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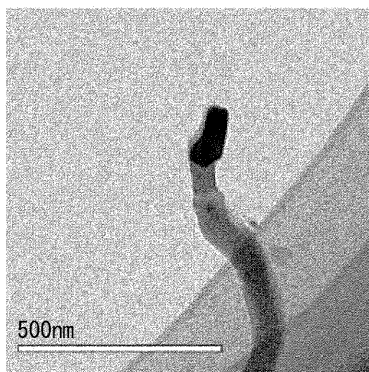
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(54) **IRON ORE PELLET PRODUCTION METHOD**

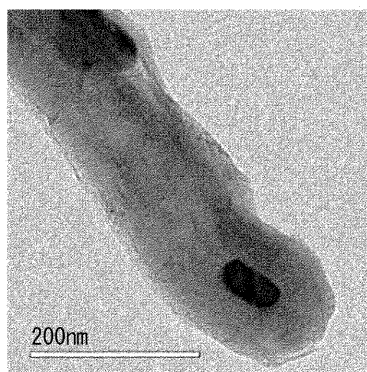
(57) Provided is a method of producing iron ore pellets that can obtain high-strength iron ore pellets and contribute to carbon neutrality. The method of producing iron ore pellets includes: a mixing process of mixing iron ore having a total Fe content of 63 mass% or less, solid carbon, binder, and auxiliary material to obtain a mixture; a granulation process of granulating the mixture to obtain

green pellets; and a firing process of firing the green pellets to obtain iron ore pellets by burning CH<sub>4</sub> gas to heat the green pellets from outside while burning the solid carbon to heat the green pellets from inside. The solid carbon includes carbon produced from one or more gases selected from the group consisting of CO<sub>2</sub> gas, CO gas, and CH<sub>4</sub> gas.

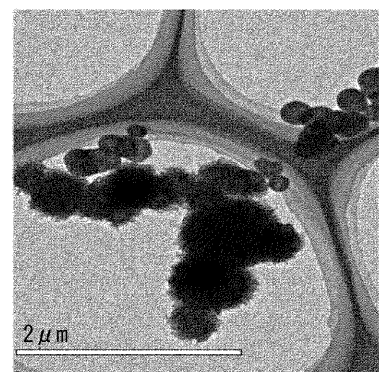
*FIG. 4A*



*FIG. 4B*



*FIG. 4C*



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**Description**

## TECHNICAL FIELD

**[0001]** The present disclosure relates to a method of producing iron ore pellets.

## BACKGROUND

**[0002]** Iron ore pellets are made from iron ore powder granulated to have properties (such as size, strength, and reducibility) suitable for feeding into a blast furnace or solid-reducing furnace. Iron ore pellets are typically produced by a process of mixing iron ore powder, binder, and optional auxiliary material to obtain a mixture, a process of granulating the mixture to obtain green pellets, and a firing process of firing the green pellets to obtain iron ore pellets. Hereinafter, pellets in granulated form before firing are referred to as "green pellets". Here, adding carbon material such as anthracite to the mixture is known, as described in Non-Patent Literature (NPL) 1. In the firing process, the green pellets are heated by burning natural gas, which is mainly CH<sub>4</sub> gas, and the heat generated by this combustion is transferred from the surface of the green pellets to the interior. This may result in insufficient heating of the pellet interior, leading to a decrease in strength. Therefore, to supplement heat inside the pellets, carbon material such as anthracite is added to the green pellets, and by burning the carbon material in the firing process, the green pellets are heated from the inside as well.

## CITATION LIST

## Non-Patent Literature

**[0003]** NPL 1: Volodymyr Shatokha, "Iron Ores and Iron Oxide Materials", IntechOpen, published July 11, 2018, p41-59

## SUMMARY

## (Technical Problem)

**[0004]** Securing green pellet strength, and therefore iron ore pellet strength, is important in order to suppress pulverization of the green pellets during handling before being put into the kiln and to suppress adherence of resulting powder to the kiln, and therefore improvements are always being sought.

**[0005]** Further, carbon-neutral steelmaking methods are desired in response to recent public demand for CO<sub>2</sub> emission reduction. In the production of iron ore pellets, the combustion of CH<sub>4</sub> gas and carbon material in the firing process is the main source of CO<sub>2</sub> emission. As a method of producing iron ore pellets without CO<sub>2</sub> emission, the use of organic/inorganic binders has also been proposed to use unfired green pellets as iron ore pellets. However, unfired green pellets have the following problems in terms of quality. Organic binders heated to high temperatures undergo carbonization or a gasification reaction with CO<sub>2</sub> ( $C + CO_2 \rightarrow 2CO$ ) that accompanies carbonization, and therefore are no longer able to exist as a binder. It is therefore anticipated that it will be difficult to achieve binder properties when organic binders are used at high temperatures. Further, silicates (sodium silicate and calcium silicate) are examples of inorganic binders, but silicic acid itself must be separated as slag from pig iron or molten steel in either the pig iron or steelmaking process, and excess heat is required to melt pig iron. Further, an issue is caused of inorganic binder being discharged as slag, that is, excess material. Further, while the sodium in sodium silicate is solid at low temperatures, it vaporizes at temperatures of 1000 °C or less, making sodium a substance better avoided as a substance to be retained in a blast furnace. There is no known carbon-neutral oriented technology based on the premise of producing iron ore pellets through a firing process.

**[0006]** In view of the above technical problems, it would be helpful to provide a method of producing iron ore pellets that can obtain high-strength iron ore pellets and contribute to carbon neutrality.

## (Solution to Problem)

**[0007]** To work towards this, the inventors focused on improving carbon material added to green pellets. That is, the inventors arrived at the idea of using solid carbon including carbon produced from one or more gases selected from the group consisting of CO<sub>2</sub> gas, CO gas, and CH<sub>4</sub> gas as the carbon material to be added to the green pellets. In this case, although CO<sub>2</sub> is generated by the combustion of the carbon material, the above greenhouse gases are consumed as raw material for the carbon material, thus contributing to carbon neutrality. Further, the inventors have found that the use of solid carbon (carbon material) including carbon produced from one or more gases selected from the group consisting of CO<sub>2</sub> gas, CO gas, and CH<sub>4</sub> gas unexpectedly increases the strength of the iron ore pellets.

**[0008]** The present disclosure was completed based on these discoveries, and primary features of the present

disclosure are described below.

[1] A method of producing iron ore pellets, the method comprising:

a mixing process of mixing iron ore having a total Fe content of 63 mass% or less, solid carbon, binder, and auxiliary material to obtain a mixture;  
a granulation process of granulating the mixture to obtain green pellets; and  
a firing process of firing the green pellets to obtain iron ore pellets by burning CH<sub>4</sub> gas to heat the green pellets from outside while burning the solid carbon to heat the green pellets from inside, wherein  
the solid carbon includes carbon produced from one or more gases selected from the group consisting of CO<sub>2</sub> gas, CO gas, and CH<sub>4</sub> gas.

[2] The method of producing iron ore pellets according to [1], wherein the solid carbon includes carbon produced from one or both of CO<sub>2</sub> gas and CO gas.

[3] The method of producing iron ore pellets according to [1], wherein the CH<sub>4</sub> gas used in the production of the carbon includes CH<sub>4</sub> gas produced from one or both of CO<sub>2</sub> gas and CO gas.

[4] The method of producing iron ore pellets according to any one of [1] to [3], wherein the carbon includes carbon produced using iron as a catalyst.

[5] The method of producing iron ore pellets according to any one of [1] to [4], wherein the fraction of the carbon in the solid carbon is 10 mass% or more.

[6] The method of producing iron ore pellets according to any one of [1] to [5], wherein the CH<sub>4</sub> gas used in the firing process includes CH<sub>4</sub> gas produced from one or both of CO<sub>2</sub> and CO gas.

(Advantageous Effect)

**[0009]** The method of producing iron ore pellets can obtain high-strength iron ore pellets and contribute to carbon neutrality.

#### BRIEF DESCRIPTION OF THE DRAWINGS

**[0010]** In the accompanying drawings:

FIG. 1 is a schematic diagram of a vertical reactor used in a carbon production test using CO gas as raw material; FIG. 2A is a photograph of sintered ore after a carbon production test, and FIG. 2B is a photograph of solid carbon after a carbon production test;  
FIG. 3A and FIG. 3B are schematic diagrams of apparatus used in a carbon production test using CH<sub>4</sub> gas as raw material; and  
FIG. 4A, FIG. 4B, and FIG. 4C illustrate TEM observation results of solid carbon obtained from carbon production tests.

#### DETAILED DESCRIPTION

**[0011]** The following describes an embodiment of the method of producing iron ore pellets. The embodiment described below is an example embodiment of the present disclosure, and does not limit configuration to the specific example described.

**[0012]** The method of producing iron ore pellets according to an embodiment of the present disclosure includes: a mixing process of mixing iron ore, solid carbon (carbon material), binder, and auxiliary material to obtain a mixture; a granulation process of granulating the mixture to obtain green pellets; and a firing process of firing the green pellets to obtain iron ore pellets by burning CH<sub>4</sub> gas to heat the green pellets from outside while burning the solid carbon to heat the green pellets from inside. The solid carbon includes carbon produced from one or more gases selected from the group consisting of CO<sub>2</sub> gas, CO gas, and CH<sub>4</sub> gas.

**[0013]** The mixture used to produce the iron ore pellets consists of iron ore, solid carbon, binder, and auxiliary material. According to the present embodiment, the iron ore includes iron ore having a total Fe content (T.Fe.) of 63 mass% or less. Iron ore having a T.Fe. of 63 mass% or less is inexpensive and suitable for production.

**[0014]** The amount of solid carbon mixed in the iron ore pellets is preferably 0.80 mass% or more relative to the iron ore content of the iron ore pellets in order to obtain a sufficient green pellet strength improving effect. On the other hand, an excess of solid carbon causes melting of the iron ore pellets due to excessive heat in the firing process. Therefore, the amount of solid carbon in the iron ore pellets is preferably 2.00 mass% or less relative to the iron ore content of the iron ore

pellets.

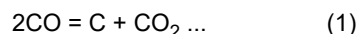
**[0015]** The solid carbon includes carbon produced from one or more gases selected from the group consisting of CO<sub>2</sub> gas, CO gas, and CH<sub>4</sub> gas. That is, carbon produced from one or more gases selected from the group consisting of CO<sub>2</sub> gas, CO gas, and CH<sub>4</sub> gas is used as the carbon material (solid carbon) added to the green pellets. In this case, although CO<sub>2</sub> is generated by the combustion of the carbon material, the above greenhouse gases are consumed as raw material for the carbon material, thus contributing to carbon neutrality. Further, the addition of carbon produced from one or more gases selected from the group consisting of CO<sub>2</sub> gas, CO gas, and CH<sub>4</sub> gas to the green pellets has the effect of increasing the strength of the green pellets and the iron ore pellets.

**[0016]** The fraction of the carbon described above in the solid carbon added to the green pellets is preferably at least 10 mass%. The fraction of the carbon in the solid carbon is more preferably at least 50 mass%. The fraction of the carbon in the solid carbon is most preferably 100 mass%. The higher the mix proportion of the carbon, the greater the effect of higher strength and the greater the contribution to carbon neutrality. When the fraction of the carbon in the solid carbon added to the green pellets is less than 100 mass%, the balance of the solid carbon may be, for example, anthracite.

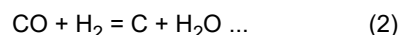
**[0017]** The solid carbon preferably includes carbon produced from one or both of CO<sub>2</sub> and CO gas. When the solid carbon includes carbon produced from CH<sub>4</sub> gas, the CH<sub>4</sub> gas used in the production of the carbon preferably includes CH<sub>4</sub> gas produced from one or both of CO<sub>2</sub> and CO gas. Reactions to produce the solid carbon from CO<sub>2</sub> gas, CO gas, or CH<sub>4</sub> gas are not particularly limited, and the following reactions are examples.

[Reactions to produce solid carbon from CO gas]

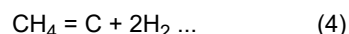
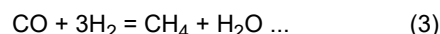
**[0018]** Solid carbon may be produced from CO gas by the Boudouard reaction indicated by reaction equation (1). The Boudouard reaction can produce solid carbon from CO gas at temperatures of about 700 °C or less.



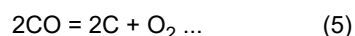
**[0019]** Solid carbon may be produced from CO gas by the reverse water-gas shift reaction indicated by reaction equation (2). The reverse water-gas shift reaction can produce solid carbon from CO gas at temperatures of about 650 °C or less.



**[0020]** CH<sub>4</sub> gas may be produced from CO gas by the methanation reaction indicated by reaction equation (3), and solid carbon may be produced from CH<sub>4</sub> gas by the pyrolysis reaction indicated by reaction equation (4). The methanation reaction can produce CH<sub>4</sub> gas from CO gas at temperatures of about 650 °C or less, and the pyrolysis reaction can produce solid carbon from CH<sub>4</sub> gas in air at temperatures of about 500 °C or more.

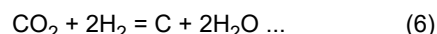


**[0021]** Solid carbon may be produced from CO gas by the decomposition reaction indicated by reaction equation (5). The decomposition reaction can produce solid carbon from CO gas under low oxygen partial pressure.

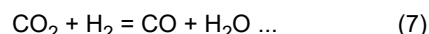


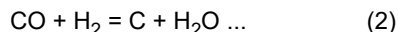
[Reactions to produce solid carbon from CO<sub>2</sub> gas]

**[0022]** Solid carbon may be produced from CO<sub>2</sub> gas by the reverse water-gas shift reaction indicated by reaction equation (6). The reverse water-gas shift reaction can produce solid carbon from CO<sub>2</sub> gas at temperatures of about 650 °C or less.

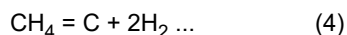
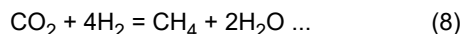


**[0023]** CO gas may be produced from CO<sub>2</sub> gas by the reverse water-gas shift reaction indicated by reaction equation (7), and solid carbon may be produced from CO gas by the reverse water-gas shift reaction indicated by reaction equation (2). The reverse water-gas shift reaction can produce CO gas from CO<sub>2</sub> gas at temperatures of about 850 °C or more.

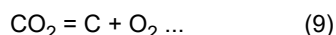




**[0024]** CH<sub>4</sub> gas may be produced from CO<sub>2</sub> gas by the methanation reaction indicated by reaction equation (8), and solid carbon may be produced from CH<sub>4</sub> gas by the pyrolysis reaction indicated by reaction equation (4). The methanation reaction can produce CH<sub>4</sub> gas from CO<sub>2</sub> gas at temperatures of about 600 °C or less.

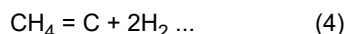


**[0025]** Solid carbon may be produced from CO<sub>2</sub> gas by the decomposition reaction indicated by reaction equation (9). The decomposition reaction can generate solid carbon from CO<sub>2</sub> gas under low oxygen partial pressure.



[Reaction to produce solid carbon from CH<sub>4</sub> gas]

**[0026]** Solid carbon may be produced from CH<sub>4</sub> gas by the pyrolysis reaction indicated by reaction equation (4).



**[0027]** The gas used for the carbon production reaction may be CO<sub>2</sub> gas, CO gas, or CH<sub>4</sub> gas alone, a mixture of two or more of the above three gases, or a mixture of one or more of the above three gases and H<sub>2</sub> gas, N<sub>2</sub> gas, or other gas. For example, a gas mixture, in vol%, of CO: 31 %, H<sub>2</sub>: 19 %, and N<sub>2</sub>: 50 % may be used.

**[0028]** Further, as the solid carbon added to the green pellets, any one of carbon produced from CO<sub>2</sub> gas, carbon produced from CO gas, or carbon produced from CH<sub>4</sub> gas may be used alone, or a mixture of two or more may be used.

**[0029]** The solid carbon is preferably produced using iron as a catalyst. It is known that iron can be used as a catalyst for the Boudouard reaction indicated in reaction equation (1) and the pyrolysis reaction of CH<sub>4</sub> indicated in reaction equation (4). Further, it is known that iron oxide can be used as a catalyst for the reverse water-gas shift reaction indicated in reaction equation (7). Therefore, when producing the solid carbon, sintered ore or directly reduced iron may be charged into a furnace, and the iron or iron oxide contained in the sintered ore or the directly reduced iron may be used as a catalyst. Further, in a high temperature reaction such as the CH<sub>4</sub> pyrolysis reaction indicated in reaction equation (4), alumina may be charged into the furnace to maintain the furnace temperature.

**[0030]** The solid carbon is preferably in fibrous form and preferably has an aspect ratio (length/diameter) of 10 or more. When the solid carbon is fibrous, the strength of the iron ore pellets is suitably obtainable. Further, the solid carbon may be in spherical form. When the solid carbon is spherical, a binder effect of fine particles is obtained and the strength of the iron ore pellets is suitably obtainable. When the solid carbon is spherical, cumulative particle size D90 is preferably from 10 μm to 50 μm. The solid carbon (carbon material) produced from one or more gases selected from the group consisting of CO<sub>2</sub> gas, CO gas, and CH<sub>4</sub> gas preferably has a carbon content of 50 mass% more, and the balance may contain Fe, FeO, and the like.

**[0031]** Bentonite is a preferred binder for the iron ore pellets, but any known or optional binder may be used, including organic and inorganic binders that provide similar effects. The amount of the binder is preferably 0.1 mass% or more relative to the iron ore content of the iron ore pellets in order to achieve sufficient effect. On the other hand, when an excessive amount of binder is included, in addition to higher production costs, the effect of the binder becomes smaller as the content increases. Therefore, the amount of the binder is preferably 4.0 mass% or less relative to the iron ore content of the iron ore pellets.

**[0032]** The iron ore pellets may be mixed with quicklime, limestone (CaCO<sub>3</sub>), dolomite (CaMg(CO<sub>3</sub>)<sub>2</sub>), or the like, as the auxiliary material. Adjustment of basicity of the iron ore pellets is made by the auxiliary material. The basicity of the iron ore pellets is calculated by the weight ratio of CaO/SiO<sub>2</sub> in the iron ore pellets. The basicity of the iron ore pellets is preferably from 0.01 to 1.5.

**[0033]** The iron ore pellets are produced by typical grinding, mixing, granulation, and firing processes. The grinding process may be carried out using a typical ball mill or other grinder. The mixing process may be carried out using a typical concrete mixer, high-speed mixer, or the like. The granulation process may be carried out using a typical pelletizer, drum mixer, or the like.

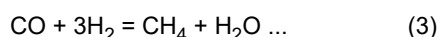
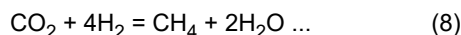
**[0034]** The Blaine number of the iron ore after the grinding is preferably around 2000 cm<sup>2</sup>/g to 4000 cm<sup>2</sup>/g. The Blaine number is measured by a Blaine air permeability apparatus as specified in JIS R 5201:2015 and represents the specific

surface area of powder. In the pellet production process, the Blaine number is used as a control index for ore particle size, with higher values indicating finer powders.

**[0035]** A granulated green pellet preferably has a particle size of about 9.5 mm to 12 mm. When the particle size of the green pellets is less than 9.5 mm, gas permeability degrades when filled into a blast furnace as fired pellets. When the particle size of the green pellets exceeds 12 mm, reducibility decreases.

**[0036]** The firing process may be carried out using a typical rotary kiln, electric furnace, or the like. In the firing process, the green pellets are heated from outside by burning CH<sub>4</sub> gas, while the solid carbon is burned to heat the green pellets from inside. Firing conditions are preferably a furnace temperature of 1200 °C to 1350 °C and a hold time at the furnace temperature of 5 min to 30 min.

**[0037]** The CH<sub>4</sub> gas used in the firing process preferably includes CH<sub>4</sub> gas produced from one or both of CO<sub>2</sub> and CO gas, in terms of a carbon neutral viewpoint. The method of producing the CH<sub>4</sub> gas is not particularly limited, but it may be produced, for example, by the methanation reaction indicated in reaction equations (3) and (8).



## EXAMPLES

**[0038]** Iron ore, binder, and auxiliary material were prepared as raw material for iron ore pellets. Table 1 lists the composition of the iron ore used. The iron ore was dried at 105 °C for 24 h and then grinding was carried out to obtain iron ore powder having a Blaine number of 2560 cm<sup>2</sup>/g and a volume average diameter of 95 μm. Bentonite was used as the binder and limestone was used as the auxiliary material.

[Table 1]

| Table 1 (mass%) |                  |                                |      |      |     |
|-----------------|------------------|--------------------------------|------|------|-----|
| T.Fe            | SiO <sub>2</sub> | Al <sub>2</sub> O <sub>3</sub> | CaO  | MgO  | LOI |
| 61.50           | 3.70             | 2.30                           | 0.07 | 0.09 | 5.4 |

**[0039]** Next, tests were conducted to produce solid carbon from CO gas or CH<sub>4</sub> gas. Details are provided below. Further, anthracite was prepared as solid carbon that was not produced from any of CO, CO<sub>2</sub>, or CH<sub>4</sub> gas. Anthracite was prepared in particle form having a particle size of 1 mm or less and a C content of 85 mass%.

[Carbon production test from CO gas]

**[0040]** A vertical reactor was used to produce carbon from CO gas. FIG. 1 illustrates a schematic diagram of a vertical reactor. An alumina support 12, alumina balls 14, and sintered ore 16 were charged in this order into a furnace core tube 10 having an inner diameter of 80 mm. Solid carbon was produced by flowing a gas comprising, in vol%, CO: 31 %, H<sub>2</sub>: 19 %, and N<sub>2</sub>: 50 % (hereafter referred to as CO mixed gas) from a gas inlet pipe 18 at 550 °C or 800 °C (heated by a heater 20 and measured with a thermocouple 22) for 3 h at a gas flow rate of 17 L/min. The iron contained in the sintered ore was used as a catalyst for carbon production.

**[0041]** After reduction, a sample was sieved through a 0.125 mm sieve to separate sintered ore and solid carbon. For the sample after reduction, 1.5 wt% was solid carbon. FIG. 2A is a photograph of sintered ore and FIG. 2B is a photograph of solid carbon after the carbon production test from CO mixed gas at 800 °C. The solid carbon was a fine powder, having a cumulative particle size measured at D10: 2.1 μm, D50: 6.61 μm, and D90: 14.8 μm. That is, 90 % or more of the cumulative number of powder particles had a particle size of 14.8 μm or less. Table 2 lists the results of the component analysis of the solid carbon.

[Table 2]

| Table 2 (mass%) |      |     |      |
|-----------------|------|-----|------|
| T.Fe            | M.Fe | FeO | C    |
| 13.6            | 7.8  | 7.2 | 72.0 |

[Carbon production test from CH<sub>4</sub> gas]

**[0042]** Carbon was produced from CH<sub>4</sub> gas using the apparatus illustrated in FIG. 3A and FIG. 3B. First, 30 g (about 20 to 30 pieces) of directly reduced iron (DRI) 32, which was reduced from 10 mm diameter iron ore pellets, was charged as a catalyst into an electric furnace 30 illustrated in FIG. 3A, and 100 % CH<sub>4</sub> gas was flowed in from a gas inlet 34 at 900 °C for 1 h at a gas flow rate of 1.0 L/min to produce solid carbon. Next, 500 g of 6 mm diameter alumina balls 38 were charged into the 80 mm diameter furnace core tube (alumina tube) 36 illustrated in FIG. 3B to form a soaking zone of about 50 mm height, and 100 % CH<sub>4</sub> gas was supplied from a gas inlet 40 at a gas temperature of 1400 (± 10) °C (heated by a heater 42) for 1 h at a gas flow rate of 1.0 L/min to produce solid carbon. The post-reduction samples from each test were sieved through a 0.125 mm sieve and respectively separated into DRI or alumina balls and solid carbon.

**[0043]** Table 3 lists the morphology and carbon content of the solid carbon and anthracite produced under each condition. FIG. 4A illustrates a TEM observation result of solid carbon formed from CO mixed gas at 550 °C, FIG. 4B illustrates a TEM observation result of solid carbon formed from CH<sub>4</sub> gas at 900 °C, and FIG. 4C illustrates a TEM observation result of solid carbon formed from CH<sub>4</sub> gas at 1400 °C. The solid carbon obtained in the reactions at 900 °C or less was fibrous and had an aspect ratio of 10 or more. The solid carbon obtained in the reaction at 1400 °C was spherical, having a particle size ranging from about 0.2 μm to 2.0 μm. This is thought to be because at 1400 °C, the high temperature makes the reaction highly reactive and the production of carbon in the gas phase occurs as the main reaction, while at 900 °C or lower, the pyrolysis reaction of CH<sub>4</sub> gas does not proceed easily and the production reaction using iron as a catalyst occurs as the main reaction.

[Table 3]

**[0044]**

Table 3

| Solid carbon type | Gas type (vol%)                                    | Temperature (°C) | Morphology  | C (mass%) |
|-------------------|----------------------------------------------------|------------------|-------------|-----------|
| Anthracite        | -                                                  | -                | Particulate | 85        |
| Solid carbon 1    | CO 31%<br>H <sub>2</sub> 19%<br>N <sub>2</sub> 50% | 550              | Fibrous     | 51        |
| Solid carbon 2    |                                                    | 800              | Fibrous     | 72        |
| Solid carbon 3    | CH <sub>4</sub> 100%                               | 900              | Fibrous     | 69        |
| Solid carbon 4    |                                                    | 1400             | Spherical   | 99        |

**[0045]** 1000 g of iron ore powder was prepared, to which various solid carbons in mix proportions listed in Table 4 relative to the amount of iron ore were added, and 1 mass% of bentonite relative to the amount of iron ore was added. Further, limestone was added to the iron ore powder with the amount of addition set so that the basicity of the iron ore pellets was 0.1, and the mixture was mixed at 20 rpm for 3 min using a concrete mixer. The mix proportions of solid carbon in No. 2 to 7 was set so that the same amount of carbon was mixed as that contained by anthracite in No. 1. The fraction of carbon in anthracite replaced by the carbon of solid carbons 1 to 4 is listed in Table 4 under "Carbon replacement fraction". Next, the mixed raw material was placed in a 1.2 m diameter pelletizer and granulation was carried out while adding water. Pellet particles having a particle size of 9.5 mm to 12 mm were collected and rolled in a pelletizer for another 10 min to obtain green pellets.

[Table 4]

**[0046]**

Table 4

| No. | Solid carbon type           | Solid carbon mix proportion (mass%) |                  | Carbon replacement fraction (mass%) | Drop strength (times) | Crushing strength (kgf) | Remarks             |
|-----|-----------------------------|-------------------------------------|------------------|-------------------------------------|-----------------------|-------------------------|---------------------|
|     |                             | Anthracite                          | Solid carbon 1~4 |                                     |                       |                         |                     |
| 1   | Anthracite                  | 1.00%                               | -                | -                                   | 5.3                   | 255                     | Comparative Example |
| 2   | Solid carbon 1 + anthracite | 0.90%                               | 0.17%            | 10%                                 | 5.5                   | 266                     | Example             |
| 3   | Solid carbon 1 + anthracite | 0.50%                               | 0.83%            | 50%                                 | 7.2                   | 270                     | Example             |
| 4   | Solid carbon 1              | -                                   | 1.70%            | 100%                                | 8.5                   | 277                     | Example             |
| 5   | Solid carbon 2              | -                                   | 1.20%            | 100%                                | 9.2                   | 268                     | Example             |
| 6   | Solid carbon 3              | -                                   | 1.20%            | 100%                                | 7.5                   | 276                     | Example             |
| 7   | Solid carbon 4              | -                                   | 0.85%            | 100%                                | 6.6                   | 266                     | Example             |

[Green pellet drop strength measurement]

**[0047]** For each of the Examples and Comparative Examples, drop strength measurements were carried out on ten green pellets, simulating conveyance, charging, and the like in actual operations. The process of dropping each of the green pellets from a height of 50 cm was repeated, and the process was terminated when a crack or fracture was observed in the green pellet. The number of times before termination (that is, the time when cracking or fracture was observed) was used as the drop strength, and the average drop strength of the ten particles is listed in Table 3.

**[0048]** Green pellets not used in the above test were charged into an electric furnace and fired. The temperature was increased at 10 °C/min in an air atmosphere, held at 1300 °C for 10 min, and then decreased at 10 °C/min. After the temperature was decreased, the sample was removed and iron ore pellets were obtained. In an actual firing process, the green pellets are heated by burning natural gas, mainly CH<sub>4</sub> gas, and an electric furnace was used in this test to simulate such heating.

[Iron ore pellet crushing strength measurement]

**[0049]** For each of the Examples and Comparative Examples, crushing strength (kgf) was measured for ten iron ore pellets using an autograph. The displacement speed was 2 mm/min, and the average values for ten particles are listed in Table 3.

**[0050]** Table 4 indicates that the Examples are superior to the Comparative Examples in both green pellet drop strength and iron ore crushing strength. In addition to the above, by replacing solid carbon from conventional anthracite with carbon produced from one or more gases selected from the group consisting of CO<sub>2</sub> gas, CO gas, and CH<sub>4</sub> gas, it is possible to contribute to a reduction in CO<sub>2</sub> emissions, clearly demonstrating the effectiveness of the present disclosure. Further, it is clear that carbon neutrality can be achieved in the production of iron ore pellets by using CH<sub>4</sub> gas produced from one or both of CO<sub>2</sub> gas and CO gas in the carbon production and firing process with a solid carbon replacement fraction of 100 mass%.

#### INDUSTRIAL APPLICABILITY

**[0051]** The present disclosure provides a method of producing iron ore pellets that can obtain high-strength iron ore pellets and contribute to carbon neutrality.

#### REFERENCE SIGNS LIST

**[0052]**

- 10 furnace core tube
- 12 alumina support

14 alumina balls  
 16 sintered ore  
 18 gas inlet pipe  
 20 heater  
 22 thermocouple  
 30 electric furnace  
 32 reduced iron ore pellet DRI  
 34 gas inlet  
 36 furnace core tube  
 38 alumina balls  
 40 gas inlet  
 42 heater

# Claims

1. A method of producing iron ore pellets, the method comprising:

a mixing process of mixing iron ore having a total Fe content of 63 mass% or less, solid carbon, binder, and auxiliary material to obtain a mixture;  
 a granulation process of granulating the mixture to obtain green pellets; and  
 a firing process of firing the green pellets to obtain iron ore pellets by burning CH<sub>4</sub> gas to heat the green pellets from outside while burning the solid carbon to heat the green pellets from inside, wherein the solid carbon includes carbon produced from one or more gases selected from the group consisting of CO<sub>2</sub> gas, CO gas, and CH<sub>4</sub> gas.

2. The method of producing iron ore pellets according to claim 1, wherein the solid carbon includes carbon produced from one or both of CO<sub>2</sub> gas and CO gas.

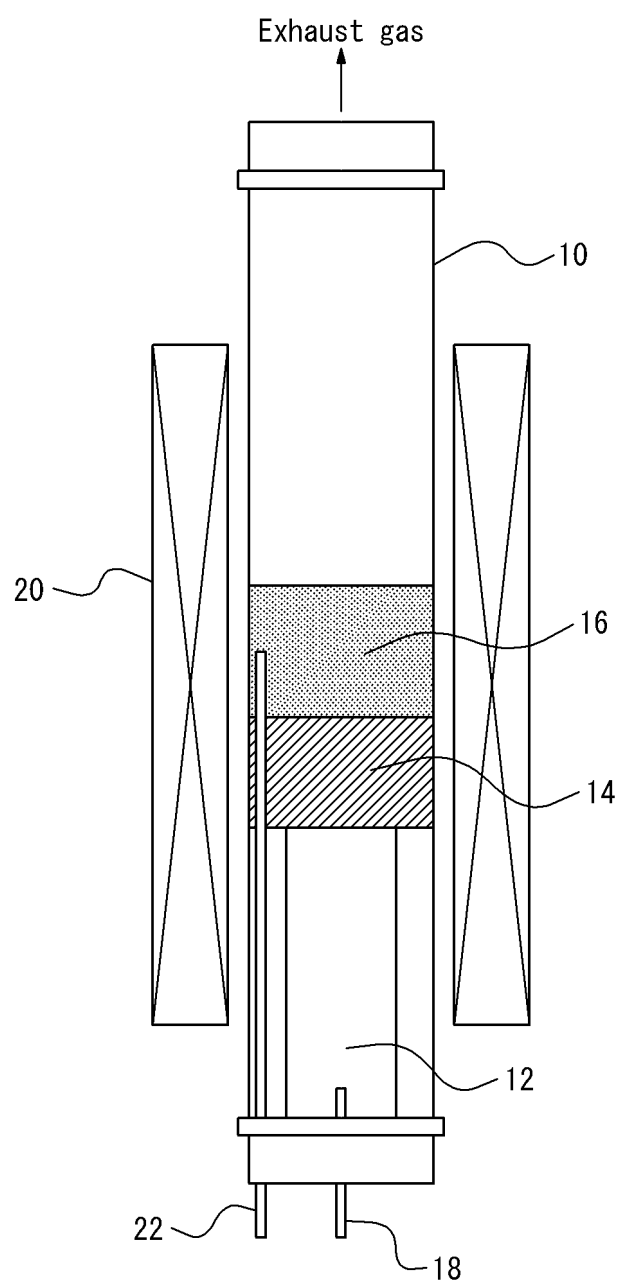
3. The method of producing iron ore pellets according to claim 1, wherein the CH<sub>4</sub> gas used in the production of the carbon includes CH<sub>4</sub> gas produced from one or both of CO<sub>2</sub> gas and CO gas.

4. The method of producing iron ore pellets according to any one of claims 1 to 3, wherein the carbon includes carbon produced using iron as a catalyst.

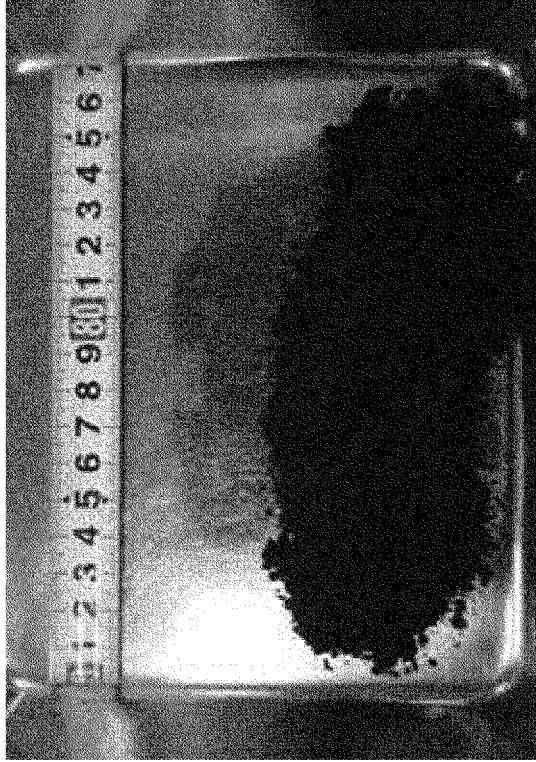
5. The method of producing iron ore pellets according to any one of claims 1 to 4, wherein the fraction of the carbon in the solid carbon is 10 mass% or more.

6. The method of producing iron ore pellets according to any one of claims 1 to 5, wherein the CH<sub>4</sub> gas used in the firing process includes CH<sub>4</sub> gas produced from one or both of CO<sub>2</sub> and CO gas.

*FIG. 1*



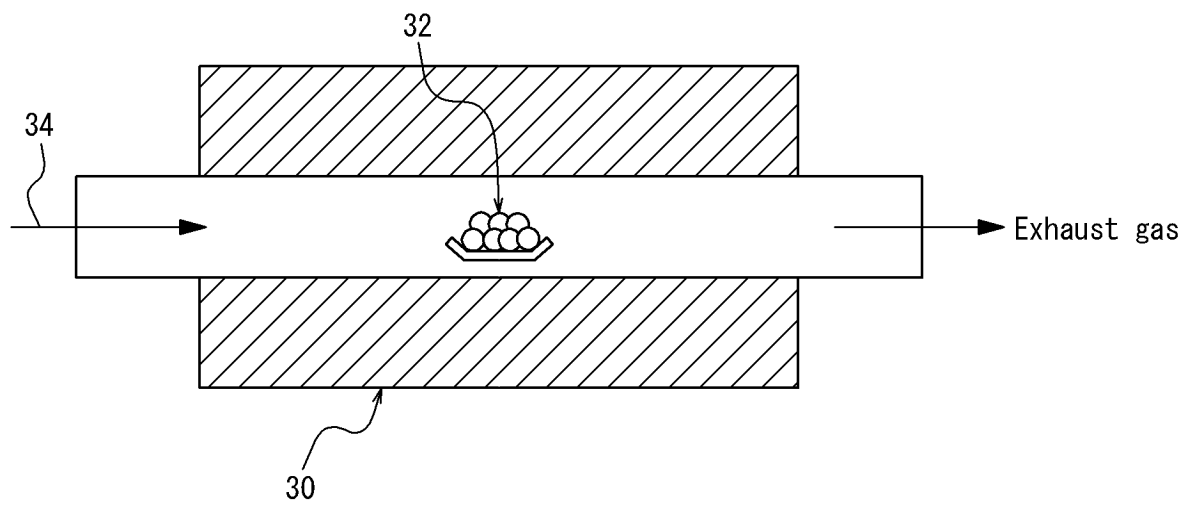
*FIG. 2B*



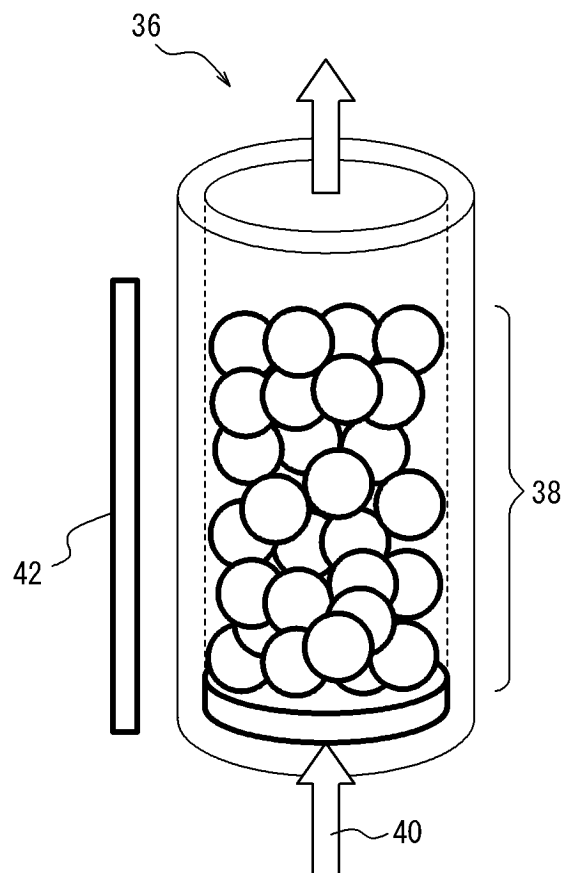
*FIG. 2A*



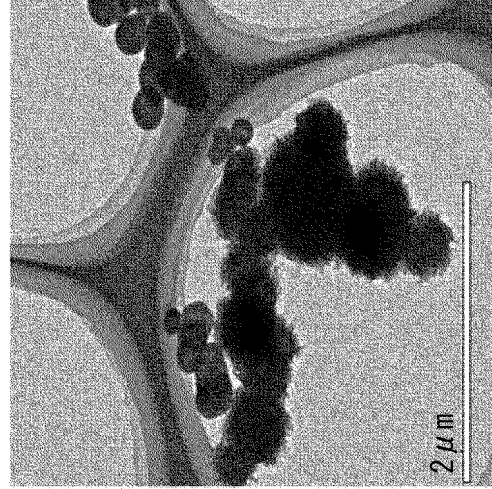
*FIG. 3A*



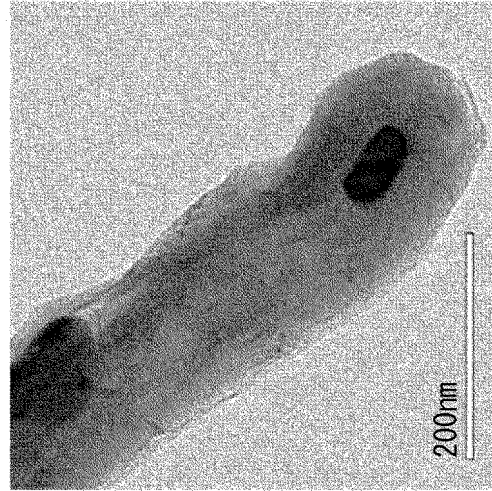
*FIG. 3B*



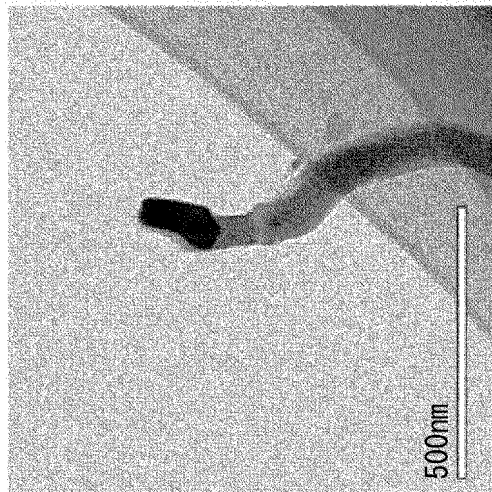
*FIG. 4C*



*FIG. 4B*



*FIG. 4A*



## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2023/025632

## A. CLASSIFICATION OF SUBJECT MATTER

*C22B 1/14*(2006.01)i

FI: C22B1/14

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)

C22B1/14

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

Published examined utility model applications of Japan 1922-1996

Published unexamined utility model applications of Japan 1971-2023

Registered utility model specifications of Japan 1996-2023

Published registered utility model applications of Japan 1994-2023

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages                                                                | Relevant to claim No. |
|-----------|---------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------|
| Y         | JP 56-123332 A (KOBE STEEL, LTD.) 28 September 1981 (1981-09-28)<br>claim (1), p. 2, upper left column, line 1 to p. 5, upper left column, line 8 | 1-6                   |
| Y         | JP 2021-165214 A (MITSUBISHI HEAVY INDUSTRIES, LTD.) 14 October 2021<br>(2021-10-14)<br>paragraphs [0002]-[0005], [0012]-[0049], fig. 3           | 1-6                   |

☐ Further documents are listed in the continuation of Box C.☒ See patent family annex.

|                                                                                                                                                                         |                                                                                                                                                                                                                                                  |
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Date of the actual completion of the international search

12 September 2023

Date of mailing of the international search report

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Name and mailing address of the ISA/JP

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Japan

Authorized officer

Telephone No.

INTERNATIONAL SEARCH REPORT  
Information on patent family members

International application No.  
**PCT/JP2023/025632**

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| Patent document<br>cited in search report | Publication date<br>(day/month/year) | Patent family member(s)                                                                                                              | Publication date<br>(day/month/year) |
|-------------------------------------------|--------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------|
| JP 56-123332 A                            | 28 September 1981                    | (Family: none)                                                                                                                       |                                      |
| JP 2021-165214 A                          | 14 October 2021                      | US 2023/0150820 A1<br>paragraphs [0002]-[0005],<br>[0014]-[0056], fig. 3<br>WO 2021/205935 A1<br>DE 112021002174 T<br>CN 115362127 A |                                      |

**REFERENCES CITED IN THE DESCRIPTION**

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**Non-patent literature cited in the description**

- **VOLODYMYR SHATOKHA.** Iron Ores and Iron Oxide Materials. IntechOpen, 11 July 2018, 41-59  
**[0003]**