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Original Research Article

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EFFECT OF OXYGEN ADDITION ON LOW-RANK COAL DRYING IN A TECHNOLOGY FOR VOLATILE GAS GENERATION

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Abstract. The article presents the results of oxygen addition effect at the low-rank coal drying in the coal heat treatment technology. *Drying, as one of the stages of the technology, is carried out by diffusion method, using an oxygen addition in a quantity of 2%. The results of the study show the possibility of intensifying moisture removal by the proposed method at the expense of intensifying oxidation processes in diffusion conditions of porous body. Coal has effective and closed porosity. The increased concentration of oxygen in air creates conditions for intensifying oxidation reactions in the effective porosity. In the matrix of the porous body of coal, heat is emitted and temperatures rise. Quantity of the heat produced by the low-temperature reaction of oxygen in coal elements (≈1.24·10^{25} W) allows to increase the temperature of the total mass of coal approximately to the 60°C. Due to the increase in temperature, the dynamic pressure in the flow through the open porosity increases (≈ 6.18·10^{-3}Pa). As a sequence, the static pressure is reduced to negative value (down to negative 100 Pa) in closed pores. The reduction of external pressure alters the gas equilibrium, which leads to the effect of destruction of capillary condensate and promotes moisture release. The final mass fraction of H_2O as a result of diffusion method of drying when the oxygen content of the air is raised during the drying process for 1 hour in a range from≈ 2.9 to 5.8 %.*

Keywords: coal, oxygen, diffusion drying, porosity, static pressure.

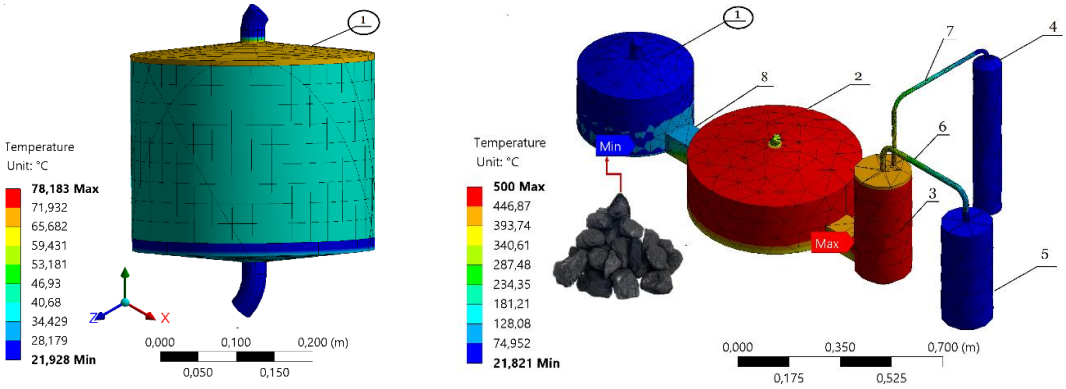
1. Introduction

The trend of development of clean coal technologies not only in Kazakhstan, but also worldwide requires finding simple solutions to obtain economically and environmentally attractive methods. In this direction, the effect could be achieved by using domestic coal reserves in the technology of the releasing volatile gases. Typically, gas generation technologies are taking stage by stage, including drying processes, processes of the thermal treatment and the releasing of the volatile gases [1,2]. Drying is one of the most responsible steps in the preparation of low-rank coals, because of the significant moisture coal content. However, such coals are highly efficient for pyrolysis, gasification and liquefaction [3], where careful preparation of the coal to processes is required. Today, well-proven coal preparation technologies with effective moisture removal, such as «evaporative drying technologies, rotary-drying, fluidized-bed drying, hot oil immersion drying, as well as non-evaporative drying technologies such as hydrothermal drying, mechanical/thermal drying, etc.», are

summarized in many sources but they [4,5] have significant shortcoming - the electric energy consumption in dryers per 1 kg of H_2O removed in a range from 300 kJ to 11000 kJ. In the direction of reducing energy costs, drying technologies are being developed for gasification using the filtration method with the use of «combined mechanical extrusion-hot air/microwave drying» taking into account slow oxidation and increased temperature [6]. Technologies have an economic advantage over the above, but are limited to an initial moisture content of up to 50%. Slow oxidation with rising temperatures within the porous coal will cause changes in all system parameters, including fragmentary changes in pore structures. The paper [7] presents studies on fragmentary changes and changes in the morphology of low-rank coals with increased temperature due to slow oxidation process in a diffusion. The authors show that the influence of the surface porous structure of coal on the oxidation process is determined by the movement of oxygen through open pores during oxygen adsorption due to increased concentration with subsequent intensification of reaction kinetics. At the same time, the authors of works [8-10] present the evolution of functional groups of different types of coal, including low rank, and the formation of volatile substances at low temperatures (up to 30°C) oxidation of alkynes, alkenes, etc. their high reactive capacity and corresponding rise in temperature from oxygen reactions. On this basis, it can be argued that in coal, as in the thermodynamic system, heat and mass exchange processes occur which do not exclude removal of moisture from the "coal-drying chamber" system. This assumption was taken as the basis of studies on the possibility of drying intensification by diffusion when using an oxygen additive in the diffusion stream. The aim of the study is to determine coal efficiency of drying by diffusion, taking into account the addition of oxygen for intensification of drying during thermal treatment of low-rank coal.

2. Material and Methods

The object of the study is a coal-drying chamber (fig.1.a) for the generating volatile gases technology (fig.1.b) and presented in more detail in [10,11] for release of CH_4, CO, H_2 in coal gasification process. In the studies, a method based on the simulation of the movement of continuous medium and discrete phase was used to determine the final moisture of coal as a porous body in the pattern of the coal-drying chamber. The drying chamber has a volume 0,0359m^3 and the surface area 1,0824 m^2. The pattern is scalable for pilot coal gas generation technology. Drying is carried out by the flow of the rising air, with the addition of oxygen in the amount of 2% of the air volume, supplied at a rate of 0.8 m/s during 1 hour through perforated openings into the coal matrix in mass 48.4 kg.



1 – coal-drying chamber; 2 – isothermal heating chamber; gas generation chamber; 4 – inert gas delivery capacity; 5 – volatile gases collection and storage capacity; 6 – volatile gas transport pipeline; 7 – inert gas pipeline; 8 – coal transporter.

Fig.1. Physico-mathematical pattern of the thermal coal treatment installation for the release of volatile gases: a – pattern of the technology; b – pattern of coal-drying chamber in technology.

The gas generation technology from coal presented (fig. 1.b) is implemented through energy activation of the element-element bonds of coal during the three main stages: drying, isothermal heating; release of volatile combustible gases [11]. The challenge of the technology is to create optimal conditions at each stage to maximize the release of the volatile gases from coal. Drying of coal involves preparing the coal with parameters that have the greatest positive effect on the subsequent process in the isothermal chamber. The scientific novelty of the research is to determine the effect of the addition of 2% oxygen to air during the drying

stage of low-grade coal through diffusion in a ball chamber of heat treatment technology. Low-rank Coke weakly sintering coal (Kazakhstan, Ekibastuz Deposit) was used as the material under investigation, corresponding to the classification of Coal Mv (coals with mechanical viscosity), the physico-mechanical characteristics were obtained experimentally in [10], which as a porous body are presented in the table. 1 and represented in [11,12].

Table1. Analytical properties of coal MV

Density, [kg/m^3]	Isotropic thermal conductivity, [W/m C°]	Specific heat, [J/kg °C]	Transmissivity, [s]	Pore volume, [m^3/kg]	Mass, [kg]
1100	0.32	1200	0.7	0.0067	76.5

To achieve the objective of assessing the effect of oxygen-added drying with the diffusion method on water removal intensity, tasks must be performed:

1. To analyze the oxygen addition (2%) in the air composition on the effect of the increase in coal temperature and dynamic diffusion flow pressure;

2. To analyze the drying intensity of coal depending on the increase in dynamic pressure and the occurrence of negative static pressure.

To solve these problems, simulation of the diffusion drying process through a coal matrix was used in the Ansys Academic Research CFD environment using the engineering modules «Flow through a porous media» and «Species transport», which allows the forecasting and control of moisture flows in relation to temperature, static, dynamic pressure, chemical reactions, species… etc. As boundary conditions, the "velocity inlet" conditions at the inlet of the drying chamber and the "pressure outlet" conditions at the outlet in a given area of geometry are taken into account, as presented in table 2 and the properties of coal and air according to tables 1 and 3.

Table 2. Boundary conditions

Velocity magnitude, [m/s]	Turbulent intensity, [%]	Turbulent viscosity ratio	Temperature, [K]	Backflow total temperature, [K]	Mass inlet fraction O_2	Mass outlet fraction O_2	Initial moisture, [%]
0.8	5	5	290	300	0.23	0,21	60

Table 3. Physical properties of airant

Density, [kg/m^3]	Specific heat, [J/kg·K]	Thermal conductivity, [W/m·K]	Viscosity, [kg/m·s]	Molecular weight, [kg/k·mol]	Standart state entropy, [J/kgmol·K]	Turbulent dissipation rate, [J/kgmol·K]
1.225	1006.43	0.0242	1.7894e-05	28.966	194336	0.007468

Table 4. Physical properties of coal

Density, [kg/m^3]	Specific heat, [J/kg·K]	Thermal conductivity, [W/m·K]	Initial temperature, [K]	Permeability, [мD]	Pore volume, (m^3/kg)
1100	802.3	0.243	290	28.966	0.00036

The computational grid parameters accepted for calculation with independent similarity are shown in Table 5. Analysis of the grid independence shows a discrepancy in the size of cells 2 times not more than 3 percent. Dynamic mesh selection for drying chamber is shown in figure 2.

Table 5. Mesh parameters

Target skewness	Inflation option			Mesh metric Element quality		Mesh metric Element quality		Pinch tolerance [m]	Sizing	
									nodes	elements
0.000009	Transition ratio	Max layers	Grows rate	Min [m]	Max [m]	Everage [m]	Standard deviation			
	0.272	40	1.2	2.0354e-0004	1m	0.57015	0.00376	3.117e-0.04	4178330	2307966

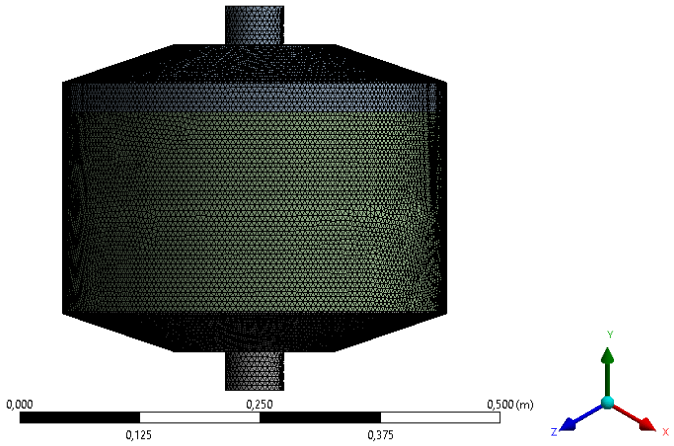


Fig.2. Dynamic mesh selection for drying chamber

In addition, under the conditions of oxygen transfer to porous space, it is necessary to take into account the parameters of the model of the porous medium. Table 6 presents the body coal porous zone conditions. Таблица 6. Porous zone conditions.

Table 6. Body coal porous zone conditions

Relative velocity resistance	Viscous resistance (Inverse absolutely permeability) in all directions [$1/m^2$]	Effective diffusion coefficient [m^2/s]	Effective thermal conductivity, [W/m·K]
Viscous resistance	3.45e+13	$4.8 \cdot 10^{-9}$	0.243

In the simulation, a kinetic-diffusive scheme of coal oxidation under self-heating is used In the simulation, a kinetic-diffusive scheme of coal oxidation under self-heating was used «Viscous Standard k-e (2-eqn) in the realizable k-epsilon model and standard wall functions. The drying time was based on the "Pseudo Time step" value of 1 second with 3,000 operations, which corresponds to the 50 min drying process.

3. Results and discussion

3.1 Analysis results and discussion of oxygen addition (2%) in the air composition on the effect of the increase in coal temperature and dynamic diffusion flow pressure.

To predict the increase in temperature and dynamic pressure of the diffusion flow in an effective coal of internal heating is due to an increase in oxygen concentration and intensification of the kinetics of reactions with heat release [13]. Low-rank coals have a developed, both open and closed porosity, which creates heterogeneity of properties such as heat conductivity, heat capacity, density, etc., in the coal matrix that determine the conditions for the intensity of oxidation reaction kinetics at a given velocity. In turn, temperature causes a change in dynamic pressure directly related to the velocity of the diffusion flow. The kinetic model of internal heating in a coal layer can be represented by the following dependencies, according to [14]:

$$(1-\varepsilon)C_c\rho_c\frac{\partial T_{ox}}{\partial t}=\frac{\partial}{\partial x}\left(\lambda\frac{\partial T_{ox}}{\partial x}\right)+Q_c k(T_{ox})C_c, \quad (1)$$

where T_{ox} (K), C_c (KJ/kg), ρ_c(m^3/kg), λ_c, Q_c is relatively concentration and temperature of oxygen, heat capacity and density of coal, heat of oxygen reaction with coal elements; k is oxygen diffusion coefficient and ε is temperature coefficient of oxygen sorption rate; λ – thermal conductivity of coal efficient coefficient (W/mK).

Temperature coefficient of oxygen sorption rate, taking into account the unevenness of the velocity distribution [15]:

$$\varepsilon\frac{\partial C}{\partial t}=\frac{\partial}{\partial x}\left[D_{ox}\frac{\partial C_c}{\partial x}\right]-k(T_{ox})C_c, \quad (2)$$

and

$$k(T_{ox})=k_0+E(T_{ox}-T_0), \quad (3)$$

where E is activation energy for the low-rank coal presented is 1 -11 kcal/mole; ε is kinetic ratio of oxygen concentration change in layer is equal $\frac{C}{C_0}$.

The concentration of oxygen at the coal surface will be determined by its air content, taking into account convective heat exchange between coal and air. If the oxygen concentration in the supplied air is considered constant, then the system can be supplemented by boundary conditions:

$$\begin{aligned}&\lambda\frac{\partial T_{ox}(0,t)}{\partial x}=\alpha\left(T_{ox}[0,T]\right)-T_0\\&C[0;T]=C_{0x}\\&T[x;0]=T_{0x}\end{aligned} \quad (4)$$

The intensity of convective heat exchange between coal and oxygen-containing air under given geometric conditions is determined by a number Nu, directly dependent on the heat exchange coefficient α (heat transfer coefficient (W/m^{2}K)), the geometric characteristics of the drying installation and the coefficient of thermal conductivity of coal. At the same time, the oxygen diffusion process in the coal layer is carried out at a given concentration, so as an initial boundary conditions it is decided on the stationary equation of the change in oxygen concentration under diffusion conditions:

$$C[x;0]=C_{ox}\exp\left(-x\sqrt{k/D_{ox}}\right) \quad (5)$$

The maximum heat reaction in the coal is observed on the surface at the highest possible concentration of oxygen. Due to the heat conductivity of coal, which directly depends on density and porosity, layers will be heated deep down according to the temperature gradient direction. On this basis it can be written:

$$\begin{aligned}&T_c[x,t]\to T_{ox};x\to\infty\\&C_{ox}[x,t]\to 0;x\to\infty\end{aligned} \quad (6)$$

Simulation showed (fig. 3. a, b) that in the process of diffusion drying zones with maximum mass fraction of oxygen (fig. 3.a) are formed, reaching 0.23 in which there is a maximum can be increase in temperature to 348 K (fig. 4) due to the emission of heat C_c = 1.24 ·10^{-25} W (fig. 3.b). But at the same time, the maximum coal temperature does not exceed the low-grade coals' self-ignition temperatures (t ≈ 130°C – 150°C). Average temperature by volume of coal during drying when the initial air temperature is set T = 18°C, consists of t ≈ 60°C. In addition, an amount of CO_2 is generated during the self-heating process, which determines whether the risk of self-ignition can be reduced.

As noted above, chemical reactions in a given temperature range occur through the reaction of oxygen to coal. Of course, the concentration of oxygen on the surface of coal plays a major role, and the addition of active oxygen will help to accelerate the oxidation process and increase temperature and pressure. The line graph presented in fig. 4 shows the dependence of temperature rise, heat reaction, oxygen concentration and dynamic pressure. Fig. 4 shows that the proposed method gradually increases coal temperature from 291 K to 355 K in different zones of the coal matrix throughout the drying process. The oxidation reaction begins with a mass fraction of oxygen on the coal surface ≈$1.42 \cdot 10^{-3}$ $O_{2/}$ per one kg of air, corresponds to the mean value of heat reaction $C_c \approx 0.62 \cdot 10^{-24}$ W.

The maximum amount of heat reaction by the flow of oxygen approximately C_c consists of ≈$1.24 \cdot 10^{-25}$ at the mass fraction of the oxygen $O_2 \approx 1.42 \cdot 10^{-3}$ O_2/ per one kg of air. The dynamic pressure growth rate in the flow reaches $P_d \approx 6.18 \cdot 10^{-3}$ Pa.

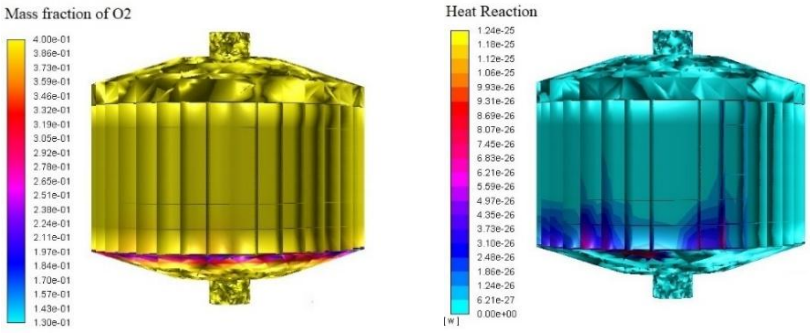


Fig.3. O_2 mass fraction and heat of oxygen reaction with coal during the diffusion drying process: a – O_2 mass fraction; b – heat of oxygen reaction with coal.

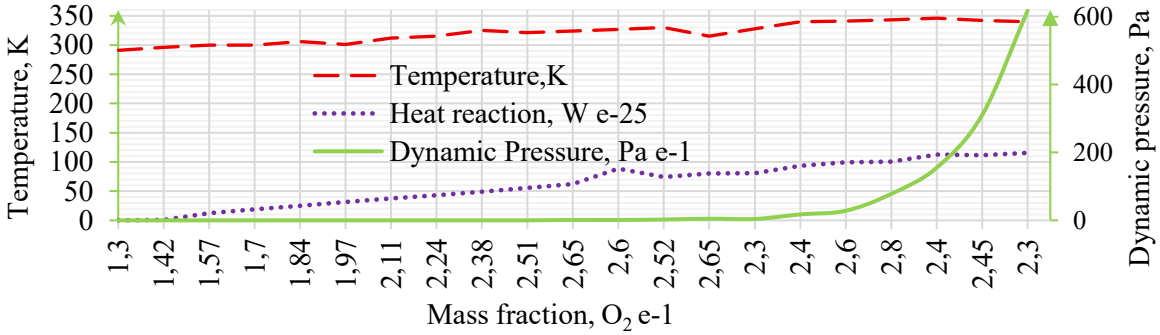


Fig.4. Dependence of temperature rise, heat reaction on the surface oxygen concentration of coal and dynamic pressure.

3.2 Analysis results and discussion of the drying intensity of coal depending on the increase in dynamic pressure and the occurrence of negative static pressure.

The intensity of the drying n process is determined by the total transport of moisture through the coal body during the process. According to the theory of heat mass exchange, the total water transfer through a porous body under diffusion conditions is defined as [15 -21]:

$$G_i = \int_F (j_F \cdot n_F) dF, \tag{7}$$

where n_F is normal vector to the surface; $j_F = f(x; y; z; \tau)$ is the flow of matter.

The density of the flow of matter (moisture) is defined by both a molecular and a convective component that occurs in this process:

$$j = j_{diff} \cdot j_{conv} = -\rho D grad m \cdot \rho m v, \tag{8}$$

where is D the diffusion coefficient of a two-component mixture consisting of water and air.

The external mass exchange is characterized by the intensity of the flow emitted from a wet coal surface, according to Dalton's Law of the partial pressure:

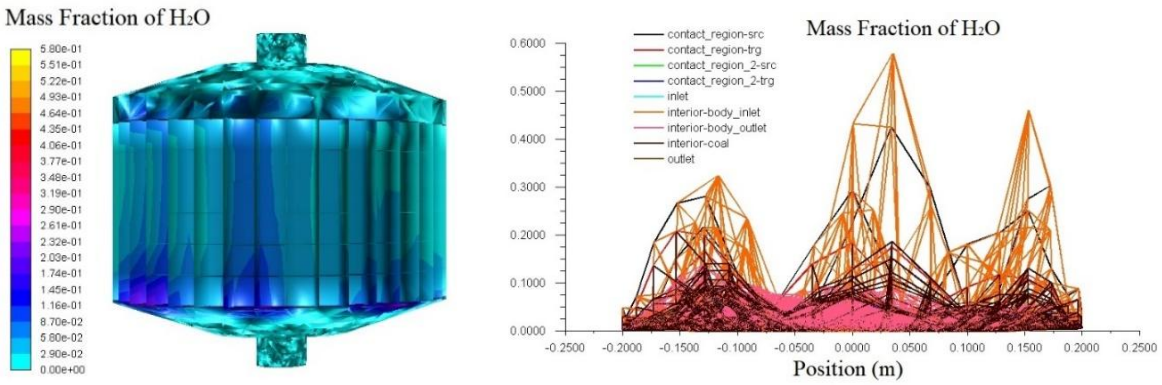
$$g_i = \alpha^{/}\left(\phi P_n - \left(\phi P_c\right)\right)dF, \tag{9}$$

where $\alpha^{/}$ is the moisture coefficient, which depends on the hydrodynamic conditions of the environment; ϕ is coefficient taking into account the reduction of the water vapour above the surface of coal partial pressure (interior body coal).

$$\phi = 0.091 \cdot 10^{\frac{293}{t_c+273}}, \tag{10}$$

where t_c is temperature of environment.

Fig. 5. a ,b shows the model of drying in a diffusion chamber, taking into account at least 23% oxygen content. According to fig. 5 (a), the mass moisture fraction in the respective coal zones may vary from 0.60 (at the interior body inlet) to 0.01 (at the interior body outlet) during drying. According to figure 5 (b), maximum mass fraction on the perimeter of the chamber is observed at the exit from the drying chamber and on the surface of the coal (interior body coal and interior body outlet). On the interior body outlet mass fraction of water consists of no more than ≈ 0.02. At the same time, at the inlet of the coal-drying chamber (interior body inlet) mass fraction of water consists of for about ≈ 0.06. The calculation model is based on the initialization and calculation of the activity of all chamber zones during the process in the total mass of coal, taking into account the total moisture. (Wet basis model)



a b

Fig.5. Mass fraction of H_2O release during the diffusion drying process: a – in common; b – on the perimeter of the chamber.

According to the abovementioned equations (7 - 9), a significant influence on the intensity of the removal of moisture from the low-grade coal matrix is caused by the magnitude of the dynamic pressure when the temperature changes accordingly [20, 21]. Low-rank coals have a closed porosity, with moisture that can exceed more than 40%. Taking into account the Kelvin's equation for a model of pore-containing matrix drying [22], Knudsen's diffusion law, and also considering the gas permeability of an unconsolidated porous medium [23] and the above represented equations (7 - 10) the water rate equation for drying low-rank coal can be presented:

$$J_w = -4\pi\varepsilon \frac{DT^{0.75}\left(P_d - P_s\right)}{RP_d} M, \tag{11}$$

where T is the medium temperature (K), R is the equilibrium gas constant, M is molar mass of water.

Figure 6 shows a line graph of the dehumidification in the coal matrix as a function of static, dynamic pressure and temperature.

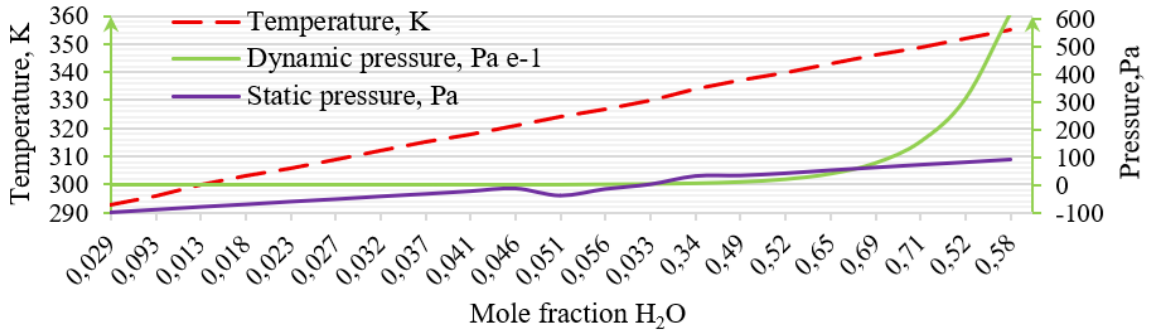


Fig.6. Dependence H_2O mole fraction from dynamic pressure and absolute temperature during the diffusion drying process.

The intensity of moisture (H_2O) release in the process increases significantly with increasing dynamic pressure in a range from $P_d \approx 2\%$ to 65% under conditions of the negative static pressure $P_s \approx$ -100 before it became positive. It is suggested that, the changing in pressure alters the gas Kelvin's equilibrium, which leads to the effect of destruction of capillary condensate and promotes moisture release [24]. Thus, it can be argued that diffusion drying with the addition of 2% active oxygen has a positive effect on drying intensity. In addition, the self-heating effect of coal at a temperature increase of up to 60 °C also has a positive impact on further heat treatment of low-rank coal. To confirm the effectiveness of this method compared to vacuum drying, the final moisture content results obtained in this study (#2) were compared with the results (#1) obtained from experimental low-grade coal formation in a ball pit diffusion chamber - dryer in the vacuum of identical gasification technology. [24-27] and presented in Table 4.

Table 7. Comparison of the simulation and experimental data of vacuum drying

Experimental group	Average density [kg/m^3]	Static Pressure [Pa]	Average final moisture [%]
#1	980	≈ -0.025	≈5.17
#1	950	≈ -0.102	≈1.03
#1	917	≈ -0.475	≈2.37
#1	1000	≈ -0.475	≈2.38
#1	1100	≈ -0.478	≈2.38
# 2	1100	≈ -0.100	≈2.9 – 5.8

Comparative analysis of results showed that the active addition of oxygen during low-rank coal drying promotes drying intensification in a similar way to the conditions for creating diffusional dryers when creating vacuum even at higher coal density.

4. Conclusion

Evaluating of the oxygen addition at the low-rank coal drying in the heat treatment technology in amount of 2% shows, that intensification of moisture removal occurs along the chain «increase in oxygen concentration – reaction with heat release – rise in temperature – increase in dynamic pressure – decrease static pressure to negative», which leads to conditions that promote internal moisture output from coal. At this:

1. The results of analysis of the influence of the addition of oxygen in the amount of 2% in the air composition on the effect of the increase in coal temperature showed the possibility of increasing the temperature of coal from 291 K to 355 K in different zones of the coal matrix due to intensification of oxidation reactions in zones of concentration increase oxygen from ≈ $1.42 \cdot 10^{-3}$ O_2/per kg of coal.

2. The results of analysis of coal drying intensity relative to temperature rise and dynamic and static pressure change showed that it is possible to dry coal with an initial humidity of 60% when moisture is finally removed to 2,9 - 5.8% due to reduced pressure contributing to the release of moisture.

Conflict of interest statement
The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

CRediT author statement
Atyaksheva A.V., Sarakeshova N. N., Rozhkova O. V., Zhumaliyeva A.K.: Conceptualization, Data Curation, Writing Original Draft; Methodology. **Zaksylyk A.M., Aitmagambetova M.B., Umirzakov R. A.:** Investigation, Software, Writing Review & Editing, Supervision. The final manuscript was read and approved by all authors.

Statement on the use of Artificial Intelligence.
During the preparation of this manuscript, artificial intelligence tools were used solely for language editing and grammatical improvement.

Data Availability Statement
The data are available upon reasonable request from the authors.

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