

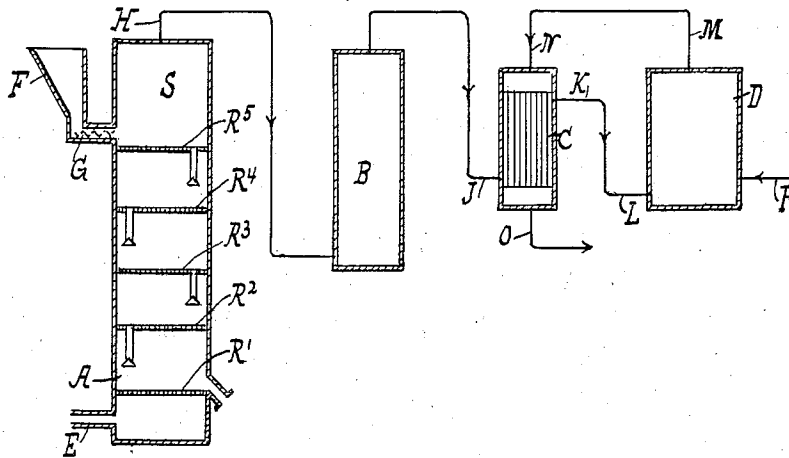
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PRODUCTION OF MIXTURES OF NITROGEN AND HYDROGEN FROM BITUMINOUS FUELS

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PRODUCTION OF MIXTURES OF NITROGEN AND HYDROGEN FROM BITUMINOUS FUELS

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The present invention relates to the production of mixtures of nitrogen and hydrogen from bituminous fuels.

It has already been proposed to subject bituminous fuels such as brown coal, lignite, peat, oil shale and the like to low temperature carbonization by passing therethrough hot gases obtained from a gasifying process. For this purpose producers having low temperature carbonization attachments are frequently employed or better still shaft furnaces which are subdivided into different treatment zones by several grates, so that the single processes are carried out separate from each other on the single grates.

It has also been proposed to split up the hydrocarbons contained in industrial or natural gases into carbon monoxide and hydrogen by incomplete combustion.

We have now found that mixtures of nitrogen and hydrogen are obtained in an advantageous manner from bituminous fuels by subjecting the fuels to low temperature carbonization by means of scavenging gases, which are obtained by gasifying the low temperature carbonization residues arranged in several layers one above the other with the aid of steam or air, which may also be enriched in oxygen, or both employed in such an amount that the layer of fuel is brought into vigorous motion on its bed and by converting the gas mixture obtained from the low temperature carbonization, after separating the condensable components, preferably by cooling down to from 20° to 25° C. into a mixture of nitrogen and hydrogen by incompletely burning the hydrocarbons contained therein preferably with air, which may also be enriched in oxygen, converting the resulting carbon monoxide with steam and separating the carbon dioxide.

If the gases obtained by the said gasification of the low temperature carbonization residues contain already sufficient nitrogen, the subsequent incomplete combustion may also be effected with oxygen.

The composition of the gasifying agent, which may consist for example of a mixture of air enriched with oxygen and steam, as well as the composition of the gas containing

oxygen necessary for the partial combustion of the hydrocarbon contained in the gas formed, as which oxygen may be employed for example, depend according to the present invention simply on the desired composition of the final gas as regards nitrogen and hydrogen and can be determined by previous calculations or by appropriate experiments.

The conversion of carbon monoxide into carbon dioxide by means of steam is carried out at temperatures ranging from 380° to 620° C., preferably between 400° and 500° C. and preferably in the presence of catalysts.

The advantages of the process according to the present invention consist not only in a considerable simplification of the apparatus; but also in the great saving of energy which results from this combination.

As special advantages of the process may be mentioned:

1. A mixture of nitrogen and hydrogen having a high content of hydrogen is obtained having any desired composition as for example the composition necessary for the synthesis of ammonia, in a continuous gasifying process, whereby the necessary heat of the gasifying and low temperature carbonization process may be covered simply by the combustion process by employing gasifying agents containing oxygen. Thus neither a preheating of the gasifying agent, nor an external supply of heat to the process is necessary.

2. The combination of the production of mixtures of nitrogen and hydrogen with the simultaneous low temperature carbonization of fuels renders possible a far reaching utilization for the low temperature carbonization of the sensible heat of steam which has remained undecomposed in the watergas reaction. As a result of the presence of steam during the low temperature carbonization, an increased yield of tar of excellent quality is obtained. The bitumen contained in the fuel is completely obtained as tar and the hydrogen in the final gas is formed simply by the watergas reaction of the small coke and by the decomposition of the hydrocarbons.

3. Apart from ashes and carbon dioxide,

only high quality final products are obtained from the fuel, firstly a mixture of nitrogen and hydrogen capable of being further worked up, for example into ammonia, and
 5 secondly a high quality tar, the entire energy content of the low temperature carbonization residues being completely utilized in the process.

The nature of this invention will be further described with reference to the accompanying drawing which illustrates diagrammatically an arrangement of apparatus suitable for carrying out the invention but the invention is not restricted to this arrangement.
 15 ment.

A shaft furnace A, which is subdivided into different treatment zones by several grates R_1 , R_2 , R_3 , R_4 , and R_5 , is supplied with fuel, as for example predried brown
 20 coal, from a bunker F through a conveyor G. A gasifying agent consisting of air enriched with oxygen and steam is introduced into the shaft furnace A below the lowest grate R_1 through a pipe E employed in such
 25 an amount that each layer of fuel is brought into vigorous motion on its bed. The small coke formed from the brown coal is thus completely gasified. The gasifying temperature at the grate R_1 amounts to about 900°
 30 C. and the distance between the grates R_1 and R_2 is adjusted so that the gasifying gas laden with undecomposed steam passes through the grate R_2 at a temperature of about 800° C. This gas mixture then effects
 35 the low temperature carbonization and the driving out of the residual water content of the coal on the grates R_2 , R_3 , R_4 and R_5 . The gas mixture laden with the products of the low temperature carbonization is freed
 40 from any dust which it has carried along with it in a dust removing plant, either a cyclone device or electrical means, arranged in the upper part S of the shaft furnace A and leaves the shaft furnace A at H at a
 45 temperature of about 250° C. The mixture of gases, water vapor and tar vapors then pass through a condensing plant B in which the anhydrous tar, benzines and low temperature carbonization water are removed
 50 from the gases by the methods usually employed in the low temperature carbonization industry. The gases leaving the condensing plant B which have been cooled to about 20° C. still contain a certain percentage of
 55 gaseous hydrocarbons. In order to remove these hydrocarbons the gas mixture is converted with a mixture of oxygen and nitrogen introduced at P into a combustion chamber D after it has been brought to a high
 60 temperature in a heat exchanger C. The gas mixture enters the heat exchanger C at J, flows therethrough in counter current to the hot gases coming from the combustion chamber D and leaves at K at a temperature of
 65 about 750° C. It then passes into the com-

bustion chamber at L. The gas mixture is strongly heated by the heat of reaction set free during the combustion and leaves the combustion chamber D at M at about 1100° C. It then enters the heat exchanger C at N and leaves it again at O cooled to 250° C. The mixed gas thus obtained is then freed from carbon monoxide in the usual manner by conversion with steam and the carbon dioxide thus formed is removed in the usual
 70 manner. The mixture of nitrogen and hydrogen which remains may be directly employed for the synthesis of ammonia without further treatment.

The following example will further illustrate the nature of this invention, but the invention is not restricted to this example.
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Example

Mid continent brown coal is worked up in
 85 the said apparatus after having been dried to about 8 to 10 per cent. The brown coal, which has been dried in the usual manner in a steam or fire-gas drying plant, has an average water content of 8.4 per cent and
 90 yields 14.9 per cent of tar.

428 cubic meters of air enriched with oxygen and having an oxygen content of 43.8 per cent and 622 kilograms of steam are employed for the gasification of each metric
 95 ton of predried brown coal.

The yield of high quality anhydrous low temperature carbonization tar amounts to 142 kilograms per metric ton of predried brown coal.
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After separating the tar, benzines and low temperature carbonization water there remain 1380 cubic meters of a gas mixture having the composition 20.2 per cent of carbon dioxide, 22.1 per cent of carbon monoxide, 36.0 per cent of hydrogen, 17.9 per cent of nitrogen, 1.0 per cent of hydrogen sulphide and 2.8 per cent of hydrocarbons.
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58.3 cubic meters of a mixture of oxygen and nitrogen containing 79.2 per cent of oxygen are necessary for the incomplete combustion of the hydrocarbons. 1418 cubic meters of a gas are then obtained having the composition 19.7 per cent of carbon dioxide, 25.5 per cent of carbon monoxide, 35.0 per cent of hydrogen, 18.7 per cent of nitrogen, 1.0 per cent of hydrogen sulphide and 0.1 per cent of methane. This gas mixture is freed from carbon monoxide by conversion with steam. After removing the
 110 carbon dioxide thus formed, about 1134 cubic meters of a mixture of nitrogen and hydrogen containing about 75 per cent of hydrogen is obtained which may be directly employed for the synthesis of ammonia.
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What we claim is:—

1. A process for the production of a mixture of hydrogen and nitrogen from bituminous fuel which comprises subjecting said fuel while arranged in layers to a low tem-
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perature carbonization, subjecting the residues from said carbonization, also arranged in layers, to a gasification by blowing upwardly through said layers of residue, a gasifying agent in an amount sufficient to bring said layers of residue into vigorous motion on their beds, the low temperature carbonization of said fuel being effected by leading the gases arising from said gasification still hot through the layers of fuel to be carbonized, separating by condensation the condensable compounds contained in the gases and vapors issuing from the low temperature carbonization, subjecting the non-condensed gases to incomplete combustion, carbon monoxide thereby being formed, treating the incompletely burned gases with steam at a temperature between 380° and 620° C., the carbon monoxide thus being converted into carbon dioxide, removing said carbon dioxide from the gases and effecting at least one of the steps of gasification and incomplete combustion by means of a gas containing nitrogen.

2. A process for the production of a mixture of hydrogen and nitrogen from bituminous fuel which comprises subjecting said fuel while arranged in layers to a low temperature carbonization, subjecting the residues from said carbonization, also arranged in layers, to a gasification by means of steam and air by blowing upwardly through said layers of residue, a gasifying agent in an amount sufficient to bring said layers of residue into vigorous motion on their beds, the low temperature carbonization of said fuel being effected by leading the gases arising from said gasification still hot through the layers of fuel to be carbonized, separating by condensation the condensable compounds contained in the gases and vapors issuing from the low temperature carbonization, subjecting the non-condensed gases to incomplete combustion with air, carbon monoxide thereby being formed, treating the incompletely burned gases with steam at a temperature between 380° and 620° C., the carbon monoxide thus being converted into carbon dioxide, and removing said carbon dioxide from the gases.

3. A process for the production of a mixture of hydrogen and nitrogen from bituminous fuel which comprises subjecting said fuel while arranged in layers to a low temperature carbonization, subjecting the residues from said carbonization, also arranged in layers, to a gasification by blowing upwardly through said layers of residue, a gasifying agent in an amount sufficient to bring said layers of residue into vigorous motion on their beds, the low temperature carbonization of said fuel being effected by leading the gases arising from said gasification still hot through the layers of fuel to be carbonized, separating by condensation

the condensable compounds contained in the gases and vapors issuing from the low temperature carbonization, subjecting the non-condensed gases to incomplete combustion, carbon monoxide thereby being formed, treating the incompletely burned gases with steam at a temperature between 380 and 620° C., the carbon monoxide thus being converted into carbon dioxide, and removing said carbon dioxide from the gases, effecting at least one of the steps of gasification and incomplete combustion by means of a gas containing nitrogen, the amounts and compositions of the gases containing nitrogen used for said gasification and for said incomplete combustion being so adjusted that the said gases freed from carbon dioxide represent a mixture which may directly be employed as initial material for the synthesis of ammonia.

4. A process for the production of a mixture of hydrogen and nitrogen from bituminous fuel which comprises subjecting said fuel while arranged in layers to a low temperature carbonization, subjecting the residue from said carbonization, also arranged in layers, to a gasification with steam and air enriched in oxygen by blowing upwardly through said layers of residue, a gasifying agent in an amount sufficient to bring said layers of residue into vigorous motion on their beds, the low temperature carbonization of said fuel being effected by leading the gases arising from said gasification still hot through the layers of fuel to be carbonized, separating by condensation the condensable compounds contained in the gases and vapors issuing from the low temperature carbonization, subjecting the noncondensed gases to incomplete combustion with air enriched in oxygen, carbon monoxide thereby being formed, treating the incompletely burned gases with steam at a temperature between 380° and 620° C., the carbon monoxide thus being converted into carbon dioxide, and removing said carbon dioxide from the gases.

5. A process for the production of a mixture of hydrogen and nitrogen from bituminous fuel which comprises subjecting said fuel while arranged in layers to a low temperature carbonization, subjecting the residue from said carbonization, also arranged in layers, to a gasification by means of steam and air enriched in oxygen by blowing upwardly through said layers of residue, a gasifying agent in an amount sufficient to bring said layers of residue into vigorous motion on their beds, the low temperature carbonization of said fuel being effected by leading the gases arising from said gasification still hot through the layers of fuel to be carbonized, separating by condensation the condensable compounds contained in the gases and vapors issuing from the low tem-

perature carbonization by cooling said gases
and vapors down to from 20-25° C., sub-
jecting the non-condensed gases to incom-
plete combustion with air enriched in oxy-
5 gen, carbon monoxide thereby being formed,
treating the incompletely burned gases with
steam at a temperature between 400 and
500° C., the carbon monoxide thus being con-
verted into carbon dioxide, and removing
10 said carbon dioxide from the gases.

In testimony whereof we have hereunto
set our hands.

CHRISTIAN SCHNEIDER.
HEINRICH BUETEFISCH.

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