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Application of thermodynamic equilibrium modelling to predict slagging and fouling tendencies of bituminous coal and wheat straw blends.

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Abstract. The combustion or co-combustion of biomass or alternative fuels is important in the energy sector because of the need to reduce the share of fossil fuels. This article is a continuation of previous studies on the behaviour of the mineral matter of selected fuels during the sintering processes. The blends of wheat straw biomass from Polish crops (WS) with bituminous coal from the Makoszowa mine (BC) were studied. The study included proximate and ultimate analysis and oxide analysis of ash blends with the following composition: 10wt% WS/90wt%BC, 25wt% WS/75wt%BC and 50wt% WS/50wt%BC. Based on the oxide content, a prediction (using FactSage 8.0 software) of the sintering process of the mixtures tested. The following parameters were determined: slag phase content, specific heat at constant pressure, and ash density. The fracture stresses tests were carried out using the mechanical test. Pressure tests were also performed using the pressure drop test method. The test results of all test methods used were compared with each other. On the basis of this comparison, a clear correlation was found between the sintering temperatures determined by the mechanical method and the pressure drop method and the physical properties of the ashes, such as density and heat capacity, as well as the chemical properties, i.e. the content of the slag phase. The results of the presented research are a valuable addition to the previous work of the authors. The goal of this work is to develop a precise and measurably simple method to determine the sintering temperature of ashes. This is an extremely important issue, especially in the case of the need to use a wide range of fuels in the energy industry.

Key words: sintering; biomass; coal; FactSage; mechanical test; pressure drop test, co–combustion

1. INTRODUCTION

The problem of reducing CO_2 emissions requires multipronged solutions. It is important not only to introduce renewable energy sources but also to increase the efficiency of energy conversion systems based on the combustion/co-combustion of fossil fuels. Historically, the main source of thermal, mechanical, and electrical energy is the chemical energy stored in fossil fuels.
Of course, currently fossil fuel combustion technologies are gradually replaced with technologies for converting wind energy, solar energy, or geothermal energy stored on the Earth, and also in the future with nuclear fusion energy [1–3]. This action is inevitable due to the problem of CO_2 emissions, but also due to the limited resources of fossil fuels [4]. Due to the need to supply many industries with continuously high electrical power in a 24h/24h cycle, completely abandoning the combustion process is practically impossible. Furthermore, nowadays, the technology to convert and store electricity obtained as a result of the renewable energy conversion process still requires a lot of research and development work [5].
And here, as a solution to the problem of the need to increase the volume of energy obtained from renewable sources in the 24 h / 24 h regime, the combustion and co-combustion of biomass and/or alternative fuels comes to mind [6–8].
Another challenge due to the increased operational risk of the fouling and slagging process is large-scale biomass combustion and/or co-combustion [9-14]. This is related to the chemical composition of the biomass. In particular, the high content of alkali metals lowers the softening point of the ash (compared to coal). It is a problem when co-combustion is realised in a boiler dedicated for coal combustion. A positive effect in the case of biomass combustion compared to coal combustion is the lower

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content of ash generated in the combustion process [15]. However, the deposition of ash on the heating surfaces of the boiler when burning biomass containing a large amount of alkali metals results in the formation of molten deposits that are difficult to remove. The reason for this process is the higher content of alkali metals that reduces the softening point of ash [16, 17]. When these layers are adhered to heat exchangers, they reduce the degree of heat transfer and, consequently, contribute to reducing the efficiency of the energy conversion process. The same thing happens in the high-temperature section. The settling ash undergoes a slagging process, creating a glassy layer on the boiler surface that is difficult to remove. Both slagging and fouling processes reduce the operational efficiency of the boiler and additionally require periodic shutdown of the boiler to remove deposits [18].

Determining the ash sintering temperature is not a simple matter because of the diverse chemical and physical structure of the ash. Both coal and biomass vary greatly depending on their source. In the case of biomass, the composition of the noncombustible substance depends both on the type of biomass and on the type of soil on which it grew. The size and shape of the ash grains also change. Therefore, a precise determination of the sintering temperature of ash from a given fuel should be made for a given batch of ash.

There are many methods based on which the sintering temperatures of coal ash or biomass are determined. The most popular method is the ash fusion test (AFT) [19–21]. This method is based on observations of the change in the shape of a sample formed from ash when it is heated until it melts. The disadvantage of this method is the relatively large impact of the observer on the result of the experiment, which translates into an accuracy in determining the sintering temperature of +/– 50°C. Another commonly used method is the calculation of oxide indices based on the determined oxide composition of the tested ash. This method is based on statistical correlations of various combinations of the basic and acid oxide content with the ash sintering temperature. Several oxide indicators have been proposed, based on which it is possible to determine whether the fuel has a high or low tendency to slagging or ashing. This is a purely statistical method that is not related to the mechanism of transformation of the mineral substance of the ash. However, this method gives satisfactory results in terms of allowing fuel division into fuels with a high/medium/low risk of slagging and fouling [22].

Methods based on observations of changes in the physical properties of the ash during the sintering process are also used. This group of methods includes: TMA Thermomechanical analysis based on the observation of shrinkage of the sample during sintering [23], a method based on the observation of changes in the thermal conductivity of ash during sintering [24], observation of changes in compressive stresses of ash samples after various thermal treatments, observation of changes in the electrical resistivity of ash [25], and other methods.

Interesting results were also obtained by analysing changes in the dissipation factor in the AC electrical method [26], as well as by analysing changes in fracture strength in the mechanical method and pressure changes in the pressure drop test method [22, 27]. The last two methods, together with the method of thermodynamic analysis of ashes during sintering, were tested for bituminous coal and wheat straw biomass. This work is a key complement to earlier mechanical and pressure drop tests conducted for wheat straw biomass and its blends with bituminous coal. This work is very important from the point of view of the methodology of the proposed measurements. Therefore, the current publication extends the research to include blends of bituminous coal and wheat straw.

The authors' goal is to complement the research and check whether the proposed methods can be considered reliable and objective.

2. MATERIALS AND METHODS

In the work presented here, the following ash samples were tested: from wheat straw (WS), from coal (BC) from the Polish mine Makoszowy, and a blend of ashes from wheat straw and coal in the following proportions: 10wt% WS/90wt%BC, 25wt% WS/75wt%BC and 50wt% WS/50wt%BC. The ash was prepared according to European standards [28]. The tested ash had a grain fraction of 200 µm according to the standard [29]. A description of the ash preparation process can be found in [22]. The preparation and measurement procedure is shown in Figure 1.

The technical analysis in the analytical state was carried out in accordance with the Polish Standards for Solid Fuels [30–33]. The composition of the ashes was determined using the Thermo iCAP 6500 Duo ICP plasma spectrometer method using ASCRM-010 as the standard substance. In the strength method,

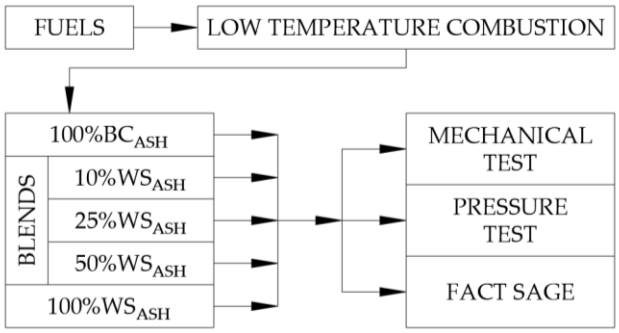


Fig.1. The preparation and measurement procedure.

cylindrical specimens with a diameter of 8-8.5 mm and a height of 10-12 mm were tested. Before each strength test, the ash samples were heated isothermally at a temperature in the range of 500 ° C to about 1000 ° C (in 50°C increments) for 4 h and then quenched at room temperature. The measurement procedure is shown in Figure 2.

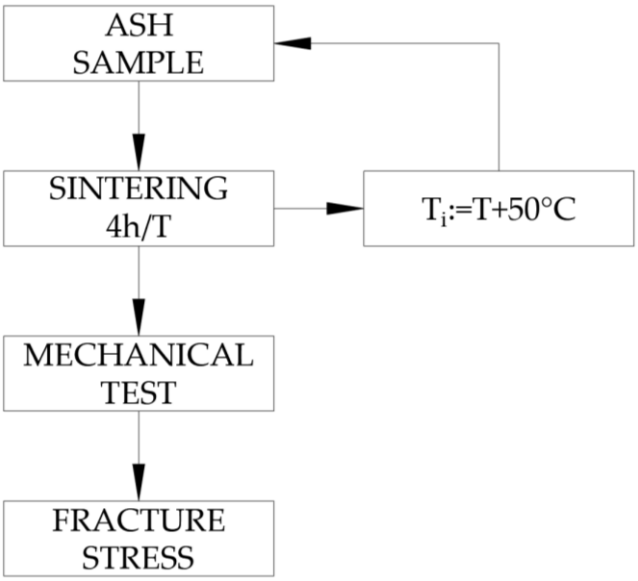


Fig.2. The mechanical test procedure.

For the pressure method, a sample was formed directly in the measuring tube in the form of a cylinder with a diameter of 10 mm and a height of 1.5 mm [22]. The measurement procedure is shown in Figure 3.

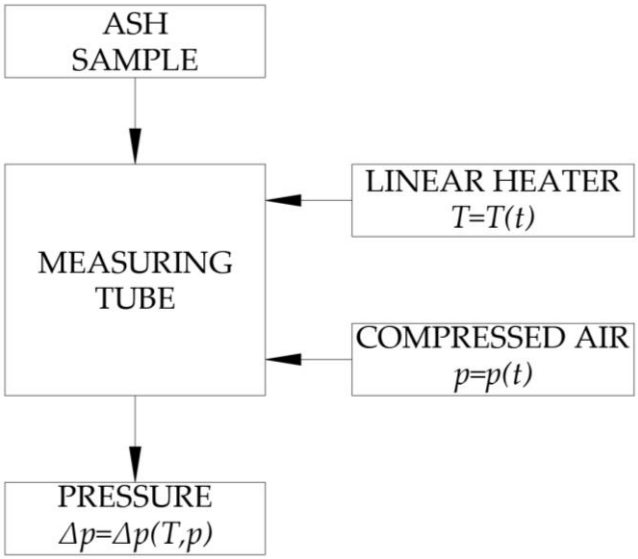


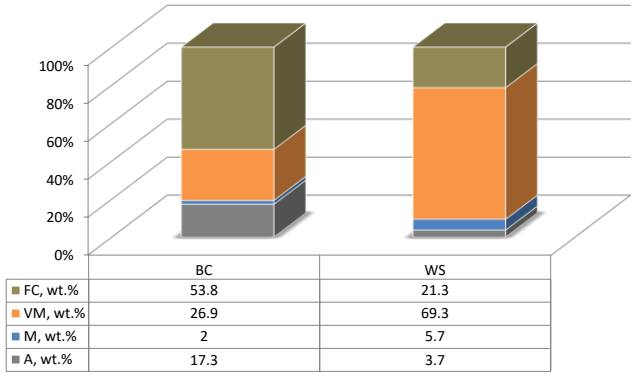
Fig.3. The pressure drop test procedure.

Each mechanical and pressure test was repeated on five samples made from the same ash. The final results are the arithmetic mean, and the uncertainty is the mean standard deviation.
The mechanical test and pressure drop test were complemented by thermodynamic analysis based on minimum Gibbs free energy calculations of nonstoichiometric systems. The analysis was performed using FactSage 8 software (https://factsage.com/). The formation of a single slag phase was assumed, and the FToxid, FTmisc, ELEM and FactPS databases were used. The atmosphere was also excluded [36]. The oxide content of the ashes was entered as input data. Thermodynamic calculations were carried out at temperatures from 500 to 1100 ° C (in 20 ° C increments) under a pressure of 0.1 MPa.

3. RESULTS AND DISCUSSION

3.1 Samples Analsyis

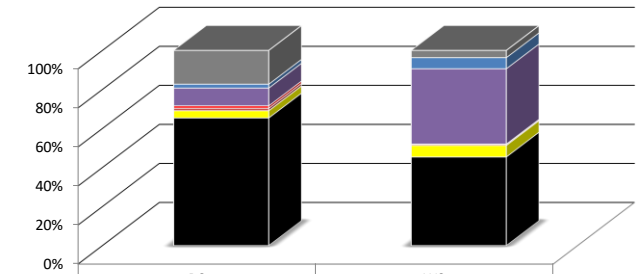
The presented work concerns research on the selected physicochemical properties of ash blends obtained from the ash of coal and biomass. It is an important supplement to the authors' previous research by examining selected physicochemical properties of blends of coal ash and biomass ash.



	BC	WS
FC, wt.%	53.8	21.3
VM, wt.%	26.9	69.3
M, wt.%	2	5.7
A, wt.%	17.3	3.7

Fig.4. The proximate analysis (A—ash, M—moisture, VM—volatile matter, FC—fixed carbon) of fuel samples on air dried basis; BC—bituminous coal, WS—wheat straw[22].

The following fuels were selected as the basis for preparing the blends: bituminous coal from the Polish Makoszowa mine and wheat straw from Polish crops. The content of ash, moisture, volatile substances and fixed carbon is shown in Fig. 4 (procedure described in [22, 34, and 35]).



	BC	WS
A, wt.%	17.3	3.7
M, wt.%	2	5.7
O, wt.%	9.1	38.6
S, wt.%	1.2	0.08
N, wt.%	1.3	0.6
H, wt.%	3.7	5.9
C, wt.%	65.4	45.4

Fig.5. Ultimate analysis (C, H, N, S, O and moisture (M) and ash (A) of the fuel samples on air dried basis; BC—bituminous coal, WS—wheat straw[22].

The content of C, H, N, S, O, ash, and moisture in these fuels is shown in Fig. 5.

The heating value (HHV) and fuel ratio (FR) for wheat straw and hard coal are shown in Fig. 6. The Fuel Ratio was calculated using the formula: FR = FC/VM [22].

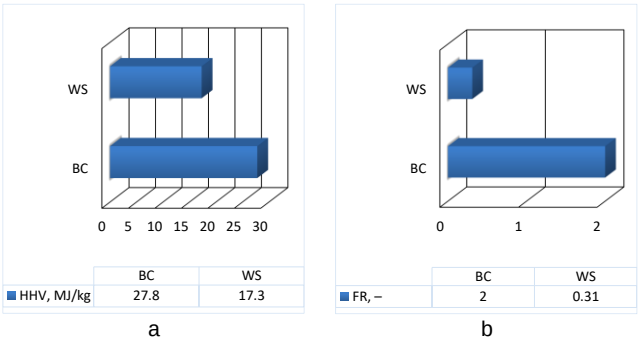
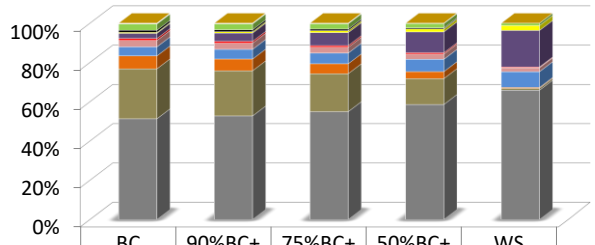


Fig.6. Characteristics of the tested fuels: (a) heating value (HHV) and (b) fuel ratio (FR)[22].

Three mixtures were prepared from hard coal ash and wheat straw ash: 90wt%BC/10wt%WS, 75wt%BC/25wt%WS and 50wt%BC/50wt%WS.



	BC	90%BC+ 10%WS	75%BC+ 25%WS	50%BC+ 50%WS	WS
SrO	0.1	0.09	0.075	0.05	0
BaO	0.2	0.18	0.15	0.1	0
Mn3O4	0.1	0.09	0.075	0.05	0
SO3	3.2	3	2.7	2.2	1.2
TiO2	1.1	0.99	0.825	0.55	0
P2O5	0.5	0.7	1	1.5	2.5
K2O	2.6	4.2	6.6	10.6	18.6
Na2O	1	0.94	0.85	0.7	0.4
MgO	3.3	3.17	2.975	2.65	2
CaO	4.6	4.94	5.45	6.3	8
Fe2O3	6.6	5.99	5.075	3.55	0.5
Al2O3	25.5	23.04	19.35	13.2	0.9
SiO2	51.4	52.84	55	58.6	65.8

Fig.7. Composition of the ash of tested samples (wt.%).

The oxide composition of hard coal and wheat straw was also determined (Thermo iCAP 6500 Duo ICP plasma spectrometer method using ASCRM-010). The oxide composition of the mixtures was calculated according to the procedure described in [22]. The results are shown in Fig. 7.

3.2 Results of pressure drop test, mechanical test, and FactSage analyse

Ash obtained by low temperature ashing of selected bituminous coal from the Polish Makoszowa coal mine (BC) and wheat straw from Polish crops (WS) and their mixtures (10wt% WS/90wt%BC, 25wt% WS/75wt%BC and 50wt% WS/50wt%BC) were examined for changes in their strength properties and changes in the microstructure of the compressed samples during their sintering process. The change in strength properties, particularly fracture stress, was examined using the strength method, while the change in microstructure was examined using the pressure method. The sintering process was carried out in the temperature range of 500°C to approximately 1100°C.

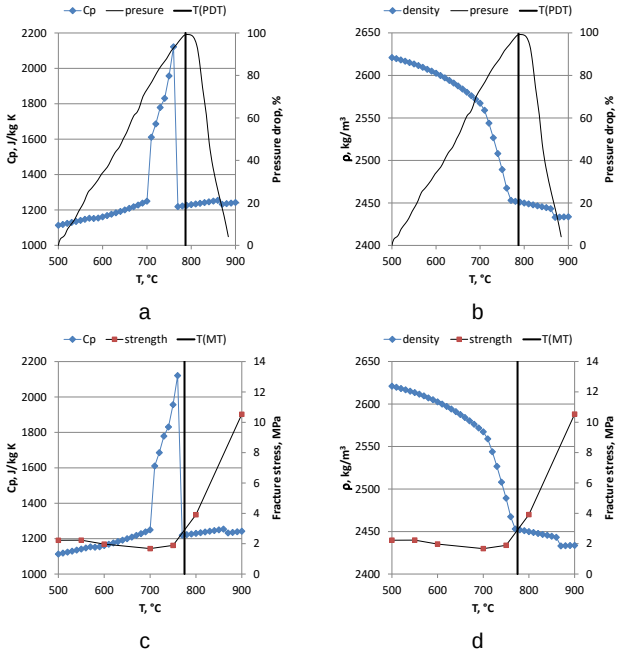


Fig.8. Wheat straw: (a) Specific heat (c_p) and pressure drop as a function of temperature; (b) density (ρ) and pressure drop as a function of temperature; (c) specific heat and fracture stress as a function of temperature; (d) density and fracture stress as a function of temperature; T(PDT) - pressure drop test) and T(MT) - the mechanical test.

The results of the pressure drop test measured during the wheat straw ash sintering process and the changes in heat capacity and also ash density (predicted using FactSage 8.0 software) are shown in Figs. 8 a and b. Looking at Fig.8 a and b, it can be seen that the temperature at which the pressure drop begins corresponds to a discontinuity in the heat capacity cp (Fig.8 a) and, at the same time, a marked decrease in the density of ash (Fig.8 b). The results for biomass end at 900°C, because above this temperature the biomass undergoes a phase transformation into a slag phase. The observed discontinuity in specific heat is related to phase transformations occurring in the ash during its sintering. The decrease in density, on the other hand, may be indicative of changes at the microstructural level of the sample. Therefore, during the sintering of wheat straw, as shown in [22], the observed changes are rather related to a change in the microstructure of the sample under study. In particular, an increase in porosity and the formation of air transport channels inside the sample are observed. Similarly, good agreement was observed between the temperature range in which there is a clear increase in fracture stress and discontinuity in heat capacity (Fig. 8c). Fig. 8d shows that a significant decrease in sample density occurs in the same temperature range as a significant increase in fracture stress.

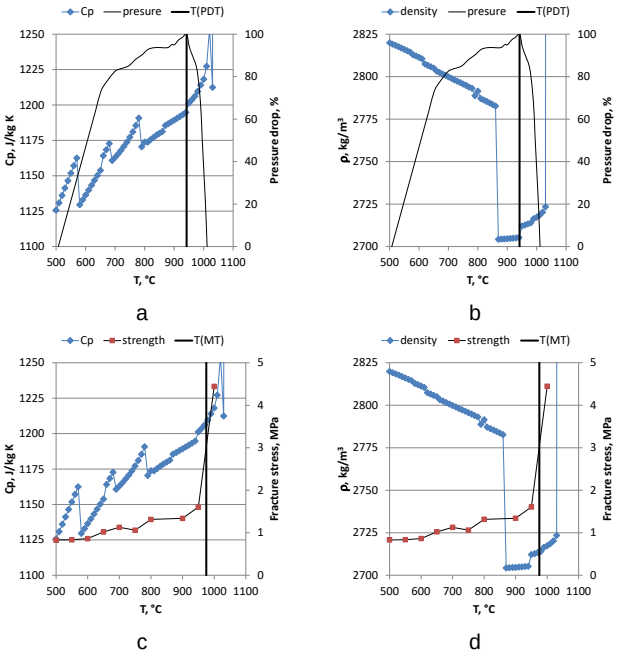


Fig.9. Bituminous Coal: (a) Specific heat (c_p) and pressure drop as a function of temperature; (b) density (ρ) and pressure drop as a function of temperature; (c) specific heat and fracture stress as a function of temperature; (d) density and fracture stress as a function of temperature; T(PDT) - pressure drop test) and T(MT) - the mechanical test.

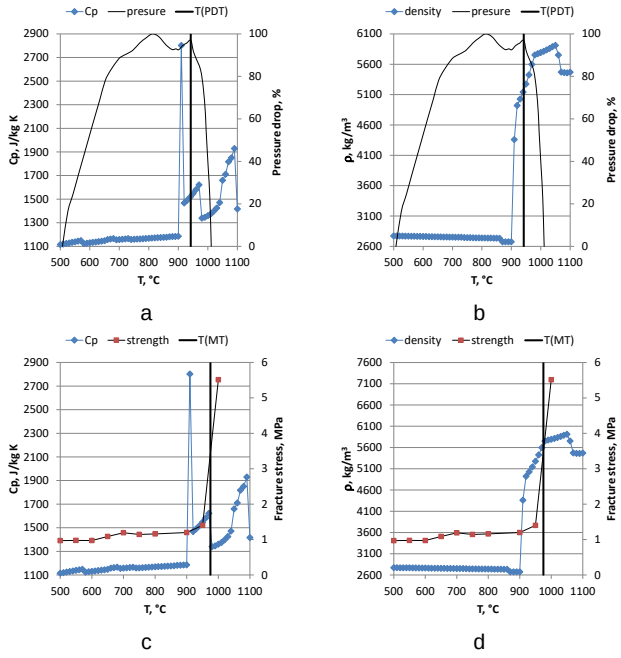


Fig.10. 90%BC+10%WS: (a) Specific heat (c_p) and pressure drop as a function of temperature; (b) density (ρ)and pressure drop as a function of temperature; (c) specific heat and fracture stress as a function of temperature; (d) density and fracture stress as a function of temperature; T(PDT) - pressure drop test) and T(MT) - the mechanical test.

Fig. 9 shows analogous relationships for bituminous coal. As shown in Fig. 9a, the sharp drop in pressure in the pressure drop method can be associated with the discontinuity of specific heat, but the course of specific heat itself in the tested temperature range has many more points of discontinuity related to phase changes occurring in the coal ash. The density changes (Fig. 9b) compared to the drop test results show the compliance of the pressure drop temperature range with the increase in the density of the bituminous coal ash sample. In this case, the pressure drop is caused by the sample shrinkage associated with the increase in its density. The increase in fracture stress during the bituminous coal ash sintering process occurs in the same temperature range in which the discontinuity of the heat capacity is observed (Fig. 9c) as well as the increase in the sample density (Fig. 9d)

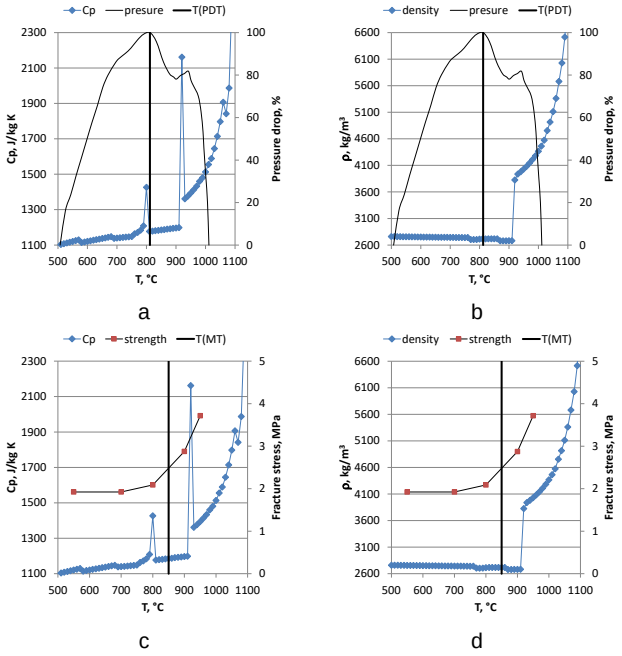


Fig.11. 75%BC+25%WS: (a) Specific heat (c_p) and pressure drop as a function of temperature; (b) density (ρ) and pressure drop as a function of temperature; (c) specific heat and fracture stress as a function of temperature; (d) density and fracture stress as a function of temperature; T(PDT) - pressure drop test) and T(MT) - the mechanical test

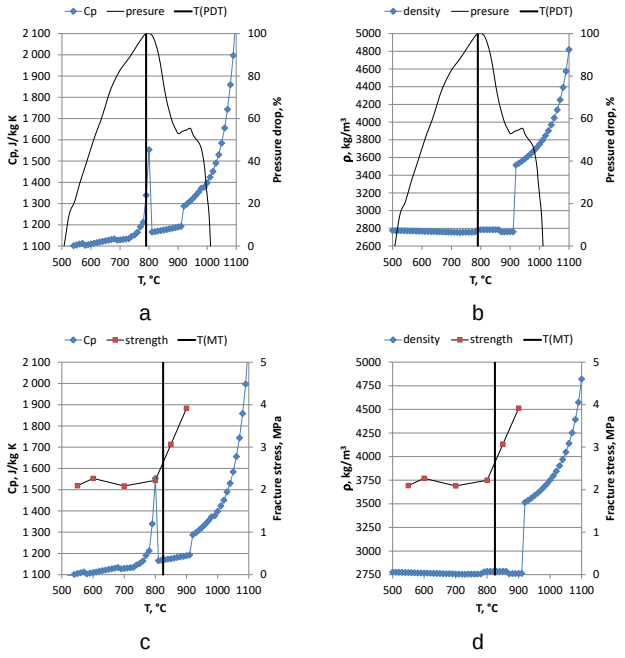


Fig.12. 50%BC+50%WS: (a) Specific heat (c_p) and pressure drop as a function of temperature; (b) density (ρ)and pressure drop as a function of temperature; (c) specific heat and fracture stress as a function of temperature; (d) density and fracture stress as a function of temperature; T(PDT) - pressure drop test) and T(MT) - the mechanical test.

Figs. 10–12 present the results of pressure drop and fracture stress tests performed during the sintering process of wheat straw and bituminous coal ash blends compared to the prediction (using FactSage 8.0) of heat capacity and density.

As we can see (Figs. 10–12 b and d), even a small addition of wheat straw to bituminous coal causes a change in the sintering mechanism in its initial phase. During the sintering of bituminous coal ash, the pressure drop is caused by the leakage caused by the shrinkage of the compressed ash sample, while in blends of WS and BC, there are changes at the microstructure level in the form of an increase in porosity and the formation of channels through which air migrates in the pressure drop method. Microscopic studies for selected coals and biomasses were conducted previously. The existence of changes at the microstructural level leading to changes in porosity (formation of channels inside the sintered ash as a result of pore merging) was found [37].

Figs. 8-12 show good compliance of the characteristic changes of pressure in the pressure drop test and fracture stress in the mechanical test, with the prediction of changes in heat capacity and ash density, during its sintering. Moreover, comparing the results shown in Figures 8-12 with those of an earlier paper (barley straw and its blends with bituminous coal) [27], there is very good qualitative agreement in all the relationships studied (cp vs. pressure drop, density vs. pressure drop, cp vs. fracture stress and density vs. fracture stress).

On this basis, as well as on the basis of the conclusions from previous research [22], the sintering temperatures of the tested ashes were determined. The results are presented in Fig. 13.

Looking at Fig. 13, it can be concluded that the 10wt% addition of wheat straw ash to bituminous coal ash practically does not affect the value of the sintering temperature for both the pressure and mechanical tests. The addition of 20wt% of wheat straw ash, causes a relatively large (compared to coal) reduction in the sintering temperature (about16%). However, the addition of 50wt% wheat straw reduces the sintering temperature by only about 3% compared to bituminous coal.

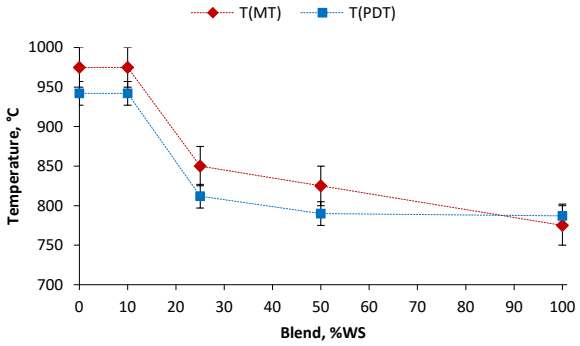


Fig.13. Sintering temperature of the blend %WS determined by the pressure drop test T(PDT) and the mechanical test T(MT).

As we can see based on the attached test results, the addition of 10wt% of wheat straw ash, does not cause a significant reduction in the ash sintering temperature, while the mechanism of transformation of the mineral substance of the sintered ash seems to be different, i.e. different nature of density changes (Fig. 10 b, d and 13).

To explain this discrepancy (similar sintering temperatures but different nature of density changes), the content of the slag phase in individual ashes was determined using FactSage 8.0 software. The results are shown in Fig.14. Looking at Fig. 14a, one can notice a similar course of slag phase formation for bituminous coal ash and the blend of 90wt%BC/10wt%WS ash in the initial phase of the sintering process: in the temperature range from 700 ° C to 900 ° C. Additionally, the growth dynamics of the slag phase in the higher range (up to approximately 910°C) is practically identical. This supports the statement that the sintering temperature is related not only to the absolute content of the slag phase but also to the dynamics of the slag phase formation.

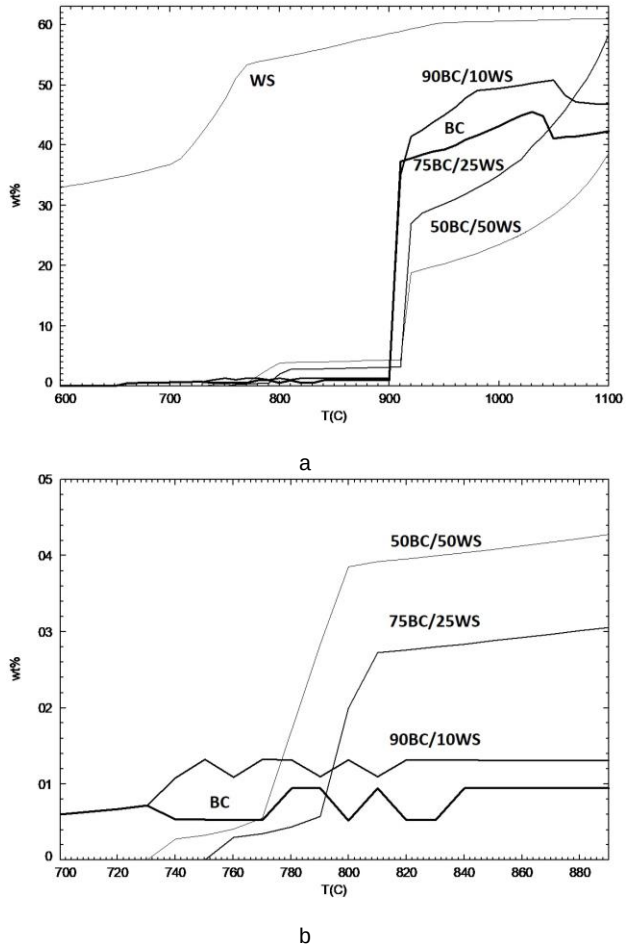


Fig.14. Phase of slag in equilibrium for bituminous coal (BC), wheat straw (WS) and the blends (a) and the initial stage of slag phase formation (b).

This is confirmed by the result for 25wt%WS/75wt%BC and 50wt%WS/50wt%BC blends. On the basis of Fig. 14, it is clearly visible that the growth dynamics of the slag phases is similar to each other. This may explain the small difference in sintering temperatures shown in Fig. 13.

6. CONCLUSIONS

Based on the studies presented, it was concluded that:

1. Equilibrium simulations (FactSage 8.0 software) confirm previous results, i.e., good correlation between the sintering temperatures determined by the pressure drop test, as well as the mechanical test with characteristic changes in heat capacity and density of sintered ash.
2. The predicted density changes (FactSage 8.0 software) density changes of all the tested samples clearly indicate that the ash sintering temperature determined in both custom tests, the mechanical test and the pressure drop test, is closely related to the initial stage of the sintering process, when the properties of the sintered ash begin to change.
3. The onset of the sintering process in selected samples of BC coal ash and WS biomass ash mixtures, a good correspondence between the sintering temperature determined by both nonstandard techniques, is manifested by an increase in fracture stress (mechanical test) and manifested by shrinkage of the samples or an increase in their porosity (pressure drop test).
4. The results presented show that the mechanism of the mineral matter transformation of fuels, especially biomass and coal / biomass blends, is very complex. It is suggested that not only chemical reactions or phase changes occur, but also changes in the microstructure of the ash occur.
5. The process of transformation of the mineral substance of fuels, particularly biomass and coal/biomass blends, has not been fully recognized up until now.

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