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EFFECTIVE DIFFUSIVITY OF MOISTURE IN LOW RANK COAL DURING SUPERHEATED STEAM DRYING AT ATMOSPHERIC PRESSURE

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The effective diffusivity of water in brown coal of Bełchatów mine was experimentally determined. The experiments were performed in superheated steam at 200°C and atmospheric pressure using slightly compressed pellets of cylindrical shape. The drying and temperature curves of the sample were used to identify diffusivity. An inverse problem was formulated and solved by the finite element method for 3D axially symmetric cylindrical geometry of the sample. A satisfactory fit of the simulated curves to experimental results was obtained. The obtained dependence of effective diffusivity on moisture content and temperature may be used in designing lignite dryers.

Keywords: diffusion, moisture, lignite

1. INTRODUCTION

Fossil fuels are so far dominating both thermal and electrical energy production all over the world. Coal and lignite share is in total ca. 30%. Low rank coal share will increase in the future. At the moment Europe is the third world largest consumer of lignite after USA and China. In Poland low rank coal's share in energy production is currently 36%. Poland has one of the largest brown coal deposits in Europe and our dependence on it is bound to increase in the future (Kasztelewicz and Kozioł, 2007; Zaporowski B., 2008). Considering that overall cost (including mining) of the energy produced from low rank coal is 20-30% lower than that of black coal, this tendency is understandable. However, all energy producers in Europe are limited by CO_2 emission quotas. In order to reduce the emission of CO_2 the efficiency of the power generation process must be improved. Drying of coal before combustion is one of the ways. Although numerous drying methods of coal were elaborated (Allardice et al., 2004; Pikoń and Mujumdar, 2007; Ross et al., 2005), superheated steam drying seems to be the most suitable for the power industry (Kudra and Mujumdar, 2009; Pakowski, 2011). Low grade steam generated in superheated steam drying can be easily incorporated into the system of the plant. Dust explosion hazard is eliminated due to oxygen-free atmosphere. Finally, there is no emission to the atmosphere since the drying cycle is fully closed.

For the superheated steam dryer design the properties of lignite necessary include sorption isotherms and effective diffusivity. Such data are either difficult to obtain in literature or nonexistent. We dealt with sorptional equilibrium of lignite in our earlier work (Pakowski et al., 2011). In this work we concentrate on the effective diffusivity of water in lignite.

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2. EARLIER WORK

Numerous papers present experimental results of drying of lignite either in air or in superheated steam. However, the model which is used for process simulation of the single grain drying is the receding core model (Chen et al., 2000, Chen et al., 2001, Looi et al., 2002). In the receding core model it is assumed that pores are open and evaporation front recedes into the particle during drying. Thus the entire solid is split into dry and wet zones. There is no moisture transport in the wet zone but only in the dry zone. Since the pores are filled with vapor there is no moisture diffusion in the pores but only the Darcy flow of vapor. The driving force for this flow is pressure difference between pressure on the evaporation front and the external pressure. Pressure on the evaporation front is produced by the vapor-wet solid equilibrium (sorptional equilibrium). The only fitted parameter needed in the model is permeability of coal structure to vapor. In the model it is assumed that the front is sharp and no moisture is retained in the solid i.e. the solid is non-hygroscopic. However, it follows from the shape of sorption isotherm that lignite is in fact hydrophilic. In such a case the evaporation front may be wide and eventually fill the entire solid. This corresponds to purely diffusional model which is often used to describe drying in solids of much finer pore structure such as polymers, fruit and vegetable tissue, paper, wood etc. Indeed, the real mechanism of moisture transport during drying may be composed of both mechanisms i.e. Darcy flow and diffusion. Such a model was developed for superheated steam drying of wood (Adamski, 2009).

3. EXPERIMENTAL

3.1. Sample preparation

Raw lignite was obtained from Bełchatów open pit mine. The sample of lignite was shipped in a double sealed bag and stored in refrigerator until needed. The raw lignite moisture content measured upon receiving the sample, by drying to constant weight at 105°C, was X = 1.2 kg/kg. Granular lignite was compressed into pellets of cylindrical shape, in a tubular mold closed at one end. The pellets were 22 mm in diameter and 35 mm high just before measurements. Always the same mass i.e. 14 g of raw lignite was used. It was noticed that during compression a certain amount of water dripped from the mold. It was then necessary to re-check the pellet moisture content. By repeating gravimetric analysis it was found that compressed pellets have moisture content X = 0.9 kg/kg, which was consistent throughout all measurements. It means that raw lignite contains at least 0.30 kg/kg of free water, which can be removed by mechanical methods.

In this procedure the apparent density of dry compressed lignite was also measured and was found equal to 580 kg/m^3.

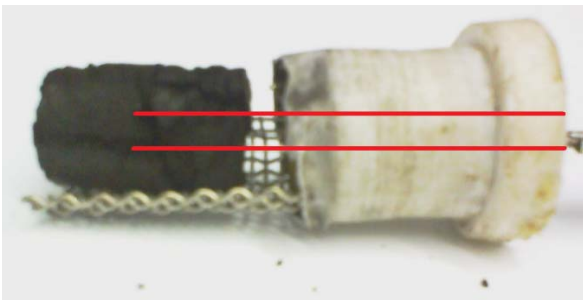

Fig. 1. Lignite pellet used for sample temperature measurements. Lines show positions of thermocouples

Two pellets were simultaneously used in each drying experiment. One served for measuring drying kinetics and was continuously weighed during drying. The second served for solid temperature measurements. The sample had two fine holes drilled: one along its axis and the other parallel to the axis 5mm away (approximately in the middle of sample radius) as shown in Fig.1. Thin (1 mm) Fe-CuNi thermocouples were tightly pressed into the holes. The whole assembly consisted of sample supporting mesh and Teflon stopper, which served for installing the sample in place.

3.2. The experimental set-up and procedure

The experimental set-up is shown in Fig. 2. It provides steady flow of superheated steam of constant temperature and velocity, in which a cylindrical sample of lignite undergoes drying. Steam is generated in a boiler (1) by two immersed electric heaters 1500W each. It passes through a butterfly valve (2) and is then superheated in an electric superheater (3) to the required temperature. The flow of steam is then equalized by a section (4) filled with stainless steel shavings. The first sample (Fig. 1) is fixed horizontally in chamber (5). It serves material temperature measurements. The second identical sample is placed in chamber (6). It is suspended in a wire mesh basket under a balance (7) and continuously weighed. Steam velocity in measuring chambers is approximately 5 cm/s and is individually measured in each run. The steam leaving the chambers is condensed and condensate is returned to the boiler. The cover has a small central hole which serves as a vent, so the internal pressure is equal to the atmospheric one. The escaping steam also centers the wire, on which the sample basket is suspended so it never touches the walls. The entire set-up is wrapped in electric heating blanket and insulated with mineral wool to protect against steam condensation on the walls. Additionally, the top removable cover has two horizontal holes in which cartridge heaters are installed. This prevents steam condensation and eventual dripping of condensate on the samples.

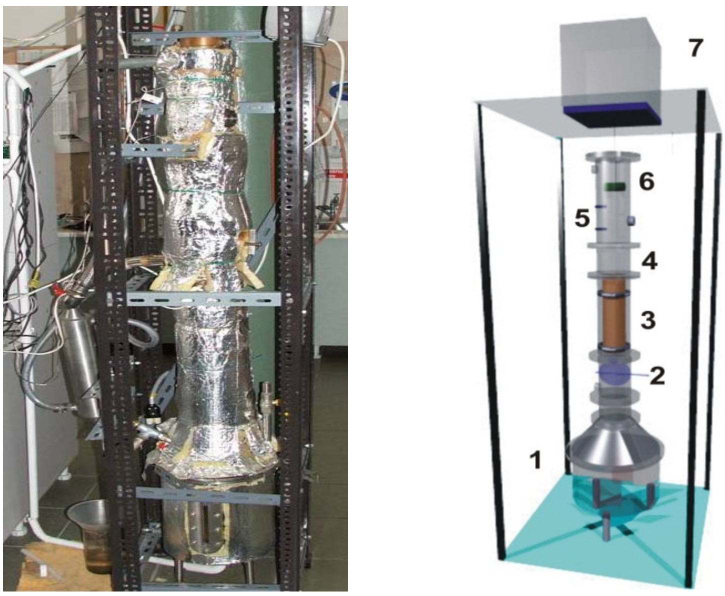

Fig. 2. Experimental set-up for superheated steam drying a) view b) schematic.
1 – boiler with two electric heaters, 2 – butterfly valve, 3 – superheater, 4 – flow equalizer,
5 – temperature measuring section, 6 –weighing section, 7 – electronic balance

3.3. Diffusivity identification

Mass transfer in porous solids during superheated steam drying is due to at least three mechanisms: Darcy flow of liquid moisture caused by generated pressure gradient, diffusion of moisture in vapor phase and diffusion of bound moisture, both caused by moisture content gradient (Pakowski, 2011). In this preliminary work, however, the effective diffusivity encompassing all the three mechanisms was used. The axially symmetric, cylindrical, three-dimensional description of mass and heat balances in the sample is as follows:

- mass balance:

$$\frac{\partial X}{\partial \tau} = D_{eff}\left(\frac{\partial^2 X}{\partial r^2} + \frac{1}{r}\frac{\partial X}{\partial r} + \frac{\partial^2 X}{\partial z^2}\right) \tag{1}$$

- heat balance:

$$\frac{\partial T}{\partial \tau} = \frac{1}{\rho_m \cdot (c_L X + c_S)} \cdot \left[\lambda_m\left(\frac{\partial^2 t}{\partial r^2} + \frac{1}{r}\frac{\partial t}{\partial r} + \frac{\partial^2 t}{\partial z^2}\right) + \rho_m D_{eff}\left(\frac{\partial^2 X}{\partial r^2} + \frac{1}{r}\frac{\partial X}{\partial r} + \frac{\partial^2 X}{\partial z^2}\right) \cdot c_L t\right] + \\ - \frac{c_L t}{c_L X + c_S}\frac{\partial X}{\partial \tau} \tag{2}$$

The initial conditions: at $t = 0$, $t = 20$ [°C] and $X = 0.9$ [kg/kg]

Boundary conditions for mass transfer:

- at cylinder axis

$$\left.\frac{\partial X}{\partial r}\right|_{r=0} = 0 \tag{3}$$

- at cylinder periphery

$$-D_{eff}\rho_m \left.\frac{\partial X}{\partial r}\right|_{r=0} = w_{Dr} \tag{4}$$

- at cylinder bases

$$-D_{eff}\rho_m \left.\frac{\partial X}{\partial z}\right|_{z=0,Z} = w_{Dz} \tag{5}$$

Boundary conditions for heat transfer (likewise):

$$\left.\frac{\partial t}{\partial r}\right|_{r=0} = 0 \tag{6}$$

$$-\lambda_m \left.\frac{\partial t}{\partial r}\right|_{r=0} = \alpha_r (t_\infty - t) - w_{Dr}\Delta h_v \tag{7}$$

$$-\lambda_m \left.\frac{\partial t}{\partial z}\right|_{z=0,Z} = \alpha_z (t_\infty - t) - w_{Dz}\Delta h_v \tag{8}$$

The solid was assumed to be isotropic. The effective diffusivity dependence on moisture content and temperature was assumed to have the following form (Pakowski Z., 2000)

$$D_{eff} = d_1 X^{d_2} \exp\left(-\frac{d_3}{RT}\right) \tag{9}$$

Drying rate w_D was calculated using the following relationship:

$$w_D = \frac{\alpha \cdot \left(t_\infty - t^*\right)}{\Delta h_v} \tag{10}$$

Where t^* is the equilibrium temperature of solid at a given moisture content and vapor partial pressure (here atmospheric). This concept is a hypothesis that was presented in an earlier work (Pakowski and Adamski, 2011). Heat transfer coefficient at cylinder periphery was estimated using available correlations for flow past a cylinder and equaled to 14.8 W/(m^{2}K). Heat transfer coefficient at cylinder bases was assumed equal to the above one.

The vapor-solid equilibrium for lignite was taken from (Pakowski et al., 2011). It has the form of the Henderson equation (11):

$$\frac{p}{P_0} = 1 - \exp\left(-a_1 X^{a_2} t^{a_3}\right) \tag{11}$$

with a_1=0.7623; a_2=1.5879; a_3=0.8392

The following material properties were used:

- Dry lignite specific heat capacity (Heidenreich et al., 1999):

$$c_S = 1150 + 2.03(T - 300) - 1.55 \cdot 10^{-3}(T - 300)^2 \quad \text{[J/(kg K)]} \tag{12}$$

- Dry lignite heat conductivity (Heidenreich et al., 1999)

$$\lambda_m = 0.19 + 2.5 \cdot 10^{-4}(T - 300) \quad \text{[W/(m K)]} \tag{13}$$

- Dry lignite apparent density (measured in this work)

$$\rho_m = 580 \quad \text{[kg/m}^3\text{]} \tag{14}$$

- Steam properties were calculated from relationships available at (Pakowski et al., 1991).

Diffusivity was identified using the results of experiments performed at steam temperature of 200°C. The kinetic curves of the solid moisture content and temperature were generated by the above model solved by FEM using COMSOL Multiphysics software. Optimization of least squares objective function was used to find parameters of the diffusivity equation (9) using the data of both drying curve and temperature curve. In this way the dependence of D_{eff} on both moisture content and temperature was taken into consideration. Steam condensation period was not taken into account as being very short.

4. RESULTS AND DISCUSSION

The experimental and computed results are presented in Figs. 3 and 4.

The initial period of steam condensation is visible as a rapid increase of solid temperature and moisture content in the experimental results. It is followed by a period of constant drying rate, where temperature remains equal to saturation temperature of steam (here 100°C). The experimental drying rate is, in fact, not really constant since a small curvature of the drying curve is observed. The drying rate begins to fall when X approximately equals 0.7. However, the temperature curves show the end of the constant rate period at X = 0.25 kg/kg. At this point the temperature begins to raise rapidly starting at the surface and penetrating the solid. Finally, the dry sample reaches the steam temperature.

These results were used to identify the parameters of the diffusivity equation (9). The finite element method, programmed in COMSOL Multiphysics was used. The obtained parameters are: $d_1 = 1.82 \cdot 10^{-7}$ m^2/s, d_2 = 0.0773 i d_3 = 3549 kJ/kmol (water). Graphical representation of this relationship is shown in Fig. 5.

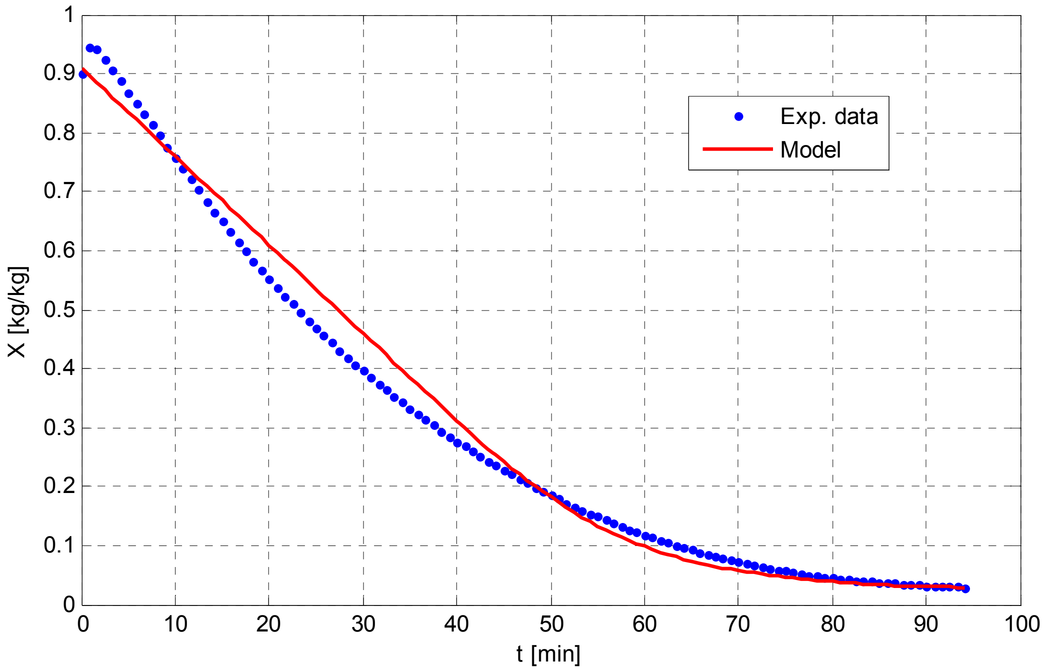

Fig. 3. Experimental and simulated drying curves. Multiple regression coefficient R^2=0.98

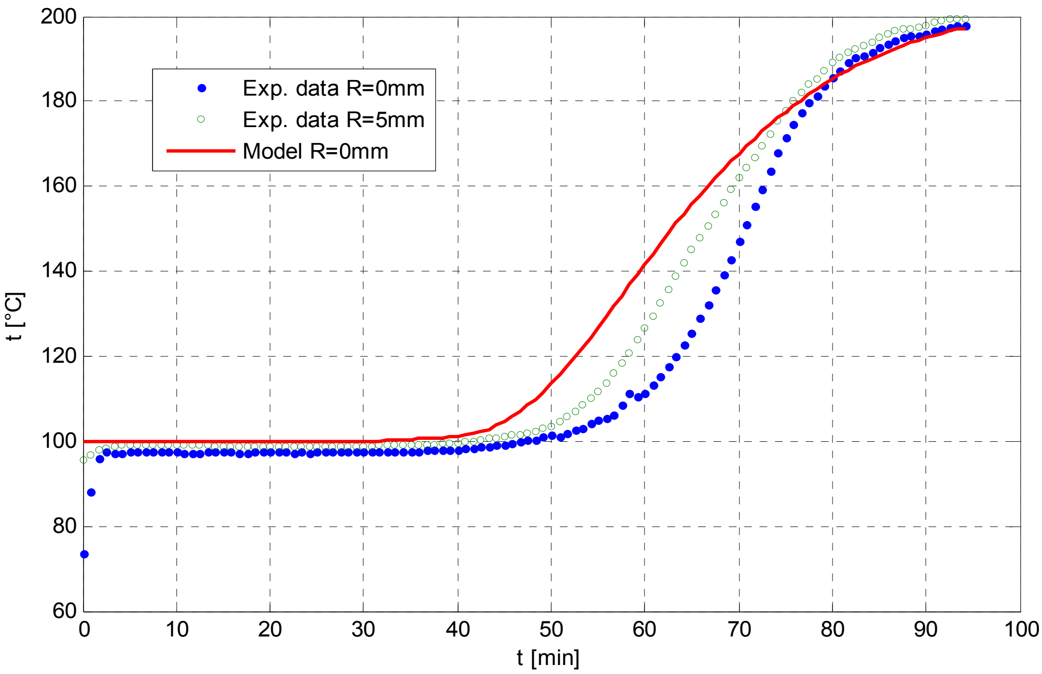

Fig. 4. Experimental temperatures and simulated temperature in the center of sample. Multiple regression coefficient R^2=0.93

A comparison of the obtained drying curve with the simulated one shows that the simulated curve has an initial straight line segment indicating the constant rate period ending approximately at $X = 0.2$ kg/kg. This is the result of the sorption isotherm indicating unbound moisture present in this range and the assumption that the solid surface instantly assumes equilibrium temperature. This is an oversimplification that needs being eliminated by a more complex model of the drying rate. Also the falling drying rate, in reality, begins earlier than predicted by the model. Further work to improve the model is required.

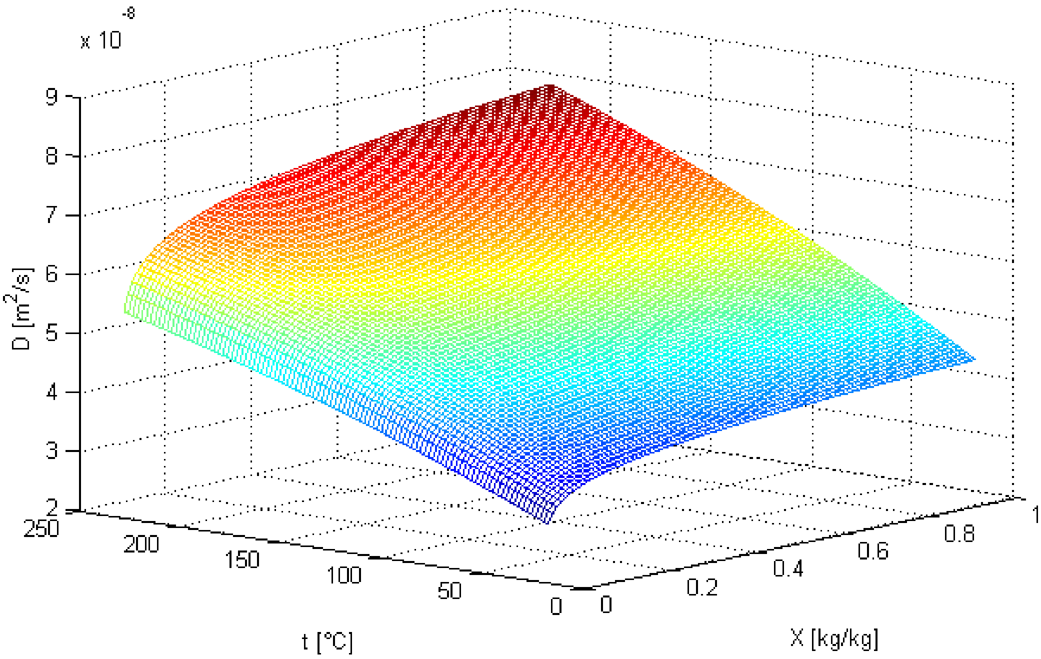

Fig. 5. Dependence of moisture effective diffusivity on moisture content and temperature

As observed in Fig. 5 the effective diffusivity here is 1-2 orders of magnitude larger than diffusivity obtained typically for solids. There are two main reasons for this: 1) at high temperature (equal and above the boiling point) moisture flow is driven by the internal pressure in the form of Darcy flow, 2) the solid is highly porous and has larger pores than typical monolithic porous solids such as wood, clay, fruit and vegetable tissue etc.

A small dependence of the diffusivity on moisture content indicates that the model is a good description of the real process.

5. CONCLUSIONS

Wet Bełchatów brown coal „as mined" contains approximately 1.2 kg/kg of moisture. This moisture content can be reduced to 0.9 kg/kg by mechanical pressing. The rest must be removed by drying. To describe the drying process by a simple model an effective diffusivity of moisture is required. At present no such data are available in literature. The results obtained in this work on the basis of the performed experiments indicate stronger dependence of the effective diffusivity on temperature than on moisture content in the experimental range of these parameters. Although the simulated results satisfactorily fit the experimental results, further refinement of the model would improve the fit accuracy. The work on this is in progress.

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SYMBOLS

c	heat capacity, J/(kgK)
Δh_v	latent heat of vaporization of moisture, kJ/(kg K)

p	partial pressure, Pa
P_0	total pressure, Pa
R	universal gas constant J/(mol K)
R	sample radius, m
r	current radius, m
T	temperature, K
t	temperature, °C
w_D	drying rate, kg/(m^{2}s)
X	moisture content, kg/kg d.m.
z	current length, m
Z	sample length, m

Greek symbols

λ	heat conductivity, W/(m K)
ρ	apparent density, kg/m^3
τ	elapsed time, s

Subscripts

L	liquid moisture
S	dry solid
m	wet solid
r	at cylinder periphery
z	at cylinder base
$\propto$	steam

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