Modelling of Gasification of Refuse-derived fuel (RDF) based on laboratory experiments

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Abstract

In this work, a model of gasification process that enables all, material and energy balance, calculation of gas composition, gas heating value and reactor temperature is presented. The model parameters were estimated by laboratory experiments. A fraction of MSW generally separated from inorganic materials and biodegradable components, so called Refuse-derived fuel (RDF), was studied by material characterization methods and laboratory scale experiments. Composition of RDF was determined by the separation of a representative sample into its basic components (paper, foils, hard plastics, textiles), which were homogenized by grounding in a cryogenic mill. All components of RDF and also a representative mixed sample of RDF were studied by thermogravimetric (TGA) analysis, differential scanning calorimetry (DSC), elemental analysis and bomb calorimetry to determine the kinetics of thermal decomposition, proximate and elemental composition, heat of the reaction and higher heating value. The pyrolysis and gasification experiments were performed in a laboratory scale reactor. The amount of tar in raw produced gas was determined. Industrial scale gasification process were studied by mathematical modeling and computer simulation. All processes, gasification with air, gasification with oxygen and gasification with both oxygen and steam were investigated under different conditions. RDF conversion of 100% was achieved by gasification with air at the air to RDF mass ratio of 2.2. However, the gas heating value was only 4.6 MJ/Nm³. Gasification of RDF using oxygen enables producing a gas with the heating value of around 10 MJ/Nm³ at the oxygen to RDF mass ratio of 0.5. By increasing the steam to RDF mass ratio, the content of H₂ and CO₂ increased, however the content of CO, reactor temperature and gas heating value decreased.

Keywords

Refuse-derived fuel, RDF, Gasification, Modelling

Introduction

Refuse-derived fuel (RDF) is a fraction of municipal solid waste (MSW) which is received after separation of inorganics and biodegradables. RDF represents a considerable part of MSW and contents mainly thus components of MSW that
represent the largest environmental challenges. Synthetic and natural polymer materials presented in RDF naturally degrade within a very long time and require very large spaces in the case of landfilling. Its heterogeneity and composition variations are important challenges for its mechanical recycling. On the other hand this fraction of MSW represents a valuable source of material and energy. Feedstock recycling methods such as pyrolysis and gasification have a big potential to be used for recovery of valuable chemicals and energy from RDF. Gasification of RDF results in the production of a combustible gas containing mainly H₂, CO, CO₂ and N₂ if air is used as the gasification agent. Some methane, light hydrocarbons, H₂S, NH₃ and tar are also present in the gas [1].

A number of power generation plants based on gasification technology are in operation worldwide. A list of operating gasification plants has been provided by Higman and Van der Burgt in [2].

Modelling of gasification enables predicting of optimal process conditions and reducing the number of experiments in the processes of design and operation. A simple equilibrium mathematical model based on minimisation of the Gibbs free energy was used by some authors to model the gasification of RDF. Materazzi et. al. [3] used the equilibrium model to evaluate a two stage waste gasification system. From their work it results that, except for H₂, the predicted composition of gas is in good coherence with the experimental data. The equilibrium model was also applied by Barba et. al. for RDF gasification [4] and Jarunthammachote and Dutta [5] for the gasification of charcoal and coconut shell. The authors found that the model data are in good agreement with the experimental ones. However, Li et. al. [6] applied an equilibrium model for coal gasification and declared that this model has some limitations in predicting gas composition. Generally, the equilibrium model provides good results at temperatures above 1000 °C and it fails at the reactor temperatures below 800 °C [7-8].

In this work, we combined the equilibrium model with some empirical correlations based on laboratory experiments to predict the carbon conversion, reactor temperature, gas composition, gas heating value and gas tar content under different types and amounts of the gasification agents. Input data for the RDF gasification model were determined by proximate and elemental analysis, calorimetric analysis and tar content measurement.

**Raw Material**

RDF provided by a Slovak waste processing company contained around 21 wt. % moisture. Composition of dried RDF was estimated by hand separation of a representative sample of RDF into its basic material categories. Four different material categories were identified: Paper 63.2 wt. %, Polyethylene foil 15.8 wt.%, Hard plastics 19.1 wt.% and Textile 2.0 wt.%.

A sample of wet RDF (5 kg) was homogenised by grounding to particles of sizes less than 1 mm. 100 g of grounded RDF was rather homogenized by grounding in a cryogenic mill. During the homogenization the moisture content of RDF decreased to 10 %.
Experimental part

Elemental analysis

Dry basis elemental composition of RDF was estimated by a Vario Macro Cube ELEMENTAR elemental analyzer. A CHNS (Carbon, Hydrogen, Nitrogen, Sulphur) module with the combustion tube temperature of 1150 °C and the reduction tube temperature of 850 °C was used. The module did not enable the determination of the Cl content. The mass of the sample was around 1 g. Results of both proximate and elemental analyses are given in Table 1.

Table 1: Proximate and elemental composition of RDF

<table>
<thead>
<tr>
<th>Component</th>
<th>Moisture</th>
<th>VM*</th>
<th>FC*</th>
<th>ASH*</th>
<th>C</th>
<th>H</th>
<th>N</th>
<th>S</th>
<th>O**</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wt. %</td>
<td>10</td>
<td>75.5</td>
<td>8.9</td>
<td>15.6</td>
<td>51.7</td>
<td>5.9</td>
<td>0.9</td>
<td>0.4</td>
<td>25.5</td>
</tr>
</tbody>
</table>

*massive free basis

**calculated to 100%

Based on the elemental composition of RDF shown in Table 2 and the molecular mass of elements, a summary formula of RDF (dry ash free) in the form of $C_aH_bO_cN_dS_e$ was proposed. The final molecular formula of the RDF used is $CH_{1.3599}O_{0.3703}N_{0.0149}S_{0.0029}$ and its molecular mass based on one carbon is 19.607 g/mol.

Thermogravimetric analysis

Using simultaneous thermogravimetric (TG)/differential scanning calorimetric (DSC) measurements (Netzsch STA 409 PC Luxx. Germany), the proximate analysis of RDF was done (Table 1). Experimental conditions were met at the linear heating rate of 5 °C min⁻¹ in the nitrogen flow of 60 ml h⁻¹. The samples were heated from 20 °C to 800 °C; at this temperature, they were maintained for around 30 minutes and then they were combusted by entering the oxygen to the system. Samples of RDF, of around 20 mg, were used in the TG/DSC measurements.

Figure 1 shows the behaviour of thermal decomposition of an RDF sample. Four different decomposition steps were observed. The first one at temperatures around 100 °C corresponds with the evaporation of water. The second decomposition region between temperatures 250-350 °C shows the decomposition of cellulose and paper fraction of RDF. In the temperature range of (400-500 °C), plastic fraction of RDF (PE, PP, PS) is decomposed. The last decomposition step at temperatures around 700 °C is probably caused by the decomposition of inorganic salts such as CaCO₃.
RDF Heating value

Higher Heating Value (HHV) of RDF was measured using an FTT isoperibolic calorimetric bomb, Fire Testing Technology Limited. Combustion of the sample took place in a calorimetric bomb under oxygen atmosphere at 30 bars. The sample mass was around 1 g. Benzoic acid was used as a standard material. To eliminate the effect of sample heterogeneity, four different samples of RDF were taken for this measurement; in addition, each measurement was repeated at least three times, the allowed variation between the measurements was 0.2 MJ/kg. As results from Table 2, average HHV was 20.81 MJ/kg.

Table 2: HHV of RDF

<table>
<thead>
<tr>
<th>Run</th>
<th>HHV [MJ/kg]</th>
</tr>
</thead>
<tbody>
<tr>
<td>RDF 1</td>
<td>21.15</td>
</tr>
<tr>
<td>RDF 2</td>
<td>21.62</td>
</tr>
<tr>
<td>RDF 3</td>
<td>22.17</td>
</tr>
<tr>
<td>RDF 4</td>
<td>18.3</td>
</tr>
<tr>
<td>Average</td>
<td>20.81</td>
</tr>
</tbody>
</table>

Tar content measurement

RDF sample of 20 g was subjected to gasification in a laboratory batch tube furnace. Air velocity was 15 l/hr and the reactor temperature varied from 700 °C to 1100 °C. The produced gases were condensed in a series of six scrubbers filled with isopropanol. Four scrubbers were kept at 30 °C and the last two ones at -20 °C. After each experiment, the content of all scrubbers was mixed and a 50 ml sample was taken for distillation. Vacuum distillation was carried out at the pressure of 10 kPa until complete distillation of isopropanol, water and light components. The amount of distillation waste represented the Tar obtained from 50 ml of the solution. The amount of Tar obtained from a 20 g sample was determined proportionally. Table 3 shows the
tar content based on the feed mass of the raw untreated gas produced by gasification of RDF versus the reactor temperature:

**Table 3: Producer gas tar content**

<table>
<thead>
<tr>
<th>Temperature [°C]</th>
<th>700</th>
<th>800</th>
<th>900</th>
<th>1000</th>
<th>1100</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tar [mg/g RDF]</td>
<td>81.5</td>
<td>71</td>
<td>61</td>
<td>36.5</td>
<td>30.5</td>
</tr>
</tbody>
</table>

**Gasification model**

The following assumptions were made in modelling the gasification process of RDF:
- steady state flow is considered inside the gasifier
- no temperature and concentration gradient exist inside the reactor
- the residence time is long enough to reach complete decomposition of RDF and unreacted part of RDF is only carbon.
- only the major species are considered in the product gases, i.e CO, CO$_2$, H$_2$, CH$_4$, H$_2$O, NH$_3$, H$_2$S, N$_2$ and Tar.

Then, the overall material balance of RDF gasification can be written as:

$$
CH_4O_N_S_{x_1} + x_2O_2 + x_3H_2O \rightarrow x_4CO + x_5CO_2 + x_6H_2 + x_7CH_4 + x_8H_2O + x_9NH_3 + x_{10}H_2S + x_{11}CH_{b1} O_{d1} N_{d1} S_{d1}
$$

where, $x_1$ and $x_2$ are the number of moles of oxygen and steam used for each mole of RDF and, $x_3$, $x_4$, $x_5$, $x_6$, $x_7$, $x_8$, $x_9$ and $x_{10}$ are the moles of each corresponding species, obtained from each mole of RDF based on a single atom of carbon. $CH_{b1} O_{d1} N_{d1} S_{d1}$ is the empiric formula of tar that was estimated in the same way as that of RDF using results of elemental analysis of tar.

A linear dependence between the tar content and the temperature based on RDF gasification experiments (Table 3) was assumed: $y = aT + b$ where $y$ represents the amount of tar in mg of tar per g of RDF and $T$ the temperature in °C. The constants $a$ and $b$ were estimated by regression of data in Table 3.

Mass balance of individual elements provides five algebraic equations, together with total mass balance, it makes six equations; however, to estimate all ten unknown stoichiometric coefficients, at least ten equations are required. The equilibrium constant reaction of four gasification reactions should be added to the model. The following reactions were selected for this study:

- Boudouard reaction &nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp; C+CO$_2$=2CO
- Shift reaction &nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp; CO+H$_2$O=CO$_2$+H$_2$.
- Methanation reaction &nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp; C+2H$_2$=CH$_4$
- Steam gasification of carbon &nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp;&nbsp; C+H$_2$O=CO+H$_2$

Equilibrium constant of individual reactions is given by:

$$
K_a = \left( \frac{P}{P^0} \right)^{\sum_i n_i \phi_i} \prod_i x_i^{n_i}
$$  \hspace{1cm} (1)

where $K_a$ is the equilibrium constant, $P^0$ the pressure at which $K_a$ is defined [Pa], $P$ the system pressure [Pa], $n_i$ the stoichiometric coefficient of component $i$, $\phi_i$ the fugacity coefficient of component $i$, and $x_i$ the molar fraction of component $i$. 

The value of equilibrium constant is calculated at constant temperature and pressure using the standard state Gibbs free energy \( \Delta r G^{298} \):

\[
K^o_{a298} = e^{-\Delta r G^{298}/RT}
\]

(2)

where \( K^o_{a298} \) is the equilibrium constant at 298 K, \( \Delta r G^{298} \) the standard Gibbs free energy \( [Jmol^{-1}] \), \( R \) the gas constant \( (R = 8.314 Jmol^{-1}K^{-1}) \), and \( T \) is the temperature [K].

Fugacity coefficients were assumed to be equal to 1 in this work. Standard Gibbs free energy was calculated based on the standard reaction enthalpy, \( \Delta r H^{298} [Jmol^{-1}] \), and the standard reaction entropy, \( \Delta r S^{298} [Jmol^{-1}K^{-1}] \) as:

\[
\Delta r G^{298} = \Delta r H^{298} - T \Delta r S^{298}
\]

(3)

The values of \( \Delta r H^{298} \) and \( \Delta r S^{298} \) were calculated using the standard reaction enthalpy and the entropy of formation:

\[
\Delta r H^{298} = \sum v_i \Delta r H_i^{298}
\]

(4)

\[
\Delta r S^{298} = \sum v_i \Delta r S_i^{298}
\]

(5)

where \( v_i \) is the stoichiometric coefficient, \( \Delta r H_i^{298} \) the standard enthalpy of formation \( [Jmol^{-1}] \), and \( \Delta r S_i^{298} \) the standard entropy of formation \( [Jmol^{-1}K^{-1}] \).

Reaction enthalpy and reaction entropy at the temperature of the system can be calculated by:

\[
\Delta r H = \Delta r H^{298} + \sum v_i c_{pi} \cdot (T - 298)
\]

(6)

\[
\Delta r S = \Delta r S^{298} + \sum v_i c_{pi} \cdot \ln \left( \frac{T}{298} \right)
\]

(7)

where \( \Delta r H \) is the reaction enthalpy at temperature \( T [Jmol^{-1}] \), \( \Delta r S \) the reaction entropy at temperature \( T [Jmol^{-1}K^{-1}] \), and \( c_{pi} \) the average molar heat capacity of component \( i [Jmol^{-1}K^{-1}] \).

In a gasification reactor, the reaction temperature is obtained by partial oxidation of the feed. The overall enthalpy balance of the reactor is:

\[
H_{RDF} + H_{O2(air)} + H_{steam} + Q_R = H_{gas} + H_{ash} + H_C + Q_{loss}
\]

(8)

where \( Q_R \) is the heat of reaction \( [J] \), \( H_{RDF} \) the enthalpy of RDF feed \( [J] \), \( H_{O2(air)} \) are the enthalpy of oxygen and air, respectively \( [J] \), \( H_{steam} \) is the enthalpy of water steam \( [J] \), \( H_{gas} \) the enthalpy of gas \( [J] \), \( H_{ash} \) the enthalpy of ash \( [J] \), \( H_C \) the enthalpy of unreacted carbon \( [J] \), and \( Q_{loss} \) represents the heat losses from the reactor \( [J] \).

Neglecting the heat losses, the heat of reaction can be calculated as the difference between the heating values of reactants and products:

\[
Q_R = m_{RDF} Q_{RDF} - \sum (-\Delta r H_i) n_i
\]

(9)

where \( m_{RDF} \) is the mass flow of RDF feed \( [kg] \), \( n_i \) is the mole flow of component \( i \) in the products \( [kmol] \), \( Q_{RDF} \) the lower heating value of RDF \( [Jkg^{-1}] \), and \( \Delta r H_i \) is the heat of combustion of component \( i \) in the products \( [Jkmol^{-1}] \).

If the RDF feed and oxygen (air) are fed to the reactor at the same temperature which is selected as the reference temperature, \( T_{ref} \), then \( H_{RDF} = 0, H_{O2(air)} = 0 \) and

\[
H_{gas} + H_{ash} + H_C - H_{steam} = (T - T_{ref}) \sum n_i c_{pi} + m_{C} \bar{c}_{pC} + m_{ash} \bar{c}_{pash} - m_{steam} \bar{c}_{psteam}
\]

(10)
where $m_{ash}$ is the mass flow of ash [kg], $m_c$ the mass flow of the remaining carbon [kg], $m_{steam}$ the mass flow of steam [kg], $\overline{c}_{pash}$ the specific heat capacity of ash [Jkg$^{-1}$K$^{-1}$], $\overline{c}_{pc}$ the specific heat capacity of remaining carbon [Jkg$^{-1}$K$^{-1}$], and $\overline{c}_{psteam}$ is the specific heat capacity of steam [Jkg$^{-1}$K$^{-1}$].

Substituing equations (10) and (9) into equation (8) for the temperature of products the following results:

$$ T = T_{ref} + \frac{m_{RDF}Q_{RDF} - \left(\sum_{i} (-\Delta H_i)n_i\right) - Q_{loss}}{\left(\sum_{i} n_i\overline{c}_{pi}\right) + m_c\overline{c}_{pc} + m_{ash}\overline{c}_{pash} - m_{steam}\overline{c}_{steam}} $$

(11)

**Results and discussion**

An MS EXCEL subroutine was used to solve the model described above. The subroutine was implemented into the ASPEN Plus simulation environment to use the component property databanks. Three different agents were considered and observed by modelling: gasification of RDF using air, gasification using pure oxygen and gasification using pure oxygen and steam. Effect of the gasification agent (air, oxygen or steam) to RDF mass ratio on the conversion, reactor temperature, gas composition and the gas heating value was observed.

1. Air gasification

Gasification of RDF with the composition described above was observed at different air to RDF mass ratios. Figure 1 shows the composition of produced gases at different air to RDF mass ratio. H$_2$ and CO are the most important components of the produced gas; the content of H$_2$ showed a maximum at $m_{(Air)}/m_{(RDF)}$=1 and that of CO at $m_{(Air)}/m_{(RDF)}$=2.2. The content of methane decreased to a value near zero at $m_{(Air)}/m_{(RDF)}$=2. However, from Figure 2 it results that at these values of $m_{(Air)}/m_{(RDF)}$, the adiabatic reactor temperature is around 850 K and the conversion of RDF is only 50-60%. A 100% conversion was achieved at $m_{(Air)}/m_{(RDF)}$=2.1 at the reactor temperature of around 950 K. The gas heating value, because of high nitrogen content, was relatively low, 4.6 MJ/Nm$^3$ and it remained practically constant for $m_{(Air)}/m_{(RDF)}$ ranging from 1.15 to 2.1, when it rapidly decreased because of the starting CO oxidation to CO$_2$. Low reactor temperature can cause high content of tar in the produced gas. But increasing the reactor temperature requires oxidation of H$_2$ and CO which results in losing the energy content of the gas. Based on this observation, the optimal value of $m_{(Air)}/m_{(RDF)}$ for the gasification of RDF studied in this work was 2.1.

It was found that the RDF elemental composition can significantly affect this optimal value of $m_{(Air)}/m_{(RDF)}$; in our previous work [9], the optimal value of $m_{(Air)}/m_{(RDF)}$ was 3.2 for an RDF sample with different compositions (higher H$_2$ content).
Figure 1. Gas composition at different $m_{\text{Air}}/m_{\text{RDF}}$ values using air as the gasification agent

Figure 2. Conversion and reactor temperature at different $m_{\text{Air}}/m_{\text{RDF}}$ values using air as the gasification agent

Tar content of raw untreated gas decreased with the increasing reactor temperature and the air to RDF mass ratio. At optimum $m_{\text{Air}}/m_{\text{RDF}}=2.1$, the mass fraction of tar was 0.033, which is very high and such a gas must be treated before using in an engine or turbine for electricity generation.
The content of H$_2$S and NH$_3$ decreased with the increasing $m_{\text{Air}}/m_{\text{RDF}}$ (Figure 3); the content of H$_2$S at $m_{\text{Air}}/m_{\text{RDF}}=2.1$ was 0.083 mol%. Combustion of such gas results in the SO$_2$ content in the flue gases of around 280 mg/Nm$^3$, which is above the EU limits for combustion of solid fuels (200 mg/Nm$^3$).

2. Pure oxygen gasification:

Oxygen blown gasification of RDF enables the production of gas with a 2-2.5 fold higher heating value. However, the gasification plant requires an air separation unit. Complete conversion of RDF was achieved at $m_{\text{O2}}/m_{\text{RDF}}=0.44$, the adiabatic reactor temperature at these conditions was around 1100 K and the gas heating value was approx. 10 MJ/Nm$^3$.

The content of H$_2$ showed a maximum at $m_{\text{O2}}/m_{\text{RDF}}=0.22$ (45 mol.%), but the conversion at this point was only 48%. The content of CO showed a maximum at $m_{\text{O2}}/m_{\text{RDF}}=0.59$, (55 mol.%), but the H$_2$ content at this point was 26 mol.% and that of CO$_2$ was 15 mol.% while its minimum value at $m_{\text{O2}}/m_{\text{RDF}}=0.44$ was 12 mol.%.

Based on this observation, Figures 4 and 5, the optimal value of $m_{\text{O2}}/m_{\text{RDF}}$ for gasification of the RDF studied in this work was 0.45. Equilibrium model based calculated gas compositions published by other authors showed a similar trend. However, experimentally measured data showed lower H$_2$ and higher CO$_2$ content of the gas. These differences are significant particularly at temperatures below 800 °C.
3. Steam and oxygen gasification:

The addition of steam to a gasification reactor enables taking care of water-gas shift reaction and steam gasification reaction of carbon. As a result of these reactions, a higher content of H2 and CO2 and lower content of CO in the produced gas were obtained. At constant \( m(O_2)/m(RDF) = 0.45 \), the gas composition, reactor temperature and
gas heating value at different $m_{\text{steam}}/m_{\text{RDF}}$ values were determined. Preheated steam with the temperature of 600 K and the pressure of 12 bar was considered. It was found that by increasing the mass ratio of steam to RDF from zero to 0.5, the content of $H_2$ increased from 38 mol.% to 46 mol.% and that of $CO_2$ increased from 12 mol. % to 23 mo.%. However, the content of CO decreased from 45 mol.% to 28 mol.% the reactor temperature decreased by 78 K and the gas heating value decreased from 9.9 MJ/Nm$^3$ to 8.5 MJ/Nm$^3$. The effect of steam addition to a gasification reactor at $m_{\text{O2}}/m_{\text{RDF}}$=0.45 is shown in Figures 6 and 7.

**Figure 6.** Effect of steam addition to gasification reactor on gas composition at $m_{\text{O2}}/m_{\text{RDF}}$=0.45

**Figure 7.** Effect of steam addition to gasification reactor on reactor temperature and gas Heating value at $m_{\text{O2}}/m_{\text{RDF}}$=0.45
Conclusion

An RDF sample with the moisture content of 10 wt. % and the elemental composition: C: 51.7 wt. %, H: 5.9 wt.%, N: 0.9 wt.% S: 0.4 wt. %, Ash: 15.6 wt. % and O: 25.5 wt.%, was gasified in a laboratory tube furnace to experimentally measure the raw untreated gas tar content. Tar content of raw gas decreased from 81.5 mg/gRDF at 973 K to 30.5 at 1373 K.

Results of experimental measurements were implemented into a mathematical model of an industrial scale gasification process and the reactor temperature, RDF conversion, gas composition and gas heating value were observed under different gasification agent to RDF mass ratios. The 100% RDF conversion was achieved when gasification with air at $m_{\text{air}}/m_{\text{RDF}}=2.1$ was applied. However, the gas heating value was only 4.6 MJ/Nm$^3$ and the reactor temperature was 950 K. The produced gas contained 18.8 mol.% of H$_2$, 19.7 mol.% of CO, 8.7 mol.% of CO$_2$, N$_2$ and a minor amount of other components. Raw untreated gas tar content was 3.3 mass %. The content of NH$_3$ did not exceed the EU limits for NOx caused by the combustion of these components, but the limit for H$_2$S was slightly exceeded.

Gasification of RDF using oxygen enabled the production of gas with the heating value of around 10 MJ/Nm$^3$ at $m_{\text{O2}}/m_{\text{RDF}}=0.45$. Under these conditions, the content of H$_2$ was 39 mol. %, that of CO was 45 mol.% and that of CO$_2$ was around 12 mol.%; the content of other components including methane was 4 mol.%. By increasing the $m_{\text{steam}}/m_{\text{RDF}}$ ratio, the content of H$_2$ and CO$_2$ increased. However, the content of CO, the reactor temperature and the gas heating value decreased.

Acknowledgement:

This work was supported by the Grant VEGA No. 1/0757/13 from the Slovak Scientific Grant Agency.

References


