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Patents Act 1952-1969

CONVENTION APPLICATION FOR A PATENT

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METALLGESELLSCHAFT AKTIENGESELLSCHAFT,

of Reuterweg 14, D-6000 Frankfurt/Main,

Federal Republic of Germany

hereby apply for the grant of a Patent for an invention entitled:

PROCESS OF MAKING BROWN COAL CONTAINING PELLETS FOR

GASIFICATION

which is described in the accompanying complete specification. This application is a

Convention application and is based on the application numbered (1)

P34 41 756.7

for a patent or similar protection made in (4) Federal Republic of Germany on 15th November 1984

address for service is Messrs. Edwards, Waters & Sons, Patent Attorneys,

50 Queen Street, Melbourne, Victoria, Australia

Dated this 13th day of November 1985.

METALLGESELLSCHAFT
AKTIENGESELLSCHAFT

by

Wayne McMaster
DECLARATION IN SUPPORT OF A CONVENTION APPLICATION FOR A PATENT OR PATENT OF ADDITION

In support of the Convention Application made by

METALLGESELLSCHAFT AKTIENGESELLSCHAFT
Reuterweg 14, D-6000 Frankfurt/Main

(hereinafter referred to as the applicant) for a Patent
for an invention entitled: PROCESS OF MAKING BROWN COAL CONTAINING PELLETS FOR GASIFICATION

We, WOLFGANG WAGNER and ANNELIESE GASTEYER, both of Reuterweg 14, D-6000 Frankfurt/Main, Federal Republic of Germany

do solemnly and sincerely declare as follows:

1. We are authorised by the applicant for the patent to make this declaration on its behalf.

2. The basic application as defined by Section 141 of the Act was made in Federal Republic of Germany on the 15th day of November 1983 by

METALLGESELLSCHAFT AKTIENGESELLSCHAFT

3. DIETER SAUTER, of Am Jungfernborn 25, 6369 Nidderau 5, and UDO ZENTNER, of Masurenweg 10, D-6100 Darmstadt, Federal Republic of Germany

are the actual inventors of the invention and the facts upon which the applicant is entitled to make the application are as follows:

The applicant is the assignee of the said actual inventors

4. The basic application referred to in paragraph 2 of this Declaration was the first application made in a Convention country in respect of the invention the subject of the application.

DECLARED at, Frankfurt/Main, Germany this 7th day of October 1985.

Metallgesellschaft
Aktiengesellschaft

To: THE COMMISSIONER OF PATENTS

Authorized: A. Wagner

[Signature]
1. A process of making brown coal containing pellets for gasification in a reactor under pressures in the range from 5 to 150 bars by means of oxygen, steam and/or carbon dioxide as gasifying agents in a process in which the pellets are fed in the reactor to the top of a fixed bed, which is gradually descending, the gasifying agents are introduced into said fixed bed from below, and the mineral constituents are withdrawn as solid ash or liquid slag from the bottom of the fixed bed, characterized in that a mixture for pelletization is produced, in which the content of fine-grained coal consists of at least 80 wt.% hard brown coal, which has a particle size not in excess of 1 mm, the mixture to be pelletized contains 10 to 15 wt.-% water and moisture and 4 to 10 wt.-% bentonite, the mixture is mixed with an addition of water as a pelleting liquid to form
moist pellets which contain 25 to 35 wt. % water and moisture, and the moist pellets are dried in a water vapor containing atmosphere and in such a manner that the core of the pellets is heated to at least 90°C before the pellets have lost one-tenth or more of their water content and that the dried pellets contain 1 to 4 wt.% water and moisture.
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pellets have
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moisture.

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PATENTS ACT 1922

COMPLETE SPECIFICATION
(ORIGINAL)

Class
Int. Class

Application Number: 4 0 5 0 9 8
Lodged:

Complete Specification Lodged:
Accepted:
Published:

Priority:

Related Art:

Name of Applicant: METALLGESELLSCHAFT AKTIENGESELLSCHAFT

Address of Applicant: Reuterweg 14, D-60900 Frankfurt/Main 50,
Federal Republic of Germany

Actual Inventor: DIETER SAUER and UDO ZENTNER

Address for Service: EDWD. WATERS & SONS,
50 QUEEN STREET, MELBOURNE, AUSTRALIA, 3000.

Complete Specification for the invention entitled:

PROCESS OF MAKING BROWN COAL CONTAINING PELLETS FOR GASIFICATION

The following statement is a full description of this invention, including the best method of performing it known to

1.
FEDERAL REPUBLIC OF GERMANY

Certificate

Metallgesellschaft Aktiengesellschaft in 6000 Frankfurt has filed a patent application entitled "Process of Making Brown Coal Containing Pellets for Gasification" with the German Patent Office November 15, 1983.

The annexed papers are a correct and exact reproduction of the original disclosure papers of that patent application.

The annexed Abstract, which is to accompany the application but is no constituent part thereof, agrees with the original filed November 15, 1983.

The application has provisionally been designated in the German Patent Office with the Symbols C 10 B 5/10 and C 10 B 57/03 of the International Patent Classification.

Munich, dated October 1, 1983

The President of the German Patent Office

by order

signature

Klinger

Seal: German Patent Office

File reference: P 34 41 766.7
Process of Making Brown Coal Containing Pellets for Gasification

This invention relates to a process of making brown coal containing pellets for gasification in a reactor under pressures in the range from 5 to 150 bars by means of oxygen, steam and/or carbon dioxide as gasifying agents in a process in which the pellets are fed in the reactor to the top of a fixed bed, which is gradually descending; the gasifying agents are introduced into said fixed bed from below, and the mineral constituents are withdrawn as solid ash or liquid slag from the bottom of the fixed bed.

Coal-containing pellets for gasification in a fixed bed are known from Published German Applications 28 51 370 and 28 53 369 and from European Patent 10 712. The gasification of granular coal in a fixed bed is explained, e.g., in Ullmanns Enzyklopädie der technischen Chemie, 4th edition (1977), Vol. 14, on pages 383 to 386.
Details of the gasification process in which the ash remains solid are apparent from U.S. Patents 3,540,867 and 3,854,895 and Published German Application 2,201,278. The modified process in which liquid slag is withdrawn has been explained in British Patent Specifications 1,507,905; 1,508,671 and 1,512,677. In said known processes a granular fuel having particle sizes in the range from about 3 mm to about 60 mm is supplied to the gasification reactor.

It is an object of the invention to provide a simple and economical process of making pellets for gasification. The pellets are required to have adequate dimensional and structural stabilities so that they withstand mechanical handling, such as rehandling, transportation and pouring, and the thermal processes involved in gasification. This is accomplished in accordance with the invention in that a mixture for pelletization is produced, in which the content of fine-grained fuel consists of at least 90 wt.-% hard brown coal, which has a particle size not in excess of 1 mm, the mixture to be pelletized contains 10 to 25 wt.-% water and moisture and 4 to 10 wt.-% bentonite, the mixture is shaped with an addition of water as a pelletizing liquid to form moist pellets which contain 25 to 35 wt.-% water and moisture, and the moist pellets are dried in a vapor containing atmosphere and in such a manner that the core of the pellets is heated to at
least 90°C before the pellets have lost one-tenth or more of their water content and that the dried pellets contain 1 to 4 wt.% water and moisture.

Owing to their water and moisture content of 25 to 35 wt.%, the initially moist pellets are plastically deformable and do not tend to disintegrate as they are rehandled and transported. But they do not sufficiently resist pyrolysis as they are heated so that they would disintegrate in the upper portion of the fixed bed in the gasification reactor. Under the conditions which are encountered there at gas temperatures of about 350 to 600 °C, the structure of the pellets is loosened to such a degree that a major part thereof is disintegrated to form fine grains and dust. If the moist pellets obtained by the pelletization are dried with purging gas outside the reactor, e.g., on a drying belt, the pellets consisting mainly of brown coal will suffer such a strength loss that they will disintegrate as they are transported.

In numerous experiments it has surprisingly been found that bentonite-bonded brown coal pellets can be transformed to structurally strong dry pellets if the pellets are dried in an atmosphere which has a high water vapor content and care is taken that even the core of the pellets being dried is heated to at least 90°C before the drying process proper begins. As a result, temperatures which are
as close as possible to the boiling temperature of water are obtained also in the interior of the pellets before a substantial evaporation of water takes place. In this manner large differences in moisture content and temperature between the surface and the core of each pellet are avoided so that the dried pellet has a high structural strength.

The pellets are preferably dried in such a manner that the atmosphere directly contacting the pellets is almost saturated with water vapor. This can be achieved in that the moist pellets are dried while they are covered in a bed consisting of hot granular solids, which are at temperatures of about 150 to 300 °C. Such a bed of hot solids may consist, e.g., of a sand bed. Alternatively, drying may be effected by means of superheated steam at temperatures of about 110 to 180 °C. The dried pellets contain water and moisture not in excess of 4 wt. %.

The hard brown coal in the mixture to be pelletized suitably has a particle size not in excess of 0.5 mm and contains 60 to 80 wt.% of a fine fraction not in excess of 0.063 mm. The hard brown coals to be used preferably have the Class Numbers 10 to 12 of the International Classification for Brown Coals. Such brown coals have in the as-mined state a total moisture content (measured without ash) not in excess of 40 wt.%.
fuel content of the pellets preferably consists only of hard brown coal without any other carbonaceous component.

The binder for the pellets may consist of any of the conventional bentonites, such as natural sodium bentonites, activated calcium bentonites, or of raw clays. The bentonite may be fed together with the brown coal to a grinding plant or may subsequently be admixed to the fine-grained brown coal in a mixer. In a preferred practice bentonite and coal are jointly subjected to a grinding process because this will result in a fine dispersion of the binder in the brown coal. In a succeeding mixer, a first moistening step is suitably performed to provide a mixture which contains 10 to 25 wt.%, preferably 12 to 20%, water and moisture. Such a mixture can be processed further without raising dust and the bentonite can take up water and swell.

The pellets are formed in the usual manner on a pelletizing disc or, e.g., in a rotary kiln, with an addition of water as a pelletizing liquid. The moist pellets thus obtained have a diameter of about 6 to 30 mm, preferably 8 to 20 mm.

If the pellets are to be gasified in a process in which liquid slag is withdrawn, it will be recommendable to add a fine-grained flux, such as lime, to the mixture to be pelletized so that the melting temperature
of the ash will be reduced. In that case the mixture to be pelletized suitably contains 3 to 15 wt.% flux. The fixed bed in the gasification reactor may consist only of the pellets, with or without flux, but the pellets may alternatively be fed to the top of the fixed bed together with granular coal having particle sizes in the range from 3 to 60 mm.

Numerous tests in which the dry pellets in accordance with the invention were processed in the gasification reactor have shown that the pellets exhibit a satisfactory behavior as they are dried, coked and gasified. During the gasification of the pellets, their carbon content decreases progressively from the outside to the inside. The progress of that decrease has been closely studied and the pellets have been found to have a high structural strength in each stage of the combustion process. This is by no means usual with pellets consisting mainly of brown coal.

In the examples which will be described hereinafter a rehandling test will be mentioned to which the pellets are subjected in order to test their stability in transit and when they are dropped several times from different elevations. That test, which simulates the manner in which the pellets are handled in practice, will be explained with reference to the drawing. The pellets are first conveyed on a first conveyor belt 1 to the lower
mixture to form a flux. The pellets may consist only of dry pellets mixed together. The range from the inside.

The dry pellets used in the gasification exhibit a satisfactory gasified.

the carbon content to the inside.

Example 1 A hard brown coal from the United States, which contains 2 wt.% ash (on a dry basis), is dried to a moisture content of about 8 to 10 by weight and is ground in a ball mill to a particle size not in excess of 0.315 mm. The brown coal already contains about 17 wt.% montmorillonite, which is that component of bentonite which is active as a binder. For this reason it is not necessary to add a binder. When the ground coal has been moistened to a water content of about 18 wt.%, pellets 8 to 16 mm in diameter are formed on a pelleting disc as the
-8-
mixture is sprayed with water. The pellets contain 28 wt.% water and moisture and have a breaking strength of 22 to 35 N per pellet.

Part of said pellets are subjected to the rehandling test described hereinbefore. This results in a formation of less than 0.2 wt.% of fines not in excess of 1 mm. But these pellets are not suitable as a feedstock for a gasification in a fixed bed because during their drying in a heated gas stream the strength of the pellets will substantially decrease and dust will be formed in large amounts. After the pellets have been dried in an air stream at a temperature of 150°C, the pellets have a residual strength of only 15 to 25 N. If such pellets are then subjected to a further drying and to a low-temperature distillation under conditions such as are encountered in the upper portion of the fixed bed in a gasification reactor, the strength of the pellets will be further reduced to 10 to 2 N. For this reason the pellets are not suitable for gasification.

A second charge of the moist pellets is dried in superheated steam at temperatures of 120°C and, in another experiment, of 150°C. In both cases the dried pellets contain about 3 wt.% water and moisture and have a strength of 60 to 80 N. They have an adequate stability for transportation and gasification.

A third charge of the moist pellets is
contain 28 wt.% water and moisture and have a strength of 22 to 26 N. When subjected to coking, they are subjected to a high temperature. This results in the pellets being reduced to 10 wt.% water and moisture and having a strength of 5 to 7 N.

The pellets are then used as a feedstock for gasification. During their formation, they are dried in an air atmosphere and have a residual moisture of 3 wt.% after they are formed in the gasification reactor. The pellets are then dried in an air atmosphere and have a residual moisture of 3 wt.% after they are formed in the gasification reactor, reduced to 10 wt.% water and moisture and are suitable for gasification.

A fourth charge of the moist pellets is dried in a sand bed, in which the weight ratio of pellets to sand is 5 : 40 and the sand is at a temperature of 200°C. The pellet-containing sand bed is placed into a drying chamber, which is at a temperature of 200°C, and is left in said chamber for two hours. The sand is subsequently removed by sieving when it is still hot. Thereafter, the pellets are cooled in a closed vessel. The dry pellets contain 1 wt.% water and moisture and have a strength in the range from 140 to 210 N. The pellets resist abrasion and the rehandling test results in a formation of less than 0.2 wt.% fines not in excess of 1 mm. After a coking of the pellets under a pressure of 25 bars, the resulting coke pellets have breaking strengths between 100 and 150 N. In the process of fixed-bed gasification, a coking takes place also in the upper part of the fixed bed. The coking test proves that the pellets dried in the sand bed have a high resistance to pyrolysis. A formation of fines during coking has not been observed. This shows that the dry pellets can be used as a feedstock for gasification in a fixed bed.
Example 2

A hard brown coal from Poland, which contains 15 wt.% ash (on a dry basis) is dried to have a moisture content of up to 10 wt.% and is then ground in a ball mill to a particle size not in excess of 0.315 mm. Because the coal does not contain a natural binder, 6 wt.% bentonite must be added. The hard brown coal and the bentonite are mixed and moistened with water in a mixture. The resulting mixture is shaped on a pelletizing disc to form pellets which are 8 to 16 mm in diameter and contain 29 wt.% water and moisture. They have a breaking strength of 20 to 30 N. The pellets are suitable for transportation in a moist state.

A first charge of the pellets is dried in an air stream at a temperature of 150°C. This results in a partial disintegration of the pellets to fragments and dust. The resulting dried pellets have a residual strength of only 4 to 5 N. An increase of the binder content to 10 wt.% and also a decrease of the drying temperature to 100°C can result only in a slight improvement in strength. The pellets are not suitable for any use.

A further charge of the moist pellets is dried in a sand bath, which is at temperatures between 150 and 220°C. The resulting dry pellets contain about 1.5 wt.% water and moisture. Having strengths of 80 to 120 N, they are suitable for transportation.
low-temperature distillation of said dried pellets under the pressure of 25 bars, which is usual for a gasification in a fixed bed, results in coke pellets having a strength of 60 to 80 N. This shows that said dry pellets are suitable in every respect as a feedstock for gasification in a fixed bed.

Results which are similar to those described hereinbefore have been obtained by the processing of other hard brown coals, mainly from the United States, in processes which are of the kind described in Examples 1 and 2 and comprise a drying in a sand bed or in a water vapor atmosphere.
1. A process of making brown coal containing pellets for gasification in a reactor under pressures in the range from 5 to 150 bars by means of oxygen, steam and/or carbon dioxide as gasifying agents in a process in which the pellets are fed in the reactor to the top of a fixed bed, which is gradually descending, the gasifying agents are introduced into said fixed bed from below, and the mineral constituents are withdrawn as solid or in liquid form from the bottom of the fixed bed, characterized in that a mixture for pelleting is produced, in which the content of fine-grained fuel consists of at least 90 wt.% hard brown coal, which has a particle size not in excess of 1 mm, the mixture to be pelleted contains 10 to 25 wt.% water and moisture and 4 to 10 wt.% binder, the mixture is shaped with an addition of water as a pelleting liquid to form moist pellets which contain 35 to 55 wt. % water and moisture, and the moist pellets are dried in a water vapor containing atmosphere and in such a manner that the core of the pellets is heated to at least 90°C before the pellets have lost one-tenth or more of their water content and where the dried pellets contain 1 to 4 wt.% water and moisture.

2. A process according to claim 1, characterized in that the moist pellets are dried while they are
covered in a bed consisting of hot granular solids, which are at temperatures of about 150 to 300 °C.

3. A process according to claim 2, characterized in that the moist pellets are dried in a sand bed.

4. A process according to claim 1, characterized in that the pellets are dried in an atmosphere which contains superheated steam at temperatures of about 110 to 150 °C.

5. A process according to claim 1 or any of the following claims, characterized in that the hard brown coal in the mixture to be pelletized has a particle size not in excess of 0.5 mm and contains 60 to 80 wt.% of a fine fraction not in excess of 0.065 mm.

6. A process according to claim 1 or any of the following claims, characterized in that the moist pellets have diameters of about 6 to 50 mm.

7. A process according to claim 1 or any of the following claims, characterized in that the mixture to be pelletized contains 2 to 15 wt.% of a fine-grained flux, particularly lime, for reducing the melting temperature of the ash.

8. A process according to claim 1 or any of the following claims, characterized in that the pellets are fed to the top of the fixed bed together with granular coal having particle sizes in the range from 60 to 60 mm.
Provisional No. 9128 L

ABSTRACT

Process of Making Brown Coal Containing Pellets for Gasification

The gasification is performed under pressures in the range from 5 bars to 150 bars with oxygen, steam and/or carbon dioxide as gasifying agents. The pellets are fed to the top of a slowly descending fixed bed in a gasification reactor. The gasifying agents are introduced into the fixed bed from below and the mineral constituents in the form of solid ash or liquid slag are withdrawn from the lower end of the fixed bed. To make the pellets, a mixture is first provided which contains at least 80 wt.% fine-grained hard brown coal, which has a particle size not in excess of 1 mm. The mixture to be pelletized contains 10 to 25 wt.% moisture and 4 to 10 wt.% bentonite. With an addition of water as a pelletizing liquid, the mixture is shaped to form moist pellets, which contain 25 to 35 wt.% water. The moist pellets are dried in an atmosphere which is rich in water vapor.
During that drying, the core of the pellets is heated to at least 90°C before the pellets have lost one-tenth or more of their water content. The dried pellets contain water and moisture not in excess of 4 wt.%. The moist pellets may be dried, e.g., in a sand bed.
END