(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
20 May 2010 (20.05.2010)

PCT

(10) International Publication Number
WO 2010/054607 A1

(51) International Patent Classification:
C10L 9/08 (2006.01) *C10B 57/04* (2006.01)

(21) International Application Number:
PCT/CZ2009/000134

(22) International Filing Date:
10 November 2009 (10.11.2009)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
PV2008-728 14 November 2008 (14.11.2008) CZ

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(81) Designated States *(unless otherwise indicated, for every kind of national protection available)*: AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IS, JP, KE, KG, KM, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PE, PG, PH, PL, PT, RO, RS, RU, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States *(unless otherwise indicated, for every kind of regional protection available)*: ARIPO (BW, GH, GM, KE, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Declarations under Rule 4.17:

— *of inventorship (Rule 4.17(iv))*

Published:

— *with international search report (Art. 21(3))*

— *before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments (Rule 48.2(h))*

WO 2010/054607 A1

(54) **Title**: METHOD FOR BROWN COAL COKE PRODUCTION USING SINGLE-STAGE THERMAL REPROCESSING

(57) **Abstract**: The method for brown coal coke production is based on the fact that brown coal with carbon content in dry matter of 60 to 75% by weight, volatile matter 40 to 60% by weight and tar 17.5 to 30% by weight is by crushing and grinding modified to the grain-size of under 0.25 mm, moistened to the total water content ranging between 10 and 25% by weight; the homogenised mixture is placed in the retort, and after its compacting it is indirectly heated to the internal temperature of the retort between 950 to 1300°C and kept at this temperature for 2 hours or more. The product is subsequently removed from the retort after cooling to the surface temperature of 400°C.

Method for brown coal coke production using single-stage thermal reprocessing

Field of Technology

This invention is based on the method for the production of brown coal coke using the thermal reprocessing of brown coal by high-temperature carbonisation, in the absence of air in the single technological stage.

Current State of the Technology

Brown coal has been considered, and in the literature was previously referred to, as a non-caking material. By low-temperature carbonisation (low-temperature coking) of brown coal using final temperatures ranging between 580 to 600°C, low-temperature coke is produced, which is characterised by its high reactivity and its tendency to spontaneous combustion (i.e. freshly produced material spontaneously oxidising in air). The produced combustible matter is fine-grained, soft and friable and has a high ash content. With increasing of grain size the ash content also increases from 15 to 35%; this is accompanied by a decline in its total heating value.

The production of coke from brown coal by high-temperature carbonisation with final coking temperatures ranging between 900 and 1050°C, has not been carried out directly due to its non-coking characteristics, however this was technologically circumvented in the manner that brown coal was, added as the strengthening agent in the quantity of 3 to 10% (non-coking proportion with 80% of its granularity modified to under 0.5 mm) to caking hard coal during the preparation of the coal charge. The coal charge is a mixture of different kinds of better or weakly-caking types of hard coal with a 90 – 93% granularity modified to under 3 mm, which constitutes the charge for the coke-oven battery.

The published JP 51135902 patent application describes an indirect method for the production of coke, in which the de-ashed and desulphurised liquid product, obtained by the application of an appropriate solvent to the brown coal, is submitted to the effects of increased temperature and pressure in a hydrogen atmosphere; the solid part is separated and mixed in the appropriate proportion with raw coal material. The mixture created in this way constitutes the charge for the coke-oven battery.

The second method for the preparation of high-temperature coke from brown coal is also indirect and it is carried out technologically in two steps. First a low-temperature coke is made from the brown coal, which is subsequently briquetted, using (or not) bonding agent. The pressed briquettes are

carbonised at high temperature between 900 to 1200°C. This technological process permits the brown coal coke production in two technological steps.

The Patent No. 236540 describes the method for improving of caking of weakly-coking coal used for the production of metallurgical coke, which is based on the fact that aluminium oxide, in the form of a powder with grain-size under 1 mm, in quantity from 0.05 to 0.5% by weight, is added as a catalyst to the raw coal prior to its coking. This method, however, is primarily focused on weakly-caking hard coal.

Other technologies for the production of brown coal coke have also been two-stage processes. The dried brown coal was finely ground and under high pressure compressed into briquettes, which were subsequently dried. These dried briquettes were carbonised at a high-temperature in a controlled temperature regime with a final temperature of 1100°C. The increase of temperature up to 650 – 700° C was slow. The gradient of the temperature increase in the interval 700 – 1100°C, was conversely rapid. This technology was in the past used in Lauchhammer, Germany.

Principle of the Invention

The above-mentioned problems concerning the production of brown coal coke have, to some extent, been eliminated by means of this invention, which is based on the fact that, according to the tests carried out, the values of the qualitative characteristics of the brown coal charged have been determined. When the brown coal matter has the carbon content in the dry matter (C^d) 60 to 75% by weight, the content of the volatile matter in the dry matter (V^d) 40 to 60% by weight, and the tar content in the dry matter (T^d_{sK}) 17.5 to 30% by weight, the matter shows caking property by high temperature carbonisation (these values have been determined by using samples of brown coal in different states, so they may mutually overlap, i.e. the carbon content in dry matter may include part of the carbon in the form of tar and part of the tar may also constitute a part of the volatile components; this results in the sum of these values not being 100%).

Brown coal of a suitable quality is brought, by crushing and grinding, to a grain-size smaller than 0.25 mm. The resulting mixture is moistened, by the addition of process water, to a final content of total water ranging between 10 to 25% by weight, and homogenised by simultaneous stirring. The mixture of brown coal, with its modified grain size, moistened and homogenised, is placed into the retort, in which it is gradually compacted. After loading and compacting the moistened charge into the retort, the retort is closed and placed into a pyrolysis furnace where the content is indirectly heated to a final internal temperature of the retort ranging between 950 to 1300°C. This temperature is then kept for 2 hours or more, depending on the geometry and other parameters of the coking retort. Through the simultaneous interaction of factors, increasing temperature and coking time, the

chemical-physical conversion of the brown coal matter and also its coking take place in the absence of air. The product of this conversion is high-temperature brown coal coke, produced in a single technological step.

Examples of Implementation of the Invention

The qualitative parameters of charging brown coal are shown in Table No. 1. The charge of brown coal with grain-size adjusted to below 0.25 mm, moistened to 12.6% of total water was charged and compacted into a cylindrical retort, which was subsequently placed into a pyrolysis furnace and heated with the temperature gradient of 9.95°C . min^{-1} until it reached an internal final temperature of the charge of 1050°C. The final temperature of the charge was kept for the following two hours. After completion of carbonisation the coke was moved from the retort having the surface temperature of 400°C (the internal temperature of the coke core ranged between 500 and 700°C). Unlike a low-temperature brown coal coke, the brown coal coke produced in this manner, at this surface temperature, is not subject to spontaneous combustion. The qualitative parameters of the brown-coal coke are defined in Table No. 2

The produced brown coal coke is grey-black, matte, lumpy and non-brittle. It has a small surface area, which amounts to 1.04 m^2. g^{-1} and 15.2% of the developed adsorption pores ranging in interval size between 44 and 65 nm.

Table No. 1 - Qualitative parameters of charging brown coal

Parameter		Value	Parameter		Value
analytical water	[% by weight]	3.43	nitrogen in dry matter	[% by weight]	0.95
ash in dry matter	[% by weight]	3.58	nitrogen in combustible matter	[% by weight]	0.99
total sulphur	[% by weight]	1.03	oxygen in dry matter	[% by weight]	15.77
sulphur in combustible matter	[% by weight]	1.07	oxygen in combustible matter	[% by weight]	16.35
carbon in dry matter	[% by weight]	72.92	volatile combustible matter in dry matter	[% by weight]	54.14
carbon in combustible matter	[% by weight]	75.63	volatile combustible matter in combustible matter	[% by weight]	56.15
hydrogen in dry matter	[% by weight]	5.75	calorific value in dry matter	[MJ . kg]	31.85
hydrogen in combustible matter	[% by weight]	5.96	calorific value in combustible matter	[MJ . kg]	33.04

Parameter		Value
tar in dry matter	[% by weight]	24.61
low-temperature coke in dry matter	[% by weight]	57.38
pyrogenetic water in dry matter	[% by weight]	7.01
gas in dry matter	[% by weight]	11.00
tar in combustible matter	[% by weight]	25.54
low-temperature coke in combustible matter	[% by weight]	55.83
pyrogenetic water in combustible matter	[% by weight]	7.27
gas in combustible matter	[% by weight]	11.36

Table No. 2: Qualitative parameters of brown coal coke, obtained in accordance with the invention

Parameter		Value	Parameter		Value
analytical water	[% by weight]	0.34	nitrogen in dry matter	[% by weight]	0.43
ash in dry matter	[% by weight]	8.01	nitrogen in combustible matter	[% by weight]	0.47
total sulphur	[% by weight]	0.58	oxygen in dry matter	[% by weight]	1.8
sulphur in combustible matter	[% by weight]	0.63	oxygen in combustible matter	[% by weight]	1.96
carbon in dry matter	[% by weight]	89.08	volatile combustible matter in dry matter	[% by weight]	1.87
carbon in combustible matter	[% by weight]	96.84	volatile combustible matter in combustible matter	[% by weight]	2.03
hydrogen in dry matter	[% by weight]	0.1	calorific value in dry matter	[MJ . kg]	30.71
hydrogen in combustible matter	[% by weight]	0.11	calorific value in combustible matter	[MJ . kg]	30.69

PATENT CLAIM

1. The method for production of coke from brown coal raw material with the content of carbon in dry matter (C^d) ranging between 60 and 75 % by weight, of volatile combustible matter in dry matter (V^d) 40 to 60% by weight and of tar in dry matter (T^d_{sK}) 17.5 to 30% by weight, which is modified by crushing and grinding to the grain size below 0.25 mm, moistened to the final content of total water in the range between 10 and 25% by weight, characterised by the feature that the homogenised mixture obtained in such manner is placed into the retort, and after its compacting indirectly heated to the final internal retort temperature ranging between 950 and 1300°C; this temperature is kept for 2 hours or more and the product is subsequently removed from the retort after cooling to the surface temperature of 400°C.

INTERNATIONAL SEARCH REPORT

International application No
PCT/CZ2009/000134

A. CLASSIFICATION OF SUBJECT MATTER
INV. C10L9/08 C10B57/04
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C10L C10B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 4 419 186 A (WIENERT FRITZ O [US]) 6 December 1983 (1983-12-06) abstract; claims 1,2,4	1
A	JP 04 091191 A (KEIHAN KK) 24 March 1992 (1992-03-24) abstract	1
A	DE 37 42 817 A1 (RHEINISCHE BRAUNKOHLENW AG [DE]) 6 July 1989 (1989-07-06) column 1, line 67 - column 2, line 5 claim 1	1
A	US 4 450 777 A (WOLFRUM ERHARD [DE] ET AL) 29 May 1984 (1984-05-29) abstract; claims	1

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents :

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"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
16 April 2010	29/04/2010

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/CZ2009/000134

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