MAGNETITE ORE

GANGUE

IRON OXIDE

A method for producing granular metallic iron by heating and reducing a raw material mixture which includes an iron oxide-containing material, a carbonaceous reductant and a Li$_2$O supplying material in a thermal reduction furnace, wherein the iron oxide-containing material includes a hematite-containing material, and the raw material mixture includes at least Fe, Ca, Mg, Si and Li as constituent elements in such a manner that a slag forms a by-product during heating and reduction contains CaO, MgO, SiO$_2$ and Li$_2$O. This method enables granular metallic iron to be produced at a high productivity even when a hematite-containing material is used as the iron oxide-containing material.
METHOD FOR PRODUCING GRANULAR METALLIC IRON

TECHNICAL FIELD

[0001] The present invention relates to a method for producing granular metallic iron, and more particularly, to a method for producing granular metallic iron by heating and directly reducing a raw material mixture including at least an iron oxide-containing material and a carbonaceous reductant in a thermal reduction furnace.

BACKGROUND ART

[0002] A blast furnace process has been mainly used as a process of producing iron from iron oxide-containing materials such as iron ore and iron oxide. On the other hand, a direct reduction iron-making process which does not use a blast furnace and instead directly reduces the iron oxide in a raw material mixture with a carbonaceous reductant has been developed and is attracting attention as an approach suited to high-mix, low-volume production on a relatively small scale (e.g., Patent Documents 1 to 4).

[0003] The direct reduction iron-making process produces granular metallic iron (reduced iron) by charging a raw material mixture including an iron oxide-containing material and a carbonaceous reductant (hereinafter, also referred to as "carbon material") such as coal or coke (alternatively, a simple compact of the mixture or a carbon material-containing compact of the mixture additionally being in the form of a pellet or briquette) onto a hearth of a movable hearth-type thermal reduction furnace (e.g., a rotary hearth furnace); applying heat or radiant heat from heating burners to the mixture in order to directly reduce iron oxide in the mixture by the carbonaceous reductant while the mixture moving in the furnace; carburizing and melting metallic iron (reduced iron) obtained by the above reduction; causing the metallic iron to coalesce into granules while separating the metallic iron from slag formed as a by-product; and then, cooling and solidifying the metallic iron.

[0004] The direct reduction iron-making process does not require a large-scale facility such as a blast furnace, and has been intensively studied in order to achieve practical use. However, to perform on an industrial scale, further improvements will need to be made in many areas, including operational stability, safety, cost-effectiveness, the quality of the granular metallic iron obtained as the product (e.g., the purity of the granular metallic iron), etc.

[0005] One of the challenges faced here is improving the productivity of granular metallic iron. This is because, even assuming direct reduction iron-making to be capable of saving space compared with existing iron-making processes such as the blast furnace process, if the productivity is poor, carrying out the direct reduction iron-making process on an industrial scale will not be feasible.

[0006] In order to increase the productivity of granular metallic iron in the direct reduction iron-making process, it is desired that the raw material mixture charged onto the hearth in a movable hearth-type thermal reduction furnace melt in a short time to increase the amount of granular metallic iron produced per unit time, and that the resulting granular metallic iron have a high ability of coalescing (in other words, that the yield of Fe which is obtained as granular metallic iron after coalescing be high with respect to the Fe content in the raw material mixture).

DISCLOSURE OF THE INVENTION


[0011] In conventional direct reduction iron-making processes, it is common to use iron ore as the iron oxide-containing material. Iron ore is known to be broadly divided into two types: hematite ore and magnetite ore. Hematite ore is an ore composed mainly of hematite (Fe₂O₃) having an oxidation number of 3. Magnetite ore is a mixture of wustite (FeO) having an oxidation number of 2 and hematite (Fe₂O₃) having an oxidation number of 3. In general, it is reported that hematite ore production accounts for about 80 to 90% of the total amount of iron ore produced, whereas magnetite ore production accounts for about 10 to 20%. Thus, as a rule, iron ore often contains hematite.

[0012] The inventors have found from their investigations that the productivity of granular metallic iron varies considerably depending on the type of iron oxide-containing material included in the raw material mixture. Specifically, they have found that when hematite ore alone is used as the iron oxide-containing material in the raw material mixture, the productivity is worse than in cases where magnetite ore alone is used. However, in conventional direct reduction iron-making processes, no attention is paid to the type of iron oxide-containing material. Even on referring to Patent Documents 1 to 4 above, nothing at all is described there concerning changes in productivity and the like depending on the type of iron oxide-containing material included in the raw material mixture.

[0013] Accordingly, there exists a need for technology which, in cases where the iron oxide-containing material includes the hematite ore that accounts for the preponderance of iron ore, is capable of producing granular metallic iron at a high productivity in the same way as when the iron oxide-containing material consists of magnetite ore.

[0014] In view of the above, an object of the invention is to provide a method for producing granular metallic iron at a high productivity when the iron oxide-containing material in the raw material mixture includes a hematite-containing material in the direct reduction iron-making process.

[0015] One aspect of the present invention is directed to a method for producing granular metallic iron, comprising: charging a raw material mixture including an iron oxide-containing material, a carbonaceous reductant and a Li₂O supplying material into a thermal reduction furnace; heating the raw material mixture and reducing iron oxide in the raw material mixture by the carbonaceous reductant to form metallic iron and to form slag as a by-product; causing the metallic iron to coalesce into granules while separating the metallic iron from the slag; and then cooling and solidifying the metallic iron, wherein the iron oxide-containing material includes a hematite-containing material, and the raw material mixture includes at least Fe, Ca, Mg, Si and Li as constituent elements in such a manner that the slag contains CaO, MgO, SiO₂ and Li₂O, the Li₂O content in the slag is 0.05% by mass or more, and the slag has a basicity [(CaO+MgO)/SiO₂] in a range of 1.5 to 1.9.
Another aspect of the present invention is directed to a method for producing granular metallic iron, comprising: charging a Li₂O supplying material and a raw material mixture including an iron oxide-containing material and a carbonaceous reductant into a thermal reduction furnace; heating the Li₂O supplying material and the raw material mixture and reducing iron oxide in the raw material mixture by the carbonaceous reductant to form metallic iron and to form slag as a by-product; causing the metallic iron to coalesce into granules while separating the metallic iron from the slag and then cooling and solidifying the metallic iron, wherein the iron oxide-containing material includes a hematite-containing material, and one of the raw material mixture and the Li₂O supplying material includes at least one element from among Fe, Ca, Mg, Si and Li as a constituent element, and the other one of the raw material mixture and the Li₂O supplying material includes at least the remaining elements as constituent elements in such a manner that the slag contains CaO, MgO, SiO₂ and Li₂O, the Li₂O content in the slag is 0.05% by mass or more, and the slag has a basicity [(CaO+MgO)/SiO₂] in a range of from 1.5 to 1.9.

The objects, features, aspects and advantages of the invention will become clearer through reference to the following detailed description and drawings.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a photograph in place of drawing which shows the morphology of the gangue included in magnetite ore.

FIG. 2 is a photograph in place of drawing which shows the morphology of the gangue included in hematite ore.

FIG. 3 is a schematic diagram showing an exemplary configuration of a rotary hearth-type thermal reduction furnace.

BEST MODE FOR CARRYING OUT THE INVENTION

The inventors have investigated the reasons for the difference in granular metallic iron productivity that arises between when magnetite ore alone is used as the iron oxide-containing material and when hematite ore alone is used. As a result, the inventors have found that magnetite ore and hematite ore differ in the morphology of the gangue included in the ore, and that this disparity affects the productivity.

The term ‘gangue’ refers to those components, of the components in ore that has been extracted from a mine (crude ore), other than the minerals containing the useful metal, and is typically composed of oxides such as Al₂O₃, SiO₂ and CaO.

The gangue within magnetite ore, as shown in FIG. 1, is present in morphology where the gangue is attached to the iron oxide. By contrast, the gangue included in hematite ore, as shown in FIG. 2, is present in a morphology surrounded by iron oxide or, conversely, in a morphology in which the gangue surrounds iron oxide. In cases where magnetite ore is used as the iron oxide-containing material, because the gangue does not surround iron oxide, when the ore is heated in a movable hearth furnace, the iron oxide portion is thought to be readily reduced. On the other hand, in cases where hematite ore is used as the iron oxide-containing material, because gangue sometimes surrounds the iron oxide, the gangue is thought to interfere with heat conduction, making it difficult for the iron oxide to rise in temperature, and also making it difficult for reduction of the iron oxide by reducing gases such as CO to proceed.

In addition, the inventors have found that because, of the gangue components, Al₂O₃ has an especially high melting point, when the Al₂O₃ included in the hematite-containing material (hematite ore) is 0.30% by mass or less with respect to the 100% by mass of the hematite-containing material, there is substantially no effect on the melting of the gangue, whereas at more than 0.30% by mass of Al₂O₃, the large proportion of Al₂O₃ in the gangue causes the gangue to melt with greater difficulty.

Accordingly, the inventors conducted further studies based on the notion that, were it possible for the gangue in hematite ore to be rapidly melted, the melting time for the raw material mixture charged into the thermal reduction furnace might be shortened, increasing productivity. As a result, the inventors have found that by deliberately including a Li₂O supplying material in the raw material mixture and setting the Li₂O content in the slag that forms as a by-product to 0.05% by mass or more, the melting point of the gangue included in the hematite ore can be lowered, enhancing the productivity of the granular metallic iron.

The melting of hematite ore is promoted by having a raw material mixture include a Li₂O supplying material, or by charging both a raw material mixture and a Li₂O supplying material into the thermal reduction furnace, in such a manner that the Li₂O content in the slag that forms as a by-product becomes 0.05% by mass or more. However, when the reduced iron obtained by reducing the melted iron oxide has a poor ability of coalescing, some of the reduced iron is taken up by the slag that forms as a by-product and ultimately discharged from the system as slag. As a result, the yield of granular metallic iron worsens, giving a poor productivity.

To address this problem, the inventors conducted additional investigations aimed at increasing the yield of the granular metallic iron and further improving the productivity without hindering the hematite ore melt-promoting effects owing to the inclusion of a Li₂O supplying material. As a result, the inventors have found that, if the raw material mixture includes at least Fe, Ca, Mg, Si and Li as constituent elements in such a way that the slag which forms as a by-product has a Li₂O content of 0.05% by mass or more and the slag has a basicity [(CaO+MgO)/SiO₂] in a range of from 1.5 to 1.9, that is, if the slag basicity [(CaO+MgO)/SiO₂] determined from the contents of CaO, MgO and SiO₂ in the slag is in a range of from 1.5 to 1.9 by having the Li₂O content in the slag that forms as a by-product be 0.05% by mass or more and also by adjusting the amounts of a CaO supplying material, a MgO supplying material and a SiO₂ supplying material included in the raw material mixture, the flow properties of the slag become better and the ability to separate the granular metallic iron from the slag improves, thus increasing the ability of the granular metallic iron to coalesce and enhancing the yield of granular metallic iron.

Accordingly, the invention has a feature that: (1) the Li₂O content in the slag is set to 0.05% by mass or more by blending a Li₂O supplying material into the raw material mixture or by charging a Li₂O supplying material to the thermal reduction furnace, and adjusting the blended amount or charged amount; and (2) the basicity [(CaO+MgO)/SiO₂] of the slag determined from the CaO, MgO and SiO₂ contents in the slag is set in a range of from 1.5 to 1.9 by adjusting the amounts of the Ca(+) supplying material, the MgO supplying...
material and the SiO₂ supplying material included in the raw material mixture or in other charging materials which are charged to the thermal reduction furnace. In this way, even when a hematite-containing material is used as the iron oxide-containing material, granular metallic iron can be produced at a good productivity.

In this invention, the Li₂O in the slag acts as a sort of melting point depressant for the hematite-containing material, and the CaO, MgO and SiO₂ in the slag having a basicity in the above-indicated range act as slag flow improvers in the presence of Li₂O. The essential features of the invention are explained in detail below.

[Hematite-Containing Material]

In the present invention, the iron oxide-containing material includes hematite-containing materials. As used herein, a “hematite-containing material” is an iron oxide source composed of hematite ore. Viewed microscopically, it is an iron oxide source containing iron oxide surrounded by gangue. Therefore, the iron oxide-containing material used may be: one composed solely of hematite ore (hematite-containing material); one composed of hematite ore and magnetite ore; or one composed of hematite ore, magnetite ore, and an iron oxide source other than these ores. For example, a material that is intermediate between hematite ore and magnetite ore may be used as the iron oxide-containing material.

No particular limitation is imposed on the content of the hematite-containing material with respect to the whole iron oxide-containing material. In conventional direct reduction iron-making processes, if even a little hematite ore is included in the raw material mixture, the malleability of the overall raw material mixture worsens, lowering the productivity. Moreover, as the proportion of the hematite ore in the raw material mixture rises, the productivity falls even further. By contrast, because the present invention is able to achieve a high productivity even when hematite ore is included in the raw material mixture, the higher the proportion of the raw material mixture accounted for by hematite ore, the more strikingly the effects of the invention are achieved. In this invention, the hematite-containing material is generally included in a ratio of preferably 50% by mass or more, and more preferably 80% by mass or more, with respect to the 100% by mass of iron oxide-containing material.

Of the gangue components included in the hematite-containing material with respect to the 100% by mass of hematite-containing material, Al₂O₃ may be included in a content of substantially 0.30% by mass or more. Because Al₂O₃ has an especially high melting point, in conventional direct reduction iron-making processes, when hematite ore containing 0.30% by mass or more of Al₂O₃ is used as the iron oxide-containing material, the gangue components do not readily melt, resulting in an increased melting time for the raw material mixture. By contrast, in the present invention, because Li₂O acts as a hematite-containing material melting point depressant, the melting time for the raw material mixture is relatively short, resulting in a relative increase in the production output of granular metallic iron per unit time. Hence, this is effective for improving the productivity in cases where a hematite ore containing a large amount of Al₂O₃ is used (e.g., in cases where the Al₂O₃ content is preferably 0.40% by mass or more, and more preferably 1.0% by mass or more with respect to the 100% by mass of hematite-containing material).

[Li₂O Content in Slag]

In the present invention, the Li₂O content in the slag that forms as a by-product when the raw material mixture is heated and reduced is set to 0.05% by mass or more. The Li₂O in the slag is effective as a hematite-containing material melting point depressant. By controlling the Li₂O content in the slag within the above range, the melting point of gangue in the hematite-containing material can be lowered sufficiently, as a result of which the solubility of the raw material mixture is increased, enabling the productivity of granular metallic iron to be improved. The content of Li₂O included in the slag that forms is preferably 0.1% by mass or more, and more preferably 0.3% by mass or more.

On the other hand, if the amount of a Li₂O supplying material included is made too large, vigorous evaporation of the Li₂O components will occur and the evaporated Li₂O will react with refractory materials within the furnace, accelerating damage. Hence, from the standpoint of extending the life of the refractory materials within the furnace, the upper limit in the Li₂O content within the slag that forms is preferably set to 12% by mass or less, more preferably 11% by mass or less, and much more preferably 10% by mass or less.

The Li₂O content in slag can be regulated by adjusting the amount of the Li₂O supplying material blended into the raw material mixture. Alternatively, regulation is possible by adjusting the amount of the Li₂O supplying material which is separately charged into the thermal reduction furnace. It is preferable to adjust the blending amount and charged amount of the Li₂O supplying material as appropriate for the gangue content, form and the like. Specifically, it is preferable to carry out adjustment while taking into consideration the proportion and type (especially the component composition), etc. of the hematite-containing material within the iron oxide-containing material.

The above Li₂O supplying material used in the present invention is a material which is charged, either alone or in admixture with other materials (e.g., the raw material mixture), into the thermal reduction furnace, and supplies Li₂O to the slag that forms as a by-product in the course of heating and reduction of the raw material mixture. The type of Li₂O supplying material is not subject to any particular limitation. Examples thereof include Li₂O and lithium carbonate (Li₂CO₃). The use of Li₂O is preferred because it has an especially large melting point depressant effect on the gangue in the iron oxide-containing material. Because Li₂O readily dissolves in water, the Li₂O dissolves in moisture within in the raw material mixture and becomes liquid, whereupon, presumably, the surface area of contact with the iron oxide-containing material increases, the reactivity rises, the melting point of gangue in the iron oxide-containing material is efficiently depressed, and the solubility is enhanced.

In the subsequently described working examples of the invention, experiments were carried out in which a Li₂O supplying material was blended into the raw material mixture, although the oxides of other metals which, like lithium, are alkali metals (e.g., Na₂O, K₂O) may be blended into the raw material mixture, either alone or in combination. This is because alkali metal oxides other than Li₂O also have a melting point depressing effect on gangue within the iron oxide-containing material and depress the melting point of the iron oxide-containing material in the same way as when Li₂O is blended. Therefore, a Na₂O supplying material (a material which is charged into the thermal reduction furnace either alone or in admixture with other materials, and supplies Na₂O to the slag that forms as a by-product in the course of heating and reduction of the raw material mixture) or a K₂O supplying material (a material which is charged into the thermal
reduction furnace either alone or in admixture with other materials, and supplies K$_2$O to the slag that forms as a by-product in the course of heating and reduction of the raw material mixture) may be used instead of the Li$_2$O supplying material. However, because the lithium oxide incurs reduction less easily than the sodium oxide and the potassium oxide, it does not readily become lithium metal and evaporate, and is thus practical. Therefore, of the alkali metal oxides, Li$_2$O is preferred.

[Slag Basicity: 1.5 to 1.9]

[0037] The slag basicity $[(\text{CaO}+\text{MgO})/\text{SiO}_2]$ is set in a range of from 1.5 to 1.9. The upper limit in the slag basicity is set to 1.9, primarily because considerations such as the productivity of reduced iron were taken into account. That is, from the standpoint solely of depressing the melting point of the slag, the slag basicity may be set higher than 1.9, thereby ultimately enabling the melting point of the final slag to be further depressed. However, if the slag basicity is made too high, the viscosity of the slag rises, worsening the flow properties of the slag, hindering agglomeration of the reduced iron, and making it difficult to obtain granular metallic iron having a suitable, nearly spherical shape. Moreover, the yield of the granular metallic iron will tend to decrease. The slag basicity is preferably adjusted to 1.85 or below.

[0038] The lower limit in the slag basicity is set to 1.5 because the flow properties of the slag worsen when the basicity falls below this value and, even if the reducing potential of the atmosphere can be maintained at a sufficiently high level, the reducing reactions fail to proceed, as a result of which reduced granular metallic iron is not obtained. The preferred basicity for working the invention is 1.6 or more; the more preferred basicity is 1.65 or more.

[MgO Content of Slag: 4 to 10% by Mass]

[0039] As described above, the present invention is a method for producing granular metallic iron by charging a raw material mixture including an iron oxide-containing material that includes a hematite-containing material, a carbonaceous reductant and a Li$_2$O supplying material into a thermal reduction furnace or by charging a Li$_2$O supplying material and a raw material mixture including an iron oxide-containing material that includes a hematite-containing material and a carbonaceous reductant into a thermal reduction furnace, and applying heat and reduction to produce granular metallic iron. The method has a feature that the raw material mixture includes at least Fe, Ca, Mg, Si and Li as constituent elements, or that one of the raw material mixture and the Li$_2$O supplying material includes at least one element from among Fe, Ca, Mg, Si and Li as a constituent element and the other one of the raw material mixture and the Li$_2$O supplying material includes at least the remaining elements as constituent elements in such a manner that the slag which forms as a by-product contains CaO, MgO, SiO$_2$ and Li$_2$O, the Li$_2$O content in the slag has a specific value or higher, and the slag has a basicity $[(\text{CaO}+\text{MgO})/\text{SiO}_2]$ in a specific range. In addition, it is preferable for the MgO content in the slag to be from 4 to 10% by mass.

[0040] As with the above-described Li$_2$O, the MgO in the slag also has a hematite-containing melting point depressing action. In addition to control of the Li$_2$O content, by also controlling the MgO content in the slag within the above-indicated range, the productivity of the reduced iron can be increased. When the MgO content in the slag is 4% by mass or more, at typical operating temperatures, crystallization of the complex oxide 2CaO·SiO$_2$ within the slag is suppressed, the flow properties of the slag increase, and the ability for the reduced iron to coalesce improves. Therefore, it is preferable to set the MgO content to 4% by mass or more. More preferably is to set the MgO content to 4.5% by mass or more, and much more preferably is to set the MgO content to 5% by mass or more.

[0041] On the other hand, when the MgO content in the slag is 10% by mass or less, MgO crystallization within the slag is suppressed, increasing the ability for the reduced iron to coalesce. As a result, the target large-grained granular metallic iron can be produced in a high yield. Therefore, the MgO content in slag is preferably set to 10% by mass or less. The upper limit in the MgO content is more preferably 9.5% by mass, and much more preferably 9.0% by mass.

[0042] The above-described slag basicity and MgO content in slag may be controlled by adjusting the amounts of the iron oxide-containing material and the carbonaceous reductant which make up the raw material mixture. The reason is that the iron oxide-containing material and the carbonaceous reductant themselves contain at least CaO, MgO and SiO$_2$. However, the gist of the present invention is not limited to a method for adjusting the proportions of the above-described raw material mixture; as is subsequently explained, the slag basicity and the MgO content in the slag may be controlled by additionally blending some “other MgO supplying material” into the raw material mixture (this is described later in detail). Because the iron ore included as the iron oxide-containing material and the coal or coke included as the carbonaceous reductant are natural products and the respective contents of CaO, MgO and SiO$_2$ vary according to the types used, it would be difficult to uniformly specify the amounts in which they are to be included. However, it is desirable to adjust these suitably while taking into account the component composition of the gangue present in the iron ore and the like included as the iron oxide-containing material and the component composition of the ash present in the coal, coke or the like included as the carbonaceous reductant.

[0043] For example, in cases where a carbonaceous powder is charged as a bed material, the slag basicity and the MgO content in the slag are controlled by adjusting the amounts of the iron oxide-containing material and the carbonaceous reductant while taking into account the constituents of the carbonaceous powder and the amounts thereof.

[0044] It should be noted that the most important features of the invention have to do with setting the Li$_2$O content in the slag that forms as a by-product and setting the basicity of the slag; the use of a carbonaceous powder which is charged onto the hearth as a bed material is not essential. However, charging a carbonaceous powder as a bed material onto the hearth is desirable because the reducing potential within the furnace can thereby be increased more efficiently and both an increase in the metallization and a decrease in the sulfur content can be more effectively achieved. To more reliably elicit such effects as a bed material, it is desirable for a granular carbonaceous powder to be applied onto the hearth to a thickness of about 2 mm or more. Moreover, by applying the carbonaceous powder as a bed material in the form of a layer having a certain degree of thickness, this bed layer serves as a buffer between the raw material mixture and the hearth refractory, or serves as a material for protecting the hearth refractory from the slag.
that forms as a by-product, and is thus useful as well for extending the life of the hearth refractory.

However, when the bed layer is too thick, problems may arise, such as the infiltration of raw material mixture into the bed layer on top of the hearth, which may keep the reduction reactions from proceeding. Hence, it is preferable to hold the layer thickness to about 7.5 mm or less.

The type of carbonaceous powder used as the bed material is not subject to any particular limitation. For example, use may be made of ordinary coal or coke which has been crushed and preferably adjusted to a suitable particle size. Alternatively, when coal is used, anthracite, which has a low fluidity and does not exhibit swelling on or sticking to the hearth, is preferred.

As used in the present invention, SiO$_2$ supplying material refers to a material which is charged into the thermal reduction furnace, either alone or in a mixed state with other materials (e.g., the raw material mixture), and supplies SiO$_2$ to the slag that forms as a by-product in the course of heating and reduction of the raw material mixture. No particular limitation is imposed on the type of SiO$_2$ supplying material. Examples thereof include the hematite ore or coal included in the raw material mixture.

As used in the present invention, MgO supplying material refers to a material which is charged into the thermal reduction furnace, either alone or in a mixed state with other materials (e.g., the raw material mixture), and supplies MgO to the slag that forms as a by-product in the course of heating and reduction of the raw material mixture. No particular limitation is imposed on the type of MgO supplying material. Examples thereof include the hematite ore or coal included in the raw material mixture (also referred to as “MgO supplying materials included in the raw material mixture” in this specification). Other examples thereof include calcined lime (CaO) and calcium carbonate (CaCO$_3$) (also referred to as “other CaO supplying materials” in this specification). The “other CaO supplying materials” may be included in the raw material mixture. In such cases, the basicity of the slag formed as a by-product and the CaO content within the slag are controlled by adjusting the amounts of the oxide-containing materials and carbonaceous reductant while taking into consideration, in addition to the raw material mixture, the component composition and included amount of such “other CaO supplying materials.” That is, in addition to iron oxide-containing materials and carbonaceous reductants which are “MgO supplying materials included in the raw material mixture” and are also “CaO supplying materials included in the raw material mixture,” the raw material mixture may include a “MgO and CaO supplying material,” such as dolomite ore. A “MgO and CaO supplying material” is a material which is charged into the thermal reduction furnace, either alone or in a mixed state with other materials (e.g., the raw material mixture), and supplies MgO and CaO to the slag that forms as a by-product in the course of heating and reduction of the raw material mixture.

The method of adding other MgO supplying materials, other CaO supplying materials, MgO and CaO supplying materials and Li$_2$O supplying materials is not subject to any particular limitation. Suitable use may be made of, for example: a method in which these materials are blended into the raw material mixture; a method in which these materials are pre-charged onto the rotary hearth, either together with, or independent of, the bed material; or a method in which these materials are charged at the same time as charging of the raw material mixture or are separately charged therefrom after above, in such a way that the slag that forms as a by-product contains CaO, MgO, SiO$_2$, and Li$_2$O, the Li$_2$O content in the slag is 0.05% by mass or more, and the slag has a basicity $[(\text{CaO+MgO})/\text{SiO}_2]$ in a range of from 1.5 to 1.9.

As used in the present invention, “CaO supplying materials” refers to a material which is charged into the thermal reduction furnace, either alone or in a mixed state with other materials (e.g., the raw material mixture), and supplies CaO to the slag that forms as a by-product in the course of heating and reduction of the raw material mixture. No particular limitation is imposed on the type of CaO supplying material. Examples thereof include the hematite ore or coal included in the raw material mixture (also referred to as “CaO supplying materials included in the raw material mixture” in this specification).
In this invention, when operation is carried out as described above using a full-scale movable hearth-type thermal reduction furnace, by raising the final basicity of the slag to about 1.9 using a MgO supplying material as the basicity regulator, the slag can be fully melted in a temperature range up to 1450°C under actual operation. This enables granular metallic iron to be produced under stable operating conditions. As a result, though there will be some degree of difference depending on the grade of coal or the like included in the carbonaceous reductant or the bed material, granular metallic iron can be produced in a short time.

Next, brief descriptions are provided of the movable hearth-type thermal reduction furnace which may be used in the invention and of the method of producing granular metallic iron using such a furnace. In addition, a mechanism capable of producing granular metallic iron is described in detail in conjunction with FIG. 3. The following diagrams are not intended to limit the invention, and may be carried out with suitable modifications without departing from the scope of the invention as described above and below, all such modifications falling within the technical scope of the invention.

FIG. 3 is a schematic view showing an exemplary configuration of a rotary hearth thermal reduction furnace, which is a type of movable hearth-type thermal reduction furnace. In order to show the internal structure of the furnace, a portion of the furnace has been cut away in the diagram to show the interior.

A raw material mixture 1 which includes an iron oxide-containing material, a carbonaceous reductant and a Li₂O supplying material passes through a material feed hopper 3 and is continuously charged onto a rotary hearth 4 of a rotary hearth-type thermal reduction furnace F. The iron oxide-containing material includes a hematite-containing material, and coal, coke or the like is typically used as the carbonaceous reductant.

No particular limitation is imposed on the form of the raw material mixture 1 when it is supplied. Generally, simple compacts obtained by compressing a raw material mixture which includes, for example, an iron oxide-containing material, a carbonaceous reductant and a Li₂O supplying material, or carbon material-containing compacts obtained by forming such a raw material mixture into pellets or briquettes, are supplied. Alternatively, a material obtained by suitably mixing an iron oxide-containing material, a carbonaceous reductant and a Li₂O supplying material may be supplied. Or a granular carbonaceous powder 2 may be supplied together with simple compacts or carbon material-containing compacts.

In the present invention, as described above, aside from the raw material mixture 1 which includes an iron oxide-containing material and a carbonaceous reductant, the thermal reduction furnace F may also be charged with a Li₂O supplying material. Where necessary, substances other than the raw material mixture (such as a MgO supplying material or a CaO supplying material) may be charged. From the standpoint of protecting the environment, it is preferable not to charge a fluorine supplying material (e.g., fluorite).

Next, the procedure when charging the above raw material mixture 1 into the thermal reduction furnace F is described. Before charging the raw material mixture 1, it is desirable to charge a granular carbonaceous powder 2 from the material feed hopper 3 onto the rotary hearth 4 and spread it over the entire hearth 4 as a bed, after which the raw material mixture 1 is charged onto this bed.

FIG. 3 shows an example in which a single material feed hopper 3 is used both to charge the carbonaceous powder 2 and to charge the raw material mixture 1. However, it is of course possible to use two or more hoppers and charge the carbonaceous powder 2 and the raw material mixture 1 separately. Although the carbonaceous powder 2 charged as the bed increases the reducing efficiency and also is highly effective for promoting low sulfuration of the granular metallic iron obtained by heating and reduction, the charging of this powder may be omitted. The type of carbonaceous powder charged as the bed is not subject to any particular limitation. Illustrative examples include coal and coke. It is desirable for the carbonaceous powder charged as a bed to be a material having a low sulfur content than the carbonaceous reductant included in the raw material mixture.

The rotary hearth 4 of the thermal reduction furnace F shown in FIG. 3 rotates in a counterclockwise direction. The speed of rotation varies according to the size of the furnace F and the operating conditions, but typically is a speed of one revolution per about 8 to 16 minutes. A plurality of heating burners 5 are provided on the sidewalls of the furnace body 6 in the thermal reduction furnace F, and heat is supplied to the hearth by the heat of combustion from these heating burners 5 or by radiant heat therefrom. The heating burners 5 may be provided on the roof of the furnace.

The raw material mixture 1 charged onto the rotary hearth 4 made of a refractory material is heated by the heat of combustion and radiant heat from the heating burners 5 as it moves circumferentially through the thermal reduction furnace F on the rotary hearth 4. Next, as it passes through the heating zone within the thermal reduction furnace F, the iron oxide within the raw material mixture 1 is reduced. Next, while separating from the molten slag that forms as a by-product, the reduced iron incurs carburization by the remaining carbonaceous reductant, melts, and granularly coalesces, becoming granular metallic iron 10. The granular metallic iron 10 is then cooled and solidified in a zone on the downstream side of the rotary hearth 4, following which it is successively discharged from the top of the hearth by a discharging device 6 such as a screw. The slag that has formed as a by-product is also discharged at this time. After these have passed through a hopper 9, separation of the metallic iron and the slag is carried out by any suitable separating means (e.g., a screen mesh or magnetic separating device). An exhaust gas duct 7 is also shown in FIG. 3.

As explained above, in the present invention, by heating and reducing, in a movable hearth-type thermal reduction furnace, a raw material mixture which includes an iron oxide-containing material, a carbonaceous reductant and a Li₂O supplying material, and having the raw material mixture include at least Fe, Ca, Mg, Si and Li as constituent elements in such a manner that the Li₂O content in the slag which forms as a by-product at that time is a specific value or higher and the slag has a basicity [(CaO+MgO)/SiO₂] in a specific range; or by heating and reducing, in a movable hearth-type thermal reduction furnace, a Li₂O supplying material and a raw material mixture which includes an iron oxide-containing material and a carbonaceous reductant, and having one of the raw material mixture and the Li₂O supplying material include at least one element from among Fe, Ca, Mg, Si and Li as constituent element, and having the other of the raw material mixture and the Li₂O supplying material include at least the remaining elements as constituent elements in such a manner that the Li₂O content in the slag which
forms as a by-product at that time is a specific value or higher and the slag has a basicity \([\frac{\text{CaO} + \text{MgO}}{\text{SiO}_2}]\) in a specific range, it is possible to produce granular metallic iron at a high productivity even when a hematite-containing material is used as the iron oxide-containing material.

**EXAMPLES**

[0065] The invention is described in greater detail below by way of examples. The following examples are not intended to limit the invention, and may be practiced with suitable modifications without departing from the scope of the invention as described above and below, all such modifications falling within the technical scope of the invention. The results shown in the following examples were obtained by carrying out tests using a small, experimental thermal reduction furnace.

[0066] Using the two types of hematite ore A and B and the one type of maghemite C shown in Table 1 as the iron oxide-containing material, and using coal as the carbonaceous reductant, these materials were mixed to give a mixture M. Table 1 shows the component compositions of the hematite ore and the maghemite ore, and Table 2 shows the component compositions of the coal (of the analysis value, “other” refers to solid carbonaceous components).

[0067] Aside from an iron oxide-containing material and a carbonaceous reductant, a binder (wheat flour) was also blended into the above mixture M. Moreover, slag basicity-adjusting auxiliary materials such as calcium carbonate (CaCO3) as a CaO supplying material and dolomite ore (composed primarily of CaCO3, MgCO3) as a MgO and CaO supplying material, and also lithium carbonate (Li2CO3) or Li2O as a Li2O supplying material, were additionally blended to give a raw material mixture (sometimes referred to below as a “blend”). Table 3 shows the component composition of the blend.

[0068] Each of the resulting blend was formed into raw-material compacts in the form of pellets. The resulting raw-material compacts were charged into a small experimental thermal reduction furnace, and subjected to heating and reduction. Coal having the component composition shown in Table 2 (carbonaceous powder) was placed on the hearth as a bed material to a thickness of about 5 mm. The temperature within the furnace was adjusted to 1450°C.

[0069] The iron oxide portion within the raw-material compacts charged onto the hearth of the thermal reduction furnace was reduced while maintaining a solid state during heating within the furnace for a period of from 10 to about 16 minutes, and the reduced iron that formed underwent melting point depression and mutually coalesced while incurring carburization by the carbonaceous powder remaining after reduction. The slag that formed as a by-product at this time also partially or substantially completely melted and mutually coalesced, resulting in separation into granular metallic iron in a molten state and molten slag. The granular metallic iron in the molten state and the molten slag subsequently cooled to temperatures at or below their melting points (specifically, cooled to about 1100°C) and solidified, and were discharged from the furnace as granular metallic iron or slag in a solid state.

[0070] At this time, after the raw-material compacts were charged into the experimental thermal reduction furnace, the situation within the furnace was visually observed and the length of time it took for all of the compacts discernible within the field of view to dissolve was measured. The measured melting completion times are shown in Table 5 below.

Letting the melting completion time in No. 1 be the norm when hematite ore A was used, and letting the melting completion time in No. 6 be the norm when hematite ore B was used, the melting completion time was rated as “Good” when it was shorter than the melting completion time serving as the corresponding norm and was rated as “NG” when it was longer.

[0071] The component compositions of the granular metallic iron that was obtained and the slag that formed as a by-product are shown in Table 4 below.

[0072] The basicity \([\frac{\text{CaO} + \text{MgO}}{\text{SiO}_2}]\) of the slag was calculated from the contents of CaO, MgO and SiO2 present in the slag. The calculated results are shown in Table 5 below. The yield (Fe(1)/Fe(0)) was calculated from the ratio of the mass of iron obtained as coalesced granular metallic iron (Fe(1)) to the mass of iron determined from blending calculations (Fe(0)).

The calculated results are shown in Table 5 below. In addition, the distribution ratio of sulfur \([\frac{S}{S^2}]\) was calculated from the content of sulfur in the granular metallic iron \([S]\) to the content of sulfur present in the slag \([S^2]\). These calculated results also are shown in Table 5 below.

[0073] Of Nos. 1 to 11, Nos. 1 to 5 are examples in which the hematite ore A in Table 1 was used, Nos. 6 to 10 are examples in which the hematite ore B in Table 1 was used, and No. 11 is a reference example in which the maghemite ore C in Table 1 was used. Nos. 1, 3 and 6 are examples in which a Li2O supplying material was not blended into the raw material mixture. The following observations can be made from Tables 4 and 5 below.

[0074] In No. 3, because the basicity of the slag that forms as a by-product is less than 1.5, the basicity being in particular about 1.4, the slag melting point decreases; hence, even without blending Li2O supplying material into the raw material mixture, the melting completion time is shorter than No. 1. However, it is apparent that, because the slag basicity is less than 1.5, the distribution ratio of sulfur is small and the yield has declined.

[0075] In Nos. 2, 4 and 5, the Li2O content in the slag is 0.05% by mass or more and the slag basicity \([\frac{\text{CaO} + \text{MgO}}{\text{SiO}_2}]\) is in a range of from 1.5 to 1.9. While maintaining a yield comparable to that of No. 1, the melting completion time is shorter than in No. 1. Moreover, it is apparent that, as the Li2O content in the slag becomes higher, the distribution ratio of sulfur tends to become higher.

[0076] In Nos. 7 to 10, the Li2O content in the slag is 0.05% by mass or more and the slag basicity \([\frac{\text{CaO} + \text{MgO}}{\text{SiO}_2}]\) is in a range of from 1.5 to 1.9. While maintaining a yield comparable to that of No. 6, the melting completion time is shorter than in No. 6. Moreover, it is apparent that, as the Li2O content in the slag becomes higher, the distribution ratio of sulfur tends to become higher and the melting completion time tends to become shorter.

[0077] As can be seen from a comparison of No. 11 with Nos. 1 and 6, when maghemite ore is used (No. 11), it is apparent that even without blending a Li2O supplying material in the raw material mixture, the melting completion time becomes shorter than when hematite ore is used (Nos. 1 and 6).

[0078] In these examples, the slag basicity and the amount of MgO in the slag are adjusted by including dolomite ore as a MgO and CaO supplying material. In the case of the blend compositions in these examples, by controlling the dolomite ore within the blend in a range of generally from 0 to 3.2% by


mass, the slag can be controlled within the basicity and MgO content ranges prescribed in the invention.

As discussed above, in the present invention, the Li₂O content of the slag is set to 0.05% by mass or more by blending a Li₂O supplying material in the mixture M, and the slag basicity \( [(CaO+MgO)/SiO₂] \) determined from the CaO, MgO and SiO₂ contents within the slag is set in a range of from 1.5 to 1.9 by adjusting the amounts of a CaO supplying material, a MgO supplying material and a SiO₂ supplying material included in the raw material mixture, thereby making it possible to shorten the melting completion time and improve productivity without lowering yield.

### TABLE 1

<table>
<thead>
<tr>
<th>Component composition of ore (% by mass)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Type</td>
</tr>
<tr>
<td>A</td>
</tr>
<tr>
<td>B</td>
</tr>
<tr>
<td>C</td>
</tr>
</tbody>
</table>

### TABLE 2

<table>
<thead>
<tr>
<th>Analysis value (% by mass)</th>
<th>Component composition of total ash (% by mass)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volatiles</td>
<td>Total ash</td>
</tr>
<tr>
<td>15.8</td>
<td>4.3</td>
</tr>
</tbody>
</table>

### TABLE 3

<table>
<thead>
<tr>
<th>Composition of blend (% by mass)</th>
</tr>
</thead>
<tbody>
<tr>
<td>No.</td>
</tr>
<tr>
<td>1</td>
</tr>
<tr>
<td>2</td>
</tr>
<tr>
<td>3</td>
</tr>
<tr>
<td>4</td>
</tr>
<tr>
<td>5</td>
</tr>
<tr>
<td>6</td>
</tr>
<tr>
<td>7</td>
</tr>
<tr>
<td>8</td>
</tr>
<tr>
<td>9</td>
</tr>
<tr>
<td>10</td>
</tr>
<tr>
<td>11</td>
</tr>
</tbody>
</table>

### TABLE 4

<table>
<thead>
<tr>
<th>Component composition of granular metallic iron (% by mass)</th>
<th>Component composition of slag (% by mass)</th>
</tr>
</thead>
<tbody>
<tr>
<td>No.</td>
<td>C</td>
</tr>
<tr>
<td>1</td>
<td>2.26</td>
</tr>
<tr>
<td>2</td>
<td>2.53</td>
</tr>
<tr>
<td>3</td>
<td>2.15</td>
</tr>
<tr>
<td>4</td>
<td>2.72</td>
</tr>
<tr>
<td>5</td>
<td>2.26</td>
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<tr>
<td>6</td>
<td>2.50</td>
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<tr>
<td>7</td>
<td>3.78</td>
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<tr>
<td>8</td>
<td>2.98</td>
</tr>
<tr>
<td>9</td>
<td>2.83</td>
</tr>
<tr>
<td>10</td>
<td>3.34</td>
</tr>
<tr>
<td>11</td>
<td>2.36</td>
</tr>
</tbody>
</table>
TABLE 5

<table>
<thead>
<tr>
<th>Ore No.</th>
<th>Type</th>
<th>Melting completion time (min)</th>
<th>Basicity (CaO + MgO)/(SiO₂)</th>
<th>Yield (%)</th>
<th>Distribution ratio of sulfur (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>A</td>
<td>14.3</td>
<td>1.77</td>
<td>95.3</td>
<td>4.3</td>
</tr>
<tr>
<td>2</td>
<td>A</td>
<td>13.0</td>
<td>1.74</td>
<td>95.4</td>
<td>4.6</td>
</tr>
<tr>
<td>3</td>
<td>A</td>
<td>12.0</td>
<td>1.41</td>
<td>85.0</td>
<td>3.4</td>
</tr>
<tr>
<td>4</td>
<td>A</td>
<td>12.0</td>
<td>1.52</td>
<td>88.1</td>
<td>3.5</td>
</tr>
<tr>
<td>5</td>
<td>A</td>
<td>11.2</td>
<td>1.65</td>
<td>94.8</td>
<td>4.8</td>
</tr>
<tr>
<td>6</td>
<td>B</td>
<td>12.3</td>
<td>1.66</td>
<td>99.6</td>
<td>7.9</td>
</tr>
<tr>
<td>7</td>
<td>B</td>
<td>10.8</td>
<td>1.84</td>
<td>102.4</td>
<td>58.0</td>
</tr>
<tr>
<td>8</td>
<td>B</td>
<td>11.5</td>
<td>1.70</td>
<td>100.9</td>
<td>17.5</td>
</tr>
<tr>
<td>9</td>
<td>B</td>
<td>11.5</td>
<td>1.74</td>
<td>100.1</td>
<td>16.2</td>
</tr>
<tr>
<td>10</td>
<td>B</td>
<td>11.0</td>
<td>1.72</td>
<td>97.0</td>
<td>33.6</td>
</tr>
<tr>
<td>11</td>
<td>C</td>
<td>10.9</td>
<td>1.71</td>
<td>103.1</td>
<td>4.1</td>
</tr>
</tbody>
</table>

[0080] As described in detail above, one aspect of the present invention is directed to a method for producing granular metallic iron, comprising: charging a raw material mixture including an iron oxide-containing material, a carbonaceous reductant and a Li₂O supplying material into a thermal reduction furnace; heating the raw material mixture and reducing iron oxide in the raw material mixture by the carbonaceous reductant to form metallic iron and to form slag as a byproduct; causing the metallic iron to coalesce into granules while separating the metallic iron from the slag; and then cooling and solidifying the metallic iron, wherein the iron oxide-containing material includes a hematite-containing material, and the raw material mixture includes at least Fe, Ca, Mg, Si and Li as constituent elements in such a manner that the slag contains CaO, MgO, SiO₂ and Li₂O, the Li₂O content in the slag is 0.05% by mass or more, and the slag has a basicity [(CaO+MgO)/SiO₂] in a range of from 1.5 to 1.9.

[0081] Another aspect of the present invention is directed to a method for producing granular metallic iron, comprising: charging a Li₂O supplying material and a raw material mixture including an iron oxide-containing material and a carbonaceous reductant into a thermal reduction furnace; heating the Li₂O supplying material and the raw material mixture containing iron oxide in the raw material mixture by the carbonaceous reductant to form metallic iron and to form slag as a byproduct; causing the metallic iron to coalesce into granules while separating the metallic iron from the slag; and then cooling and solidifying the metallic iron, wherein the iron oxide-containing material includes a hematite-containing material, and one of the raw material mixture and the Li₂O supplying material includes at least the remaining elements as constituent elements in such a manner that the slag contains CaO, MgO, SiO₂ and Li₂O, the Li₂O content in the slag is 0.05% by mass or more, and the slag has a basicity [(CaO+MgO)/SiO₂] in a range of from 1.5 to 1.9.

[0082] In this invention, because Li₂O in a content of 0.05% by mass or more depresses the melting point of the hematite-containing material, shortening the melting time of the raw material mixture, and because slag having a basicity [(CaO+MgO)/SiO₂] of from 1.5 to 1.9 and high flow properties increases the ability of the reduced iron to coalesce, thus improving the yield of granular metallic iron, even in cases where the iron oxide-containing material includes a hematite-containing material, granular metallic iron can be produced at a high productivity.

[0083] In these methods, the hematite-containing material used may be a material which includes 0.30% by mass or more of Al₂O₃ with respect to the 100% by mass of hematite-containing material. Even in cases where hematite ore containing 0.30% by mass or more of Al₂O₃, which has an especially high melting point compared with other gangue components, is used, Li₂O lowers the melting point of the gangue within the hematite ore, making the melting completion time of the raw material mixture shorter than in conventional direct reduction iron-making processes. Hence, this invention is able to maintain a high productivity.

[0084] In these methods, it is preferable for the MgO content in the slag to be from 4 to 10% by mass. If the MgO content in the slag is within this range, the MgO lowers the melting point of the hematite-containing material, and also is able to increase the ability of the reduced iron to coalesce while maintaining the high flow properties of the slag.

[0085] Use may be made of at least one compound selected from among Li₂O and Li₂CO₃ as the above Li₂O supplying material.

[0086] In these methods, the above raw material mixture may additionally include dolomite ore, or may additionally include at least one compound selected from among CaO and CaCO₃, and may additionally include at least one compound selected from among MgO and MgCO₃. Alternatively, in these methods, dolomite ore may be charged into the thermal reduction furnace, at least one compound selected from CaO and CaCO₃ may be charged into the thermal reduction furnace, and at least one compound selected from among MgO and MgCO₃ may be charged into the thermal reduction furnace. By having the raw material mixture include at least one material from among dolomite ore, CaO, CaCO₃, MgO and MgCO₃, by charging the raw material mixture and at least one material from among dolomite ore, CaO, CaCO₃, MgO and MgCO₃ into the thermal reduction furnace, in such a manner that the slag basicity [(CaO+MgO)/SiO₂] falls in a range of from 1.5 to 1.9, it is possible to respond to fluctuations in the CaO content and MgO content due to type of hematite-containing material used as the iron oxide-containing material and the type of coal or coke used as the carbonaceous reductant and produce granular metallic iron at a high productivity.

[0087] Moreover, the slag that forms as a by-product in the methods of the invention has a Li₂O content of 0.05% by mass or more, and the slag has a basicity [(CaO+MgO)/SiO₂], as determined from the CaO, MgO and SiO₂ contents of the slag, of from 1.5 to 1.9.

INDUSTRIAL APPLICABILITY

[0088] It is possible to produce granular metallic iron at a high productivity even when the iron oxide-containing material includes hematite-containing material by the method for producing granular metallic iron of the present invention.

1. A method for producing granular metallic iron, comprising:
   charging a raw material mixture comprising an iron oxide-containing material, a carbonaceous reductant, and a Li₂O supplying material, into a thermal reduction furnace;
   heating the raw material mixture and reducing iron oxide in the raw material mixture with the carbonaceous reductant to form metallic iron and to form slag as a byproduct;
causing the metallic iron to coalesce into granules while separating the metallic iron from the slag; and then cooling and solidifying the metallic iron,

wherein the iron oxide-comprising material comprises a hematite-comprising material,

and the raw material mixture comprises at least Fe, Ca, Mg, Si, and Li, as constituent elements in such a manner that the slag comprises CaO, MgO, SiO₂, and Li₂O.

Li₂O content in the slag is 0.05% by mass or more, and the slag has a basicity, (CaO+MgO)/SiO₂, in a range of from 1.5 to 1.9.

2. A method for producing granular metallic iron, comprising:

charging a Li₂O supplying material and a raw material mixture comprising an iron oxide-comprising material and a carbonaceous reductant into a thermal reduction furnace;

heating the Li₂O supplying material and the raw material mixture and reducing iron oxide in the raw material mixture with the carbonaceous reductant, to form metallic iron and to form slag as a by-product;

causing the metallic iron to coalesce into granules while separating the metallic iron from the slag; and then cooling and solidifying the metallic iron,

wherein

the iron oxide-comprising material comprises a hematite-comprising material, and

one of the raw material mixture and the Li₂O supplying material comprises at least one element iron among Fe, Ca, Mg, Si, and Li, as a constituent element, and the other one of the raw material mixture and the Li₂O supplying material comprises at least the remaining elements as constituent elements in such a manner that the slag comprises CaO, MgO, SiO₂, and Li₂O.

Li₂O content in the slag is 0.05% by mass or more, and the slag has a basicity, (CaO+MgO)/SiO₂, in a range of from 1.5 to 1.9.

3. The method according to claim 1, wherein the hematite-comprising material comprises 0.30% by mass or more of Al₂O₃ with respect to the 100% by mass of hematite-comprising material.

4. The method according to claim 1, wherein MgO content in the slag is from 4 to 10% by mass.

5. The method according to claim 1, wherein the Li₂O supplying material is at least one compound selected from the group consisting of Li₂O and Li₂CO₃.

6. The method according to claim 1, wherein the raw material mixture further comprises dolomite ore.

7. The method according to claim 1, wherein the raw material mixture further comprises at least one compound selected from the group consisting of CaO and CaCO₃.

8. The method according to claim 1, wherein the raw material mixture further comprises at least one compound selected from the group consisting of MgO and MgCO₃.

9. The method according to claim 2, wherein the hematite-comprising material comprises 0.30% by mass or more of Al₂O₃ with respect to the 100% by mass of hematite-comprising material.

10. The method according to claim 2, wherein MgO content in the slag is from 4 to 10% by mass.

11. The method according to claim 3, wherein MgO content in the slag is from 4 to 10% by mass.

12. The method according to claim 9, wherein MgO content in the slag is from 4 to 10% by mass.

13. The method according to claim 2, wherein the Li₂O supplying material is at least one compound selected from the group consisting of Li₂O and Li₂CO₃.

14. The method according to claim 3, wherein the Li₂O supplying material is at least one compound selected from the group consisting of Li₂O and Li₂CO₃.

15. The method according to claim 9, wherein the Li₂O supplying material is at least one compound selected from the group consisting of Li₂O and Li₂CO₃.

16. The method according to claim 2, wherein the raw material mixture further comprises dolomite ore.

17. The method according to claim 3, wherein the raw material mixture further comprises dolomite ore.

18. The method according to claim 9, wherein the raw material mixture further comprises dolomite ore.

19. The method according to claim 2, wherein the raw material mixture further comprises at least one compound selected from the group consisting of CaO and CaCO₃.

20. The method according to claim 2, wherein the raw material mixture further comprises at least one compound selected from the group consisting of MgO and MgCO₃.