Technology organic and inorganic substances

The object of this study is the process of roasting iron ore pellets. The study solves the task of replacing fossil fuel with plant-based fuel in order to reduce environmental load and ensure the stable quality of pellets, which is necessary for use in blast furnaces.

The influence of biofuel content at a given temperature and air speed on the strength of pellets after roasting was studied. As a result of the research, it was established that the fuel content has a decisive effect on the strength of pellets. Among all types of fuel that were investigated, pellets with the addition of sunflower husks and wood had the highest strength that meets the requirements for blast furnace melting of 200 kilograms. The use of wheat straw and charcoal does not make it possible to completely replace solid fuel in the layer of pellets.

The results show that the use of up to 0.36 % of sunflower husk makes it possible to increase the strength of burned pellets compared to samples without biofuel content. Adding all other considered types of fuel reduced the strength of the pellets.

These results are explained by the different content of lignin, cellulose, and hemicellulose, which determines the characteristics of the biomass. The high content of cellulose and hemicellulose allows for high hydrophilicity due to the high number of OH groups and positively affects the formation of raw pellets. Volatile substances released during the combustion of biofuel contribute to the formation of spherical pores, as well as their uniform distribution, which prevents the propagation of cracks under load.

Research results make it possible to establish the optimal roasting mode, decrease harmful emissions, and bring down costs by reducing fossil fuel consumption

Keywords: pellet roasting, wheat straw, sunflower husk, soft wood, charcoal

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DETERMINING THE IMPACT OF DIFFERENT TYPES OF BIOFUELS ON THE QUALITY OF IRON ORE PELLETS

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1. Introduction

The steel industry is the second largest industrial sector responsible for a significant portion of global greenhouse gas emissions. Steel plants account for 7 % to 9 % of global CO2 emissions from fossil fuel consumption. On average, in 2018, every ton of steel produced accounted for 1.85 tons of CO₂. With the demand for steel projected to grow in the coming decades, reducing emissions in this industry is a major challenge. Conventional energy efficiency measures can reduce emissions by only 25–40 % of the average for each ton of steel produced. To achieve further reductions, breakthrough technologies are needed, such as the use of hydrogen or biomass as reducing agents [1]. One of the processes of steel production is the preparation of metallurgical raw materials, namely the roasting of iron ore pellets. One of the ways of using fossil fuel when burning pellets is to add it to the charge. The use of biomass as an alternative source of energy is the most relevant trend in reducing harmful emissions in the metallurgical industry.

Agricultural production has a unique potential to provide the production process with alternative sources of energy that can be obtained from by-products of crop production. According to the 2018 estimate, the amount of conditional fuel obtained from by-products of crop production is 24,738.3 thousand tons. This amount of fuel could replace the use of 26,845.6 thousand tons of hard coal. The considered types of biomass – sunflower husks, wheat straw, wood, and charcoal obtained by pyrolysis are relevant types of biomass for Ukraine [2].

Devising a technology for roasting pellets using various fuels of plant-based origin will make it possible to reduce dependence on non-renewable resources and reduce emissions of harmful substances. When using fuel of plantbased origin, the total emissions into the atmosphere will be zero since the amount of carbon oxides released during the roasting of biomass is equal to the amount that the plants absorbed in the process of photosynthesis. Therefore, investigating the possibility of using alternative fuel

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of plant-based origin in the process of roasting pellets is a relevant task.

When roasting pellets on a conveyor machine, heating is carried out by burning gas above the layer, which causes uneven heating of the pellets along the height of the layer and, as a result, their different strength. More uniform heat treatment along the height of the layer can be achieved by using a combined fuel: solid, which is added to the charge, and gaseous or liquid, which is burned above the layer. However, the use of solid fuel in the charge in the production of pellets could cause problems because of the possibility of heat formation in the lower layers during roasting on the conveyor machine, which occurs as a result of local excess heat and more melt formation. This leads to a decrease in the quality of the roasted product and the performance of the roasting machine. This poses the task of determining the optimal content of solid fuel in pellets, which may differ depending on the type of solid fuel used.

2. Literature review and problem statement

Pellets with the addition of fuel of plant origin are used in the blast furnace and the direct reduction process. These are new raw materials for iron production. They consist of carbonaceous biomaterial powder, iron ore powder, and a small amount of binder materials. Carbonaceous biomaterials used in pellets can be used raw or after heat treatment. Pellets with the addition of biofuel must meet the minimum requirements for mechanical strength for a blast furnace. The production of these pellets requires special research [3].

In general, there are different ways of using biomass in metallurgical processes to replace fossil fuels. The steel industry is mainly focused on the use of biomass as an alternative source of energy, as well as a reducing agent [3]. The use of biofuel in metallurgical processes faces a number of problems. One of the main ones is low heat of combustion, high humidity, and a significant share of volatile components in raw biomass, which makes it less efficient compared to conventional coal. To solve these tasks, pre-treatment of biomass, in particular torrefaction or pyrolysis, is necessary. After these processes, the fixed carbon content can increase to 50 % or more, which improves its usability.

For example, in [4], the possibility of adding cow manure during the production of pellets and its effect on the strength of roasted pellets were studied. The study has shown that the addition of 1 % cow dung provides for the strength of roasted pellets at the level of 357 kg/pellet. The porosity of the pellets was 25.36 %, which meets the industrial requirements for ensuring good recoverability in blast furnaces. When increasing the amount of cow dung to 1.75 %, a decrease in strength is observed due to an increase in the content of volatile substances that contribute to the formation of cracks inside the pellets. However, when the additive is further increased to 2 %, the strength increases again. This was explained by the improvement of the fusion of particles, which is caused by the release of heat during the combustion of biomass. The main problem is that during roasting the volatile substances contained in cow dung caused the formation of microcracks in the pellets and reduced the cold strength of the pellets. Although this problem is partially solved at higher addition percentages due to particle coalescence, initial weakening of the granules remains a noticeable issue.

Work [5] considers the replacement of anthracite fine coal with eucalyptus charcoal in the process of producing iron ore pellets. The main attention is paid to the assessment of physical and mechanical properties of pellets when using coal with different content of volatile substances. Strength indicators when using coal with a low content of volatile substances turned out to be higher than when using coal with a high content of volatile substances. This is due to the fact that coal with a low content of volatile substances decomposes at higher temperatures, which contributes to better compaction of pellets. A problem with increasing the percentage of fossil fuel substitution with biomass is that fuels with higher volatile matter content can decompose prematurely, creating cracks in the pellets. More research is needed to establish the exact temperatures at which these processes occur and their effect on the quality of the final product. The distribution of energy inside the pellets during combustion can be uneven, especially when using biomass with a larger particle size. This factor affects the strength of the pellets, and more research is needed to understand how particle size affects the uniformity of heat distribution and the final physical properties of the pellets.

Paper [6] evaluates the possibility of replacing mineral coal with carbon-containing residues from leather waste in the process of roasting pellets. The results showed that adding pyrolyzed skin residues to the batch increases the compressive strength of the roasted pellets. Microstructural analysis showed that pellets made using hide residues have a more uniform pore distribution and a lower concentration of large pores compared to pellets made using coal. This also explains the increase in their strength. Another explanation for the increased strength is that the presence of the collagen fraction contained in the skin remnants may act as a binding material for the particle surfaces. The problem that reduces the efficiency of biomass use is the decrease in pellet strength due to increased porosity. The authors note that although increasing the fixed carbon content could improve the uniformity of heat distribution within the pellets, excessive carbon content leads to increased porosity, which in turn may cause cracks and reduce the strength of the pellets.

Study [7] investigates the possibility of using rice husk as a renewable and carbon-neutral fuel instead of conventional coking fine coal in the pellet production process. The porosity of the pellets increases significantly with the addition of rice husk. The porosity of the pellets without the addition of rice husk was 6.4 % while with the addition of 1.8 % rice husk, the porosity reached 26.35%. The compressive strength increased with an increase in the content of rice husk up to 1.44 %; however, with a further increase in the content of rice husk, the strength decreased due to an increase in the volume of pores. The study showed the presence of hematite, magnetite, and wurtzite phases in the pellets. These iron-containing phases contribute to increasing the strength of the pellets. Pores and spongy zones are found to be rich in iron, while gray zones are rich in calcium, aluminum, and silica. This indicates mass transfer inside the pellets during the roasting process. Replacing coking coal with rice husk in the pellet production process has shown significant advantages in increasing the porosity and strength of the pellets. The problem with adding rice husk to iron ore pellets is the increase in porosity, which can negatively affect the mechanical properties. There is also an uneven distribution of temperature during roasting, which can cause the formation of pores of uneven sizes in different parts of the pellets.

In the context of impact of the addition of plant-based fuel on the strength of iron ore raw materials, the agglomeration process can be considered. Paper [8] investigates the influence of biocoke on the process of agglomeration of iron ores and the properties of agglomerate strength. The research is aimed at replacing conventional coke with biocoke made using wood pellets at different carbonization temperatures (950 °C and 1100 °C). It is shown that biocoke carbonized at a temperature of 950 °C has a good potential for use in the process of sintering iron ores. The use of biocoke with a biomass content of up to 15 % does not negatively affect the agglomeration process and the properties of the final product, the strength indicators remain at the level of conventional coke fuel. The task is to further increase the share of biomass. The use of biocoke in the amount of 30-45 % leads to a deterioration in the quality of the agglomerate due to a change in the thermal conditions of the process and an increase in the porosity of the agglomerate, which reduces its strength.

Work [9] shows the possibility of replacing up to 25 % of coke with charcoal and up to 50 % with walnut shells during agglomeration, while maintaining acceptable agglomerate strength indicators. Despite the positive results, there are unsolved issues related to the low efficiency of using some types of biomass, such as sunflower husks, due to its low bulk weight and the porous structure of the agglomerate. An option to overcome these difficulties is the preliminary preparation of biomass, in particular, its pressing to increase the volumetric weight and improve the roasting conditions.

Our review of studies on the replacement of fossil fuels in the production of pellets with fuels of plant origin demonstrated the promising nature of this technology and the need for its further development. More attention is paid to the use of biomass as a reductant in the works of researchers, so it is possible to note the lack of data on the use of biomass as a solid fuel in the charge of pellets for a blast furnace. Among the considered experiments, on can see that mainly biomass is

subjected to preliminary treatment, for example, pyrolysis. There are almost no data on the use of raw biomass. Studies examining the possibility of using charcoal in roasting pellets did not consider a complete replacement of fossil fuels in the pellet layer, but only a partial one. That is, the use of fuel of plant-based origin mixed with fossil fuel, for example, anthracite or coke, which is conventionally used in the technology of adding solid fuel to pellets.

3. The aim and objectives of the study

The aim of this study is to establish the possibility of using sunflower husks, wheat straw, wood, and charcoal as a solid fuel in a pellet charge. This would make it possible to reduce the cost of fossil fuels, which have a higher cost and a negative impact on the environment.

To accomplish the aim, the following tasks have been set:

 to conduct an experimental roasting of pellets with the addition of biomass of each species; to build regression equations for the strength of the resulting pellets;

 to establish the influencing factors and their nature on the strength of pellets; - to define optimal roasting parameters: biomass content, temperature, air speed for each type of biomass;

– to analyze the micro- and macrostructure of the resulting pellets in order to analyze the distribution of pores and identify the phases formed in the roasting process.

4. The study materials and methods

The object of our research is the process of roasting iron ore pellets. The subject of the study is the effect of biomass content in pellets on their mechanical strength. The main hypothesis of the study assumes that there is a possibility of replacing the solid fossil fuel used in the pellet charge with biomass while maintaining the necessary metallurgical characteristics for further use in the blast furnace. Increasing the biomass content of the pellets would have a positive or negative effect on their mechanical strength, depending on the type and amount of biomass used. The work assumes that all samples have a homogeneous composition, except for changes in the biomass content.

Based on the results of our review of the literature, relying on the experience of adding biomass to the charge of pellets, the biomass content (F_c) is investigated in the range of 0–1 %. Roasting temperature T=1150–1250 °C, air velocity $V_{air}=0-0.6$ m/s. The output variable is the strength of roasted pellets (*M*). For the study, 4 types of fuel of plantbased origin, which are relevant for Ukraine, were chosen: wheat straw, sunflower husks, wood industry waste, charcoal. Each type of plant-based fuel was crushed to a fraction with a maximum size of 0.1 mm. Then the fuel was dried in a drying cabinet at a temperature of 105 °C over 24 hours. To investigate the possibility of using plant-based fuels for roasting pellets, a charge with the following composition was prepared: iron ore concentrate, bentonite (Table 1), as well as one of the four types of plant-based fuels under investigation. Moistening of mixtures was carried out over 100 % of the mass of dry materials, by adding 5 % of water from the mass of the mixture. Bentonite was added in the amount of 0.5 % to all types of charge.

Table 1

Chemical composition of metallurgical concentrate and bentonite [5, 7]

Materials	Total Fe, %	FeO, %	$\mathrm{Fe}_{2}\mathrm{O}_{3},\%$	SiO ₂ , %	$\mathrm{Al}_2\mathrm{O}_3,\%$	CaO, %	MgO, %	PL ¹ , %
Iron ore concentrate	65.88	28.27	62.71	6.44	0.30	0.17	0.26	1.85
Bentonite	4.1	0	6.08	62.28	13.56	1.7	1.94	10.34

Note: ¹PL – puncture losses.

Next, cylindrical briquettes with a diameter of 10 mm, a height of 10 mm, and a weight of 3.8 grams were formed from each of the obtained samples of the charge. Forming of briquettes was carried out on a hydraulic press "MS-1000" with an effort of 1 ton/briquette. Next, the resulting briquettes were dried in a drying cabinet at a temperature of 105 °C for two days. Briquettes were roasted in an electric resistance furnace SUOL-0.25.1/12.5-I1 in an air atmosphere for 20 minutes. The roasting temperature in accordance with the experimental plan was 1150, 1200, 1250 °C. The briquettes were fed into the working space in the furnace in a ceramic shuttle. The temperature in the working space of the furnace was measured by a thermocouple. The speed of the air flow was selected in such a way that the decrease in temperature in the working space of the furnace was not too great and did not have an effect on the temperature factor during roasting. The speed of the air flow was measured at the entrance to the working space of the furnace using a manual anemometer.

Next, the compressive strength of the roasted briquettes was measured on the FP-100/1 breaking machine.

Using a Neophot-21 microscope, macro and microstructures were examined, at magnification x50 and x500, respectively; images were acquired.

To determine the strength of pellets based on the strength of the briquettes, an experimental dependence was used:

$$F_{\text{pellet}} = 0.45 \times F_{\text{briquette}},$$
 (1)

where F_{pellet} is pellet strength, $F_{\text{briquette}}$ is briquette strength. The resulting strength is calculated for pellets with a diameter of 12 mm.

> 5. Results of research on the impact of biofuel on pellet strength

5. 1. Regression equation for the strength of roasted pellets depending on the content of biofuel, temperature, and air speed

In the course of the experiment on determining the effects exerted on the strength of roasted pellets by such roasting parameters as temperature, solid fuel content of plant-based origin, data were obtained.

The study was conducted according to the non-composite plan of the second order by Box-Behnken for three factors, which makes it possible to derive a regression equation in the form of a full quadratic polynomial. The interval of variation and the values of the factor levels are given in Table 2. The matrix of the experiment for three factors is given in Table 3. According to the experiment plan, the amount of work to obtain samples of roasted pellets using solid fuel of plant-based origin is 15 experiments for each type of fuel of plant-based origin under investigation. The experiment is active.

Table 2

Values of natural factors for levels

Factor	Easter ID	Factor levels			
No.	Factor ID	-1	0	1	
1	Solid fuel content, %	0	0.5	1	
2	Roasting temperature, °C	1150	1200	1250	
3	Air speed, m/s	0	0.3	0.6	

The least squares method was used to construct the regression equations. Equations are given in natural form.

Pellets using charcoal:

$$M = 21,110 + 22 \cdot F_c - 34.0 \cdot T - 2,669 \cdot V_{air} + +80.5 \cdot F_c^2 + 0.01383 \cdot T^2 + 562 \cdot V_{air}^2 - -0.262 \cdot F \cdot T + 147.6 \cdot F_c \cdot V_{air} + 1.896 \cdot T \cdot V_{air},$$
(2)

 $R^2{=}95.53~\%,~R^2~{\rm (adj)}{=}87.48~\%,~{\rm F-value}{=}11.87,~{\rm P-value}{=}0.007,$

where F_c is the fuel content of plant-based origin, %; *T* – roasting temperature, °C; V_{air} – air speed, m/c.

Table 3

Box-Behnken second-order non-composite plan, for 3 factors

Experiment No.	Fuel content, %	Roasting temperature, °C	Air speed, m/s	
1	0	1150	0.3	
2	1	1150	0.3	
3	0	1250	0.3	
4	1	1250	0.3	
5	0	1200	0	
6	1	1200	0	
7	0	1200	0.6	
8	1	1200	0.6	
9	0.5	1150	0	
10	0.5	1250	0	
11	0.5	1150	0.6	
12	0.5	1250	0.6	
13	0.5	1200	0.3	
14	0.5	1200	0.3	
15	0.5	1200	0.3	

Pellets using wood:

$$M = -16,603 + 1,418 \cdot F_c + 27.79 \cdot T - 239 \cdot V_{air} + +30.8 \cdot F_c^2 - 0.01146 \cdot T^2 + 123 \cdot V_{air}^2 - -1.377 \cdot F_c \cdot T + 375.5 \cdot F_c \cdot V_{air} + 0.038 \cdot T_c \cdot V_{air},$$
(3)

 R^2 =95.79 %, R^2 (adj)=88.22 %, F-value=12.65, P-value=0.006.

Pellets using wheat straw:

$$M = -14,042 + 223 \cdot F_c + 24.1 \cdot T - 1,306 \cdot V_{air} - -72.3 \cdot F_c^2 - 0.01017 \cdot T^2 - 23 \cdot V_{air}^2 - -0.262 \cdot F_c \cdot T + 136.2 \cdot F_c \cdot V_{air} + 1.053 \cdot T_c \cdot V_{air},$$
(4)

 $R^2=92.16$ %, R^2 (adj)=78.05 %, F-value=6.53, P-value=0.026.

Pellets using sunflower husks:

$$M = -52,094 + 534 \cdot F_c + 87.7 \cdot T - 2,628 \cdot V_{air} - -386.6 \cdot F_c^2 - 0.03676 \cdot T^2 - 792 \cdot V_{air}^2 - -0.266 \cdot F_c \cdot T + 196 \cdot F_c \cdot V_{air} + 2.56 \cdot T_c \cdot V_{air},$$
(5)

 R^2 =93.49 %, R^2 (adj)=81.76 %, F-value=7.97, P-value=0.017.

On the basis of standardized regression coefficients, Pareto tables were constructed for each type of biofuel showing the effect of different factors on the dependent variable. The standardized coefficient in regression analysis is used when the data are standardized so that the variances of both the dependent and independent variables are equal to 1. This makes the coefficient dimensionless and allows comparison of the effects of different variables. The standardized coefficient shows how the dependent variable will change when the predictor changes by one standard value.

5.2. Factors of influence and their nature on the strength of pellets

In this analysis, the F_c , T and V_{air} factors were coded as A, B, and C, respectively. Standardized effects (Fig. 1–4) show how strongly each factor affects pellet strength. To visualize the direction of influence of various factors, the main factors are plotted in Fig. 5–8.







Fig. 2. Pareto table of standardized effects for pellets with wood



Our results show the influence of variables on the strength of pellets. The fuel content variables have the greatest influence, and in the case of sunflower husks, the quadratic fuel content variable. Therefore, it was decided to focus attention on explaining those factors that have the greatest influence. In the future, it is planned to investigate the influence of other variables that also affect strength.



Fig. 4. Pareto table of standardized effects for sunflower husk pellets



Fig. 5. Effect of charcoal content on pellet strength



0,0 0,2 0,4 0,6 0,8 1,0 Fuel content, %



Fig. 8. Effect of sunflower husk content on pellet strength

5.3. Optimum roasting parameters when using different types of biofuel

To establish optimal roasting parameters, partial derivatives from regression equations were used for each factor. Derivatives were equated to zero and a system of three equations was solved. In this way, the value of the local maximum in the studied interval of variation of each factor was found. The optimization parameter was the compressive strength of pellets. The following technological conditions of the pellet roasting process were determined (Table 4).

Table 4

Technological conditions of the pellet roasting process with the addition of fuel of plant origin

Type of fuel	% fuel	Т	Vair	Strength, kg/pellet
Charcoal	0.19	1,150	0	234
Wood	1	1,150	0.6	213
Straw	0,1	1,183	0	202
Sunflower husks	0,8	1,200	0.37	238

The strength of pellets over 200 kilograms would allow their melting in a blast furnace.

5. 4. Micro- and macrostructure of roasted pellets

After roasting the pellets, in order to analyze the resulting structure, microstructural and macrostructural analyses were carried out and the following images were acquired (Fig. 9).

Fig. 9 shows images of samples without the use of fuel, as well as samples with the addition of sunflower husks as the most promising type of fuel considered.

6. Discussion of results related to the influence of biofuel on the strength of pellets

 R^2 and R^2 (adj) of all resulting regression equations (2) to (5) have high values of 80 % or more. A high R^2 value indicates the percentage of variation explained by the model, and an adjusted R^2 (adj) takes into account the number of factors in the model. These are good indicators, indicating that the model has a high explanatory power. A Fisher test (*F*) value significantly greater than 1 indicates that the model has significant explanatory power. A low p-value (less than 0.05) means that the probability of obtaining such results by chance is small. This indicates high statistical significance of the model. Hence, the models are well suited to explain the variation in the dependent variable and are statistically significant.

The plots of effect of charcoal content (Fig. 5) show that the dependence of the strength of pellets on the charcoal content has a very steep slope, the strength indicators tend to 0 at the maximum charcoal content of 1 %. The Pareto table for charcoal (Fig. 1) indicates the decisive influence of this variable on the resulting indicator. Such dependences can be explained by the fact that charcoal, firstly, has a high content of volatile substances that begin to be released at relatively low temperatures, compared to fossil coal [6]. As a result of this process, microcracks are formed in the pellet even before the main roasting temperatures are reached, so during roasting the pellet does not reach the required strengthening values. Studies have shown a steady decrease in the compressive strength of the pellets with an increase in the proportion of charcoal in the mixture with fossil coal. The role of the content of volatile substances is especially emphasized, a clear dependence has been established that precisely with the growth of the share of charcoal with a high content of volatile substances, the compressive strength of the pellets decreases.

Another reason that can explain such a sharp decrease in the strength of pellets with the addition of charcoal is the hydrophobicity of this material. Hydrophobicity of coal is explained by its structural and chemical characteristics. The main reason for the hydrophobicity of coal is its high content of aromatic compounds, such as polycyclic aromatic hydrocarbons, which have strong hydrophobic properties. These compounds have a tendency to form $\pi-\pi$ interactions, which leads to a higher sorption of hydrophobic organic compounds on the coal surface compared to other types of organic matter [10].



Fig. 9. Images of the macro and microstructure of roasted pellets: a - T = 1,200 °C, $V_{air} = 0$ m/s, without fuel, x50; b – without fuel, x500; c – sunflower husk 1 %, T = 1,250 °C, $V_{air} = 0.3$ m/s, x50; d – sunflower husk 1 %, T = 1,250 °C, $V_{air} = 0.3$ m/s, x500; 1 - gray areas; 2 - pores; 3 - spongy zones; 4 - white zones

Analyzing the diagram of the influence of factors, one can see that the wood content in the layer of pellets (Fig. 2, 6) still has the greatest influence on strength. But the content of this type of fuel does not have such a pronounced effect in comparison with charcoal. This can be explained as follows. Unlike charcoal, untreated wood does not have such pronounced hydrophobic properties. Raw biomass consists of three main components: cellulose, hemicellulose, and lignin. Cellulose is a polymer consisting of long glucose chains that form strong micro- and macrofibrils connected by hydrogen bonds. These bonds and the presence of hydroxyl groups (-OH) make cellulose hydrophilic, as these groups easily interact with water molecules, forming hydrogen bonds [11]. Wood has a lower heat of combustion, which stabilizes the temperature regime compared to charcoal, which has a much higher heat of combustion.

Analyzing the diagram of the standardized effects of straw and the influence of straw content (Fig. 3, 7), one can see that the amount of wheat straw content in the pellet layer has a sharply negative effect on strength after roasting.

This can be explained by the fact that wheat straw has a high concentration of hydrophobic waxes on its surface. These waxes form a hydrophobic layer consisting of a cutin coating and wax particles. Hydrophobic waxes create boundaries between straw particles, which leads to a decrease in the strength of the pellets. These waxy layers prevent the formation of strong interparticle connections, limiting the interaction through hydrogen bonds and the formation of solid bridges [12]. It can be assumed that the presence of a cutin coating caused a significant decrease in the strength of the pellets compared to wood that does not have similar waxy coatings. The release of a significant amount of volatile substances further enhanced the strengthening effects during roasting. Possible ways to overcome this negative effect are the use of smaller fractions, which makes it possible to improve its interaction with other components of the mixture and increase the contact area between particles. One can also use different methods of removing hydrophobic waxes from its surface.

Analyzing the diagram of the standardized effects of sunflower husk (Fig. 4), one can see that the quadratic coefficients of the fuel content and temperature, as well as the standard coefficient of the fuel content, have a decisive influence on the strength of the pellets after roasting. The resulting dependence is atypical in the context of the previously discussed types of fuel of plant-based origin. This means that the use of sunflower husks has an extreme dependence of pellet strength on solid fuel content (Fig. 8). The peak strength of the roasted pellets is observed when the sunflower husk content is approximately 0.4 %.

This can be explained by the fact that the pore size distribution is comparatively narrower in the case of using sunflower husks. This may be due to the uniform distribution of the sunflower husk, which causes a minimal temperature gradient throughout the pellet. A narrow range of pore size distribution contributes to the high strength of granules [13], improves the efficiency of packing inside the porous solid matrix [14]. This leads to a decrease in the stress concentration around the pores, contributes to the homogeneity of the material properties, which leads to the development of isotropic characteristics in the porous solid structure [7]. The formation of various phases, such as spongy zones, white zones, and gray zones, can be observed on the images of the macro- and microstructure (Fig. 6). In gray zones, slag-forming materials are concentrated, the iron content in these zones is less than 10 %. Instead, spongy areas, pores, and white areas are rich in iron. Iron content in white areas is 72 %. The increase in compressive strength can be associated with the formation of a zone matrix rich in iron and a uniform distribution of the matrix throughout the granule [7]. It can also be noted that the shape of the pores in the case of the addition of plant-based fuel has a more spherical shape compared to the samples without the addition of solid fuel. Spherical pores can interfere with crack propagation.

Also, when analyzing the properties of sunflower husks, one can see that sunflower husks have a lower lignin content compared to woody types of biomass, which explains the different efficiency of using these types of fuels as a component of pellets. According to [15], the content of lignin in sunflower husk is 15.5 %, while the content of lignin in the group of gymnosperms, which accounts for about 80 % of the world production of lumber [16], reaches 40 % [17]. Lignin provides a hydrophobic surface that allows trees to transport water to a height and contributes to mechanical strength that allows the tree to support a large weight [18]. It should also be noted that in addition to the fact that lignin is a hydrophobic substance in itself, the complex structure of lignin prevents the interaction of hydrophilic chemicals with cellulose [18]. It should be noted that in work [7], in which an increase in the compressive strength of pellets was observed with the addition of fuel of plant-based origin, the fuel added, namely rice husk, also has a low percentage of lignin – 7 % [19].

Instead, the content of cellulose and hemicellulose in sunflower husk is 56.5 % and 28 %, respectively [19]. Cellulose and hemicellulose contain a significant amount of hydroxyl OH groups. Hydroxyl groups are hydrophilic groups and can increase the water-holding capacity of raw pellets [20]. For example, in [21] it is noted that the OH groups of starch ensure effective adsorption with the surface of bentonite sheets, which leads to an increase in the hydrophilicity of bentonite. In particular, the electronegativity of hydroxyl groups was used to explain the chemical adsorption of these groups on aluminum (Al^{3+}), which is located at the edges of the montmorillonite layers in bentonite. The low electronegativity of Al3+ promotes its interaction with oxygen and further formation of chemical bonds with anionic functional groups of organic binders. This leads to an increase in the negative charge on montmorillonite and an improvement in its dispersion, as well as a decrease in the size of bentonite particles and an increase in surface area [21].

Our study has certain limitations that should be taken into account when interpreting the results and their practical application. First, the experiments were conducted under laboratory conditions using small-sized briquettes (diameter 10 mm, height 10 mm), which may not fully reflect the behavior of pellets under industrial conditions. This can affect the reproducibility of results when scaling up the process. Second, the ranges of input data such as biofuel content (0–1 %), roasting temperature (1150–1250 °C), and air velocity (0–0.6 m/s) were limited. Therefore, the results are adequate only within these ranges and may not be reproduced beyond them. In addition, biomass properties can vary depending on the batch of raw material, which can affect the stability and repeatability of the results.

As regards the shortcomings of our study, one may note the limited number of studied types of biofuels and their concentrations. This limits the possibility of generalizing the results and their application to a wider range of biofuels. Also, the economic aspects of biofuel use, such as the cost of raw materials, logistics and the impact on overall production efficiency, were not taken into account. To eliminate these shortcomings, future research should expand the range of studied biofuels and their concentrations, include economic analysis, and optimize measurement techniques using stateof-the-art equipment.

Further development of the research may consist in scaling up the experiments to industrial conditions, which would make it possible to evaluate the practical efficiency of using biofuel in pellets. The effects of other types of biomass and their mixtures could also be investigated, as well as the long-term effects of using biofuels on the operation of blast furnaces. Construction of mathematical models for the biofuel roasting process would help in predicting the results and optimizing the process. However, difficulties may arise on this path, such as the diversity of biomass properties, difficulties in the technical implementation of biofuel integration into existing technological processes, as well as the difficulty of mathematical modeling of complex physicochemical processes that occur during the roasting of pellets with biofuel.

7. Conclusions

1. As a result of the study, regression equations were derived for four types of biofuels: charcoal, wood, wheat straw, and sunflower husks. These equations describe the dependence of pellet strength on biomass content, roasting temperature, and air velocity. For sunflower husks, it was established that the maximum strength of the pellets is achieved at a certain optimal content of biofuel. This can be explained by the fact that when using sunflower husks, the pores have a more uniform and narrow size distribution. This is probably due to the uniform distribution of sunflower husks in the pellet, which leads to a minimal temperature gradient throughout the volume. The presence of a narrow range of pore sizes helps increase the strength of the granules. Another factor in the strengthening of pellets using sunflower husks is the composition of the biomass, namely the low content of lignin.

2. Analysis of standardized regression coefficients and constructed Pareto tables revealed that the biofuel content is the most significant factor affecting pellet strength. It was found that the addition of charcoal and wheat straw significantly reduces the strength of the pellets due to their hydrophobic properties and high content of volatile substances. On the other hand, sunflower husk has an extreme character of influence, where at the optimal content (about 0.36%) the strength of the pellets increases compared to the control samples without biomass. This differs from known results and is explained by the peculiarities of the chemical composition of the sunflower husk, in particular, the low content of lignin and the high content of hydrophilic components.

3. For each type of biofuel, the optimal content, roasting temperature, and air speed have been determined, which provide for the strength of pellets over 200 kg/pellet, suitable for use in a blast furnace. In particular, the optimal conditions for sunflower husks are as follows: biofuel content, 0.8 %; temperature, 1200 °C; air speed, 0.37 m/s; under which the strength of pellets reaches 238 kg/pellet. This makes it possible to practically use sunflower husks and wood as an alternative to fossil fuels, which differs from conventional approaches and helps reduce costs and environmental impact.

4. After analyzing the macro- and microstructure of the pellets, it was established that the use of biofuel causes the development of mass transfer processes, as well as the formation of zones of concentration of iron and slag-forming elements. The formation of a matrix structure with an even distribution of iron-rich zones helps increase the strength of the pellets, especially when using sunflower husks. It was also found that the pores have a predominantly spherical shape, which prevents the propagation of cracks under load and additionally increases the compressive strength. These structural features provide a new understanding of the mechanisms for pellet strengthening when using the considered types of biofuel.

Conflicts of interest

The authors declare that they have no conflicts of interest in relation to the current study, including financial, personal, authorship, or any other, that could affect the study, as well as the results reported in this paper.

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Data availability

All data are available, either in numerical or graphical form, in the main text of the manuscript.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies when creating the current work.

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