

Test stands with DTF (drop tube furnace) are considered to be the ones, which simulate the process and conditions of coal combustion in a way very much resembling the actual conditions of industrial pulverized coal boilers (Su et al. 2001). This explains the popularity of these stands and their quick adaptation and utilization in studies on single particles.

The main advantages of tube reactors are a high rate of fuel heating, 10^4 – 10^5 K and the possibility of obtaining very high temperatures in the reactor, up to 2000°C. Besides, the fuel is introduced into the furnace in a dynamic form of particles suspended in a gaseous medium. Thanks to that, after the selection of an adequate stream of the fed fuel, it is possible to carry out research on a single particle or on a cloud of dust (Su et al. 2001).

The laboratory tube furnace of 2 m length and 70 mm diameter is used for studies on coal particles in the Institute of Process Engineering and Power Plant Technology (IVD) in Stuttgart. In this reactor it is possible to conduct research on single coal particles also at increased pressure. In the work of Reichelt et al. (1998) the methodology applied in such types of experiments is described, as well as the findings from research conducted for two types of coal: hard coal and brown coal, fraction 160–250 μm . Fig. 15 shows a schematic diagram of the tube reactor located at IVD. In the studies presented in (Reichelt et al. 1998), the gas prior to being introduced into the furnace is initially heated up to the temperature of 1473 or 1773 K. The fuel is fed in a flux of nitrogen through a cooled probe. It is assumed that particles burn independently, suspended in the stream of carrying gas. The estimated number of particles in a volume unit of gas has been estimated as less than 3 particles per cm^3 . Through the side-ports of the furnace, the optical fibre of a two-colour pyrometer is inserted, which captures the radiation signal emitted by a particle passing through the reactor. Based on the magnitude of the measured signal, the temperature of the particle is determined. Knowledge of the temperature and signal of the pyrometer response leads to the determination of the cross-section of particle A_p and further to the calculation of the equivalent diameter D_p (diameter of an equivalent circle).

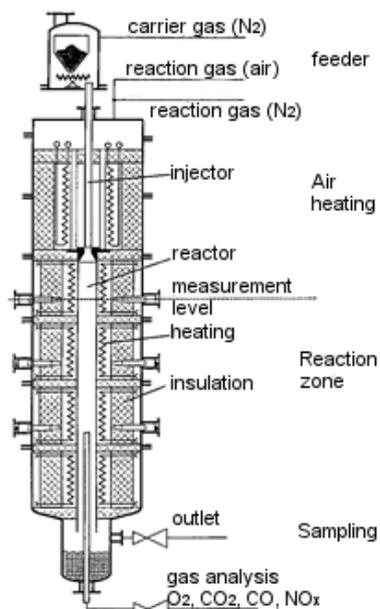


Fig. 15. IVD research stand for coal combustion investigation under elevated pressure (Reichelt et al. 1998)

Graphs presenting the measured parameters – particle diameter and its temperature (Fig. 16A and B) were drawn for the hard and brown coal and were tested in two temperatures

of gas 1473 and 1773 K. The obtained results gathered in two typical clouds. One of them comprised particles, which had the same temperature as the gas surrounding them – these particles did not react and were in equilibrium with their surroundings. In this group no correlation was found between the particle diameter and its temperature. The points in the second cloud, located higher, correspond to the particles which actually were burning when going past the visual field of the optical fibre. Only these particles were taken under consideration in the further analysis of data. On the graphs, the regression lines for the dependence: the diameter of the burning particle against its temperature are indicated. This correlation proved to be strongly negative in the case of brown coal (what means that particle surface temperature is smaller, if the particle is bigger), and weak or non-existing in the case of hard coal. However, in respect of the latter fuel, a wider particle-size distribution has been noticed, which evidently resulted from the plasticization of particles due to the increasing internal pressure on account of rising temperature (Fig. 16C). This effect was more evident in experiments conducted in lower gas temperatures. A similar plasticization of particles was not noticed in the case of brown coal particles. The common feature of findings in respect to both the tested fuels was, however, a significant fraction of particles with smaller particle-size distribution than the lower parameter of output coal screen ($< 160 \mu\text{m}$). This result can be explained by the burn-out of particles or the disintegration/explosion of particles during combustion. In consequence of particle explosion, the number of smaller particles in the reactor is increased [acc. to (Levendis et al. 2011) ca. 5 to 30 smaller particles are formed out of one big particle]. The influence of partial pressure of oxygen on the temperature of particles was also tested. Its amount was changed in two ways – through the change of total gas pressure in the reactor or by changing the concentration of oxygen in the mixture. The first method of increasing the partial pressure of oxygen has resulted in only a slight rise of the particles' temperature. On the other hand, the higher concentration of O_2 in the gas mixture caused a rapid rise of the particles' temperature (Fig. 16D).

Experiments conducted in drop tube furnaces in IVD provide interesting information concerning coal combustion under increased pressure. As the authors mention, first of all this data can be applied in newly developed technologies for pressure conversion of solid fuels. Nevertheless, as Chen and others state (Chen, Yong, Ghoniem 2012), the technology of high pressure pulverized coal combustion for power generation purposes seems to be very promising these days. In this case, the results given by Reichelt and others can be the source of direct information on the processes, which the coal particle undergoes in T-p conditions.

The tests with particles falling in a drop tube furnace were also carried out in Northeastern University in Boston. The latest publications from the scientists from Boston (Khatami, Levendis 2011; Levendis et al. 2011; Khatami et al. 2012) present experiments, which are part of a larger research project, focused on providing information regarding the coal combustion process in oxy-fuel combustion technology (Khatami et al. 2012).

The test stand at Northeastern University (Fig. 17) is an example of a short tube furnace, dedicated in the first place to

research on single coal particles. The hot part of the reactor constitutes only 25 cm of the tube, out of which 7 cm is visible to the observers – thanks to the application of a transparent quartz tube, fitted into the furnace. The interior of the

reactor is heated to a constant temperature of 1400 K by means of radiating incandescent molybdenum heating elements.

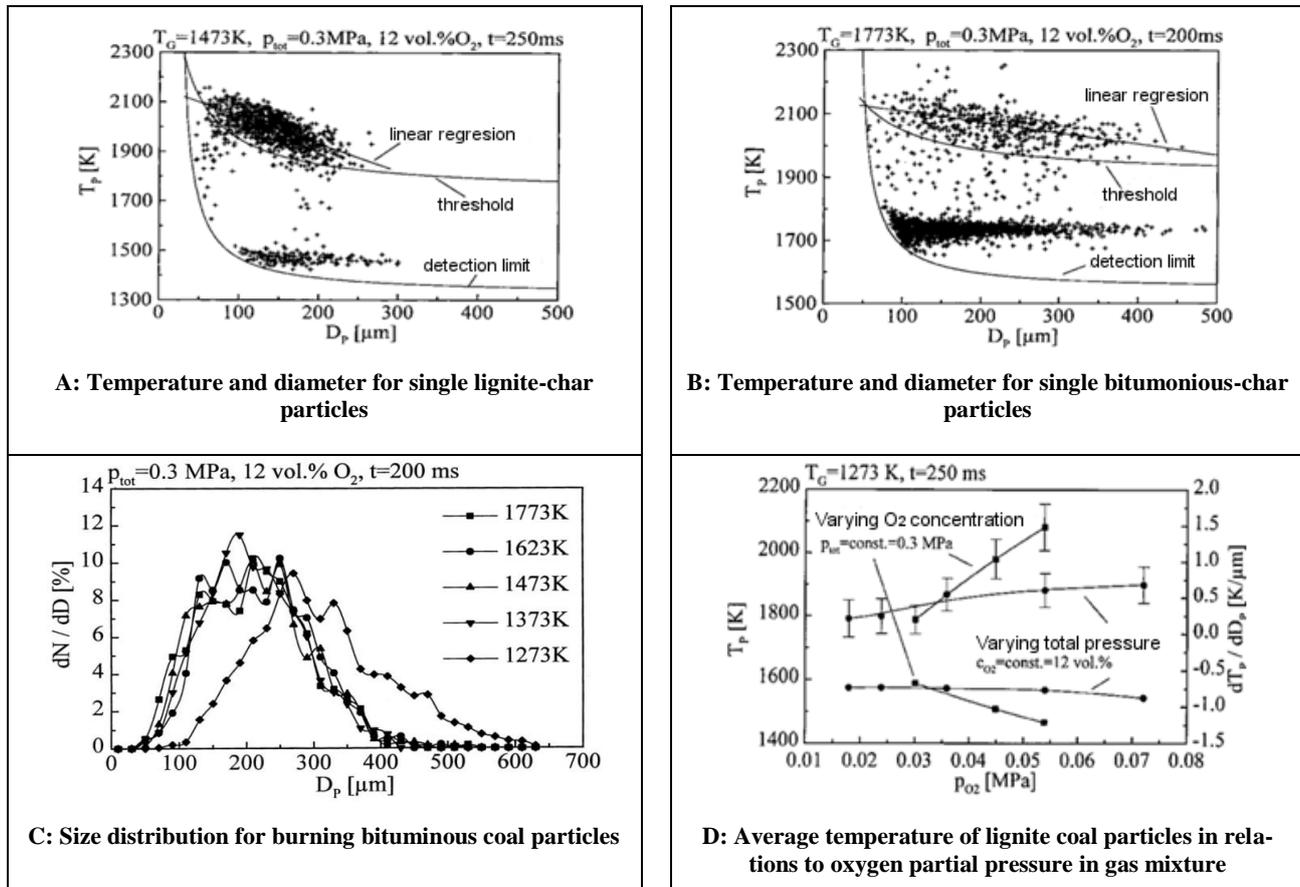


Fig. 16. Results of experiments carried out in DTF (Reichert et al. 1998)

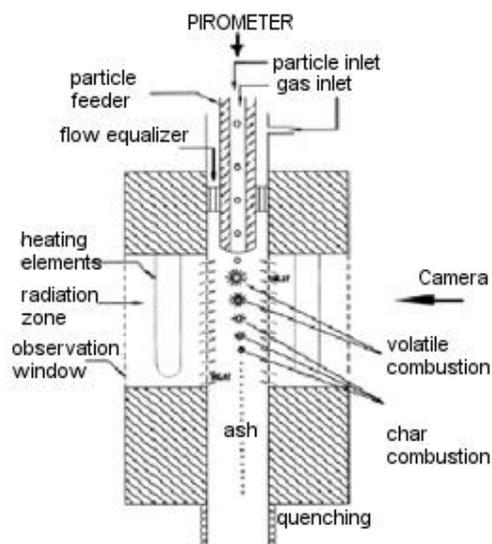


Fig. 17. Research stand for single particle investigation at Northeastern University, Boston (Levendis et al. 2011)

The fuel particle is first attached to a thin needle, which is placed in a particle feeder, located in the upper part of the furnace. Gas flows through the particle feeder to the furnace, and – after releasing the particle from the needle – acts as the

carrier of particles to the reactor space. Progress of the experiment is recorded with a high speed camera with a speed of 1000 frames/s. A three-colour pyrometer, located over the particle feeder is used for measuring particle surface temperature.

The work of Levendis and others (2011) focused on the comparison of the combustion of different types of fuel, carried out in air. The 75–90 μm fractions of the following fuels were tested: highly volatile hard coal, coking coal, brown coal and sugar cane bagasse. Thanks to the films, recorded with high speed, different types of combustion were observed for different fuels. Individual movie frames recorded for selected experiments are shown in Fig. 18.

At the beginning of the experiment, simultaneously with heating, gaseous components are released from highly volatile hard coal particles. Then volatiles ignite, enveloping the whole particle with a flame (Fig. 18A). According to the authors, there are many particles of burning soot in this kind of flame, because it is very bright, and its diameter is ca. three times bigger than the particle diameter. The movement of the particle down along the reactor causes the stretching of flame, which resembles to authors a falling meteorite. After the depletion of the gaseous volatile matter, long-lasting burning of char begins, which is observed as a very bright glowing of the particle. In the case of coking coal particles,

the occurrence of short-lived volatile flame was noted in just a few tests (Fig. 18B-i). Homogeneous combustion in these cases lasted a few milliseconds, and the formed flame was smaller and less bright when compared to the flame generated by highly volatile hard coal particles. After extinguishing the

flame, the fragmentation of the particle took place resulting with the formation of 5–10 smaller fragments, which burnt heterogeneously. In the remaining experiments (Fig. 18 B-ii), the particle disintegrated first, only after the ignition took place, individually for each of the smaller fragments.

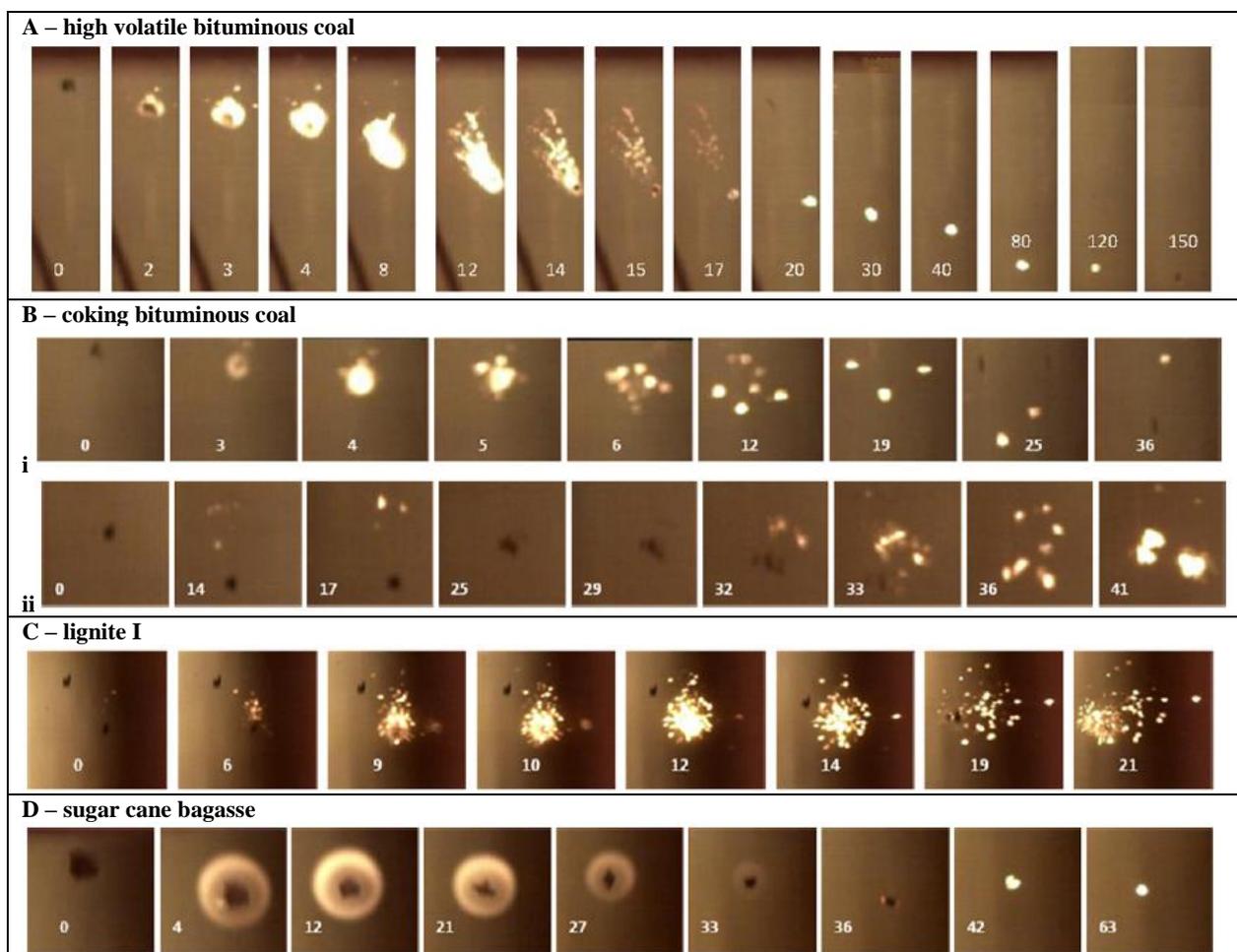


Fig. 18. Pictures from experiments of different fuels particle combustion in air. In pictures, time from the particle appearance in reactor is indicated, ms (Levendis et al. 2011)

The combustion of brown coal particles was distinguished because of the fast disintegration of the particles into a large number of smaller fragments that took place right after particle degassing began (Fig. 18C). The authors point out that this was not the effect of moisture evaporation from the particle, because the tested fuels were dried before the experiment. After ignition, ca. 50–100 different fragments of particles burning in a few millimeters, an almost spherical space was observed, which for the authors visually resembled fireworks. The fuel produced out of the sugar cane bagasse, as with every biomass fuel, contained a large amount of volatile matter. Combustion of a particle of such fuel, like in the case of highly volatile hard coal, took place in two stages. (Fig. 18D). At first, the released volatile matter burnt homogeneously forming a bright, spherical and quite transparent flame. Next, the char burnt heterogeneously, shrinking and brightly glowing. What is significant, the measurements done at the same time with a pyrometer did not detect any radiation signal emitted by the biomass flame. The authors attribute this to the lack of soot particles in such flames.

In another study (Khatami et al. 2012) researchers from Northeastern University presented the continuation of their research. This time, the aim of their work was focused on the investigation of combustion processes for four different coals and one biomass in modified atmospheres of O_2/CO_2 and O_2/N_2 . The research was done with the same experimental temperature and with the use of the same fuels, as before. Thus the results of both published works (Levendis et al. 2011; Khatami et al. 2012) and all of the conducted series of experiments can be compared.

The experiments performed by Khatami and others (2012) imply that the increase in oxygen concentration in the mixture with nitrogen results in the rise of a flame and burning char particle temperature. When the amount of O_2 was increased, the particles (for all the tested fuels) burnt more intensively, and the volatiles tended to ignite closer to the particle surface, than it was observed in air. Replacement of nitrogen in the gaseous mixture by CO_2 made the combustion process in the case of all the tested fuels less intense. At oxygen concentration below 30% in a mixture with CO_2 , the

majority of the tested particles did not even ignite (Fig. 19). The authors observed ignition in these experimental conditions (mixture: 27% O₂/73% CO₂) only in cases when the gas flow was stopped and the particles actually fell freely in the reactor. The rise of oxygen concentration in the mixture with

CO₂ increased the probability of ignition occurrence and contributed to more intensive (brighter) combustion. In spite of this, the presence of CO₂ usually led to the suppression of the volatile flame (observed in experiments with N₂) resulting with particle heterogeneous combustion.

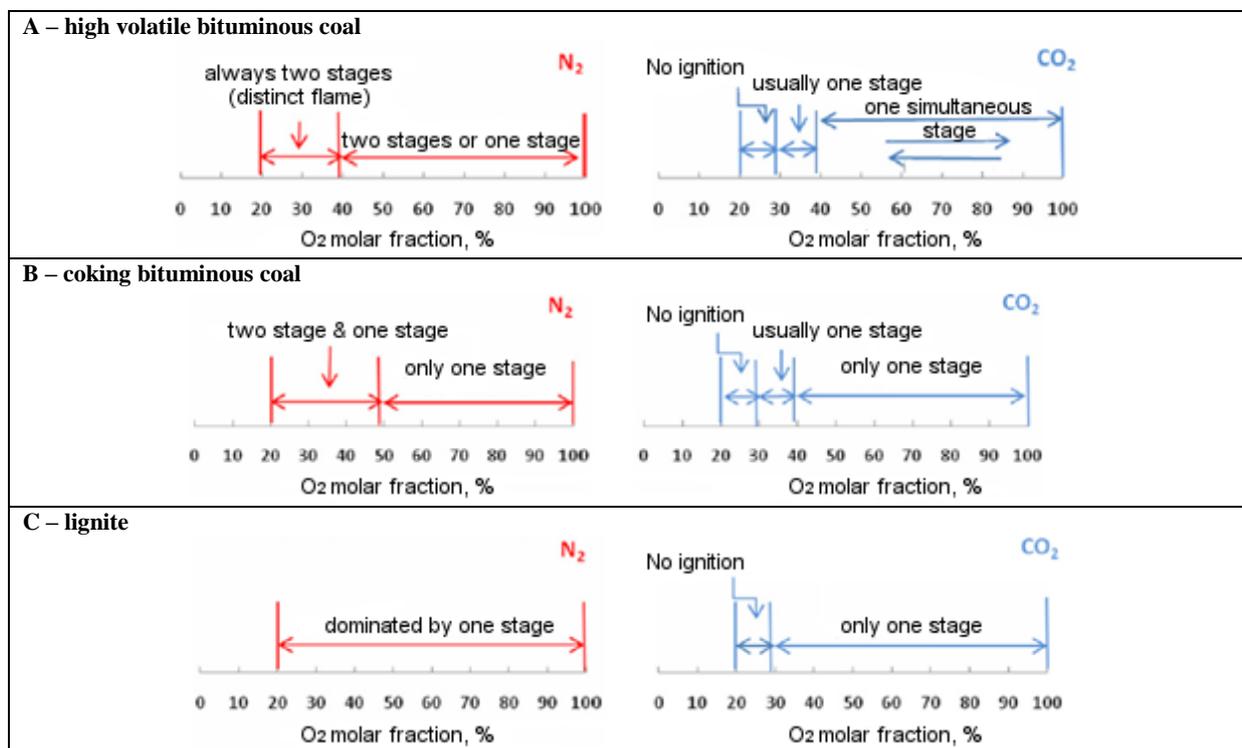


Fig. 19. Comparison of results from coal particles combustion in different gaseous atmospheres with specification of combustion mechanisms (two-stage combustion: homo- and heterogeneous, one-stage combustion: heterogeneous or no ignition) (Khatami et al. 2012)

In addition to valuable conclusions regarding the combustion of different types of fuel, the research done by Levendis and Khatami (Levendis et al. 2011; Khatami et al. 2012) shows that the measurement methodology used in studies on single particle combustion of different fuels should not be limited to just one measuring instrument. The results obtained from the pyrometer suggest that biomass fuel burns without the formation of a flame. An essential complementation of the collected data turned out to be the movies recorded with the high-speed camera. Thanks to these, researchers were able to precisely analyze the combustion process throughout the duration of the experiment. The records of particle temperature changes do not necessarily reflect the character of partial processes taking place during combustion.

The test stand with the drop tube furnace at Northeastern University in Boston, like in Sandia, the entrained flow reactor in Livermore, is a stand assigned for studies on single fuel particles. Owing to the application of a two- or tri-colour pyrometer and a high-speed camera, the findings of experiments conducted in two American research centers provide the most comprehensive information describing the processes occurring during coal particles combustion. Experiments performed and presented by Levendis and others (2011), as well as by Khatami and others (2012) have contributed to a better understanding of the differences between air and oxy-fuel combustion.

3.9. Test-stands with particle attached to a quartz needle

The first stand with particle attached to a quartz needle was constructed at the Wrocław University of Technology. The scheme of the research stand (Fig. 20) as well as the methodology of the performed experiments were presented by Karcz and Zembruski (1975). The research was continued later by Rybak (1981). Also currently, there are experiments conducted at the Wrocław University of Technology focused on the investigation of the single particle combustion process in oxy-fuel conditions.

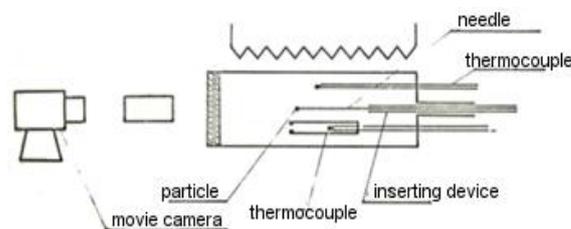


Fig. 20. Scheme of research rig at PWr with particle glued to quartz needle (Rybak 1981)

The furnace chamber, made of a ceramic tube of 30 mm diameter, is closed by a quartz sight-glass on one end, and by inserting a device used for introducing the needle with the particle, on the other. The tube is placed in an electric

resistant furnace with a maximum heating temperature of 1573 K. The coal particle is glued to the tip of the quartz needle with cement. Before the measurement, the needle is fixed into a piston type inserting device, moving along a guide. The progress of the experiment is recorded by a camera. Based on the movie, the particle temperature in the experiment is determined, with the use of the photopyrometric method (Karcz, Zembrzusi 1975; Rybak 1981).

Karcz and Zembrzusi (1975) tested the few petrographic varieties of brown coal. Their publication presents experiments carried out on about 1000 particles with diameters in the range of 90–1500 μm . The obtained results were used to determine the kinetic constants for tested fuels. From the series of 1000 experiments, the combustion time of char was isolated and then presented as a function of the particle size (Fig. 21). The authors indicate a distinct correlation between char combustion time and the petrographic composition of coal (Karcz, Zembrzusi 1975).

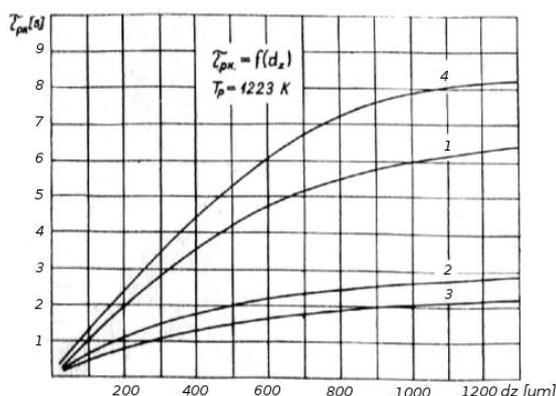


Fig. 21. Time of char combustion as a function of particle size at 1223 K experimental temperature (Karcz, Zembrzusi 1975):
1 – smudge coal, 2 – ‘piropissy’ lignite, 3 – xyloid coal, 4 – fusain

A similar test stand with the particle held on a quartz needle was designed at the Warsaw University of Technology (Fig. 22). Coal particle, with diameter of 0.5–1.5 mm was held on a quartz needle, which was placed between two electrically heated nets. Net heating rate was estimated at 10^3 K/s, with a maximum obtained temperature of 1373 K. Experiments were conducted in modified mixtures of oxygen and nitrogen. The process of particle combustion was recorded by a photomultiplier. Research performed at the mentioned stand was presented by Golec (1989). The objective of his study was to investigate the heating process influence and the coal particle structure influence on the progress and dynamic of coal combustion. The experimental results were used to develop the author’s mathematical model of the combustion process, taking into consideration the influence of the above mentioned parameters.

Golec, in his work, extensively describes the visual character of coal volatile matter combustion, distinguishing four phases:

I – initial, short phase (0–100 ms) of calm and release of volatile;

II – rapid ignition and non-uniform combustion, characterized by the outflow of strong jets of gas from particle surface (100–200 ms);

III – uniform combustion of volatiles in spherical diffusion flame with simultaneous particle swelling (the longest phase 200–420 ms);

IV – final phase: explosive release of volatile matter from the plastic particle, observed as a series of micro-explosions near the particle surface, with simultaneous extinguishing of flame outside the explosion area.

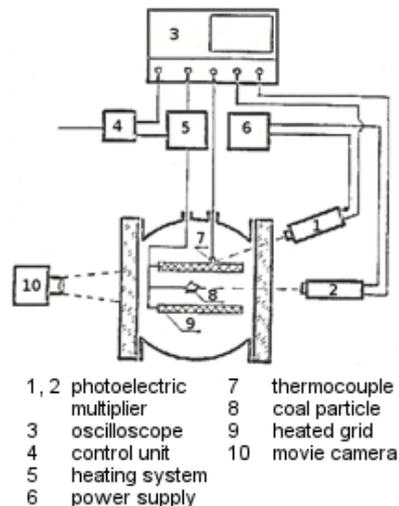


Fig. 22. Scheme of PW research rig with a particle suspended on the quartz needle (Golec 1989)

The observed multi-phase combustion process of volatile matter is explained by Golec as the remodeling of the internal structure of coal, which takes place during particle combustion (Golec 1989).

Experiments with particles held on or glued to the needle are easier to perform and the obtained results are easier to analyze than results and experiments performed with particles moving during the combustion process. The fixed location of the tested object is an unquestionable advantage in the case of the application of optical measurement methods. Yet, in some researchers’ opinions, the support of the particle (quartz needle, thermocouple) may disturb the combustion process under investigation. Tang and others (2010) argued that a thermocouple can catalyze the ignition of small particles [Essenhigh and others (Essenhigh, Misra, Shaw 1989) present a contrary view]. They also highlighted that the proper temperature measurement of particle surface is only possible when the entire surface of the sensor junction is in contact with the tested object. To ensure proper contact between the particle surface and thermocouple, even the application of very thin sensors, implies the elimination of the finest – and at the same time – the most interesting particle fractions. Therefore, it can be concluded that the usage of thermocouple as a support for the particle is justified only when sufficiently large particles are tested. During the combustion of larger coal particles, the catalytic impact of sensors on ignition can be considered negligible, and the large area of the particle’s surface allows for appropriate contact between the junction and the particle. In the case of using a quartz needle, it should be kept in mind that such support may receive too much heat from the burning particle, and the glue applied for attaching the particle to the needle may contain reactive components,

such as alkalis, which accelerate the processes of combustion, even if just trace amounts.

4. SUMMARY

Experiments performed with a single coal particle provide interesting information essential for the proper description of fundamental processes taking place during the combustion of coal, such as heat and mass exchange or reaction kinetics.

Since the first studies on single coal particles (Cassel, Liebmann 1959), several interesting test stands, dedicated to this type of research have been designed and improved upon. It should be noted, that differences in the measurement methodology and the conditions of the conducted experiments contribute greatly to diverse results and occasionally lead to contrary conclusions (Chen, Yong, Ghoniem 2012; Sami, Annamalai, Wooldridge 2001; Williams, Pourkashanian, Jones 2001). For example, experiments concerning coal oxy-fuel combustion carried out at low temperatures show comparable rates of the coal combustion reaction both in mixtures O_2/N_2 , as well as in O_2/CO_2 . Whereas studies conducted at high temperature and with increased O_2 concentration to determine the reactions of coal in CO_2 atmospheres are considerably slower than the combustion of coal in N_2 atmospheres.

Likewise, differences in the construction of test stands result in discrepancies obtained while comparing results origin from different test. For example, in experiments carried out in drop tube furnaces the flowing coal particle is heated directly by hot gas. In the case of heated grid reactors [presented eg. in (Qiao et al. 2010)], the particle is heated by conduction and radiation from the grid, with simultaneous heat transfer to the cooling gas. As a consequence of stand design differences, the ignition temperature determined in the heated grid reactor is higher than the ignition temperature obtained from Shaddix's (Shaddix, Molina 2009) experiments in drop tube furnace (Chen, Yong, Ghoniem 2012).

The experimental conditions must comply as accurately as possible with the assumptions of a theory or a model, to which they will be later referred to. Attempts to compare results obtained from different test stands should be undertaken very carefully. Despite the progress and implementation of more and more sophisticated measurement techniques in scientific research, several questions concerning the combustion process of single coal particles still do not full answer important questions.

Acknowledgements

The article was prepared based on research conducted for a Ph.D dissertation and was funded under the statutory activity of the Central Mining Institute, Poland.

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