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METHOD OF PRODUCING METALLURGICAL COKE

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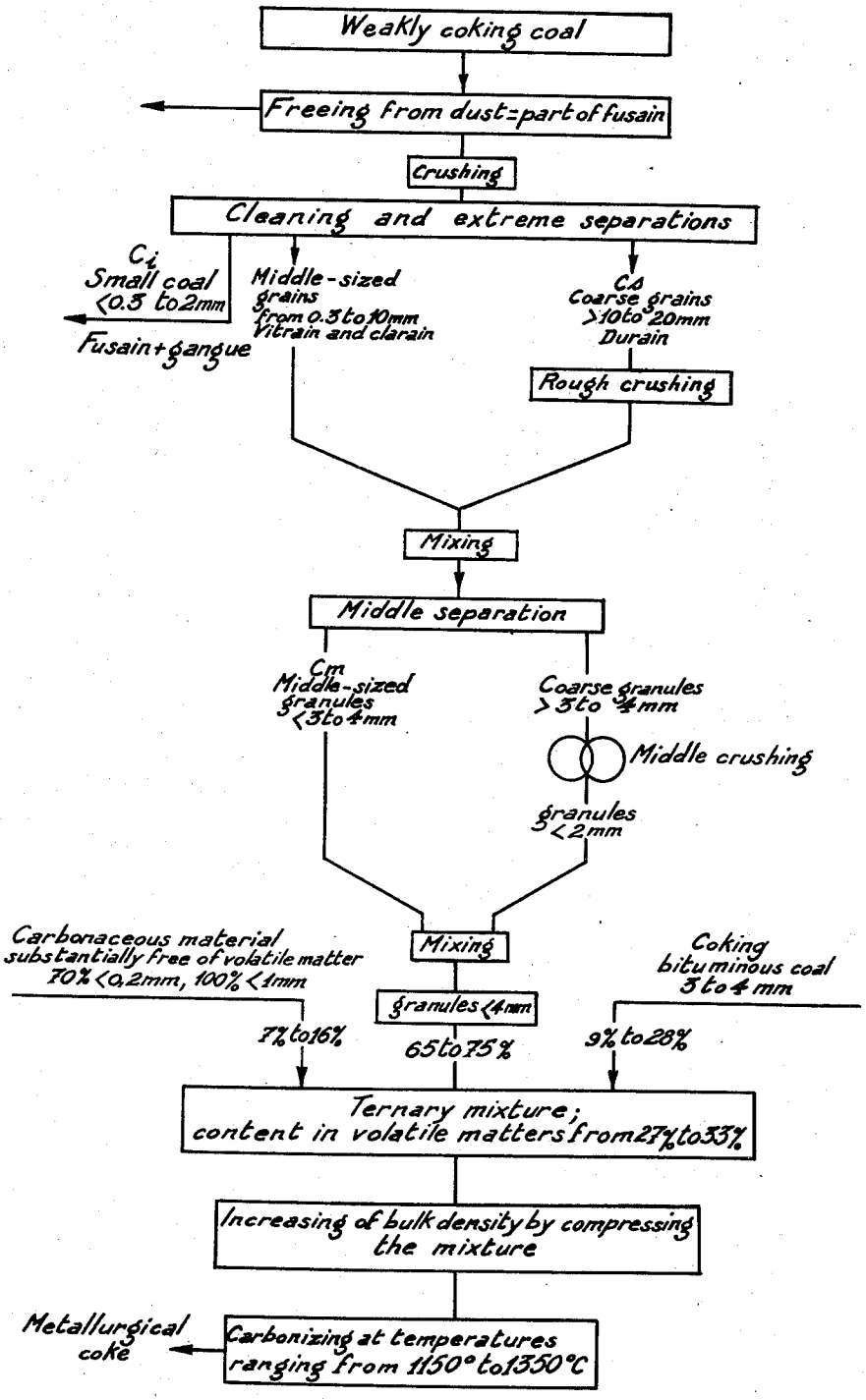

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# METHOD OF PRODUCING METALLURGICAL COKE

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10 Claims. (Cl. 202—33)

This application is a continuation-in-part of application Serial No. 222,530, filed April 23, 1951 (now abandoned), entitled: Method of Producing Metallurgical Coke.

Weakly coking coals of low bitumen content from the Lorraine Basin in France called "gras A," "gras B" and "flambants gras" pertaining to the high volatile bituminous coal category and high-volatile bituminous coals of inferior categories, and similar coals have hitherto been considered as unsuitable for producing metallurgical coke in spite of their fusibility. As a matter of fact, cokes obtained by carbonizing these coals without a special preliminary treatment do not offer a sufficient resistance to crushing.

For the sake of clearness in the present specification and as the groups of coals concerned are classified in subgroups it is deemed necessary to indicate the conditions in which the indices defining these sub-groups are found.

The amount of volatile materials is determined in accordance with the French Standards AFNOR No. 03–004. This is based on a test carried out in a crucible of enamelled porcelain either of the so-called Bayeux variety or of dull platinum. This crucible has a top diameter substantially equal to the height and a bottom diameter substantially equal to one-half of the height, the latter and the top diameter ranging from 30 to 45 mm.

For heating the crucible an oven adapted to maintain therein a constant temperature of 960° C.±10° C. is used. The crucible should be heated with a progressively increasing temperature having a substantially linear characteristic of about 100° C. per minute, the final temperature measured inside the crucible and at the contact of its lower face being fixed at 960° C. and reached after a heating period ranging from 9 to 11 minutes. This final temperature is maintained for 5 minutes. A sufficiently reducing atmosphere should be kept around the crucible in order to prevent the sample from burning during the test.

The test is carried out by heating the crucible to full red and allowing it to slowly cool down with its cover in a dryer. After measuring the weights of the crucible and cover assembly a test sample weighing 1 gram is taken from the mass of the coal to be tested and this sample is spread to a uniform layer on the bottom surface of the crucible. This sample is then wetted with 30 drops of a liquid containing 50% of water and 50% of pure alcohol, these drops being uniformly distributed over the entire surface. Then the crucible fitted with its top is introduced into the heating device and the heating step is carried out in a reducing atmosphere as set forth above. The crucible is finally taken from the oven and allowed to cool in the dryer, and after removing any trace of dust which might possibly adhere to the external walls the crucible is weighed within 60 minutes at the latest.

The swelling index is found according to the French standard method AFNOR No. 11–001 which is similar to the British method B.S.S. 1016–1942 and also to the American method A.S.T.M. D720–46.

Resistance to abrasion and to splitting is conveniently



determined by means of the Micum drum test (Mission Inter-Alliée de Contrôle des Usines et des Mines) adopted in France and Belgium since 1924 for the testing of Rhur cokes. It is performed by means of a hollow drum with a smooth inner surface constructed of sheet steel of a thickness of 5 mm., and closed at both ends and mounted with its axis horizontal. It is 1 metre in diameter and 1 metre long and has secured to the inside surface at regular intervals four angle irons extending parallel to the axis and projecting radially 100 mm. from the surface. Introduction into the drum of the coke to be tested is effected by way of a curved hinged door 500 x 600 mm. provided with securing means. By means of an electric motor operating through reduction gearing the cylinder is rotated at a speed of 25 revolutions a minute about its axis in the direction of the unencumbered sides of the angle irons.

The operator is provided with three sheet metal sieves 4 or 5 mm. thick and 700 x 500 mm. in size, perforated respectively with circular holes of 50 mm., 40 mm. and 10 mm. Each sieve is arranged in a wooden hopper 20 cm. high. There is also a wooden box for receiving the crushed coke of the same size as the hopper for the sieves.

The method of operation is as follows: The sample of 50 kgs. of coke of a size greater than 50 mm. is introduced by hand into the drum and the door is then secured. The drum is then subjected for 4 minutes to rotation at a speed of 25 revolutions a minute i.e. 100 revolutions per test. After the apparatus has come to rest the door is opened and the complete contents of the drum including dust are extracted. These contents are passed two or three times through the sieve with 40 mm. holes then through the sieve with 10 mm. holes. The fractions of coke of a size greater than 40 mm. and of a size less than 10 mm. are weighed separately. Four tests are made successively on four distinct samples each of 50 kgs.

In order to express the results, calculation is made for each test of the percentages of coke greater than 40 mm. and less than 10 mm. The arithmetic mean of the results obtained with the four samples represents the final result on which the quality of the coke examined can be judged.

The percentage of coke from 0–10 mm. in size called the Micum index M10 or tumbler non-stability on 10 mm. screen, characterises the resistance to abrasion and indicates the proportion of the coke which is capable to act as a disturbing element for the working of the blast-furnaces, more particularly by its binding with their lining. The percentage of coke of a size greater than 40 mm. called the Micum index M40 or tumbler stability on 40 mm. screen characterises the resistance to crushing and splitting.

A good metallurgical coke should have an index M40 substantially equal to, or higher than, 80 and an index M10 at the most equal to 8.

As already stated the groups of coals concerned are divided into sub-groups according to their characteristics:

(i) Coals of the type "gras A" in Lorraine and approximately of the non coking high volatile A bituminous coal category, having a percentage of volatile matter ranging from 27% to 36% (calculated on pure and dry coal), a swelling index ranging from 6 to 9, an oxygen content comprised between 6% and 8% and Micum indexes M40 between 35 and 40 and M10 between 6 and 8.

(ii) Coals of the type called "gras B" in Lorraine and approximately of the non coking high volatile B bituminous coal category, having from 35% to 42% of volatile matter, a swelling index ranging from 4 to 7, a percentage

of oxygen ranging from 8% to 10% and Micum indexes M40 between 25 and 35 and M10 between 6 and 8.

(iii) Coals of the type called "flambants gras" in Lorraine and approximately of the non coking high volatile B bituminous coal of lower grades and high volatile C bituminous coal categories, both of which have from 36% to 45% of volatile matter, a swelling index of 2 to 5, from 9.5% to 12.5% of oxygen and Micum indexes M40 between 20 and 30, and M10 between 6 and 8.

Of course, the above figures are given solely as rough limits and this invention is chiefly concerned with coals of sub-groups called "gras B" and particularly with coals of sub-group called "flambants gras" as hereinabove defined.

The high volatile matter and oxygen contents of said coal sub-categories show that they relate to coal pertaining to supplies of recent formation which as well known necessitate a quick heating law, i.e. a high temperature, in order to attain a certain degree of fusion.

Further the high volatile matter content and the small Micum indexes M40 of said coal sub-categories indicate that said coals when carbonized are subjected to a notable shrinkage causing important cracks. The practice shows moreover that their bitumen content is very low and that said bitumen is not thermostable. Therefore when said coals are carbonized for manufacturing coke the total loss in fluidity of their bitumen is attained before their volatile matters have wholly escaped so that when said bitumen solidifies the mass of coke cracks under the action of the driving off of the gases which have not escaped from said mass when said bitumen was fluent.

The main object of the present invention is to provide a process for remedying the lack of thermostable bitumen of said coals in order to manufacture from said coals a metallurgical coke having Micum indexes M40 substantially equal to, or higher than, 80 and M10 lower than 8, i.e. a coke complying with all the requirements of the metallurgy of iron.

For this purpose, the small coal of said sub-categories of coal is submitted to a dedusting operation for freeing it from free fusain by a mechanical preparatory treatment which may consist in pneumatically or hydraulically removing mud and dust from the coal. Afterwards by a crushing operation followed by a size-grading screening said dedusted small-coal is freed from almost the whole fusain remaining therein. Then a ternary mixture is prepared, said mixture having a percentage of volatile matter ranging from 27% to 33%, preferably from 29% to 30%, and comprising as main component from 65% to 75% by weight of said small-coal practically freed from fusain, and as secondary components from 9% to 28% by weight of additional coking bituminous coal having a volatile matter content of 18% to 25%, an oxygen content less than 5% and Micum indexes M40 higher than 80 and M10 lower than 8, and from 7% to 16% by weight of small grains of a carbonaceous material having practically no volatile matter. Said mixture is subjected to a compression step in order to attain a bulk density ranging from 1,000 kg./m.$^3$ to 1,100 kg./m.$^3$; then it is carbonized at a temperature comprised between 1,150° C. and 1,350° C. according to the characteristics of its components.

The use of the aforesaid coking bituminous coal as an additional coal has for its object, on the one hand, to supply to the mixture the thermostable bitumen lacking in the basic coal and, on the other hand, to decrease the volatile matter content of said mixture with respect to that of said basic coal.

The utilization of a carbonaceous material having practically no volatile matter as a secondary component for the mixture decreases the volatile matter content of said mixture with respect to that of the basic coal and adds to said basic coal an indifferent material having, as a consequence of its process of manufacture, a state of surface which necessitates the minimum quantity of bitumen to be coated thereby.



The percentages of the three components of said ternary mixture are so chosen in accordance with the actual characteristics of the basic and additional coals that the volatile matter content indicated for said mixture is obtained while taking into account the thermostable bitumen content which warrants good melting conditions. If the volatile matter content is increased, during the carbonization a great shrinkage takes place which, while less than that occurring during the carbonization of the basic coal alone, is sufficiently important for causing notable cracks for the thus manufactured coke. If on the contrary said content is decreased the mixture behaves as a lean coal, such as anthracite for example, does not melt and gives a pulverulent coke. In both cases the coke which is thus manufactured is useless for metallurgical purposes.

The compression step to which the ternary mixture is subjected has for its object to bring the grains of the basic coal nearer to each other up to an interpenetration thereof so that the leaking of the bitumen in fluid state between said grains during the carbonization step already ensures their mutual binding. It is to be noted that the conventional bulk density for a coke oven charge ranges from 700 to 750 kg./m.$^3$.

The carbonization temperatures are determined by the nature of the basic coal, which, as hereinabove indicated, is extracted from coal supplies of recent formation and therefore necessitates a quick heating law in order to attain a certain degree of fusion. If the carbonization temperature decreases the manufactured coke is a badly melted coke having a Micum index M10 substantially higher than 8, i.e. a coke which is friable and of inferior quality for metallurgical purposes.

The following description relating to a specific manner in which the method of this invention may be carried out is given solely by way of example and should not be construed as limiting the scope of the invention as set forth in the claims attached thereto.

The accompanying drawing showing a flow sheet discloses the steps of said method.

After the weakly coking coal defined hereinabove has been treated in some known dry or hydraulic way in order to remove the gangue and the free fusain therefrom, it is freed from any particles below a certain size $C_l$ to be determined according to the fusain content of the coals used, this size being however as small as possible in order to eliminate only detrimental excess coal. With the conventional washing equipments and when small coals are used, a size of about 1 mm. is generally suitable. Said particles or small coal as well known consist in fusain.

Then any coal grains above a certain size $C_s$ selected between the limit sizes of 10 mm. and 20 mm. are separated from the coal mass. These coarse grains consisting in durain are roughly crushed to a size below $C_s$ and added again to the portion of middle-sized grains comprised between sizes $C_l$ and $C_s$ which consists in vitrain. The aggregate product comprised between these last sizes is then divided into two fractions through an intermediate cut $C_m$ ranging from 3 to 4 mm.

No further sizing is required for the fraction of middle-sized granules comprised between sizes $C_l$ and $C_m$, but the other fraction of coarse granules is crushed to a size smaller than 2 mm. and then added to the first fraction.

As a result of these various steps a product is obtained wherein all granules have a size below $C_m$ without causing the necessary crushing operations to reduce the smaller granules to powder.

The thus obtained product is characterised in that it contains practically no fusain. Said last component of the basic coal is eliminated with a view to obtain a mixture which is free from a constituent having a substantial volatile matter content and necessitatng a substantial

amount of bitumen for its complete incorporation in said mixture, said constituent being a non-fusible indifferent material having a structure with a very great and irregular surface for a given volume, such as the structure of charcoal.

This product is the first component of the ternary mixture to be carbonized.

The second component is coking bituminous coal, for example coal from the Pas-de-Calais, Ruhr or even Saar-Lorrainese Basins, of a size smaller than 4 mm. and preferably 3 mm. This coal contains 18% to 25% of volatile matter, preferably 19% to 21%, an oxygen content lower than 5% and Micum indexes M40 higher than 80 and M10 lower than 8.

The third component of the mixture is granular coke breeze or anthracitic coal culm of a size not above 1 mm. and comprising at least 70% of granules smaller than 0.2 mm. The more this third component is crushed, the better the results obtained, more particularly if granules have a surface as smooth as possible.

The ratios in which the above-stated components are admixed may vary to a certain extent provided the amount of volatile matter in the mixture is kept within the above limits.

By way of example, the following formulae may be resorted to, the various percentages being established in weight:

*Formula I*

| | Percent |
|---|---|
| Coal of the type called "gras B" in Lorraine and approximately of the non cokable high volatile B bituminous coal category | 75 |
| Coking bituminous coal (as from Drocourt in the Pas-de-Calais district) | 9 |
| Crushed coke breeze | 16 |

*Formula II*

| | |
|---|---|
| Coal of the type called "gras B" in Lorraine and approximately of the non cokable high volatile B bituminous coal category | 48 |
| Coal of the type called "flambants gras" in Lorraine and approximately of the non cokable high volatile B bituminous coal of inferior categories or high volatile C bituminous coal category | 24 |
| Small coal from coking bituminous coal (as from the Pas-de-Calais district) | 16 |
| Crushed coke breeze | 12 |

*Formula III*

| | |
|---|---|
| Coal of the type called "flambants gras" in Lorraine and approximately of the non cokable high volatile B bituminous coal of inferior categories or high volatile C bituminous coal | 65 |
| Small coal from coking bituminous coal (as from the Pas-de-Calais district) | 28 |
| Crushed coke breeze | 7 |

The mixture, after being homogenized by conventional means, is then introduced into a rammer charger in which it is transformed by compression into a cake weighing several tons which is then charged into the coke oven.

The heating temperature, ranging from 1,150° C. to 1,350° C., is rated according to the proportions of the ternary mixture as indicated in the above formulae. Thus, the temperature is from 1,150 to 1,250° C. for Formula I, 1,250 to 1,300° C. for Formula II, and 1,275 to 1,350° C. (preferably 1,300° C.) for Formula III. It is of particular importance that a suitable temperature be observed in the case of Formula III or any similar formulae.

It will be readily understood that alterations may be brought to the process described hereinabove by way of example, without departing from the spirit and scope of the invention. Thus, in particular, the oven may be charged directly without subjecting the ternary mixture to a preliminary compression operation; although not so satisfactory, an appreciable result may be obtained in this case, provided the percentage of additional bituminous coal is increased or, for instance, doubled.



What I claim is:

1. A method of producing metallurgical coke having Micum indexes M40 substantially equal to 80 and M10 lower than 8, from weakly coking coals, which coals have moreover a low bitumen content, a high percentage of volatile matter ranging from 27% to 45%, a swelling index according to the ASTM D720–46 method of from 2 to 9 and a high oxygen content ranging from 6 to 12.5%, such as Saar-Lorraine coals of the types consisting of "gras A," "gras B" and "flambants gras," which comprises freeing from dust a portion of said weakly coking coal, crushing said dust-freed portion, separating from the crushed coal small coal containing fusain and gangue, crushing the remaining part of the crushed coal into small grains, preparing a ternary mixture having a percentage of volatile matter ranging from 27% to 33% and comprising as main component from 65% to 75% by weight of the thus obtained small grains and as secondary components from 9% to 28% by weight of additional coking bituminous coal having a volatile matter content of 18% to 25%, an oxygen content less than 5% and Micum indexes M40 higher than 80 and M10 lower than 8, and from 7% to 16% by weight of small grains of a carbonaceous material substantially free from volatile matter, mechanically increasing the bulk density of said ternary mixture, and carbonizing said ternary mixture with increased bulk density at a temperature between about 1150° C. and 1350° C.

2. A method of producing metallurgical coke, having Micum indexes M40 substantially equal to 80 and M10 lower than 8, from weakly coking coals which coals have moreover a low bitumen content, a high percentage of volatile matter ranging from 27% to 45%, a swelling index according to the ASTM D720–46 method of from 2 to 9 and a high oxygen content ranging from 6 to 12.5%, such as Saar-Lorraine coals of the types "gras A," "gras B" and "flambants gras," which comprises freeing from dust a portion of said weakly coking coal, crushing said dust-freed portion, separating the crushed coal into small coal containing gangue and fusain, into middle-sized grains containing vitrain and clarain and into coarse grains containing durain, eliminating the small coal for practically removing all of the fusain of said coal, roughly crushing the coarse grains, mixing the middle-sized grains and the roughly crushed coarse grains, separating the middle-sized granules of the mixture thus obtained, crushing the remaining coarse granule fraction of said mixture, mixing the crushed coarse granule fraction with the middle-sized granules of said mixture, preparing a ternary mixture having a percentage of volatile matter ranging from 27% to 33% and comprising as main constituent from 65% to 75% by weight of the granules obtained after said second mixing operation and as secondary components from 9% to 28% by weight of additional coking bituminous coal having a volatile matter content of 18% to 25%, an oxygen content less than 5% and Micum indexes M40 higher than 80 and M10 lower than 8, and from 7% to 16% by weight of small grains of a carbonaceous material substantially free of volatile matter, mechanically increasing the bulk density of said ternary mixture, and carbonizing said ternary mixture with increased bulk density at a temperature comprised between about 1150° C. and 1350° C.

3. A method according to claim 2, wherein the dimensions of the small coal range from 0.3 to 2 mm., those of the coarse grains from 10 to 20 mm., while the upper dimensions of the middle-sized grains range from 3 to 4 mm.

4. A method according to claim 2, wherein the coarse granules obtained by the second separation are crushed to granules smaller than 2 mm.

5. A method according to claim 2, wherein the coking

bituminous coal is incorporated in the ternary mixture in granules having a size below 4 mm.

6. A method according to claim 2, wherein the carbonaceous material substantially free from volatile matter is incorporated in the ternary mixture in granules having a size not exceeding 1 mm and comprising at least 70% of granules smaller than 0.2 mm.

7. A method according to claim 2, wherein the carbonaceous material substantially free from volatile matter is formed of coke breeze.

8. A method according to claim 2, wherein the carbonaceous material substantially free from volatile matter is formed of anthracitic coal culm.

9. A method according to claim 2, wherein the ternary



mixture has a percentage of volatile matter ranging from 29% to 30%.

10. A method according to claim 2, wherein after the compression step the bulk density is comprised between 1,000 and 1,100 kg./m.$^3$.

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