Experimental and theoretical investigation of wood pellet shrinkage during pyrolysis

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Gasification of wood pellets is confronted with the problem of adhesion of fuel in the pyrolysis zone. In order to investigate the cause and regularities of fuel adhesion, experimental and theoretical studies of wood pellets thermal deformations were carried out. Experimental studies were performed by measuring thermal deformation of a wood pellet in an electrical reactor from 300 to 1 000 °C temperature in an inert atmosphere. The studies showed that the pellet radius decreased by 10% from 300 to 500 °C temperature, and by 20% from 500 to 1 000 °C temperature. It was also established that the wood pellet expands to 15% from 300 to 830 °C temperature at the beginning of fuel thermal deformation.

The paper also presents the theoretical studies of wood pellets thermal deformations. The theoretical calculations were performed by Matlab using a one-dimensional pyrolysis model of wood pellets that includes the experimental data and the observed expansion of wood pellets. Simulation results were compared with the experimental ones. Modelling and experimental results showed that the thermal deformation of wood pellets values was different by 2–5%.

Key words: wood pellet, pyrolysis, temperature, thermal deformation, numerical model, radius

INTRODUCTION

Renewable fuel resources are increasingly used for electricity and heat production. With the growing demand for these stocks and their price, need to look for ways to use low-quality biofuels arises. One of the ways is gasification [1]. During this process, it is possible to get a higher quality gaseous fuel that can be used in industry. Using granulated fuel for the gasification process, there are created conditions in which the wood pellets clump together moving from the pyrolysis reactor zone to the gasification zone and clog the reactor resulting in inhibition of the gasification process. One of the reasons influencing adhesive bonds of the fuel particles are changes of wood pellets at high temperatures when the fuel particle shrinks due to the chemical reactions taking place inside, caused by high temperatures. Thermal deformations of the wood pellets affect the fuel movement, mixing and the gasification process time. In order to avoid clogging of the reactor it is necessary to know the regularities of this phenomenon, which are investigated by experimental and theoretical studies on the fuel particles changes at high temperatures during pyrolysis, combustion and gasification processes.
An experimental study of wood pellet changes by inserting a sample to a perpendicular pyrolysis reactor is presented in the paper [2]. Experiments were carried out in the range of 650 and 850 °C temperature. The published results show that a 20 mm diameter sample reduced by 10.9% of its initial diameter at 650 °C temperature and by 14.7% at 850 °C temperature. Other authors [3] provide the results of wood pellets changes obtained from 365 to 700 °C temperature. Wood pellets of 10 mm diameter were used for the experimental study. Authors’ [3] published results differ from these in the article [2]: the wood pellet shrinks by 20% in 365 °C, by 30% in 600 °C, and by 24% in 700 °C temperature. Mostly ambient temperature fuel falls into the gasification reactor and the gasification reactor temperature field (the drying zone temperature up to 100 °C, the pyrolysis zone up to 500–700 °C, and the combustion zone up to 1000–1400 °C) varies within wider range than mentioned in the works due to the processes of chemical reactions and heat transfer. During granular biomass gasification the combustion zone temperature is only 1000 °C due to high aerodynamic drag of the granular layer. In order to describe the thermal deformation of fuel particles it is necessary to determine phenomena occurring in pelletized fuel in the gasification process temperature range. This paper presents an experimental and theoretical study of wood pellets shrinkage in a pyrolysis reactor at constant temperature in the range from 300 to 1000 °C.

The theoretical study of wood pellets changes is carried out using the finite difference method (FDM) [4]. The finite difference method is based on the approximation: partial fluxion equations are replaced with partial difference equations. This finite difference approximation is the algebraic form and the target variable value is determined from the adjacent node values of the solution area. The model tracks movement of each individual particle, interaction with other particles and elements of the system, and processes in the particles. In this way each individual particle parameters are obtained by modelling and it allows to investigate biofuel as a fraction or as a particle system. The numerical model formulated by the discrete element method could be used for the granular biofuel combustion system in industrial applications [5, 6].

RESEARCH METHODOLOGY

Experimental method

Experimental studies were performed by measuring shrinkage of a wood pellet during pyrolysis in an electrical furnace from 300 to 1000 °C temperature. A quartz tube (length 1 m, diameter 0.05 m, wall thickness 0.003 m) was inserted in the electrical furnace SUOL-025.1/12.5-11. One end of the quartz tube is supplied by the heated nitrogen, which flows through the tube to other end of the pipe which sticks out from the oven. Nitrogen flow is controlled by a rotameter (Fig. 1). The wood pellet is placed in the middle of the quartz tube when the furnace and nitrogen are heated to the desired temperature (temperature is measured by K-type thermocouple). After that the sample is captured with a photo camera Canon Powershot SX30 IS. The photo camera has integrated wide-angle (24–840 mm) lens, which focus automatically on the sample. Before the experiment, the sample is weighed with KERN EW scales which accuracy is 0.01 g. The wood pellets diameter is measured with a Vernier caliper which provides precision to 0.05 mm.

Nitrogen flow selection according to the sample size is an important value for the wood pellet changes recording quality. If nitrogen flow is too low, the capturing sample changes are out of the sight due to the slow diffusion of gas evolution from wood pellets. Therefore nitrogen flow is supplied from 4 to 10 l/min according to the size and weight of the sample. If the particle diameter is up to 6.5 mm and weight up to 0.3 g, the supplied nitrogen flow is from 4 to 6 l/min. Where the particle mass is up to 0.6 g, the nitrogen flow is increased to 8 l/min. The nitrogen flow of 10 l/min is used for larger than 6.5 mm diameter particles.
At the end of the pyrolysis process, the recording is stopped and the wood pellet is moved closer to the quartz tube end where it is left to cool to ambient temperature (20 °C). The cooled wood pellet is pulled out to measure its diameter and mass.

The measurements were performed from 300 to 1 000 °C temperature by 100 °C step. The high-resolution (1 280 × 720 pixels) recorded videos of wood pellet changes were analyzed using the graphics editing program GIMP. Each 150th recorded video frame was converted into a photo and the wood pellet diameter was measured with a digital ruler, which provides the size in pixels. The value of the measured sample diameter with the digital ruler was compared with the value of the sample diameter measured with the Vernier caliper at the initial time. The compared values were expressed in millimetres by the principle of proportion. In order to reduce errors due to occurrence of digital processing, the measurements of the wood pellet diameter were repeated 3 times in the GIMP program.

Theoretical method
A theoretical study of wood pellets thermal deformation during pyrolysis is made using the numerical model published in the papers [7–9]. The main equation of pyrolysis simulation is described by the following:

\[
\frac{dT}{dt} = \frac{\lambda}{\rho \cdot C_p \cdot r^2} \frac{\delta^2 T}{\delta r^2} + \frac{2 \lambda}{r} \frac{dT}{dr} + \frac{1}{c_p \cdot r} \sum_i \left( H_i \frac{d\alpha_i}{dt} \right) + \frac{1}{c_{p,f} \cdot r} \sum_j \left( H_j \frac{d\alpha_j}{dt} \right),
\]

where \( \lambda \) is thermal conductivity (W/m · K), \( T \) is particle temperature (K), \( \rho \) is the density of the particle (kg/m³), \( C_p \) is specific heat (J/kg · K), \( r \) is particle radius (m), \( H_i \) is the amount of heat during the mass loss reaction \( i \) (J/kg), \( H_j \) is the amount of heat during the mass conversion reaction \( j \) (J/kg).

The main pyrolysis model equation (1) indicates the spherical shape of the particle. A more detailed analysis of the main equation is provided below.

1) The kinetic equations describe the particle mass loss due to the release of volatiles during the pyrolysis process:

\[
\frac{d\alpha_i}{dt} = A_i \exp \left( \frac{-E_i}{RT} \right) (1-\alpha_i)^n,
\]

\[
\frac{d\alpha_j}{dt} = A_j \exp \left( \frac{-E_j}{RT} \right) (1-\gamma_j)^n,
\]

\[
\alpha_i = \Sigma \omega_i \alpha_i,
\]

where \( A \) is the Arrhenius constant (s⁻¹), \( E \) is activation energy (J/mol), \( R \) is the universal gas constant (J/mol · K), \( T \) is absolute temperature (K), \( \alpha \) is mass loss in relative units, \( n \) defines the reaction sequence, the index \( i \) indicates the mass loss reaction, the index \( j \) indicates the mass conversion reaction. Kinetic data for these reactions is taken from the literature [10–14].

2) Changes of the wood particle are altering the parameters such as specific heat (5) and thermal conductivity (6) during pyrolysis:

\[
C_{p,f} = \frac{(\alpha_{T,f} - \alpha_{T})}{\alpha_{T,f}} C_{p,f} + \frac{\alpha_{T}}{\alpha_{T,f}} C_{p,c},
\]

\[
\lambda = \frac{(\alpha_{T,f} - \alpha_{T})}{\alpha_{T,f}} \lambda_c + \frac{\alpha_{T}}{\alpha_{T,f}} \lambda,
\]

where \( C_p \) is specific heat of the sample (J/kg · K), \( C_{p,c} \) is specific heat of char (J/kg · K), \( \lambda \) is thermal conductivity of the sample (W/m · K), \( \lambda_c \) is thermal conductivity of char (W/m · K), \( \alpha \) is the mass loss factor in relative units.

3) The heat transfer takes place on the particle surface due to convection and radiation and it is described by the following equation (7):

\[
-h \frac{dT}{dr} \bigg|_{r=R} = \varepsilon \sigma (T^4_R - T^4_{Bulk}),
\]

where \( h \) is the heat transfer coefficient (W/m² · K), \( \varepsilon \) is the emissivity coefficient, \( \sigma \) is the Stefan–Boltzmann constant (W/m² · K⁴), \( T_R \) is particle surface temperature (K), \( T_{Bulk} \) is bulk temperature (K).

Heat exchange in the centre of the particles is described below by the equation:

\[
-h \frac{dT}{dr} \bigg|_{r=0} = 0.
\]

4) Shrinkage of the wood pellet occurs when the volatiles are released from the solid phase. The shrinking factor \( \delta \) is used for the numerical model and it is found from the mass and volume ratio (9):

\[
\delta = \frac{(V_0 - V_f)}{(m_0 - m_f)} \cdot \frac{(V_0)}{(m_0)},
\]

where \( V_0 \) is the initial particle volume (m³), \( V_f \) is the residual particle volume (m³), \( m_0 \) is the initial particle mass (kg), \( m_f \) is the residual particle mass (kg).

The shrinkage factor \( \delta \) is a nondimensional value, which describes the relationship between mass and volume. The greater the shrinkage factor, the greater the shrinkage of the particle.

5) The pyrolysis reactor volume and the initial conditions of the process are described by equations (10–12), which define the material and energy balance:

\[
m_{Bulk} = \rho \left[ \frac{V_0}{1 - V} \right] + \left[ \frac{V_0}{1 - V} \right],
\]

\[
m_{Bulk} C_{p,g} \frac{dT_{Bulk}}{dt} = C_{p,g} (T_{in} m_{in} - T_{out} m_{out}),
\]
QT = \dot{m}_n \cdot T_{\text{pg}} \cdot \rho_{\text{pg}} \cdot (T_{\text{in}} - T_{\text{amb}}), \quad (12)

where \( m_{\text{Bulk}} \) is bulk mass (kg), \( \rho_{\text{pg}} \) is gas density (kg/m³), \( V \) is reactor volume (m³), \( V_p \) is pellets volume (m³), \( c_{\text{pg}} \) is specific heat of nitrogen (J/kg ⋅ K), \( m_n \) is influent nitrogen mass flow (kg/s), \( m_{\text{out}} \) is effluent nitrogen mass flow (kg/s), \( T_{\text{in}} \) is influent gas temperature (K), \( T_{\text{out}} \) is effluent gas temperature (K), \( Q_T \) is energy amount (J), \( t \) is process time (s), \( T_{\text{amb}} \) is ambient temperature (K).

6) The pyrolysis model equations are adjusted according to the experimental methodology. The additional equation (13) is used to simulate the particle expansion for modelling the pyrolysis process from 300 to 830 °C temperature:

\[
d\frac{dV}{dt} = \frac{dV_0}{dt} \cdot (1 + \varepsilon), \quad (13)
\]

where \( \varepsilon \) is the expansion factor, which varies from 0.2 to 0.025 linearly from 300 to 830 °C temperature.

Parameters such as the pyrolysis reactor diameter and length, ambient temperature, thermal conductivity of the walls, emission coefficient, nitrogen flow, temperature of nitrogen gas and the reactor, sample diameter, weight and quantity (in this case, one particle) from the experimental study are defined in the numerical model. As the porosity of the particles is not known, the equation of the shrinkage factor (9) is modified as follows:

\[
\delta = \frac{(\delta_{\text{in}} - \delta_{\text{f}})}{\delta_{\text{in}}} / \frac{m_0}{m_f}, \quad (14)
\]

RESULTS AND DISCUSSION

Experimental results

In the end of the experiment measured wood pellet mass and diameter changes at different temperatures are shown in Fig. 2. As it can be seen, the final mass of the pellets monotonically increases growing the heating temperature by the equation (15):

\[
\frac{(m_0 - m_f)}{m_0} = 0.1 + 0.9 \cdot \left(1 - e^{-\frac{T - 300}{700 - 300}}\right). \quad (15)
\]

Changes of the pellets diameter do not distribute evenly by equation (16). It depends on the heating temperature: shrinkage is increasing till 700 °C, but it is decreasing when the heating temperature grows over 700 °C temperature till 1 000 °C. The final diameter of pellets can be described as follows:

\[
\frac{(D_0 - D_f)}{D_0} = \left(T - 200\right)^2 \cdot e^{-\left(T - 400\right) / 200}. \quad (16)
\]

When the wood pellet is placed in the pyrolysis reactor, the thermal deformation of the sample proceeded for 100 seconds in the 300–400 °C temperature range. The particle expanded to 2.7% of the initial diameter at the beginning of the pyrolysis process. The expanded particle started to shrink after 300 seconds and it decreased by 12.5% of the initial diameter. The diameter changes versus time at different temperatures are shown in Fig. 3. The experiment lasted 800 seconds in the 300–400 °C temperature range till

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![Fig. 2. Residual pellet mass ((m_0 - m_f)/m_0) and diameter ((D_0 - D_f)/D_0) changes at different heating temperatures](image-url)
the changes of the wood pellet were invisible by a photo camera. In this temperature range the particle mass loss was up to 14% (Fig. 2).

When pyrolysis temperature was grown from 400 to 500 °C, the expansion of wood pellets became more intensive (see Fig. 4). The pellet radius increased to 3.1% in 400 °C and 7% in 460 °C temperature. Along with the intensifying changes of the particle size mass loss increases (see Fig. 2). The changes of the particle size have stabilized by 330 s due to the increased heating temperature.

The thermal deformation of the wood pellet in the 500–600 °C temperature range is presented in Fig. 5. The expansion of pellets was also noticeable in this temperature range. Due to the increased temperature up to 570 °C the pellet radius expanded to 11% of the initial radius. The experiment duration reduced to 240 seconds because of higher heating temperature.

The expansion of the pellet increased to 20% of the initial pellet radius when the heating temperature was raised to 600–700 °C temperature. At this temperature the pellet began to expand in the first 50 s after the placement into the pyrolysis reactor. The expanded pellet started to shrink after 50 s of the expansion process. The wood pellet expansion and shrinkage are displayed in Fig. 6. The pellet stopped to exchange after 125 s. The experiment results of the 800–900 °C temperature range are also shown in Fig. 6. The expansion of wood started to decrease when heating temperature was above 800 °C. The sample grew only to 10% of the initial radius. Also the pellet expansion was observed after 10 s from the beginning of the experiment.

Fig. 3. Wood pellet changes from 300 to 400 °C temperature

Fig. 4. Wood pellet changes from 400 to 500 °C temperature
and lasted only 10 seconds. In this temperature range mass loss was 90% of the initial mass (Fig. 2).

The experimental results received in the 900–1,000 °C temperature range are shown in Fig. 7. In this temperature range, the wood pellet expansion was no longer visible. The wood pellet shrank from 15 to 20% depending on heating temperature after 100 seconds from the start of the experiment. The changes of the pellet stabilized after 100 s of shrinkage. According to the given mass loss and the pellet shrinkage it is seen that the pellet volume decreased from 15 up to 20% when temperature raised from 500 up to 1,000 °C.

The results show that the pellet diameter changes depend on the pyrolysis temperature and heat transfer in the particles. The heat flow is higher from the reactor environment to the particle and the volatile release mechanism is also different at higher temperatures. We believe that the expansion of the pellets is due to a slow heat transfer which influences volatiles release from pellets. When the particle heats evenly at low temperatures (till 300 °C), the pressure of water vapour and volatiles compounds do not reach a critical value inside the particles and the pellets surface is not destructed by water and volatile diffusion from it. Higher heating temperature (over 300 °C) causes the pellets to quickly overheat resulting in failure of water and volatiles diffusion from the pellet. In this way, water vapour and volatiles accumulate inside the pellet and internal pressure is growing near the pellet surface. When the internal pressure is too high, it destroys the surface structure of the pellets by expanding the pellet and freeing accumulated water vapour and volatiles. Shrinkage of pellets begins after volatiles evaporation.
However, the pellets expansion decreases above 700 °C temperature, possibly due to faster particle heating. The particle overheats so quickly that emitted compounds from the surface layer decompose by high temperatures and the way for volatiles evaporation is open from the deeper layers. The expansion phenomenon is no longer detected after 800 °C temperature.

The wood pellet shrinkage lasts for 50 seconds above 900 °C temperature due to higher volatile evaporation.

**Theoretical results**

The thermal deformations of the particles were simulated to the same conditions as mentioned in the Experimental method part before using the numerical model described in the Theoretical method section. The wood pellet diameter changes obtained by numerical simulation (theoretical) and the experimental study in the 300–400 °C temperature range are compared in Fig. 8. The simulation underestimated extreme pellet changes obtained during the experiment, however, the residual pellets radius differs about 2% of the experimental results.

Theoretical and experimental results obtained in the temperature range from 400 to 500 °C are shown in Fig. 9. The start of pellet shrinkage simulated by the numerical model was identical to that measured in the experiment. Simulated pellet expansion was identical to the experimental result. The expansion time varies in simulations, even the speed of expansion was close to the experimental one. However, the residual pellets radius was simulated only with 5% difference from the experimental results.

Similar results were obtained in the temperature range from 500 to 600 °C (Fig. 10). The simulated time of wood
pellet changes was different from that obtained in the experiment, even the start time of changes was the same as in the experiment. However, the residual pellets radius was identical to the experimental one.

The theoretical calculations in the 600–700 °C temperature range are graphically presented in Fig. 11. The modelled beginning of pellet changes did not match the experimental results, but its attribution was identical to the experimental one. Furthermore, the simulated radius of residual pellets was identical to the experimental results when the expansion phenomenon of the wood pellet was estimated to the numerical model.

According to the experimental data simulations were performed at higher temperature (830 °C) (Fig. 12). The attribution of simulated pellets changes was identical to the experimental one. However, the theoretical residual particle radius varied about 5% of the experimental results. The expansion phenomenon was not estimated in the numerical model due to higher temperature (870 °C) and theoretical results correlated well with the experimental ones (Fig. 12).

The theoretical results in the 900–1 000 °C temperature range are shown in Fig. 13. It also complied with the experimental results.

The residual pellets diameter depended on pellet expansion, which is calculated by (13) equation and estimated shrinkage factor values from (14) equation in the numerical model. As can be seen from the results, the simulated shrinkage of wood pellets was identical to the experimental one when the expansion phenomenon was integrated to the numerical model. However, if the expansion phenomenon was not included in the model, the simulated particle changes of diameter varied by 20% and the residual pellets diameter differed by 30% from the experimental results in the 300–800 °C temperature range.
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Fig. 11. Numerical results of wood pellet changes between 600 °C and 700 °C temperature

Fig. 12. Numerical results of wood pellet changes between 800 °C and 900 °C temperature

Fig. 13. Numerical results of wood pellet changes between 900 °C and 1000 °C temperature
CONCLUSIONS

Experimental and theoretical investigations of wood pellet shrinkage during pyrolysis can be summarised by these statements:

- The experimental investigations of palletized wood pyrolysis at the temperature from 300 to 800 °C showed that the wood pellet expanded at the beginning of pyrolysis and only after it began to shrink. The expansion of the pellet up to 20% of its initial size depending on the pyrolysis temperature was determined.

- The expansion of particles extends the duration of volumetric changes and affects the size of residual pellets.

- The theoretical results correlate well with the experimental ones at a temperature over 800 °C if the expansion phenomenon is not included in the numerical model.

- If the expansion of wood pellets is estimated in the numerical model, the simulation results are close to the experimental results between the temperature ranges of 400 to 800 °C. The simulated changes of the wood pellet differed from experimental ones by 30% at lower temperatures, but the residue sizes are identical in both investigations.

Received 17 December 2013
Accepted 18 March 2014

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ЭКСПЕРИМЕНТАЛЬНОЕ И ТЕОРЕТИЧЕСКОЕ ИССЛЕДОВАНИЕ ИЗМЕНЕНИЯ ДРЕВЕСНЫХ ГРАНУЛ В ПРОЦЕССЕ ПИРОЛИЗА

Резюме

Газификация древесных гранул сталкивается с проблемой слипания гранул в зоне пиролиза. Экспериментальные и теоретические исследования древесных топливных гранул пиролиза были сделаны для расследования причин древесных гранул торчащие и определить закономерности него. Экспериментальные исследования были проведены измерения тепловой деформации топливных частиц во времени в интервале температур 300–1 000 °С и в инертной атмосфере в реакторе электрической пиролиза. Исследования показали, что диаметр гранулы уменьшается на 10 % в интервале температур от 300 до 500 °С и на 20 % в интервале температур от 500 до 1 000 °С. Было также установлено, что в интервале температур от 300 до 830 °С диаметр гранулы из-за тепловой деформации увеличивается до 15 %.

Также представлены теоретические исследования древесных гранул тепловой деформации. Теоретические расчеты были сделаны на пакете программирования MATLAB с помощью пиролиза одномерной модели, которая описывает эксперимент и учитывает расширение древесных гранул. Результаты моделирования сравнены с экспериментальными.

Ключевые слова: древесных гранул, пиролиз, температура, тепловая деформация, численная модель, радиус