

MATHEMATICAL MODELLING OF LIGNITE COMBUSTION IN A BED An One-Dimensional Approach

by

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Thermal plant design and choice of suitable technology requires the understanding of the fuel thermo-chemical properties. The reactions that consist of different phases may be approached in two ways. One is to determine the kinetics of each individual reaction and its interaction with the environment. The other method is to use an integrated approach, when, based on the results, the summary formula for the behaviour of combustible matter is obtained. This is an empirical approach. Despite possible objections, particularly regarding its accuracy, the narrow range of applicability, etc., the attractiveness of this procedure is evident due to direct results, simplicity and far lower costs to the experiment. That is the reason why this paper focuses on determining the kinetic parameters of the integrated model of group reactions that are part of the combustion of lignite at low temperatures. The paper develops a simple mathematical model of the combustion process for lignite in bed which is then confirmed by modelling the experimental results.

Key words: lignite, combustion, mathematical model, experiment, solid fuel

Introduction

The aim of this paper is to suggest a mathematical model of lignite combustion in a bed. As noted in [1], *modelling and simulation are always target driven processes*, so, before making any decisions about how to approach this task, it is necessary to have a clear idea of the problem and the purpose of the potential models.

The best model is the simplest model that still serves its purpose, that is, which is still complex enough to help solve the original problem, Velten [2].

Solid fuel combustion in fixed bed is practically the oldest technique of thermo-chemical conversion so it is not surprising to find a multitude of works in the literature in this area. The initial correlation between the intensity of combustion (heat output of combusted fuel – the amount of heat released per unit of time) and the flow of fresh air was published by Rosin and Kayzer [3] as a result of experimental tests. This model is based on the hypothesis that the resulting rate is limited by a slower process, and that is the diffusion rate of oxidant (oxygen) through the boundary layer from the free flow to the surface of the reaction against the chemical reactions. Following this, the resulting combustion rate is equal to the diffusion rate, which is in engineering applications examined through experimental procedures. The theoretical basis for this hypothesis based on the theory of the boundary layer was formed by

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Mondiez [4], and shortly afterwards, Spalding *et.al* [5] elaborates this idea within a consistent theoretical context. It seems that, as far as combustion of coal in a fixed bed is concerned, the interest of industry for this class of problems ceased, so that today it is almost impossible to find work related to this topic.

An overview of various models for the combustion of biomass at the mechanical grate is given in [12]. A moving grate [6-8] is a typical example of combustion in a fixed bed (the grate *carries* a bed through the combustion chamber, while the bed does not move). In contrast, other mechanized grates (*ibid.*) cause displacement in the fuel bed. The mechanical displacement in the fuel significantly changes the burning situation and it is quite obvious that each specific case requires a new model. However, combustion in the elemental segment of bed can provide information about the possible behaviour of fuel for various cases. This can also provide information for composing a suitable fuel mixture for a particular combustion chamber [9].

On the other hand, over the last 30 years (and more) the complete combustion engineering practice related to solid fuels has dramatically changed [10]. Besides permanent changes in economy and politics, agents of this change are also related to the significant advances in science and the knowledge about the characteristics and methods of characterization of fuel. In addition to traditional analysis: proximate and ultimate analysis, ash elemental analysis, *etc.*, there is growing emphasis on the information on the structure and reactivity of different types of coal and other solid fuels ranging from biomass and peat to waste coal and petroleum char. The methods of analysis increasingly include measurement of pyrolysis and oxidation, as well as kinetics, inorganic reactivity, and much more.

The method presented in this paper belongs to the group of thermal mechanical/gravimetric analysis (TMA, TGA). Classical TGA analysis [11] is performed with samples < 1 g, so that the results reproduce more kinetics parameters of chemical reactions, whereas the effects of diffusion are negligible. The mathematical model which is developed and presented here directly builds on the hypothesis of the dominant influence of diffusion in the real bed combustion. These technologies aid in fuel characterization as well as in the analysis of combustion mechanism.

In contrast to the approaches presented in [12-14], which develop more or less complicated models that try to reproduce reality as closely as possible, in this work we propose a fast method, that is adapted for specific fuel and for specific conditions and is simplified as much as possible. As modern industrial practice is daily confronted with significant fluctuations in fuel quality, such a test would improve the quality of plant operation.

Mathematical model

Phenomenological characteristics of lignite combustion

The fact that lignite is the predominant type of coal in the Balkan region is the reason why it is used in this paper. Proximate analysis of working fuel [10] when a rough separation is done on the pit indicates a high moisture content (possibly > 40%), a large proportion of volatiles (50-60%, dry and ash free of the combustible mass), lower content of char residue and a small to moderate mineral matter content. The prime of which it originated is mostly vegetable, with a small admixture of animal organisms. Heating value (net calorific value) ranges from 23.000 to 25.000 kJ/kg (dry, ash free).

The temperature at which the devolatilisation process begins is slightly higher than the drying temperature (130-170 °C), and ends at 1000-1100 °C. The initial condensation

transformation of carbonaceous matter is followed by the separation of water molecules, and then CO and CH₄. At temperatures above 500 °C more complex hydrocarbons and oxygen compounds are separated. Sulphur is partially separated in the form of hydrogen-sulphide. Kinetics of devolatilisation process (amount and composition of volatiles) depends on various factors: the degree of carbonification (rank of coal), petrographic composition, the quantity of sulphur and other minerals, as well as the degree of oxidation of the fuel.

Volatiles generally have a major impact on ignition, and consequently, on the whole combustion process. The higher their content, the easier it is to ignite the fuel and provide spacious, long and bright flames. The fuel with such properties is referred to as highly-reactive fuel. Fuels with low content of volatiles – low-reaction fuels – are more difficult to ignite, burn more slowly and give a brief flame. This last fact warns that *it is not possible to burn any fuel in any combustion chamber in a high quality and efficient manner*. The knowledge of the particular situation gives crucial guidance for composing the right fuel that will produce the desired results.

A fuel with specific characteristics, such as lignite, is essentially a solid porous material. The combustion of this fuel, which is also rich in volatiles with low-temperature separation, certainly presents a very specific problem. When such a fuel is subjected to intense thermal action of an external source, this initially causes decomposition of the surface layer of the material that results in the release of gaseous products (volatiles) and the formation of the solid (char) on the surface of the fuel. The possibility of the liquid fraction is by definition a priori not taken into account. The volatiles are mixed with the surrounding air and burn at some distance above the surface of the fuel, while the pyrolysis zone moves inwards, under the char, with the progress of the temperature field. For steeper temperature gradients within the material, pyrolysis zone will also be wider.

In the first stage of the process when the release of volatiles is intensive so that the diffusion processes limit the speed of combustion, the flame zone is formed in the gas space above the fuel bed. The penetration of the reaction front into the solid bed leads to char layer becoming wider, increasing the diffusion resistance of volatiles through the pores of char and reducing the amount of volatiles that reach the flame zone. For this reason volatile flames come closer to the surface. At one point the flames can be transferred to the surface of char residue, and then in the interior of solids. Then, the second type of combustion starts, *i. e.* the combustion in the pores.

As clearly stated previously, the centre of research and primary focus of this paper is on the low-temperature area dominated by pyrolytic reactions, although, due to the presence of oxygen, the influence of oxidative processes must also be considered.

Model of static relations

In real life situations, in which hundreds of elementary reactions happen, partialisation (as described earlier by the phenomenological characteristics), is recommended if it is fundamentally important for problem solving. Also, this is only possible when the entire problem is defined, *i. e.* when the device – combustion chamber is completely concretized. General method of solving problems, which is applied in this work, introduces operations with the net effect of a simple reaction type.

Combustion is a process of uniting two matters, *i. e.*, a process of synthesis. Pyrolysis is a process of decomposition, *i. e.*, a process of analysis. Without further going into the complexity of the phenomenon, it was found that pyrolysis dominates at low temperature reactions, thus for the basic model of all phenomena the decomposition equation is postulated in the form:



The corresponding single stage stoichiometric equation with this model is in the form:



Since the elemental composition of the fuel is known, as well as the composition of the solid residues, the composition of volatiles can be determined from the condition of conservation of the number of atoms. Based on this, it is possible to determine all of the suspended stoichiometric coefficients.

In the case when the equation is expressed in terms of 1 kg of fuel, the relation between the initial and final states can be written in the weight proportions:

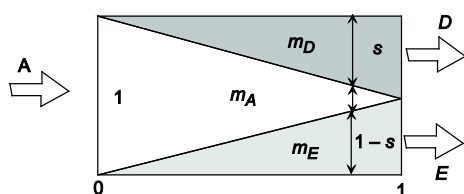
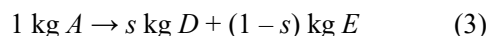


Figure 1. Schematic representation of the model pyrolysis – decomposition of the volatile fuel and solid residue

where s is the mass of volatiles liberated per unit mass of fuel. The model of thus described pyrolytic reaction is schematically shown in fig. 1.

Due to the conservation of mass, if the process is stationary, and if there are no sources or sinks of matter in the system, a mass proportion of the fuel at any stage of the reaction can be expressed as a function of the products according to:

$$X_A = 1 - X_D - X_E \quad (4)$$

During the reaction, the mass proportion of unreacted fuel is changed from 1 to 0, while the proportion of volatiles changes from 0 to s . At the same time the proportion of the solid changes from 0 to $(1 - s)$. The proportions of mass fraction are linear formations:

$$\frac{X_E}{X_{E1}} = \frac{X_D}{X_{D1}} \quad (5)$$

and are functions of the degree of progress of the reaction, wherein the ratio can be set:

$$X_{E1} = 1 - s \quad (6)$$

and after the completion of the reaction (state 1):

$$X_{D1} = s \quad (7)$$

Solving eqs. (5)-(7), it is possible to get a look-dependence of mass fraction of residual unreacted fuel from separated volatiles:

$$X_A = 1 - \frac{X_D}{s} \quad (8)$$

In the physical sense, this idealization allows us to establish the relations between the real phenomena and the model of decomposition of the resulting substance in the two products. In the model only three substances are identifiable: fuel, volatiles and solids.

The kinetics of chemical reactions

Chemical reaction of *real rate* gradually breaks down the molecules of a substance, and simultaneously generates molecules of other substances. The term *reaction rate* means a

change in the amount of some substance that is present in a volume, per time unit. If the amount of matter is expressed as mass fraction, the rate of chemical reactions of the i component can be expressed in the following form:

$$\dot{m}_i = \rho \frac{dX_i}{d\tau} \quad (9)$$

On the basis of the law of conservation of matter for the pyrolysis reaction, following the stoichiometric eq. (2), there is a relation between the rate of formation and disappearance of certain components:

$$-\frac{\dot{m}_A}{\nu_A} M_A = \frac{\dot{m}_D}{\nu_D} M_D = \frac{\dot{m}_E}{\nu_E} M_E \quad (10)$$

Relationships (10) give the relations that enable the determination of any parameter of the process by measuring the changes in the mass fraction of only one component.

Mathematical model of the lignite combustion

Starting from the equation for 2-D gas diffusion, we can obtain changes in the mass of unburned fuel that is a function of time, *i. e.*, the rate of change of fuel mass over time. The general form of the equations of gas diffusion can be presented in the form of continuity equation of mass transport:

$$\frac{\partial \rho}{\partial \tau} + \frac{\partial}{\partial x}(\rho w_x) + \frac{\partial}{\partial y}(\rho w_y) = 0 \quad (11)$$

In the case of the specified materials (substances) the eq. (11) becomes:

$$\frac{\partial(\rho X_i)}{\partial \tau} + \frac{\partial(\rho w_x X_i)}{\partial x} + \frac{\partial(\rho w_y X_i)}{\partial y} = \frac{\partial}{\partial x} \left(\rho D_i \frac{\partial X_i}{\partial x} \right) + \frac{\partial}{\partial y} \left(\rho D_i \frac{\partial X_i}{\partial y} \right) + \dot{m}_i \quad (12)$$

where i are specifies substances (oxygen, volatile, and products of chemical reactions).

Volumetric rate of matter formation \dot{m}_i , may be a complicated function. Unfortunately, real chemical reactions in coal are generally multistep reactions, and cannot be adequately described by simple Arrhenius formula. The kinetics of particular reactions depends on the chemical, physical, and thermo-physical conditions of combustion and basically it is not well explained.

In this article, an extremely idealized 1-D, uniform system of combustion of porous materials will be considered. Oxygen comes into contact with fuel, solely by diffusion to the flame location, where it reacts immediately. The location of the flame is determined by the co-ordinate x and is moved from the surface of the porous bed ($x = 0$) to the depth of the porous bed ($x = L$). We will assume that the flame front (*i. e.*, reaction location) moves at a constant rate, and that density and diffusion coefficient have a constant value.

In this simplified case, diffusion eq. (12) takes the following form:

$$\rho D_i \frac{\partial^2 X_i}{\partial x^2} + \dot{m}_i = 0 \quad (13)$$

The application of such a law on the volatile gives:

$$\rho D_D \frac{\partial^2 X_D}{\partial x^2} + \dot{m}_D = 0 \quad (14)$$

It then follows:

$$d\left(\rho D_D \frac{dX_D}{d\tau} \frac{d\tau}{dx}\right) = -\dot{m}_D dx \quad (15)$$

As, according to eq. (9) the flux of volatiles per unit of volume is equal to the change in the mass fraction per time:

$$\dot{m}_i = \frac{\partial}{\partial \tau} (\rho X_i) = \rho \frac{\partial X_i}{\partial \tau} \quad (16)$$

According to the assumption, the rate of displacement of flame front is constant:

$$w = \frac{dx}{d\tau} = \text{const.} \quad (17)$$

Inserting eqs. (16) and (17) in eq. (15) we obtain the form of law in the differential equation form:

$$d\left(D_D \dot{m}_D \frac{1}{w}\right) = -\dot{m}_D dx \quad (18)$$

By the separation of variables and by integration:

$$\int_{\dot{m}_{D0}}^{\dot{m}_D} \frac{d\dot{m}_D}{\dot{m}_D} = -\int_0^\tau \frac{w^2}{D_D} d\tau \quad (19)$$

we obtain the dependence of the logarithm of the mass flux of the separated volatiles upon time:

$$\ln\left(\frac{\dot{m}_D}{\dot{m}_{D0}}\right) = -\frac{w^2}{D_D} \tau \quad (20)$$

That is, an exponential function of the mass flux (the rate of chemical reactions) of volatiles per time:

$$\dot{m}_D = \dot{m}_{D0} \exp\left(-\frac{w^2 \tau}{D_D}\right) \quad (21)$$

Including the relation eq. (10) in eq. (21) and shortening the stoichiometric coefficients, it follows that the formula can also be transformed for fuel, and then the rate of reduction of the unreacted fuel is:

$$\dot{m}_A = \dot{m}_{A0} \exp\left(-\frac{w^2 \tau}{D_A}\right) \quad (22)$$

Now we can obtain the final form of the mathematical model that describes the reduction of unreacted fuel in the combustion process.

By the separation of variables and integration in the boundaries from a point in time, τ , to the end of combustion process ($\tau \rightarrow \infty$), we get the final form of the mathematical model of the still unreacted fuel up to the moment, τ :

$$m_A = m_{A0} \exp\left(-\frac{w^2 \tau}{D_A}\right) \quad (23)$$

Current value of the mass concentration of the still unreacted fuel is an exponential function of time and is in direct correlation with the total remaining solid mass that can be measured by a simple scale. With the increase of combustion time the mass concentration of unreacted fuel decreases from the initial value X_{A0} ($\tau = 0$) and tends to 0 when τ tends to infinity.

Equation (23) is the central hypothesis of the paper and a hypothetical law of the changes of the mass of the solid phase which needs to be experimentally tested.

Formal simplex w^2/D is a function of summary characteristics of fuel in a bed: type of coal (coal rank), granulation, porosity, density, moisture content, calorific content and quality of ash, *etc.*, and also of thermo-physical condition in the space of combustion chamber above the burning bed: temperature, air and flue gases velocity, *etc.* The project of the specific experiment is aimed to determine which parameters should be varied, and which should be kept at a certain (projected) level.

Experimental tests

In order to confirm the model, it was necessary to compare the obtained results with the results obtained in measurements in real combustion chamber. One cycle of measurement, *i. e.*, testing from ignition to the extinction was done only on a single charge of 4 kg lignite sample in the 200 mm high bed [15].

Material and experimental set-up

This experimental study used an adapted domestic furnace (which is normally used for residential heating). The cross section of the combustion chamber and the combustion grate was 200×200 mm, while the draft of the furnace with its longitudinal section and cross section is shown in fig. 2.

The furnace was placed on the scales with automatic adjustment level which allowed the necessary precision in measurements. For the purposes of measurement standard laboratory instrumentation (laboratory class precision) was used.

The fuel sample used in the experiment was air-dried Koluvara lignite, lump size 25-0 mm (*nut*). Its proximate analysis is given in tab. 1, and its ultimate analysis in tab. 2.

Fire wood was used for kindling and its net heating value was $H_d = 11400$ kJ/kg. Petroleum (fuel oil) was used for initial firing from ashtrays. Net heating value of petroleum was $H_d = 42700$ kJ/kg. One charge, which was used for the experiment, in total, consisted of:

Lignite	4000 g
Wood	100 g
Petroleum	45 g
Total	4145 g

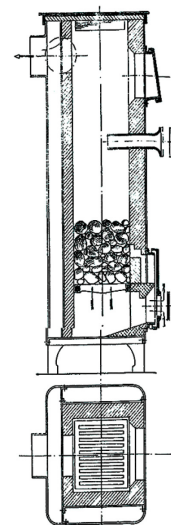


Figure 2. Cross-section of the combustion chamber and the combustion grate [15]

Table 1. Proximate analysis of lignite
(%-mass, air dried)

W [%]	A [%]	V [%]	C _{fix}	H _d [kJkg ⁻¹]
10.51	11.92	44.31	32.73	20338

Table 2. Ultimate analysis of lignite
(%-mass, air dried)

C [%]	H [%]	S [%]	N [%]	O [%]
50.18	4.19	0.47	0.59	22.08

Before inserting the measured quantities of fuel in the combustion chamber and starting the experiment, the furnace was heated to operating temperature. Regular testing of a working point ran from the moment of ignition until the end of combustion. The experiments were carried out without any intervention by the stoker, from the beginning to the end of the experiment.

The results – measured combustion reaction rates

The measurements of mass change during the test are given in the diagram in fig. 3. As is shown, the data measurements were taken for every 200 g of burned material and only at the end of the process, the values were reduced to 150 g and 20 g. The data set was measured in the total of twenty points. The same chart also shows the functions of mass loss rate during the experiment.

During the experiment other process parameters (temperature and composition of flue gases, [15]) were also recorded; however, they are irrelevant to this report.

Estimation of parameter for combustion rate equation

Now, the experimentally obtained results can be easily modelled by the derived eq. (23) and thus, determine exponent w^2/D_A for the particular combustion conditions and fuel characteristics. The estimation of the parameters is formulated as an optimization problem which is minimized with respect to the parameter. The function expresses the sums of squares of the measured and calculated mass loss curve. Using the simple method for the given conditions, in the case of the fuel used in the experiment, the following value is calculated (MATLAB):

$$\frac{w^2}{D_A} = 0.0341 \quad (24)$$

When the measured and calculated values of mass loss, from eqs. (23) and (24), are inserted in the joint diagram in fig. 4, a good agreement between the simulated curve and the actual results can be observed.

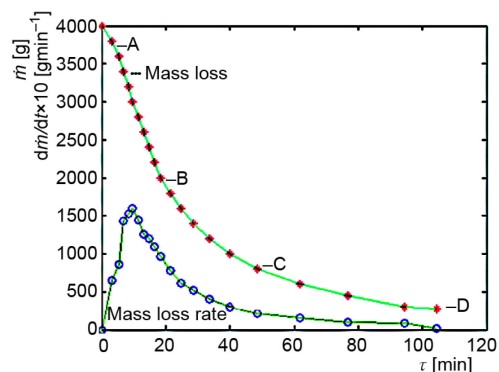


Figure 3. Measured mass loss and mass loss rate during the time

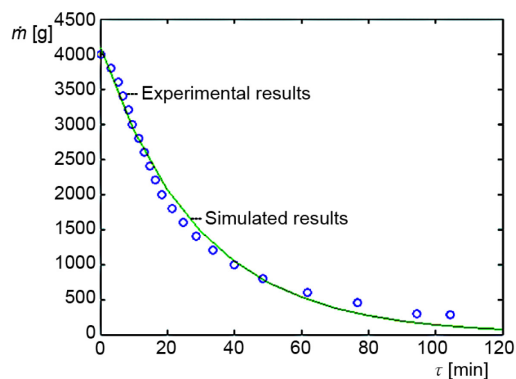


Figure 4. Experimental and simulated results of mass reduction

Discussion

In order to explain the combustion process, the combustion curve which was obtained in the experiment will be analysed. According to the curve shape, it can be divided into three sections (fig. 3.).

- The first part of the curve, from point A to point B, starting with a gentle slope (as a result of kindling), and then a steeper slope to the τ -axis indicates the fast fuel devolatilization process at the beginning of the combustion process when the mass of the fuel on the grate is almost halved. On the combustion curve, point B marks the moment when 2000 g of fuel burned, which was after 18 min 30 seconds.
- After the moment B, the process slows down, and this continues up to the point C. The second part of the curve, that is the part marked BC, has a particular curve with a relatively small radius of curvature. This part of the curve appears as a link between the sections AB and CD.
- At the end of the process, the fuel burns up to the point D, when the test is completed. The third part of the curve, namely section CD, is similar to the typical shape of the curve of char burning. This part of the curve, with a very slight curve, gently sloping toward the τ -axis, indicates that the process finishes relatively slowly. During the third stage, which lasted 54 min 45 seconds, 520 g of fuel burned.

The combustion curve also suggests that the maximum speed of combustion of the entire fuel happened during the first phase AB. During the third phase, which lasted 54 min 45 seconds only 520 g of fuel was burned, whereby the highest temperature of the fire was recorded, *i. e.* it provided the best conditions for the rapid reaction of the heterogeneous combustion of carbon in the form of char.

It can be concluded from the previous phenomenon flow analysis of the lignite burning sample, that the release of volatiles to a large extent limits the overall kinetic of coal combustion. More accurately, it creates a volatile layer (cloud) that limits the diffusion of oxygen to combustible substances.

As already noted in the *Introduction*, today's models tend to capture as many phenomena as possible, which inevitably leads to complexities. A 2-D model of straw combustion presented in [8] demonstrated that modelling can provide information about aspects such as temperature profile of the fuel bed, combustion rate and the produced chemical species which would not be available otherwise. But such a model requires the use of high-speed computer in order to obtain acceptable results in a short time. In addition, it requires the knowledge of the characteristics and parameters that are unavailable in today's practice. This is of course not acceptable in industrial practice.

The group of simple models in [16] assumes that the process is comprised of two parallel reactions with the respective pre-exponential factors and activation energies. Each sub-reaction describes the pyrolysis of volatile components with distinct reactions kinetics. A similar procedure was demonstrated in [11], but the obtained approximation was inadequate. Optimization problem of determining the combustion curve parameters is a little more complicated, however, when it comes to diffusion there is no physical basis for such approximation.

Since the content of volatiles in coal varies, (primarily as a function of age) it is logical to expect that the parameters of diffusion in eq. (23), are also variable. For this reason, recommendations for future research is to test first of all, the impact of coal rank (as well as other solid fuels) and then the other factors such as moisture content, granulation (porosity) and air flow rate through the bed.

Finally, to definitely prove the hypothesis, a statistically relevant number of experiments (at least seven) for each factor should be performed.

For this, it is also necessary to build a specially designed experimental facility as was done, for example [17, 18].

Conclusions

The hypothesis is set in this paper that the resulting combustion rate, which is directly correlated with the change in the mass of unburned fuel, is limited by the diffusion of the oxidant (oxygen) through the boundary layer from the free flow to the reaction surface. This hypothesis leads in a simplified and 1-D interpretation to the results in the form of simple exponential dependency of function of fuel mass per time.

The experimental method presented in the article belongs to a group of TMA, TGA analysis which should be done with macroscopic samples. The results of the experiment involving the combustion of 4 kg of lignite sample showed good agreement with the characteristics of the model.

These technologies enable the characterization of fuel, and can be used for analysing combustion in different processes (mechanisms) for starting the reaction layer. For a quick evaluation of the combustion kinetics, without going into the details of the process, this type of modelling provides fast, efficient, and useful solutions, a fact which is very important in industrial practice.

To definitely prove the hypothesis, it would be necessary to perform statistically relevant number of experiments for each factor. In addition, building a specially designed experimental facility would also be required.

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Nomenclature

A	– fuel, in fig. 1
A	– ash content, [%-mass], in eqs. (2) and (3), and tab. 1
C_{fix}	– fixed carbon
D	– volatile, in fig. 1 and eqs. (2) and (3)
E	– solid residue, in eqs. (2) and (3), and fig. 1
Hd	– lower (net) heating value, [kJkg^{-1}], in tab. 1
L	– bed height, [ms]
M	– molecular weight, [kgmol]
\dot{m}_i	– reaction rate, [$\text{kgm}^{-3}\text{s}^{-1}$]
s	– the stoichiometric ratio of volatiles and fuel mass, eq. (3)
w	– flow rate (x- or y-direction), [ms^{-1}], eq. (11), flame locations velocity, [ms^{-1}], eq. (18)
X	– the mass fraction

Greek symbols

τ	– time, [min]
ρ	– density, [kgm^{-3}], eq. (11)
ν	– stoichiometric coefficients, eq. (2)

Subscripts

A	– fuel
$D, D1$	– volatile, volatile at the end of the process
$E, E1$	– solid residue, solid residue at the end of the process
i	– species index
x, y	– Cartesian co-ordinates

Acronyms

TGA	– thermogravimetric analysis
TMA	– thermomechanical analysis

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