Spontaneous ignition of wood, char and RDF in a lab scale packed bed

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ABSTRACT

Many municipal waste combustors use preheated primary air in the first zone to dry the waste. In most cases the preheat temperature does not exceed 140 °C. In previous experiments it is found that at temperatures around 200 °C, in some circumstances, self- or spontaneous ignition can be achieved. Using preheated air can be a powerful tool to control the ignition and combustion processes in a waste combustion plant. To use this tool effectively, the influence of the preheated air on the fuel bed needs to be well understood. The present work is done to investigate in a systematically way the spontaneous ignition behaviour of a packed bed heated with a preheated air stream. Experiments on a lab scale packed bed reactor are carried out for various fuel types. Because MSW is an highly inhomogeneous fuel, wood and char are used as model fuels. To include the inhomogeneous character of MWS, also experiments are carried out with RDF. Parameters such as primary air flow velocity and temperature, addition of inert material, moisture content of the fuel (wood chips) and particle size (char) have been changed to see their effect on the spontaneous ignition temperature and on the minimum air temperature needed for ignition. The spontaneous ignition temperature is defined as the bed temperature at which a transition takes place from a negligible or slow fuel reaction rate to a rapid oxidation of either the volatiles or the solid fuel without an external source such as a spark or a flame. The minimum or critical air temperature is defined as the lowest air temperature at which ignition can be obtained. It is found that the type of fuel has influence on the ignition temperatures. Besides both the critical air temperature needed for the spontaneous ignition and the spontaneous ignition temperature increase with an increase in the primary air velocity (between 0.1 and 0.5 m/s) and increasing the added inert fraction (between 0 and 40 wt%), irrespective of the fuel type. The effect of air flow velocity and temperature and also the effect of inert on both the critical air temperature and the spontaneous ignition temperature can be explained qualitatively by using Semenov's analysis of thermal explosions. Semenov's theory is quantitatively applied to predict the spontaneous ignition and the critical air temperatures for wood.

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1. Introduction

As the standard of living improves, the generation of municipal wastes and the depletion of natural resources increases steadily. Due to lack of space, combustion of municipal solid waste (MSW) forms a suitable alternative to dumping for many parts of the world, in particular highly densely populated ones, despite the associated (assumed) negative effects on the environment [1]. But waste combustion is more than just preventing the waste from being dumped, also energy is produced by burning waste. Combustion of municipal solid wastes on a grate is one of the dominant processes in municipal solid waste combustion (MSWC). However, grate firing is not only used for MSWC but also for combustion of other solid fuels, such as wood and other biofuels, e.g. straw or chicken-manure, to produce heat and power. For optimal process control, it is vital to understand the burning process inside the bed [2].

Many MSWC suppliers are using preheated primary air. The general accepted explanation is that the preheated air dries the fuel and the ignition will take place earlier on the grate [3]. When the temperature of the primary air is approximately 100 °C only drying occurs. When it is around 200 °C it enters the temperature range of pyrolysis, this is where drying and pyrolysis can occur simultaneously and this can lead to spontaneous ignition [4]. By preheating the primary air to these temperatures, the waste can deliberately be ignited in the zones where this is needed. On the other hand, by using preheated air, the waste can be ignited in zones where ignition is unwanted. To be able to control the ignition of a fuel bed by preheated air, the effect of some key parameters such as primary air flow rate and temperature are investigated in this work. Besides, mechanisms which are responsible for the ignition of the fuel bed by preheated air are investigated.

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In the present work spontaneous ignition is defined as the transition from a negligible or slow fuel reaction rate to a rapid oxidation of either the volatiles or the solid fuel without an external source such as a spark or a flame. It is described using the sudden rise in temperature of the bed as an indicator. The bed temperature from which this rise starts, is called spontaneous ignition temperature. The minimum air temperature needed for spontaneous ignition to occur is called critical air temperature and the elapsed time required to produce a sustainable burning reaction is called ignition time.

Not much research is done on this topic. Van Kessel et al. [2,3] investigated the combustion of waste with preheated primary air in a lab scale batch reactor. However, their work does not deal with spontaneous ignition of the fuel bed due to the preheated air. Also research is done on the ignition of single particles in a heated air stream by Kuo and Hsi [5], but this work focuses on the mode of ignition (flaming or glowing) and the ignition time. The research which shows the most resemblance with this work is the research on self-heating, self-ignition and thermal runaway done by Semenov [6] and Frank-Kamenetskii [7]. Often their analyses are applied to coal and char [8] and cellulosic materials [9]. These works show that coal can spontaneously ignite with ambient temperatures well below 100 °C. Cellulosic materials need a higher temperature to ignite. Times to ignition are in the order of days or even longer. In our work, the used temperatures are higher and different processes may occur.

Since the combustion characteristics are difficult to measure in full-scale plants, the moving bed is simulated by a batch type fixed bed reactor. Although the heat and mass transfer in transverse direction and additional particle mixing by the grate affect the combustion performance, the heat and mass transfer in vertical direction is the dominant factor. Therefore, a lab scale packed bed reactor can be used as a simplified system to simulate the moving bed. The time elapsed in the fixed bed corresponds to the residence time of fuel in the moving bed as has been described by Gort [10].

This article presents the effect of primary air flow velocity, particle size, moisture content and addition of inert to the fuel bed on the spontaneous ignition behaviour of several solid fuels. Because MSW is very inhomogeneous in both composition and geometry it is difficult to isolate the influence of fuel parameters on the ignition behaviour, therefore wood is used as a model fuel. To see the effect of volatile content, char is used as a model fuel as well. The inhomogeneous character of MSW is included in the experiment by using RDF. First, to give a theoretical background, an analytical evaluation of spontaneous ignition is given. This evaluation is based on Semenov's analysis of thermal explosions [6]. After this the experiments which are carried out are described and the results from these experiments are presented and discussed. The results of the experiments are evaluated in the light of the theoretical background and Semenov's analysis is applied to predict the spontaneous ignition and critical air temperatures for wood. Finally conclusions are draw.

2. Spontaneous ignition theory

Semenov's analysis of thermal explosions [6] is used as a theoretical background on spontaneous ignition of packed beds. In Semenov's analysis, the radial (or horizontal) temperature gradients are assumed to be flat, but this will not be the case in a waste incinerator. However, when a small part of the waste layer is considered (for example the drying zone where preheated primary air is usually applied) the horizontal temperature gradient can be assumed to be flat in that area. Note that in the classic Semenov's analysis, the system transfers heat to the surroundings through the reactor wall of which the thermal resistance is much lower than the one of the fuel bed. In our case, the system interacts with a uniformly preheated airflow over the total surface area of interest, so this system can be treated as zero dimensional. To be able to properly use Semenov's analysis, the outer temperature of the particles should not differ too much from the interior temperature. In the experiments used to measure the ignition temperatures of wood a slowly increasing air temperature is used. Due to this slowly increasing air temperature the times to ignition (at least 40 min) are much longer than the time for the core of the 1 cm thick particles to heat up to 225 °C with an air temperature of 230 °C (about 3 min). This shows that at the moment of ignition the temperature inside the particles can be regarded uniform. The base of this analysis is formed by an energy balance of the fuel bed. In our case, this balance consists of two parts:

- The heat gained by the reactions;
- the convective heat transfer between the primary air flow and the fuel bed.

The heat gained by the reactions can be either delivered by exothermic pyrolysis or oxidation of the fuel. In Fig. 1 these two energy streams are plotted as a function of the fuel bed temperature. The thick curved line denotes the heat gained by the reactions (Q_{react}). It is assumed that the reaction rate is determined by the kinetics. For low temperatures this is a save assumption. The three parallel straight lines denote the convective heat transport from the fuel bed to the air stream for three primary air stream temperatures (Q_{conv}). These lines cross the x-axis at the air temperature (T_0). When Q_{conv} is negative, the fuel bed is heated by the primary air flow.

Now depending on the air temperature, ignition might or might not take place. In situation (a) in which the air temperature is too low to ignite the fuel bed is denoted by the light grey line. At the start of this situation, the temperature of the fuel bed is low and the hot air stream heats the fuel bed. The reaction is still very slow and not much heat is produced by the reaction. When the temperature of the fuel is higher than T_0, the air stream cools the fuel. However, the heat produced by the reaction is larger than the heat lost to the air stream and the fuel will continue to heat up. When the fuel passes point 1, the heat lost to the air stream gets larger than the heat produced by the reaction and the fuel will be cooled by the air stream. Point 1 is a stable point and it has to be noted that the temperature in this point is higher than the temperature of the air stream even if there is no ignition.

When the air stream temperature (T_0) is chosen high, the heat lost to the air stream is always smaller than the heat produced by the reaction and ignition will occur. This is situation (c) in
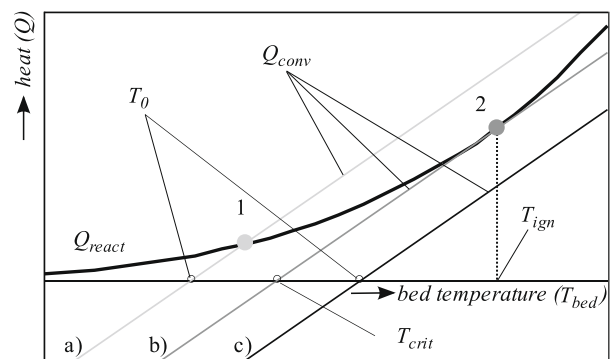

Fig. 1. Heat produced by the reaction (thick curved line) and the heat lost (positive values) or gained (negative values) due to convective heat transfer by the air stream for three values of T_0. Adopted from [6].

Fig. 1. However, the moment of ignition can hardly be determined and thermal runaway is encountered from the moment the experiment starts. So in that sense, ignition occurred from the very moment the experiment started.

The threshold for ignition is given by situation (b) (the dark grey line). The heat lost by the air stream is never larger than the heat produced by the reaction, so eventually ignition will occur. This situation gives the lowest air stream temperature at which ignition will occur. The temperature at point 2 is a critical temperature and the air temperature T_0 corresponding to this situation is the critical air temperature (T_{crit}). It has to be noted that at point 2, the slopes of the two lines are equal and a small deviation in either the reaction heat or the convective heat transport can result in a relative large difference in the spontaneous ignition and critical air temperature.

It is possible to change the convective cooling by altering the heat transfer coefficient. This can be done by changing the flow velocity of the air stream. When this velocity is increased, the heat transfer coefficient will increase as well and the heat lost by convective cooling will be more (the straight lines in the figure will become steeper). A higher T_0 is needed to ignite the fuel. When the air velocity is very low, the reaction rate for oxidation reactions (such as char combustion) will be determined by mass transport and this analysis will not be valid anymore. Besides changing the air velocity, also the particle size can be changed to influence the heat transfer coefficient. For smaller particles, the heat transfer coefficient will be higher and again a higher T_0 is needed to ignite the fuel bed.

3. Experimental

3.1. Experimental setup

The experiments were carried out at the laboratories of the Dutch research institute TNO in Apeldoorn. The schematic diagram of the experimental apparatus is shown in Fig. 2, the reactor is a lab scale packed bed reactor. The height of the reactor is 1350 mm with an inner diameter of 150 mm. The reactor is insulated with glass fiber on the outside. The grate is located at the bottom of the reactor and consists of a distributor plate made of ceramic. Four thermocouples (Pt/Rh, type S) are used to monitor the temperature of primary airflow and temperature inside the bed at different heights. The primary air can be heated up to 275 °C and the air flow rate can be given up to 50 m³/h. Every 20 s temperature and flow rate signals are send to a computer. The primary air is fed from the bottom of the fixed bed reactor through the grate. The air is preheated by an electric heater, installed in the supply line. The reactor is placed on a balance to monitor the mass of the fuel bed. Samples of the flue gases are extracted and analyzed.
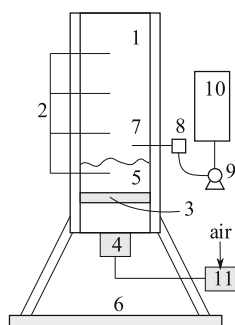

Fig. 2. Schematic overview of the experimental setup. (1)-reactor; (2)-thermocouples; (3)-grate; (4)-air flow heater; (5)-fuel bed; (6)-balance; (7)-gas suction probe; (8)-heated sample line; (9)-suction pump; (10)-gas analyzers; (11)-air flow controller.

The lower two thermocouples are positioned in the bed close to the grate. The thermocouples are protected from convection as well as radiation (after ignition) by small tubes covering the end of the thermocouples.

3.2. Materials

3.2.1. Wood chips

The wood chips consist of a mixture of wood species resulting from forest cutting. Wood mainly consists of cellulose, hemicellulose and lignin and in general they cover respectively 40–60, 20–40 and 10–25 wt% [11]. The pyrolysis and combustion processes of wood are complex because of the simultaneous degradation of the three components. The decomposition temperature range of the components is 315–400 °C, 220–315 °C and 150–900 °C, respectively [11]. Because lignin has such a wide temperature range for decomposition, it decomposes less easily than hemicellulose and cellulose. For temperatures below 300 °C the wood decomposition is mainly attributed to hemicellulose.

For some experiments, the wood chips moisture content has been changed to the desired value to study the effect of moisture content on the critical air temperature and spontaneous ignition temperature. This is done by adding some water assumed that by means of diffusion it will spread through the wood homogeneously [4].

The proximate and ultimate analysis of the wood chips is given in Table 1 and in Fig. 3 a picture of the wood chips is shown. Because the wood chips are shredded forest residue, the composition can vary. The largest dimension of the chips is about 40 mm.

3.2.2. Char

Two types of wood char have been used in the experiments. The first type of char is called commercial char. This char is generally used in barbeque's. The exact production process and the initial material are unknown but it is known that the fixed carbon content of this char is at least 82%. To study the particle size effect it has been cut into the desired size manually (small: 10–15 mm, medium: 15–30 mm and large: 30–40 mm).

The second type of char is called lab char. This type of char is made in our laboratory by keeping the wood chips in nitrogen at 350 °C for 3 h. The resulting char has a volatile content of around 40%, which is notably higher than the volatile content of the commercial char.

The commercial char as well as the inert material used in the experiments are illustrated in Fig. 3. In Table 1, as an indication a proximate and ultimate analysis of char is given.

3.2.3. Refuse derived fuel (RDF)

Refuse derived fuel is made of the paper, plastics, wood and other organic components from municipal solid waste. The compo-

Table 1
Proximate and ultimate analysis of the wood chip, wood charcoal (from literature) and RDF.

	Wood chips	Char [16]	RDF
%C	50.0	92	50
%H	6.56	2.45	6.5
%O	41.5	3	30
%N	0.34	0.53	0.8
%S	0.29	1	0.3
%vol.	82.34	9.4	75
%ash	1.38	1	
%moist (a.r.)	9	6.0	3
H_b (MJ/kg) (d.b.)	19.9	34.4	21
H_l (MJ/kg) (d.b.)		33.9	20

Fig. 3. Some of the used fuels. Upper left to right: small char (10–15 mm), medium char (15–30 mm), large char (30–40 mm). Lower left to right: wood chips (10–40 mm), stones (10 mm).

nents are dried and pelletized to pellets of 16–18 mm. The proximate and ultimate analysis of the used RDF is listed in Table 1.

3.3. Experimental procedures and programme

All the experiments with wood and commercial char have been performed considering 300 g and 250 g of fuel, respectively. To find the critical primary air temperature and the spontaneous ignition temperature the following three methods can be used:

3.3.1. Constant temperature method

In this method the air is heated quickly to a constant value. When no ignition takes place, the fuel is removed and a fresh batch is put into the reactor. A new experiment is started with a slightly higher air temperature. When ignition is found, the current air temperature is the critical one. This method is suited to find the critical air temperature and the ignition time. However, for wood and char, the spontaneous ignition temperature can not be found with this method. This will be further explained in Section 5.1. The RDF, lab char and some wood experiments are carried out with this method.

3.3.2. Slowly increasing temperature method

This method is basically the same as the previous one. However, the air is heated slowly to the desired value. Consequently, with this method it is not possible to determine the ignition time. In contrary to the previous method, this method is suitable to find the spontaneous ignition temperature. Besides, the critical air temperature can be found as well. The char experiments are done following this method.

3.3.3. Stepwise increasing temperature method

In contrary to the other two methods, in this method the fuel is not removed from the reactor when no ignition takes place. Instead the air temperature is increased by steps of 10 °C when no ignition takes place. With this method the spontaneous ignition temperature and the critical air temperature can be found. It has to be made sure that the temperature history does not influence the results. For wood this is the case and this method gives the same results as the previous method. This method is used in the wood experiments.

More comments on the effect on the ignition parameters of the three methods will be given in Section 5.1.

To study the effect of primary air flow velocity on the critical air temperature and spontaneous ignition temperature a number of experiments have been carried out at five air flow velocities (0.1, 0.2, 0.3, 0.4 and 0.5 m/s). Besides, the effect of inert in the fuel has been investigated by substituting a portion of the sample with inert (stone).

Finally, to see the effect of the size of the commercial char particles on the critical air temperature and on the spontaneous ignition temperature and time, three sample sizes are prepared (small, medium and large) by cutting the char manually into the desired size. Overall, more than 100 experiments have been carried out.

4. Results and discussion

4.1. Wood experiment results

4.1.1. The effect of primary air flow velocity

Fig. 4(a) shows the spontaneous ignition temperature as a function of primary air flow velocity at a constant moisture content for wood. Fig. 4(b) shows the critical air temperature. It can be seen that both the critical temperature needed for spontaneous ignition to occur and the spontaneous ignition temperature increase with increasing primary air flow velocities. This is an expected result, since for higher air velocities more heat is lost to the air flow due to a better convective heat transfer and a higher air temperature is needed to ignite the wood. This is also described in Section 2. With the constant temperature method, the ignition time is found to be around 5 min (see Fig. 15).When no ignition took place, the bed temperature was about 5 °C higher than the air flow temperature for all air velocities. This indicates there is an exothermic process going on indeed. Despite this exothermic process, no thermal runaway is encountered. This situation is denoted in Fig. 1 by point 1.

Especially for the lowest air flow velocity, the measurement error is distinct. This can be caused by the sensitivity of the location of point 2 in Fig. 1 as is explained in Section 2. When the air flow is low, the heat transfer coefficient is low as well and the lines denoted by Q_{conv} in Fig. 1 are close to horizontal. The more horizontal the lines, the bigger the effect of a minor deviation of the reaction heat or convective heat transfer are. In general for wood the reproducibility of the results is fairly good and the measurement error is smaller than 10%.

4.1.2. The effect of inert in the sample

Experiments have been performed to investigate the effect of inert on the spontaneous ignition temperature of the wood. For this purpose, stones are used as inert mixed with wood chips in 20 and 40 wt% (inert) proportion for air flow velocities of 0.2 and 0.3 m/s. The results are compared to the normal situation without inert.

As it is depicted in Figs. 5(a) and 5(b), increasing the percentage of inert in the mixture will increase the spontaneous ignition temperature and critical air temperature. This is due to the fact that the inert absorbs more energy given by the primary air without producing heat. To compensate that and to start the ignition more energy is needed, so the ignition temperature increases. This can also be explained with the help of Fig. 1: by adding inert to the fuel bed, the heat produced by the reaction decreases. This means that the line Q_{react} in Fig. 1 will be less steep and a higher air temperature is needed to ignite the fuel. Also the spontaneous ignition temperature (point 2) will be higher. For the last mixture of inert with wood chips (40 wt%), ignition occurs late for an air flow velocity
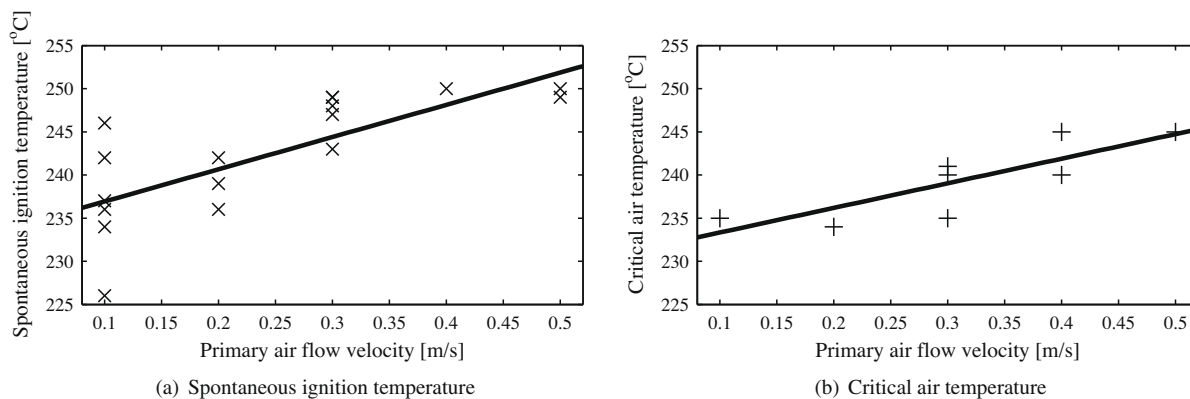
(a) Spontaneous ignition temperature   (b) Critical air temperature

Fig. 4. The measured spontaneous ignition (a) and critical air temperature (b) for wood chips as a function of the air flow velocity.

of 0.2 m/s and no ignition occurs for the air flow velocity of 0.3 m/s within 3 h. This is probably caused by the fact that, at higher flow velocities the loss due to the convection are higher than at lower air flow velocities.

4.1.3. The effect of moisture

To investigate the effect of the moisture on the spontaneous ignition temperature of wood chips samples, the wood chips samples were moistened by spraying the needed amount of water on it and storing them for two days to make the water diffuse through the sample. Experiments with a fuel moisture contents of 9–12.7, 18–20, 30 and 50 wt% have been carried out. In all experiments of moisture content lower than 50 wt% no change in critical air and spontaneous ignition temperatures has been shown.

However, no ignition could be obtained for wood chips with a moisture content of 50% on a 150 min experiment. The grate temperature was only 1 °C higher than the primary air temperature compared to others which were 5–7 °C higher. Since drying moist wood chips will take more energy, this inhibits the devolatilization step and further the exothermic reactions occurred by the decomposition of wood. Yet, if the fuel bed had been kept for a longer time at this condition, (temperature and air flow velocity) it would probably ignite.

On the whole, with these moisture contents only a few experiments have been performed, therefore, it is not possible to get a reliable relationship between the fuel moisture content and spontaneous ignition temperature. However, it is reasonable to expect no change in spontaneous ignition and critical air temperatures with varying moisture content. Only the ignition time does change.

4.2. Char experiment results

An understanding of the oxidation rate of chars at low temperature is important for predicting self-heating and spontaneous ignition temperatures. Factors affecting the spontaneous ignition of char are: the primary air flow velocity, primary air temperature, particle size, the production method of the char and the addition of inert to the sample. The effects of these factors on the spontaneous ignition characteristics have been investigated.

In Figs. 6 and 7 a typical constant temperature measurement result for the experiments for char can be seen. Fig. 6 shows the mass of the char and the bed temperature during the experiment and Fig. 7 shows the concentrations of several flue gases. It takes some time before the gas samples reach the analyzers, so there is a delay in the measured gas concentrations. In this case, the measurement for the bed temperature shows an ignition time of 1.5 min, which is significantly shorter than the ignition time for wood, which was 5 min (see Fig. 15). Ignition can be identified by the increase in

temperature from Fig. 6, but also the sudden decrease of O_2 and the increase in CO_2 show that combustion is taking place (Fig. 7). Note that these results are obtained by a constant air temperature. The spontaneous ignition and the critical air temperature are obtained by using a slowly increasing air temperature.

4.2.1. The effect of primary air flow velocity

The effect of air flow velocity on the spontaneous ignition characteristics of commercial char samples has been examined. Fig. 8(a) shows the measured spontaneous ignition temperature of medium size char particles. Because for char, most of the experiments are slowly increasing temperature experiments, the critical air flow temperature is determined in an other way compared to the wood experiments. Fig. 8(b) shows the minimum measured (out of minimal four experiments for each air velocity) air temperature at which ignition occurs. It also shows the maximum air temperature at which no ignition occurs. The reproducibility of the results for char are not as good as the ones for wood.

The figures show that both the spontaneous ignition and the critical air temperature for char are significantly lower than the ones for wood. This is probably caused by faster exothermic reaction mechanisms for char at lower temperatures. As can be seen in Fig. 8(a), the spontaneous ignition temperature of the commercial char increases with the flow velocity. For high air flow velocity experiments, the air temperature reaches the set air temperature in the heat controller quickly though, the heat transfer to the char and even more important: inside the char, may not be that quick due to a high Nusselt number. This means that the inside of the particles is still cold and no reaction takes place there yet, while the temperature of the outside of the particles (which is the measured temperature) can be high already. For low air velocities, the Nusselt number is smaller and the temperature distribution inside the particles will be more constant. This means that for relative low measured outer temperatures the particles will be able to start the runaway reactions.

Despite the clear trend, not all batches of char behaved the same. From Fig. 8(a) it can be seen that when ignition takes place, this is in all cases at a temperature below 190 °C. However, some batches of char did not ignite, even when the bed was at a temperature of 250 °C. This means not all batches of char are the same and may be a small variation in geometry or mineral content results in a significant different ignition behaviour. It is also remarkable that the lowest temperatures at which ignition occurred in Fig. 8(b) are always lower (or equal for 0.4 m/s) than the maximum temperature at which ignition did not take place. This shows that the reproducibility of the char experiments is low, probably due to variations in the composition and geometry of the char. With MSW the reproducibility can be expected to be even lower. With
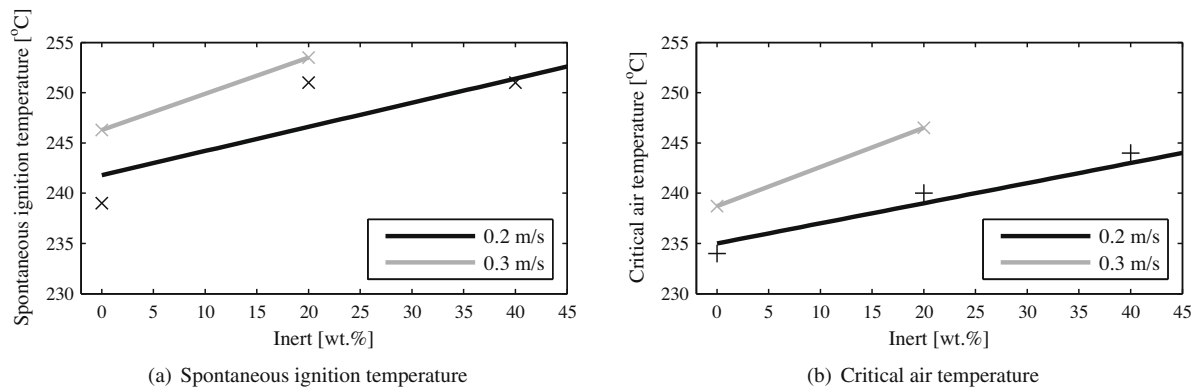
(a) Spontaneous ignition temperature

(b) Critical air temperature

Fig. 5. The measured spontaneous ignition (a) and critical air temperature (b) for wood chips as a function of the mass percentage inert for two air velocities.
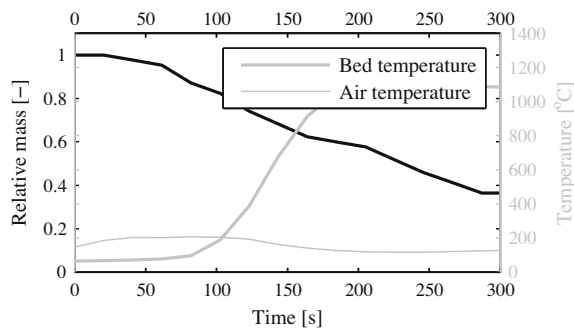

Fig. 6. Measured bed mass (black line) and air and bed temperatures (grey lines) as a function of time for lab char. The superficial air velocity is 0.1 m/s and the air temperature is 200 °C.
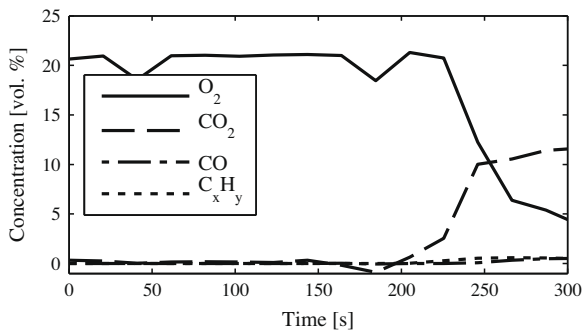

Fig. 7. Measured concentration of ,O_2,CO_2,CO and C_xH_y as a function of time for lab char. The superficial air velocity is 0.1 m/s and the air temperature is 200 °C.

this knowledge, the trend line in Fig. 8(b) has to be interpreted as the minimal air temperature at which spontaneous ignition could occur.

When no ignition took place, the bed temperature of the char was lower than the air temperature. This indicated that there is an endothermic process going on at temperatures lower than the spontaneous ignition temperature.

4.2.2. The effect of addition of inert to the sample

To see the effect of inert on the spontaneous ignition, again the two flow velocities (0.2 and 0.3 m/s) with small particle size have been considered. As it is depicted in Fig. 9, an increase of the spontaneous ignition temperature with increasing inert content has been measured. This is may be due to the absorption of heat by the inert thus there is not enough energy to start the spontaneous

ignition. Ignition did not occur for an air velocity of 0.3 m/s and 40% inert.

4.2.3. The effect of particle size

In determining whether or not spontaneous ignition occurs, the particle size of commercial char may be an important factor. In general, the rate of oxidation is depending on the external surface area per unit volume of the coal particles. The surface area increases with decreasing particle size.

The spontaneous ignition for small size particles occurs at temperatures about 5 °C lower than for medium size particles. The lower spontaneous ignition temperatures can be caused by a larger specific external surface area of the smaller particles, resulting in faster oxidation of the char. However, the difference is not clearly seen due to the fact that the small and medium particle size range is not differing much.

4.3. RDF experiment results

In Fig. 10 typical mass and bed temperature measurement results for the experiments for RDF can be seen. The ignition time in this experiment is about 40 min, which is an order longer than in the case of wood and char. This can be caused by the fact that the RDF has a higher density (500 kg/m³ versus about 200 kg/m³ for wood and even less for char), so the RDF does not heat up as fast as wood or char. The air temperature does not have a clear influence on the ignition times, however, when the air velocity is reduced to 0.1 m/s, the ignition times are around 80 min. This supports the idea that the heating of the fuel determines the ignition time, not the kinetics.

For RDF there is a typical pre-ignition stage which lasts from 2000 s until ignition at 2250 s in this case. This behaviour is probably caused by the different decomposition mechanisms for the several components found in RDF.

Fig. 11(a) shows the spontaneous ignition for RDF as a function of primary air velocity. Fig. 11(b) shows the critical air temperature as a function of the air velocity. Like with wood and char, the increasing trend with air velocity is observed here. It has to be noted that the spontaneous ignition temperature cannot be determined precisely due to the pre-ignition stage. However, Fig. 10 shows that the bed temperature is higher than the air temperature at ignition.

4.4. General experimental observations

When two individual experiments with wood chips are carried out successively, an increase in critical primary air temperature and spontaneous ignition temperature has been noticed system-
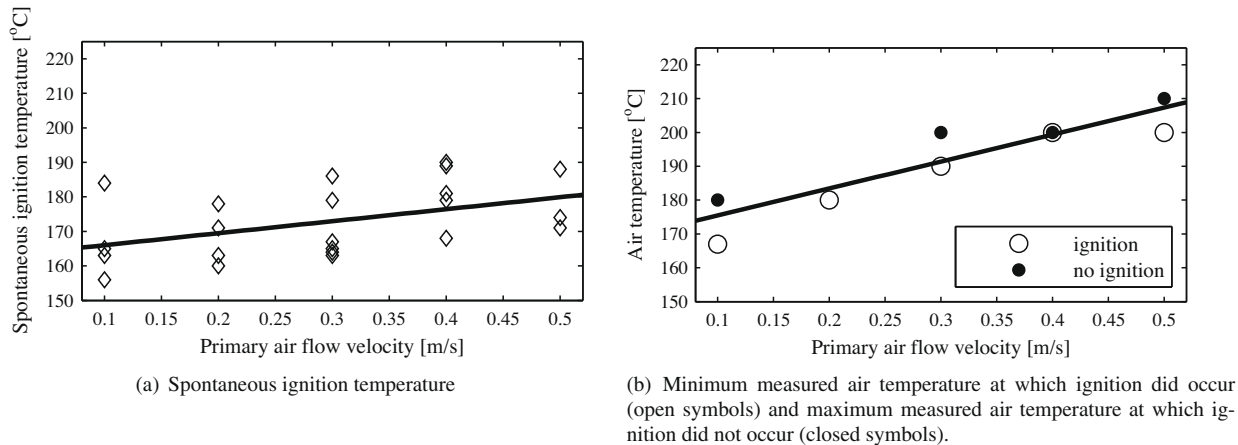
(a) Spontaneous ignition temperature

(b) Minimum measured air temperature at which ignition did occur (open symbols) and maximum measured air temperature at which ignition did not occur (closed symbols).

Fig. 8. The measured spontaneous ignition (a) and critical air temperature (b) for commercial char as a function of the air velocity.
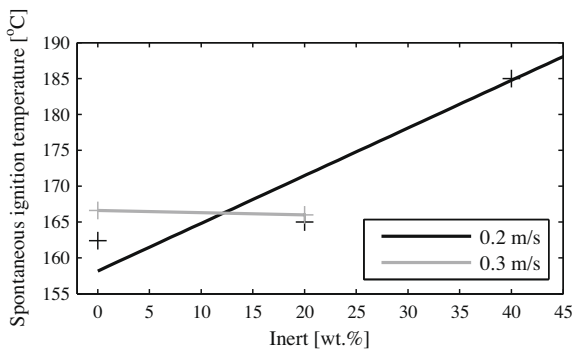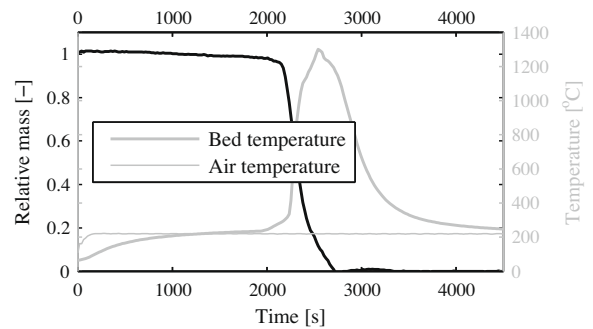

Fig. 9. Measured spontaneous ignition temperatures for small size commercial char as a function of the mass percentage inert for two air velocities.

Fig. 10. Measured bed mass (black line) and air and bed temperatures (grey lines) as a function of time for RDF. The superficial air velocity is 0.3 m/s and the air temperature is 220 °C.

atically. Particularly, the effect will be pronounced if the first experiment is done with low air flow velocities. It is noted that a significant amount of unburned volatiles are emitted. These volatiles will condense on the reactor wall. In the second experiment, the condensed tars will absorb energy to evaporate and a higher temperature is needed to deliver enough energy to ignite the fuel. To investigate this more, two consecutive experiments in two air flow velocities (0.2 and 0.3 m/s) have been carried out. It is found that both the critical air temperature and the spontaneous ignition temperature are in general 5 °C higher when a wood experiment is preceded by another wood experiment. To avoid this effect on the measurement of the temperatures, after the first wood experiment one experiment with char has been carried out to burn the condensed tars from the reactor wall. The char is only used to clean the reactor and no data is used from these runs. It has been shown that the measured temperatures for wood are more reproducible when an experiment with char is performed between the two wood experiments. It has to be mentioned that a deviation of 5 °C in the temperatures are within the measurement error.

In some of the experiments done with commercial char, a significant delay of the spontaneous ignition has been observed. In general, for all experiments the initial ignition does not occur at the exact location of one of the thermocouples inside the fuel bed. In this case the ignition front has to travel a certain distance through the bed to be sensed by one of the thermocouples and the measured ignition time will be an over-estimation. Because the wood ignition is accompanied with a flame which can be sensed by the higher thermocouples easily, this delay is

expected to be minimal for wood. However, the char ignition is exclusively heterogeneous and the delay in ignition time can be significant. This is especially the case with the large char particles.

To study the process behaviour after ignition has occurred with primary air preheating on wood, two experiments at different air flow velocities with more than 1 kg wood chips have been carried out in the laboratory setup. This experiment has been conducted, since in every other experiment only 300 g of wood has been used which covers the first two thermocouples only. Hence, this process behaviour cannot be seen.

The first experiment is carried out at an air flow velocity of 0.2 m/s and primary air temperature of 230 °C. At this flow velocity the ignition first occurs at 20 cm from the grate. In this case the thermocouple at 5 cm above the grate shows ignition next. After that the high-temperature front travels upwards. The duration of the conversion process after the breakthrough is small (5–15 min), compared to the time before the breakthrough 40 min. Fig. 12 shows the temperatures measured by the thermocouples at the specified bed heights for this air flow velocity.

The second experiment was carried out at an air flow velocity of 0.3 m/s and primary air temperature of 230 °C, at this flow velocity the spontaneous ignition starts in the bed at 35 cm from the grate. The spontaneous ignition is shown by the sudden increase of temperature at the thermocouple at this height. After that, the thermocouple below (at 20 cm above the grate) and above (at 50 cm above the grate) show an increase in temperature. Finally, the thermocouple located 5 cm above the grate shows an increase in
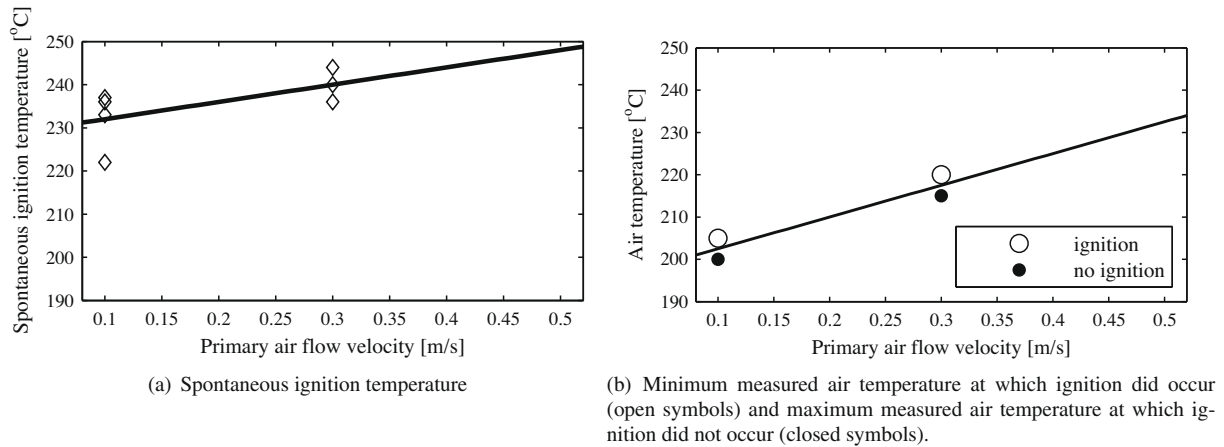
(a) Spontaneous ignition temperature

(b) Minimum measured air temperature at which ignition did occur (open symbols) and maximum measured air temperature at which ignition did not occur (closed symbols).

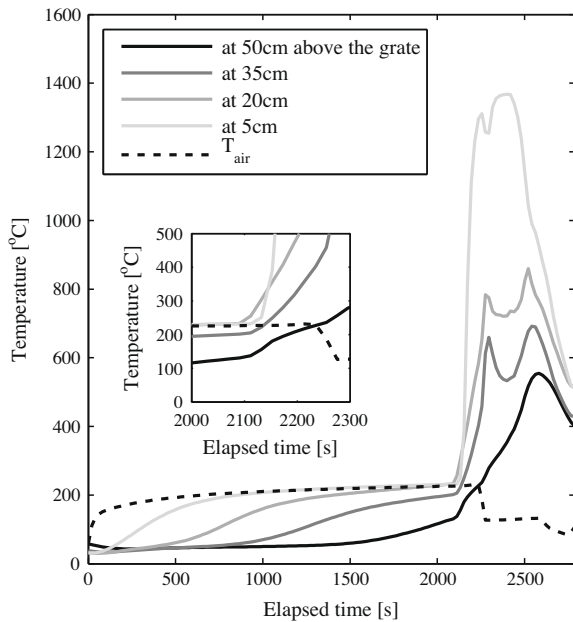**Fig. 11.** The measured spontaneous ignition (a) and critical air temperature (b) for RDF as a function of the air velocity.

Fig. 12. Measured temperatures in a deep fuel bed of wood chips. Air flow velocity is 0.2 m/s. Air flow temperature is 230 °C. The inset shows the moment of ignition in more detail.

temperature. Fig. 13 shows the temperature measured by the thermocouples at the specified bed heights for this air flow velocity. The time taken for the ignition at 0.3 m/s is slightly higher than 0.2 m/s. This may be due to the cooling effect by the higher air flow velocity.

From the above experiments, we may say that:

- The initial ignition does not occur at the grate, but 20–35 cm above the grate. After the initial ignition, a reaction fronts travels upwards and another reaction front travels downwards.
- For a flow velocity of 0.2 m/s, the reaction front travels quickly downwards, due to the presence of dry fuel and already formed char in the bottom of the fuel bed. Besides, close to the grate, the air is still oxygen-rich. However, for the flow velocity of 0.3 m/s this front does not travel downwards fast.

It is not clear why the ignition does not take place at the grate. It can be caused by the inhomogeneous composition and geometry of

the fuel bed. This can also be the cause of the measurement error for both wood and char for the experiments described earlier. As it is described in Section 2, a small deviation in reaction or convective heat transfer parameters can cause a relative large deviation in spontaneous ignition and critical air temperatures. The deep bed experiments show that it is difficult to predict at which height in the fuel bed the ignition will take place.

An experiment with wood chips was carried out with an air flow of 0.3 m/s and a temperature of 230 °C. No ignition took place and the bed temperature stabilized and after a while, the air flow was shut off. Fig. 14 shows the bed temperature and mass after the air flow was stopped. A band-stop filter is applied on the mass signal to eliminate frequencies between 4 and 40 mHz. From the rising bed temperature and the decreasing mass it can be concluded that there is an exothermic reaction emerging after the air flow is turned off. This behaviour can be explained with the help of Fig. 1. With the air flow still on, the stable point 1 is reached and the bed is cooled by the air flow. When the air flow is now stopped, the convective cooling ceases and the exothermic reaction is able to proceed. The measured behaviour shows that the exothermic reaction proceeds without oxygen so it is a pyrolysis reaction. If this pyrolysis reaction was the only reaction, the temperature should have increased exponentially. However, a new stable temperature of around 380 °C is reached. This shows that besides the exothermic pyrolysis reaction also one or more endothermic pyrolysis reactions are going on at higher temperatures (above 300 °C). In Section 5.2 the pyrolysis reactions are described into more detail.

5. Evaluation

5.1. Differences in experimental methods

The different results from the different methods described in Section 3.3 will be explained with the help of the results of the wood experiments.

A typical measurement result with the constant air temperature method for wood can be seen in Figs. 15 and 16. Fig. 15 shows the mass of the fuel and the bed temperature during the experiment and Fig. 16 shows the concentrations of several gases. It takes some time before the gas samples reach the analyzers, so there is a delay in the measured gas concentrations. The measurement for the bed temperature shows an ignition time of 5 min. Note that this is with an air velocity of 0.1 m/s. Again, ignition is indicated by both the temperature increase from Fig. 15 and the decrease and increase of oxygen and carbon dioxide respectively (Fig. 16).
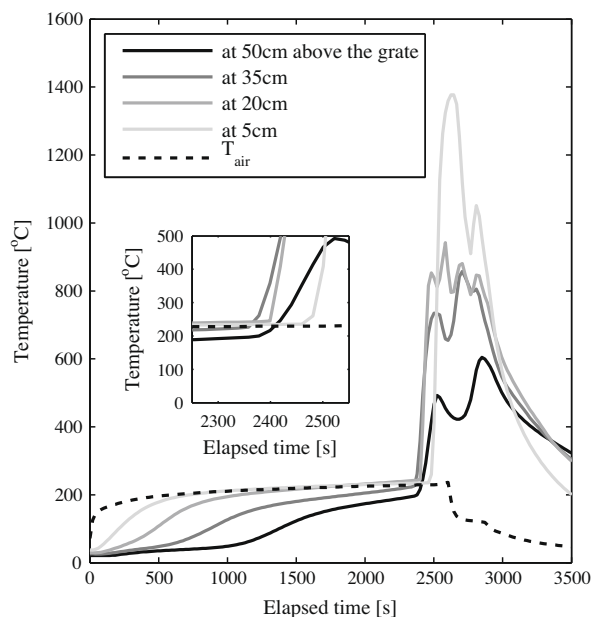
Fig. 13. Measured temperatures in a deep fuel bed of wood chips. Air flow velocity is 0.3 m/s. Air flow temperature is 230 °C. The inset shows the moment of ignition in more detail.
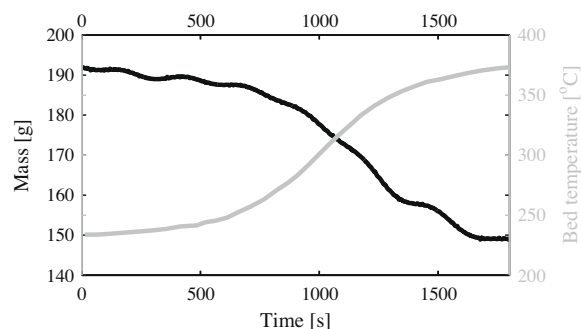

Fig. 14. Measured temperature and fuel bed mass as a function of time. A band-stop filter is applied on the mass signal to eliminate oscillations between 4 and 40 mHz. The experiment was started with an air flow velocity of 0.3 m/s and a temperature of 230 °C. At $t = 0$ the air flow was turned off.

It can be seen that the bed temperature is only 150 °C when it ignites. However, this is not the spontaneous ignition temperature of the wood. The temperature measurement does not show a clear ignition point. Because the heat lost to the air stream is always smaller than the heat produced by the reaction, no clear moment of ignition can be defined. The thermal runaway (which is ignition in our case) starts as soon as the experiment starts (black line in Fig. 1). If the air temperature is chosen closer to the critical air temperature, this effect will be less pronounced and if the air temperature is close enough, the spontaneous ignition temperature can be measured, but this a time consuming procedure. This illustrates that the constant temperature method is not suitable for determining the spontaneous ignition temperature.

An example of an experiment carried out with the slowly increasing air temperature method is shown in Fig. 17 for 0.3 m/s. By slowly increasing the air temperature (T_0 in Fig. 1) point 2 in Fig. 1 is reached slowly and can be determined accurately. The first notable difference with the constant air temperature experiment is that the time to ignition is at least an order of magnitude longer. This is inherent to the measurement method, because the
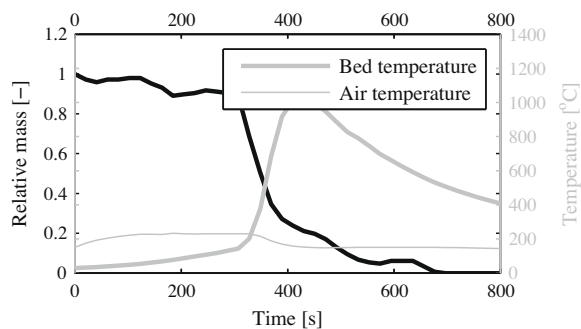

Fig. 15. Measured bed mass (black line) and bed and air temperatures (grey lines) as a function of time for wood chips. The superficial air velocity is 0.1 m/s and the air temperature is 230 °C.

bed is slowly brought to its runaway point. This shows that this method is not suitable for measuring ignition times. The second important difference is that the bed temperature is higher than the air temperature at ignition, besides, the start of the runaway (ignition) can be seen clearly at 4285 s and a temperature of 247 °C. However, more experiments are needed to determine the critical air temperature.

The critical air temperature can be determined together with the spontaneous ignition temperature in a single experiment when the stepwise increasing air temperature is used. The result from such an experiment is shown in Fig. 18. The critical air temperature is 240 °C and the spontaneous ignition temperature is 249 °C (which is almost the same as in the slowly increasing temperature method).

5.2. Heats of reaction

In reality, the shape of the heat of reaction line is not as simple as depicted in Fig. 1. Wood reacts complex, but the kinetics of several types of wood is well documented. As has been posed in the materials section, wood pyrolysis is often modeled by the sum of the pyrolysis of its main components (i.e. hemicellulose, cellulose and lignin) [12,11,13]. The kinetics of these pyrolysis reactions are not differing too much in literature. Hemicellulose is the most reactive and decomposes between around 220 °C and 315 °C. Cellulose decomposes between 315 °C and 400 °C and lignin decomposes slowly over a wide temperature range (160–900 °C) [11]. So for temperatures below 300 °C, wood degradation can be attributed to hemicellulose degradation for a great extent. Although the kinetics of these three reactions are fairly well known, values for the heats of reaction are not found in literature. However, Yang et al. [11] found that hemicellulose and lignin pyrolysis are exothermic and cellulose pyrolysis is endothermic. The overall heat of pyrolysis has been measured by Rath et al. [14]. The most important conclusions that can be drawn from the papers from Yang et al. and Rath et al. [11,14] are:

- wood pyrolysis is slightly exothermic at temperatures between 200 °C and 325 °C mainly due to the hemicellulose pyrolysis;
- between 320 °C and 380 °C the endothermic cellulose pyrolysis determines the heat of reaction;
- the degradation of wood for temperatures up to 300 °C can be attributed to the hemicellulose degradation.

From this it can be concluded that the spontaneous ignition temperature of wood will be found at least above 200 °C, which is also found in the experiments. Note that the reaction heat comes from an exothermic pyrolysis reaction. This means that also elevated temperatures can be reached without oxygen at all. At
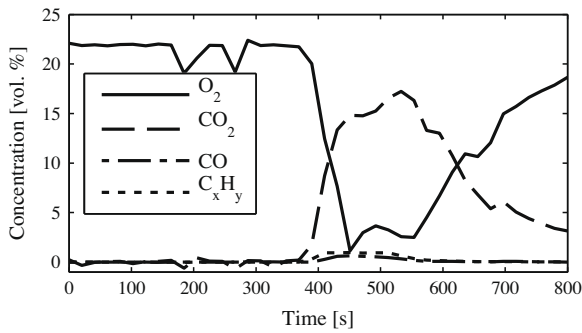
Fig. 16. Measured concentration of O_2, CO_2, CO and C_xH_y as a function of time for wood chips. The superficial air velocity is 0.1 m/s and the air temperature is 230 °C.
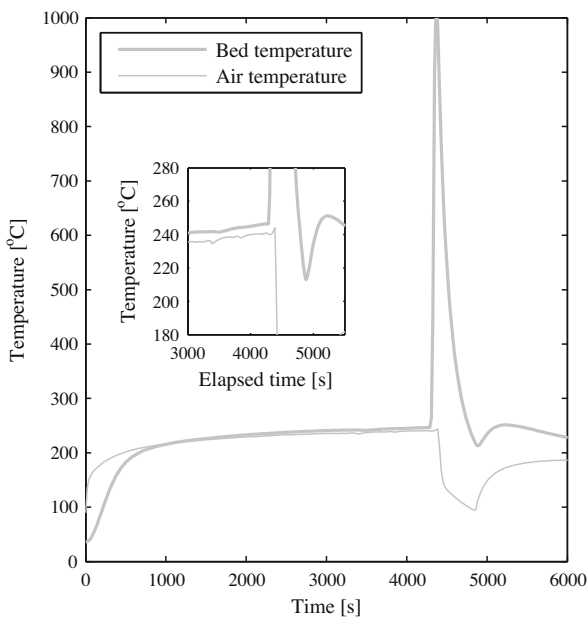

Fig. 17. Measured bed and air temperatures as a function of time for wood chips and a low heating rate. The superficial air velocity is 0.3 m/s and the air temperature is 240 °C.

Fig. 18. Measured bed and air temperatures as a function of time for wood chips and a low heating rate. The superficial air velocity is 0.3 m/s and the air temperature is 220–230–240 °C.

around 320 °C the endothermic cellulose pyrolysis starts and the temperature of the fuel bed will stabilize without oxygen (with oxygen the fuel would have been ignited already). This behaviour is seen in the experiment where the air flow is stopped after no ignition took place (see Fig. 14).

For wood charcoal, no data is found in the literature for the temperature ranges considered in the measurements. The heat of reaction is measured at the University of Twente using differential scanning calorimetry (DSC) for three types of wood charcoal in air. The charcoal samples are produced by keeping wood pellets for 3 h at either 350 °C, 500 °C or 650 °C in a nitrogen environment. The results can be seen in Fig. 19. It can be seen that the charcoal interaction with oxygen switches from endothermic to exothermic between 150 °C and 200 °C. As with wood, from this data it can be concluded that the spontaneous ignition of charcoal cannot be lower than 150 °C. This statement is supported by the experiments. These heat of reaction profiles could also explain why the bed temperature is lower than the air temperature at temperatures below the spontaneous ignition temperature, since the reaction is shown to be endothermic at these temperatures. This might lead to the idea that the temperature where the reaction gets exothermic is close to the spontaneous ignition temperature. The DSC results
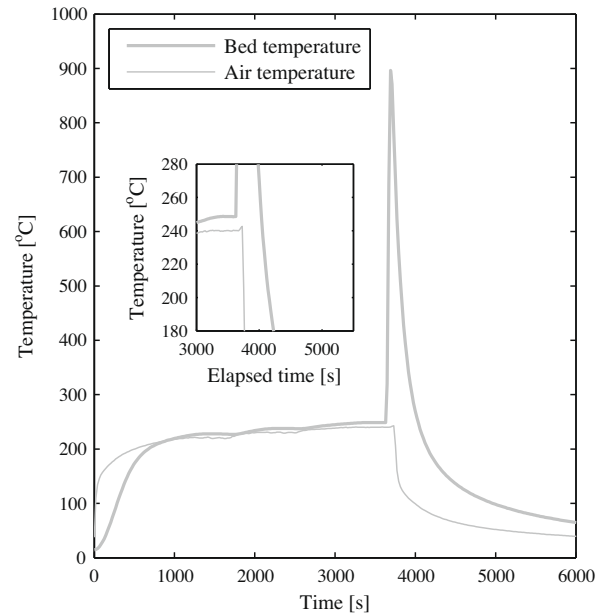
show that the temperature at which this happens is strongly dependent on the production temperature. This can be an explanation for the wide spread of the results for the char experiments.

5.3. Application of Semenov's analysis

It appears that Semenov's analysis is able to describe the ignition phenomena in a packed bed qualitatively. But when the heat produced by the reaction and the convective heat losses depicted in Fig. 1 are known, Semenov's analysis can be used to determine the spontaneous ignition and critical air temperature also quantitatively. In this section Semenov's analysis is applied on the ignition of a packed bed of wood.

Semenov's analysis assumes the reacting system to be at a uniform temperature, this assumption has to be validated for the experiments. Thermocouple readings on top of the grate and at 5 cm above the grate in the bed differ only 2 °C after 11 min of heating a wood bed with 200 °C air flowing at 0.3 m/s. Figs. 17 and 18 show that the ignition times in case of wood are in the order of 1 h. This shows that there are no vertical temperature gradients just before ignition. The fuel bed is wide enough to assume that the horizontal gradients can be neglected as well. So the assumption that the fuel bed is at a uniform temperature before ignition is justified. The convective heat transfer flux is expressed as:

$$\dot{q}''_{conv} = h(T_a - T_{bed})A_s h_{bed}, \qquad (1)$$

in which h is the heat transfer coefficient and A_s is the specific surface area of the packed fuel. The heat transfer coefficient h can be found by the Nusselt–Reynolds relation according to Bird et al. [15].

As has been said before, for temperatures below 300 °C the decomposition of wood is determined by the pyrolysis of the hemicellulose. The heat gained by the reaction can be expressed as:

$$\dot{q}''_{react} = k_{hc} \exp\left(-\frac{E_{hc}}{RT}\right) \rho_{hc} \Delta H h_{bed}. \qquad (2)$$

ΔH is the heat of reaction and h_{bed} is the total height of the fuel layer in the reactor, ρ_{hc} is the density of the hemicellulose in the wood,
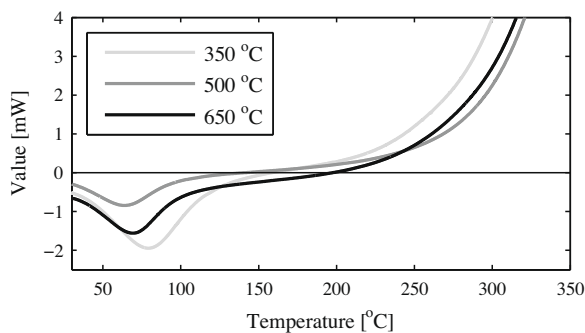
Fig. 19. Heats of reaction of three types of char as a function of temperature.

k_{hc} is the pre-exponential factor, E_{hc} is the activation energy, R is the ideal gas constant and T is the temperature of the wood. Because before ignition, a uniform temperature can be assumed throughout the fuel bed, the height of the fuel bed is not important.

Most of the kinetic data found in the literature does not focus on the low temperatures encountered in the spontaneous ignition experiments. However, Órfão et al. [13] have carried out TG analyses on wood, cellulose, hemicellulose and lignin at low temperatures. They give the kinetics for the pyrolysis of the three wood components in nitrogen but they also found that in air the pyrolysis reactions occur at lower temperatures. They do not mention the kinetic data for hemicellulose in air, but it can be found by fitting an Arrhenius equation on their TGA curve. The result is: $E_{hc} = 107$ kJ/mol and $k_{hc} = 8.9 \cdot 10^8$ 1/s.

The heat gained by the reaction (Eq. (2)) is plotted in Fig. 20 with the thick black line as a function of the bed temperature. The convective heat losses are (Eq. (1)) plotted for two air velocities (0.1 and 0.5 m/s) with the two thin lines. In all two situations the critical air temperature and the ignition temperature of the fuel bed can be seen. Also the measured ignition temperatures and the critical air temperatures are shown for these two air velocities. All two situations compare to situation (b) in Fig. 1. The figure shows that the model is able to predict the critical air temperatures well. However, it over-predicts the measured spontaneous ignition temperatures by 5–10%. There can be several reasons for the over-prediction. A straightforward reason is that the solid temperature is predicted, but the actual measured temperature is the gas temperature between the solids. With solid phase runaway, the gas temperature is always lower than the solid temperature so the spontaneous ignition temperature is measured too low. The over-prediction of the measured spontaneous ignition temperature can also be caused by inaccuracy in the kinetic data. Not much kinetic data is available at these relatively low temperatures. Besides, the fuel used in the experiments is forest residue and the exact composition of this fuel is unknown. Because the predicted values are very sensitive to the kinetics, an over-prediction of less than 10% is realistic. Because the trends and the phenomena seen in the experiments can be predicted by the theory, it is believed that the model is physically sound. To make more accurate predictions, more research on the details of the reactions is needed.

6. Conclusions

Several conclusions can be drawn from the experiments and analyses carried out in this work. According to the objectives of the work, the conclusions can be categorized in three main parts: (1) general results, (2) influence of key parameters and (3) spontaneous ignition mechanisms. The general conclusions which can be drawn from the experiments are summarized in Table 2. In the experiments the effect of several key parameters on the ignition
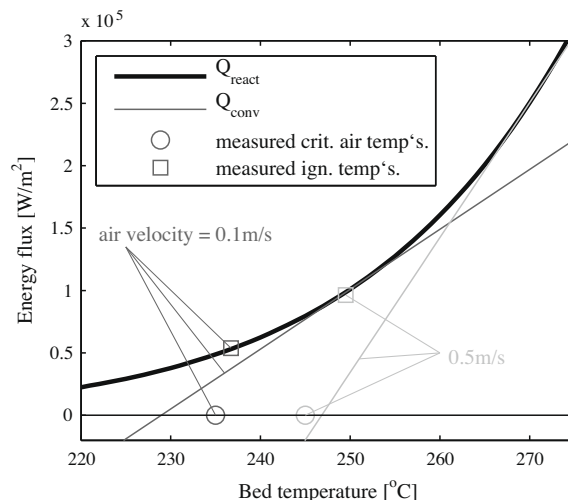

Fig. 20. Semenov's analysis applied to the spontaneous ignition of wood for three air velocities.

Table 2
Measured critical air and spontaneous ignition temperatures and ignition times for wood, char and RDF.

	Wood chips	Char	RDF
T_{crit} (°C)	230–245	170–200	205–200
T_{ign} (°C)	230–255	155–190	220–245
ign. time (min)	5	2	60

behaviour is investigated. The results can be summarized as follows:

- The primary air velocity has a significant effect on both the critical air temperature and the spontaneous ignition temperature. Increasing the primary air flow velocity increases both the temperatures. For high flow velocities the critical air temperature and spontaneous ignition temperature tends to become constant.
- Addition of inert to the wood and char fuel bed increases the spontaneous ignition temperature as well as the critical air temperature needed for the ignition.
- No significant influence of moisture content (up to 30 wt%) on spontaneous ignition behaviour for wood chips has been found.
- No significant difference in spontaneous ignition behaviour between small (10–15 mm) and medium (15–30 mm) sized char particles has been measured. No reliable results could be obtained with large (30–40 mm) sized char particles.

With the help of Semenov's analysis of thermal explosions the mechanisms determining the spontaneous ignition behaviour are derived. The main conclusions are:

- The spontaneous ignition behaviour is determined by the combination of convective heat transfer between the primary air flow and the fuel bed and the heat gained by the reactions in the fuel bed.
- For wood, the heat gained by the reactions is determined by the exothermic hemicellulose pyrolysis. This causes the spontaneous ignition temperature to be higher than the critical air temperature.
- For all experiments with char, the spontaneous ignition occurs at a temperature below the air temperature irrespective of the air flow velocity. If ignition did not occur, the bed temperature

will be below the air temperature. This indicates that until ignition the reactions in the char are endothermic. This idea is supported by DSC measurements on char.

- RDF shows a typical pre-ignition stage. This is probably caused by the inhomogeneous composition of this fuel.

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