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The effect of drying of bituminous coals on their coking properties

Key words

Bituminous coals, coking properties, coal drying

Abstract

The paper presents results of investigations of the effect of coking coals drying up to the moisture content 4 and 6% on their coking properties. The coals were dried in the atmosphere of nitrogen and air at the temperatures of 150°C and 210°C. No significant deterioration of coking properties of the dried coals was recorded.

Introduction

Quality of coke, and, especially, its mechanical properties and reactivity, are influenced mainly by coal quality and the way the coal charge is prepared for the coking process. By and large, the properties of coal charge determine coke quality, but with the limited potential of domestic coking coal resources and increasing quality requirements of main coke buyers, i.e. blast-furnace men, the problem of adopting appropriate technology of preparing coals and blends to be introduced to the coking chamber becomes very important.

In respect of technology, preparation of coal charge for the coking process involves:

— crushing or, if necessary, screening and crushing of coals in order to obtain optimal grain size composition of coal blend,

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- increasing the bulk density of coal charge inside the coking chamber by stamping or briquetting,
- preliminary thermal preparation (full drying, partial drying and preheating of coal charge).

The last of the above mentioned operations aims at restricting or eliminating coal drying inside the coking chamber. After unsuccessful attempts (technical difficulties) at introducing the technology of filling coking chambers with dry coal charge and pre-heated coal charge, there remains the interesting technology of coal charge partial drying up to the total moisture content of 5-6%. Japanese cokemaking experience (Tateoka 1992; Uchmylova 1991), which involved introduction of this technology known as CMC (Coal Moisture Control), at several coking plants, shows that drying of coal charge up to the above-mentioned moisture content has beneficial effects, e.g. an increase in the production capacity of the coke oven battery, a reduction of heat consumption for the coking process, an improvement of coke quality and operating conditions of the brickwork of coking chambers as well as a decrease in the amount of coking wastes and impurities contained in them. Keeping moisture content in the coal blend at the level of 5—6% does not cause problems which would occur while coking of a fully dried and pre-heated coal charge, connected mainly with an excessive dust emission, contamination of by-products, particularly tar, as well as with the danger of an excessive coking pressure leading, in extreme cases, to the damage of the coking chamber walls.

At present, the process of coking coal drying (CMC) most often takes place in tubular dryers, in which heat is transferred indirectly to the coal charge using a heat exchanger (the heating agent is steam) and the moisture which evaporated from coal is removed from inside the tubes by a stream of air, subsequently subjected to dust separation. Coal blend drying can also be conducted directly with combustion or inert gas in fluidization, flue or plate dryers.

One of the problems which occur during thermal dewatering of coal is the possibility of surface oxidation which leads to the deterioration of standard indices describing coking properties. Examinations of the effect of deep drying of coking coals in various time-temperature conditions and with various oxygen concentrations in the drying agent on the deterioration of coking properties have been widely discussed in literature.

Little data on the subject is available as regards the process of drying up to the total moisture content in coal equal to 4—6% and, therefore, this work aims at providing such data through appropriate investigations of Polish bituminous coals.

1. Experimental

In order to investigate the process of coal drying, special equipment was designed. It is shown in Fig. 1. The gas used for coal drying was proportioned from cylinder I with the use of a reducing valve, and the flow rate was set at $200 \, \text{dm}^3/\text{h}$ and controlled by rotameter 2. Preheating of the drying gas agent up to the set temperature was carried out in steel tube 3

with a filling improving the heat exchange, placed in an electric furnace with horizontal heating chamber 4. The temperature of the drying gas agent was measured with a thermocouple NiCr-Ni situated at the inlet to the reactor 5, in which the process of coal drying took place. A vertical steel drying reactor with 60 mm in diameter and 310 mm in height was heated with the use of tubular oven 6. The oven served for keeping the temperature of the reactor at the same level as the temperature of the drying gas agent. In the upper part of the reactor, a cover was located, which enabled filling it with the examined coal sample of 120 g. Coal, dried in a solid bed, rested inside a tube on a metal sieve. The temperature in the bed was measured by means of a NiCr-Ni thermocouple inserted through an opening in the above-mentioned cover of the reactor — 7. The coal drying operation was controlled with a glass assembly containing a receiver and a cooler 8, in which the water vapour leaving the reactor condensed.

The dynamics and the effects of drying are influenced, among others, by granulation of the dried material and, therefore, before the proper examinations it was necessary to determine representative grain-size distribution of coal, corresponding to that applied at

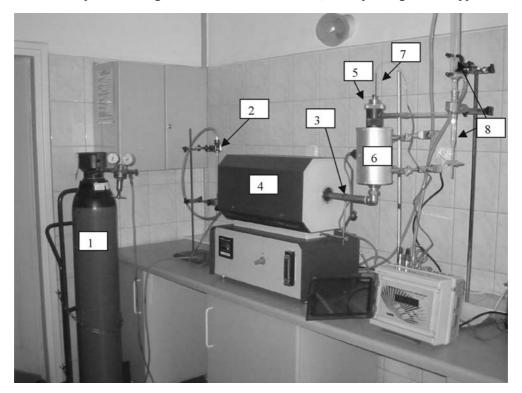


Fig. 1. Laboratory equipment for coal drying

1 — cylinder with drying gas agent, 2 — rotameter, 3 — tube for preheating of drying agent,

4 — electric furnace, 5 — reactor, 6 — tubular oven, 7 — thermocouple,

8 — glass assembly containing a receiver and a cooler

Rys. 1. Stanowisko laboratoryjne do podsuszania węgli

Grain-size distribution of industrial coal blends for the top charging system and grain composition of coals used for the examinations

TABELA 1 Uziarnienie mieszanek wsadowych w systemie zasypowym oraz skład ziarnowy węgli stosowanych w badaniach

| Size fraction | Share of size fraction [%] | | | | | | |
|---------------|----------------------------|----------------|----------------|-------------------------|--|--|--|
| [mm] | coking plant A | coking plant B | coking plant C | laboratory examinations | | | |
| > 3.15 | 12.3 | 20.7 | 16.5 | 16.5 | | | |
| 1.0—3.15 | 22.4 | 22.4 | 28.0 | 24.3 | | | |
| 0.5—1.0 | 16.9 | 15.6 | 18.6 | 17.0 | | | |
| < 0.5 | 48.9 | 41.3 | 36.9 | 42.2 | | | |

Polish coking plants. Examinations of grain-size distribution of coal blends took place at three biggest coking plants, marked with letters A, B and C (Table 1), since only there it is possible to introduce the technology of drying of coal blend for coking in coke oven batteries of the PWR-63 type, running in the top-charging system. The examinations enabled calculation of the average grain-size distribution used subsequently by the authors in their own investigations (the last column of the Table 1). However, it should be pointed out that the upper size of grains was limited to 6.3 mm. In order to evaluate the impact of drying of the examined coals on their coking properties, most widely used standard indices were selected, i.e.:

- Roga Index (RI),
- Swelling Index (SI),
- Maximal expansion pressure p_{max} ,
- Audibert Arnu dilatometer test, i.e. softening point ($t_{\rm II}$), maximum contraction temperature ($t_{\rm III}$) and final swelling temperature ($t_{\rm III}$), as well as contraction (a) and dilatation (b).

To supplement the characteristics of initial coal samples, proximate analysis of those samples was carried out, involving determination of moisture (W^a) , ash (A^a) and volatile matter content (V^{daf}) .

2. Results and discussion

The examinations focussed on low and high volatile bituminous coals which are used as components of coal blends for metallurgical coke production. A representative selection of these coals consisted of three samples of low volatile bituminous coals from Jas-Mos, Zofiówka and Borynia coal mines, and two samples of high volatile bituminous coals from Budryk and Krupiński coal mines. The results of the proximate analysis and an evaluation of coking properties for the initial samples of these coals are shown in Table 2. It can be inferred

Characteristics of coals used for the examinations

TABELA 2 Charakterystyka węgli użytych do badań

| | Proximate analysis | | | | Coking properties | | | | | | |
|-----------|--------------------|-------|-------|--------|-------------------|-----------------|--------------|------------------------|----------------------|--------------------------|---------------------------|
| Coal | W^a | A^a | Vdaf | | | | dila | tometer | test | | |
| Coai | [%] | [%] | [%] | SI [-] | RI [-] | <i>a</i> [%] | <i>b</i> [%] | t _I [°C] | t _{II} [°C] | t _{III} [°C] | p _{max} [kPa] |
| Budryk | 1.6 | 7.9 | 33.98 | 6.5 | 84 | 29 | +21 | 380 | 424 | 455 | 17.4 |
| Krupiński | 2.4 | 8.1 | 39.06 | 4.5 | 73 | 34 | +10 | 370 | 420 | 449 | 13.3 |
| Borynia | 1.2 | 5.4 | 26.85 | 8.0 | 83 | 36 | +156 | 370 | 429 | 484 | 120.8 |
| Zofiówka | 1.3 | 6.8 | 26.44 | 8.5 | 80 | 30 | +119 | 393 | 428 | 512 | 84.6 |
| Jas-Mos | 1.3 | 6.8 | 23.31 | 8.5 | 78 | 21 | +40 | 405 | 446 | 488 | 46.4 |

that the selection of the examined coals represents different coal ranks and correspondingly different coking properties, from coal with the highest rank from Jas-Mos coal mine acting as the semi-leaning component in coal blends, through very good low volatile bituminous coals with medium rank from Zofiówka and Borynia coal mines, to high volatile bituminous coal with lowest rank from Krupiński coal mine, representing average coking properties.

Initial coal samples with the above-mentioned grain composition were wetted up to the total moisture content $W_t^r = 10\%$ and, subsequently, using the equipment described above, subjected to drying up to two levels of moisture content — 6 and 4%. The type of the drying agent and its temperature constituted process variables. For the purposes of the examinations, two extreme variants of the drying gas were applied, i.e. inert nitrogen and air, the latter being a gas with a great oxidation potential. For each of the variants of the drying gas, two values of drying temperature were used, i.e. 150 and 200°C, which obviously resulted in different drying time.

Overall results characterising the effect of the drying process on coking properties (the dilatometer test was restricted to dilatation index (b) and the temperature range of coal plasticity $(t_{\text{III}} - t_{\text{I}})$) are shown in Tables 3—6. A comparison of the results concerning the properties of the initial samples (Table 2) and dried samples (Tables 3—6) shows that the impact of this process on the change in the coking properties of coals is rather small. The value of dilatation (b) is the parameter most easily influenced by changes in properties caused by drying.

In order to establish the significance of effect of drying on the change in coking properties of the examined coals, a statistical analysis of the obtained results was carried out.

The first variant involved testing of the significance of effect of the drying process, to which the examined coals were subjected under the conditions described above, on the values of the parameters characterising coking properties. Statistical hypotheses were tested by comparing the value of the calculated *t*-statistics:

$$t = \frac{\bar{x} - x_o}{S} \cdot \sqrt{n' - 1} \tag{1}$$

where:

 x_o — value of the evaluated parameter in the initial coal (before drying),

 \bar{x} — mean value of the evaluated parameter for samples after drying,

S — estimator of the standard deviation,

n — number of samples of dried coal (n = 8),

with critical value of the Student's *t*-test for the confidence level of 0.95 and for k = n - 1 degrees of freedom ($t_{\alpha=0.05;k=7} = \pm 1.895$). The results of the statistical analysis conducted in such a way are shown in Table 7.

Theory of Testing of Statistical Hypothesis was used to evaluate the impact of the following factors on coking properties:

- final moisture content in the dried coal $W_t^r = 6$ and 4%,
- drying agent: nitrogen and air,
- temperature of drying: 150 and 210°C.

TABLE 3
Coking properties of coals dried in the temperature of 150°C in the atmosphere of nitrogen
TABELA 3
Właściwości koksotwórcze węgli suszonych w temperaturze 150°C w atmosferze azotu

| | Moisture | SI | D.I. | Dilatom | n | |
|-----------|----------------|-----|------------------|-----------------|--|---------------------------|
| Coal | content [%] | [-] | <i>RI</i> [-] | <i>b</i> [%] | t _{III} —t _I [°C] | p _{max} [kPa] |
| D 1 1 | 4 | 5.5 | 75 | +35 | 73 | 12.8 |
| Budryk | 6 | 5.5 | 72 | +3 | 76 | 16.4 |
| T | 4 | 4.5 | 74 | +5 | 65 | 4.5 |
| Krupiński | 6 | 4.5 | 69 | +3 | 57 | 10.6 |
| | 4 | 7.5 | 80 | +133 | 87 | 106.1 |
| Borynia | 6 | 8.5 | 82 | +130 | 88 | 72.7 |
| 7 6 1 | 4 | 8.5 | 82 | +147 | 119 | 104.8 |
| Zofiówka | 6 | 8.5 | 81 | +155 | 99 | 108.6 |
| | 4 | 7.5 | 80 | +31 | 77 | 59.7 |
| Jas-Mos | 6 | 7.5 | 77 | +37 | 85 | 18.8 |

TABLE 4
Coking properties of coals dried in the temperature of 210°C in the atmosphere of nitrogen
TABELA 4
Właściwości koksotwórcze węgli suszonych w temperaturze 210°C w atmosferze azotu

| | Moisture | GI. | D.I. | Dilatom | | |
|-----------|-------------|-----------|------------------|--------------|---------------------------------------|---------------------------|
| Coal | content [%] | SI [-] | <i>RI</i> [-] | <i>b</i> [%] | t _{III} —t _I [°C] | p _{max} [kPa] |
| D 1 1 | 4 | 6.0 | 65 | -4 | 72 | 14.3 |
| Budryk | 6 | 6.5 | 71 | +25 | 75 | 16.5 |
| TZ ./ 1. | 4 | 4.5 | 72 | +9 | 65 | 7.6 |
| Krupiński | 6 | 4.5 | 67 | 0 | 47 | 5.2 |
| ъ . | 4 | 8.5 | 80 | +130 | 88 | 148.8 |
| Borynia | 6 | 8.0 | 82 | +129 | 91 | 152.9 |
| 7.57.1 | 4 | 8.0 | 83 | +136 | 92 | 117.4 |
| Zofiówka | 6 | 8.0 | 83 | +146 | 86 | 107.1 |
| | 4 | 7.5 | 75 | +35 | 83 | 29.6 |
| Jas-Mos | 6 | 8.0 | 78 | +35 | 85 | 32.6 |

TABLE 5
Coking properties of coals dried in the temperature of 150°C in the atmosphere of air

TABELA 5
Właściwości koksotwórcze węgli suszonych w temperaturze 150°C w atmosferze powietrza

| | Moisture | SI | RI | Dilatom | n | |
|-----------|-------------|-----|-----|-----------------|--|---------------------------|
| Coal | content [%] | [-] | [-] | <i>b</i> [%] | t_{III} — t_{I} [°C] | p _{max} [kPa] |
| D 1 1 | 4 | 6.5 | 75 | +21 | 88 | 16.4 |
| Budryk | 6 | 6.5 | 72 | +19 | 77 | 20.6 |
| ., ., . | 4 | 5.0 | 73 | +5 | 69 | 7.6 |
| Krupiński | 6 | 4.5 | 68 | +1 | 51 | 10.9 |
| ъ . | 4 | 8.0 | 80 | +127 | 88 | 136.4 |
| Borynia | 6 | 8.0 | 80 | +118 | 90 | 63.7 |
| 7.5/1 | 4 | 8.5 | 82 | +147 | 119 | 128.5 |
| Zofiówka | 6 | 8.5 | 80 | +153 | 82 | 91.8 |
| | 4 | 7.0 | 75 | +25 | 80 | 40.7 |
| Jas-Mos | 6 | 6.5 | 77 | +23 | 73 | 42.7 |

TABLE 6
Coking properties of coals dried in the temperature of 210°C in the atmosphere of air

TABELA 6
Właściwości koksotwórcze węgli suszonych w temperaturze 210°C w atmosferze powietrza

| | Moisture | GI. | n. | Dilatome | | |
|-----------|----------------|------------------|------------------|-----------------|---------------------------------------|---------------------------|
| Coal | content [%] | <i>SI</i> [-] | <i>RI</i> [-] | <i>b</i> [%] | t _{III} —t _I [°C] | p _{max} [kPa] |
| D 1 1 | 4 | 6.0 | 72 | +20 | 86 | 8.0 |
| Budryk | 6 | 6.0 | 72 | +11 | 80 | 12.0 |
| ** '/ 1 ' | 4 | 4.5 | 70 | +2 | 76 | 4.4 |
| Krupiński | 6 | 4.5 | 71 | +7 | 62 | 3.8 |
| | 4 | 8.5 | 80 | +116 | 83 | 111.6 |
| Borynia | 6 | 8.0 | 81 | +128 | 88 | 91.7 |
| _ ~ | 4 | 8.5 | 82 | +134 | 86 | 109.7 |
| Zofiówka | 6 | 8.0 | 80 | +123 | 91 | 108.4 |
| | 4 | 8.0 | 79 | +27 | 67 | 49.5 |
| Jas-Mos | 6 | 7.5 | 81 | +25 | 82 | 38.5 |

TABLE 7
Testing of the significance of effect of the drying process on coking properties of the examined coals

TABELA 7
Analiza statystyczna istotności oddziaływania warunków suszenia na właściwości koksotwórcze badanych wegli

| Coal | <i>RI</i> [-] | <i>SI</i> [-] | <i>t</i> _{III} — <i>t</i> _I [°C] | <i>b</i> [%] | p _{max} [kPa] |
|-----------|---------------|------------------|--|--------------|---------------------------|
| Budryk | -1 | -1 | 0 | -1 | 0 |
| Krupiński | -1 | 0 | -1 | -1 | -1 |
| Borynia | -1 | -1 | -1 | -1 | 0 |
| Zofiówka | 1 | -1 | -1 | -1 | 1 |
| Jas-Mos | 0 | -1 | 0 | -1 | 0 |

⁰ — insignificant effect on the examined parameter, -1 — significant effect on the decrease in the value of the examined parameter, 1 — significant effect on the increase in the value of the examined parameter).

^{0 —} nie wpływa w sposób istotny na badany parametr, –1 — wpływa w sposób istotny na obniżenie wartości badanego parametru, 1 — wpływa w sposób istotny na zwiększenie wartości badanego parametru).

TABLE 8

In this case, hypotheses were tested by comparing the value of t-statistics:

$$t' = \frac{\overline{x_i} - x_o}{S} \cdot \sqrt{n' - 1} \tag{2}$$

where:

 $\overline{x_i}$ — mean value of the evaluated parameter for samples dried on the *i*-level of the given factor,

n' — number of coal samples dried on the i-level of the given factor (n' = 4),

with critical value of the Student's *t*-test for the confidence level of 0.95 and k'=n'-1 degrees of freedom ($t_{\alpha=0,05;k=3}=\pm 2,353$). The results of statistical analysis conducted in this way are shown in Tables 8 and 9.

The results of the statistical analysis showed that drying of all the examined coals resulted in a decrease in the value of dilatation index b which turned out to be most easily influenced by changes in coal properties caused by drying. In the case of the remaining indices, the drying process led to a decrease in SI (except for the coal from Krupiński coal mine) and

volatile bituminous coals

TABELA 8

Analiza statystyczna istotności oddziaływania warunków suszenia na właściwości koksotwórcze węgli

gazowo-koksowych: Budryk i Krupiński

Testing of the significance of effect of drying conditions on coking properties of Budryk and Krupiński high

| Coal | Factor | Value | <i>RI</i> [-] | <i>SI</i> [-] | t _{III} —t _I [°C] | <i>b</i> [%] | p _{max} [kPa] |
|-----------|--------------------------|----------|---------------|---------------|---------------------------------------|-----------------|---------------------------|
| | £1 W! | 6% | -1 | 0 | 1 | 0 | 0 |
| | final W_t^r | 4% | -1 | 0 | 0 | 0 | 0 |
| | | nitrogen | -1 | 0 | 0 | 0 | 0 |
| Budryk | atmosphere | air | -1 | 0 | 1 | 0 | 0 |
| | temperature of drying | 150°C | -1 | 0 | 0 | 0 | 0 |
| | | 210°C | -1 | -1 | 0 | 0 | 0 |
| | final W_t^r | 6% | -1 | 0 | -1 | -1 | -1 |
| | | 4% | -1 | 0 | -1 | -1 | -1 |
| ., ., . | . 1 | nitrogen | 0 | 0 | -1 | -1 | -1 |
| Krupiński | atmosphere | air | 0 | 0 | -1 | -1 | -1 |
| | temperature | 150°C | 0 | 0 | -1 | -1 | -1 |
| | of drying | 210°C | -1 | 0 | -1 | 0 | -1 |

TABLE 9

Testing of the significance of effect of drying conditions on coking properties of Borynia, Zofiówka and Jas-Mos low volatile bituminous coals

TABELA 9 Analiza statystyczna istotności oddziaływania warunków suszenia na właściwości koksotwórcze węgli ortokoksowych: Borynia, Zofiówka i Jas-Mos

| Coal | Factor | Value | RI [-] | SI [-] | t _{III} —t _I [°C] | b [%] | p _{max} [kPa] |
|----------|--------------------------|----------|--------|--------|---------------------------------------|-------|------------------------|
| | C 1 HV | 6% | -1 | 0 | -1 | -1 | 0 |
| | final W_t^r | 4% | -1 | 0 | -1 | -1 | 0 |
| D : | . 1 | nitrogen | -1 | 0 | -1 | -1 | 0 |
| Borynia | atmosphere | air | -1 | 0 | -1 | -1 | 0 |
| | temperature | 150°C | -1 | 0 | -1 | -1 | 0 |
| | of drying | 210°C | -1 | 0 | -1 | -1 | 0 |
| | final W_t^r | 6% | 0 | 0 | -1 | 0 | 1 |
| | | 4% | 1 | 0 | 0 | -1 | 1 |
| 7 6 / 1 | atmosphere | nitrogen | 1 | 0 | 0 | -1 | 1 |
| Zofiówka | | air | 0 | 0 | -1 | -1 | 1 |
| | temperature of drying | 150°C | 0 | 0 | 0 | -1 | 1 |
| | | 210°C | 1 | -1 | -1 | -1 | 1 |
| | final W_t^r | 6% | 0 | -1 | 0 | -1 | 0 |
| | imai w _t | 4% | 0 | -1 | 0 | -1 | 0 |
| | | nitrogen | 0 | -1 | 0 | -1 | 0 |
| Jas-Mos | atmosphere | air | _ | -1 | 0 | -1 | 0 |
| | temperature | 150°C | 0 | -1 | 0 | -1 | 0 |
| | of drying | 210°C | 0 | -1 | 0 | -1 | 0 |

in the temperature range of coal plasticity (except for coals from Budryk and Jas-Mos coal mines). The effect of drying, under the conditions described above, on the values of RI and p_{max} was comparable with analytical error. Also, for the applied intervals of variability, no significant impact was recorded of any of the examined factors (i.e. final moisture content in dried coal, type of drying agent and temperature of drying) on the change in coking properties of the dried coals, both low and high volatile bituminous coals.

Conclusions

In conclusion, it can be stated that the examinations of drying of selected bituminous coals, representative of Polish coal resources, up to the moisture content possible to achieve

on industrial scale, showed that this process did not exert a significant effect on the deterioration of coking properties of coals. Consequently, there is no danger of the drying process having a detrimental effect on coke quality as far as deterioration of coking properties of coal charge is concerned. There occur noticeable differences in this respect between the processes of deep drying by the Precarbon Coaltek Method or others, and partial drying of coal blend (CMC process). In the former, intensive thermal preparation and the presence of oxidizing component lead to a significant or, sometimes, even drastic deterioration of coking properties of the dried coal. In the case of drying, the process of water evaporation from inside the coal grains is not completed, and the issuing water vapour forms, most probably, a kind of a protective layer preventing, or, at least, restricting, the access of oxygen and, consequently, the course of the oxidation reaction and its effects, i.e. the deterioration of coking properties. In view of the above-mentioned points, thermal pre-treatment through partial drying of coal blends has only beneficial effects (the growth of the bulk density of coal charge and the decrease of temperature gradient within the carbonised coal charge) creating conditions for improving coal quality and increasing efficiency of the coking process.

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ODDZIAŁYWANIE PROCESU PODSUSZANIA WĘGLI KOKSOWYCH NA ICH WŁAŚCIWOŚCI KOKSOTWÓRCZE

Słowa kluczowe

Węgle koksowe, właściwości koksotwórcze, podsuszanie

Streszczenie

W artykule przedstawiono wyniki badań wpływu podsuszania węgli koksowych do zawartości wilgoci 4 i 6%, na ich właściwości koksotwórcze. Węgle podsuszano w atmosferze azotu oraz powietrza przy temperaturach 150 i 210°C. Nie stwierdzono istotnego pogorszenia właściwości koksotwórczych podsuszanych węgli.