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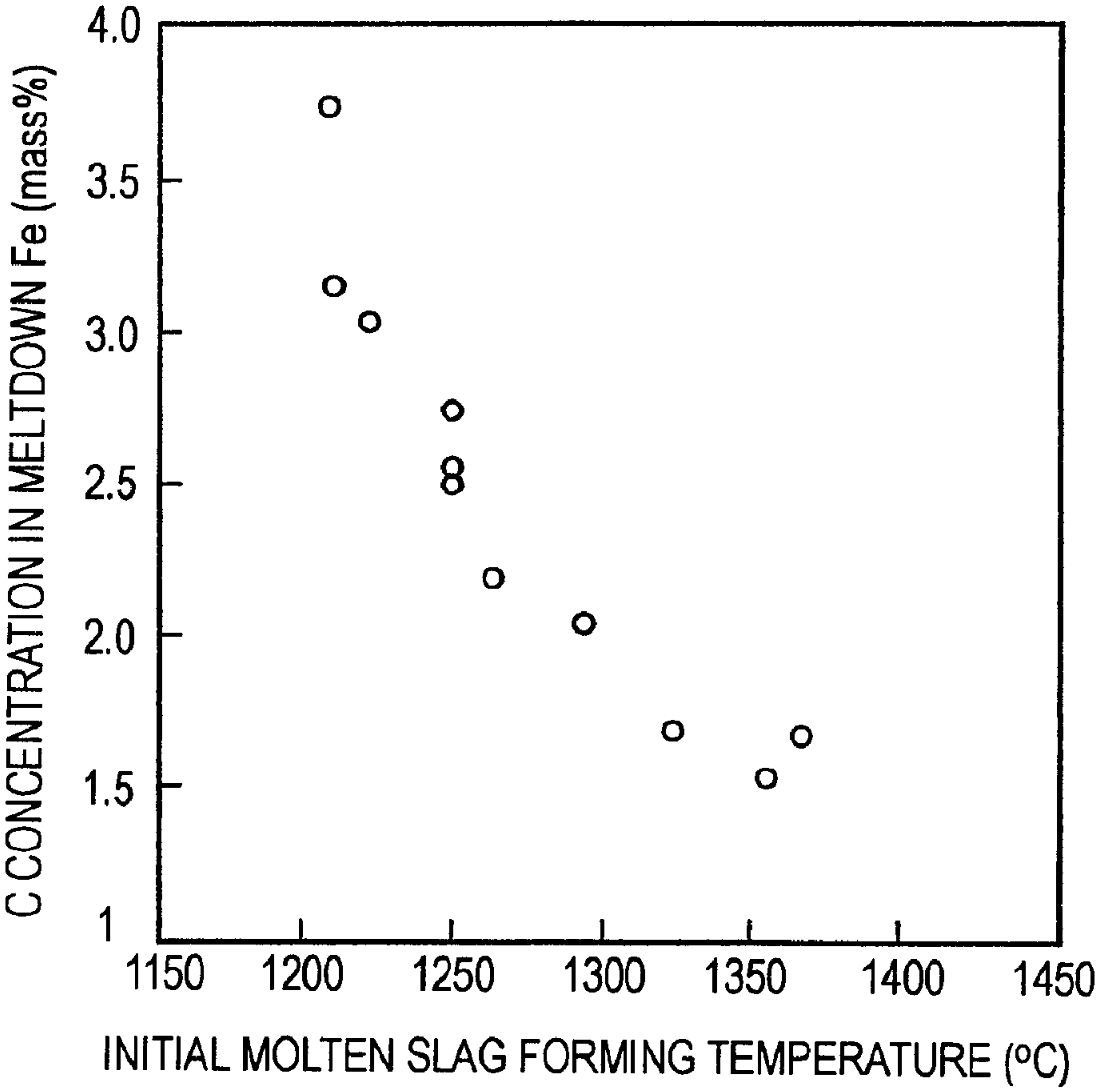

(57) Abrégé/Abstract:

A method for manufacturing metallic iron from a raw material mixture containing a carbonaceous reducing agent and an iron oxide-containing material, has a step of determining a target temperature of initial molten slag formation corresponding to a

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predetermined target carbon concentration in the metallic iron, the initial molten slag containing a gangue component, an unreduced iron oxide, and an ash component of the carbonaceous reducing agent, and being first produced in the raw material mixture by heating thereof; a step of preparing the raw material mixture producing a composition of the initial molten slag corresponding to the target temperature; and a step of heating the raw material mixture to reduce and melt the raw material mixture and to produce the initial molten slag. By this manufacturing method, metallic iron having a target carbon concentration can be efficiently manufactured.

ABSTRACT

A method for manufacturing metallic iron from a raw material mixture containing a carbonaceous reducing agent and an iron oxide-containing material, has a step of determining a target temperature of initial molten slag formation corresponding to a predetermined target carbon concentration in the metallic iron, the initial molten slag containing a gangue component, an unreduced iron oxide, and an ash component of the carbonaceous reducing agent, and being first produced in the raw material mixture by heating thereof; a step of preparing the raw material mixture producing a composition of the initial molten slag corresponding to the target temperature; and a step of heating the raw material mixture to reduce and melt the raw material mixture and to produce the initial molten slag. By this manufacturing method, metallic iron having a target carbon concentration can be efficiently manufactured.

DESCRIPTION

METHOD FOR MANUFACTURING METALLIC IRON

Technical Field

The present invention relates to an improvement in technique for manufacturing metallic iron by heating and reducing an iron source such as an iron ore using a carbonaceous reducing agent such as coke, and more particularly, the present invention relates to a technique for efficiently manufacturing metallic iron having a predetermined carbon concentration. In this technique, an iron oxide is reduced and is simultaneously carburized, and metallic iron produced thereby is efficiently separated from slag-forming components which are mixed, for example, in a raw mineral ore as gangue components and the like.

Background Art

One of the inventors of the present invention proposed a method disclosed in Patent Document 1 as a new type direct iron-making method, and research for improving the above direct iron-making method has also been carried out thereafter.

This method is a technique to produce metallic iron by heating and reducing a raw material mixture containing a carbonaceous reducing agent and iron oxide. In this method, after a metallic iron shell is formed and grown by reducing iron oxide using a reducing gas derived from a carbonaceous reducing agent, the reduction is further advanced in a solid state until the iron oxide is not substantially present inside the metallic iron shell, followed by further continuous heating to make slag, which is produced inside, flow out of the metallic iron shell, so that the metallic iron is separated from the slag.

When the above method is carried out, by melting the metallic iron shell formed by heating and reducing, the molten slag present inside the metallic iron shell may be made to flow out thereof. In order to melt the metallic iron shell, the melting point of the metallic iron shell may be decreased by dissolving carbon derived from a carbonaceous reducing agent present inside the metallic iron shell into metallic iron (this phenomenon may be called "carburization" in some cases).

In this method, after the slag flows out of the metallic iron shell, and metallic iron obtained thereby and produced slag are then cooled and solidified, metallic iron solidified in the form of particles may be separated using a magnetic separator or a sieve while the slag is ground, or the solidified metallic iron and the produced slag may be melted by heating, followed by separation using the difference in specific gravity. As a result, metallic iron can be obtained having a high purity of 95 mass percent or more or 98 mass percent or more.

In addition, as a method for promoting the separation of metallic iron by controlling the composition of produced slag, which is performed when a direct-iron making method as described above is carried out, various proposals have been made.

For example, in Patent Document 2, a method for separating granular metallic iron from low-melting point slag containing FeO has been disclosed in which steel mill waste is used as an iron oxide source and is mixed with a carbonaceous reducing agent (hereinafter referred to as a "carbonaceous material" in some cases) and an additional raw material (slag-forming agent) so that a produced slag composition is adjusted to have a CaO/SiO_2 ratio (degree of basicity) in the range of 1.4 to 1.6 on a mass basis, followed by heating to 1,250 to 1,350°C for reduction to form granular metallic iron.

However, this method is a method using steel mill waste as an iron oxide source. In addition, adjustment of the degree of basicity of a slag-forming component used in this method is performed when raw materials are mixed together, and behavior of slag produced in heating and reducing and that of iron oxide contained therein have not been sufficiently investigated.

In addition, in Patent Document 3, a method has been disclosed in which the degree of basicity of a slag-forming component in a raw material is controlled in the range of 0.4 to 1.3, and at least one third of the time for heating and reducing performed on a hearth is controlled in the range of 1,200 to 1,350°C so as to set the reduction degree of iron to 40% to 80%, followed by melting a reduced product.

The adjustment of the degree of basicity used in this method is performed by calculation when raw materials are mixed together. However, by this method, influence of unreduced iron oxide contained in a raw material on the production of molten slag, dynamic behavior of iron oxide contained in molten slag, influence of the iron oxide on a meltdown state of metallic iron produced by reduction, and the like have not been investigated at all.

In addition, the inventors of the present invention have further carried out intensive research, and as a result, a technique disclosed in Patent Document 4 was proposed. The basic concept of this invention is that, when metallic iron is manufactured by heating and reducing a raw material mixture containing a carbonaceous reducing agent and iron oxide, by controlling a liquid fraction in a solid-liquid coexisting phase of multi-component slag which is produced in reduction and melting of the raw material mixture and which contains gangue components, carburization and melting of solid metallic iron to be produced are facilitated. In particular, the technique is characterized in that besides the control of a meltdown temperature of metallic iron, the carbon content (carbon concentration, hereinafter the same expression as the above will be used) of metallic iron to be produced is controlled.

However, when the liquid fraction of slag in a solid-liquid coexistence state is controlled by this method, the operation becomes complicated as described below. That is, it is necessary that the relationship of the liquid fraction with the temperature and/or the composition of a slag-forming component contained in the raw material mixture be obtained beforehand, the amount of a raw iron oxide component and that of an ash component in a carbonaceous reducing agent, which are mixed into molten slag, be adjusted, and in addition, the additional amount of a slag-forming component, which is additionally added whenever necessary, be also adjusted. Hence, through further improvement of this method, it has been desired to develop a method for efficiently manufacturing metallic iron having a target carbon concentration by easier operation.

In addition, in order to understand a carbonization phenomenon of iron in a blast furnace, one of the inventors of the present invention carried out intensive fundamental research on reduction of iron oxide under the presence of molten slag and dynamic behavior of pig iron (Fe-C) which is produced by reduction and which dissolves carbon by carburization, and as a result, the following phenomenon was confirmed and was disclosed in Non-Patent Document 1.

This Non-Patent Document 1 relates to reduction of iron oxide in a blast furnace and carburization behavior of reduced iron (metallic iron) produced by the reduction. In this document, as shown in a schematic view of Fig. 1, behavior has been disclosed in which iron oxide (FeO) is reduced in molten slag (S) by a carbonaceous material (G: graphite) and in which produced metallic iron (Fe) is carburized. In particular, iron oxide (FeO) in molten slag (S) is reduced and carburized by carbon (C) derived from a carbonaceous reducing agent (that is, carbonaceous material, G) to produce carburized molten iron (Fe-C). Accordingly, the concentration of FeO in slag which is in contact with a carbonaceous material is decreased and becomes different from the concentration of FeO in slag which is in contact with reduced iron. Hence, the carburized molten iron (Fe-C) tends to move in a direction apart from the carbonaceous material; hence, it rapidly moves in a direction towards solid reduced iron (S-Fe) and adheres thereto to form an unified body, thereby carburizing the solid reduced iron.

However, it has not been understood how the phenomenon in a blast furnace described above can be used to improve the above direct iron-making technique developed by the inventors of the present invention.

Patent Document 1: Japanese Unexamined Patent Application Publication No. 9-256017

Patent Document 2: Japanese Unexamined Patent Application Publication No. 10-147806

Patent Document 3: Japanese Unexamined Patent Application Publication No. 2000-45008

Patent Document 4: Japanese Unexamined Patent Application Publication No. 2005-48197

Non-Patent Document 1: ISIJ International, Vol. 44 (2004), No. 12, pp. 2033 to 2039

Disclosure of Invention

The present invention has been conceived in consideration of the above circumstances, and an object of the present invention is to provide an iron-making method having more excellent operation performance and operation efficiency than that of the direct iron-making method disclosed in the above Patent Document 4 that was developed by the inventors of the present invention.

That is, the present invention is a method for manufacturing metallic iron from a raw material mixture containing a carbonaceous reducing agent and an iron oxide-containing material, and the above method has a step of determining a target temperature of initial molten slag formation corresponding to a predetermined target carbon concentration in the metallic iron, the initial molten slag containing a gangue component, an unreduced iron oxide, and an ash component of the carbonaceous reducing agent, and being first produced in the raw material mixture by heating thereof; a step of preparing the raw material mixture producing a composition of the initial molten slag corresponding to the target temperature; and a step of heating the raw material mixture to reduce and melt the raw material mixture and to produce the initial molten slag.

In one aspect, the present invention provides a method for manufacturing metallic iron from a raw material mixture containing a carbonaceous reducing agent and an iron oxide-containing material, comprising: a step of determining a target temperature of initial molten slag formation corresponding to a predetermined target carbon concentration in the metallic iron, the initial molten slag containing a gangue component, an unreduced iron oxide, and an ash component of the carbonaceous reducing agent, and being first produced in the raw material mixture by heating thereof; a step of preparing the raw material mixture producing a composition of the initial molten slag corresponding to the target temperature, wherein in the preparation step, a slag-forming component is mixed with the iron oxide-containing material and the carbonaceous reducing agent; and a step of heating the raw material mixture to reduce and melt the raw material mixture and to produce the initial molten slag, wherein a FeO content of the initial molten slag is in the range of 35% to 50% in the SiO_2-Al_2O_3-FeO system.

Brief Description of the Drawings

Fig. 1 is a schematic view conceptually showing the movement of molten iron oxide and a carbon carrier function of molten iron in molten slag containing carbon, discovered by one of the inventors of the present invention.

Fig. 2 is a multi-component phase diagram of CaO, SiO_2, Al_2O_3, and FeO, which are primary components of molten slag.

Fig. 3 is graph showing an example of the relationship between the metallization degree and unreduced FeO remaining amount in heating and reducing.

Fig. 4 is a graph showing the change in reduction degree (referred to as "metallization degree" some times in this specification) with time for heating and reducing.

Fig. 5 is a graph showing the influence of the content of iron oxide (FeO) in slag on an initial molten slag forming temperature and a CO gas generation start temperature.

Fig. 6 is a graph showing the influence of the content of iron oxide (FeO) in slag on an initial molten slag forming temperature and a CO gas generation start temperature.

Fig. 7 is a graph showing the relationship between an initial molten slag forming temperature and a carbon concentration of meltdown metallic iron (metallic iron product) obtained under the above temperature condition.

Best Mode for Carrying Out the Invention

Hereinafter, the present invention will be described in detail. As described above, the most significant feature of the present invention is that when a raw material mixture containing an iron oxide-containing material, such as iron ore, iron oxide, or a partially reduced material thereof, and a carbonaceous reducing agent, such as coke or coal, is heated to manufacture metallic iron by reduction and melting, a "target temperature of initial molten slag formation" corresponding to a predetermined target carbon concentration in metallic iron is determined, and the raw material mixture producing the initial molten slag corresponding to the target temperature is prepared and is then further heated to form the initial molten slag; that is, in other words, by controlling the forming temperature of the initial molten slag, the carbon concentration of obtained metallic iron is controlled. In the present invention, the initial molten slag is slag which is first produced in the raw material mixture and which contains gangue components, unreduced iron oxide, and an ash component in the carbonaceous reducing agent.

While the inventors of the present invention have carried out research to further improve the technique disclosed in the above Patent Document 4, it was conceived that when the phenomenon disclosed in the above Non-Patent Document 1, that is, reduction of iron oxide and carburization behavior in molten slag in a blast furnace, is effectively used, the iron-making method disclosed in Patent Document 4 may be further improved. In the iron-making method used in the Patent Document 4, in order to efficiently separate and recover high purity metallic iron at a lower temperature, it is important to melt solid reduced iron at a lower temperature, and in addition, it is also important how efficiently molten slag (hereinafter referred to as "by-product slag" in some cases), which is produced as a by-product in a heating and reducing process, is separated. For this purpose, it is important how rapidly carburization of solid reduced iron is performed for melting thereof (in this specification, a phenomenon in which solid reduced iron is melted by carburization is called "meltdown of reduced iron" in some cases).

Accordingly, the inventors of the present invention considered the probability that if the technique disclosed in the above Non-Patent Document 1 is used for direct iron making, when molten reduced iron produced by carburization following reduction using a carbonaceous material in a molten slag moves in a direction towards solid metallic iron in accordance with the above phenomenon and is then unified therewith, the molten reduced iron may be used as a carrier carrying carbon towards the solid metallic iron, and based on this assumption, research was further conducted.

As has thus been described, the method (disclosed in Patent Document 4) developed by one of the inventors of the present invention is based on new finding at the time that indicates a close relationship between the liquid fraction of by-product slag and the meltdown of metallic iron, and this method is to perform the control using a new concept, that is, the liquid fraction in a solid-liquid coexistence phase of by-product slag, without melting the entire amount of the by-product slag. According to this method, when the liquid fraction is properly controlled, carburization of solid metallic iron produced by heating and reducing can be performed at a lower operation temperature, and as a result, the melting point thereof can be rapidly decreased. In addition, since meltdown of metallic iron can be performed at a lower temperature by this method, separation from by-product slag can be efficiently performed at a low temperature, and in addition, the carbon concentration of metallic iron, which has a large influence on the quality of a metallic iron product, can also be controlled.

A significant technical feature of the above invention filed in the past is that when metallic iron is manufactured by heating, reducing, and melting the above raw material mixture, the fact confirmed from the state of by-product slag, the carburization state of produced metallic iron, and the molten state thereof in a system containing a carbonaceous reducing agent is effectively used. That is, a phenomenon is used in which when a carbonaceous reducing agent is present together with slag in a molten state, molten metallic iron produced from molten slag having fluidity has a carrier-like function, carries carbon, and is then rapidly brought into contact with the surface of solid metallic iron, and thereby carburization of the solid metallic iron is efficiently performed. In addition, an effective carburization promotion effect by coexistence of a carbonaceous reducing agent and molten slag, as described above, is not limited only to the case in which the entire amount of slag is in a molten state, and when the liquid fraction of slag in a solid-liquid coexistence state is properly controlled, the carburization of solid metallic iron is promoted, and the meltdown temperature can be decreased.

In this embodiment, the liquid fraction indicates a mass fraction of liquid, which is located between a solidus line and a liquidus line under a certain temperature condition, in a solid and a liquid (that is, in two phases, the solid phase and the liquid phase) and is determined by a thermodynamic equilibrium relationship of a multi-component system containing SiO_2, Al_2O_3, CaO, and MgO, which are primarily derived from gauge components contained in raw materials, and FeO as a main component derived from an iron source.

This liquid fraction can be quantitatively obtained by observing the behavior of a raw material mixture when it is heated, reduced, and melted using a high-temperature laser microscope, followed by performing image analysis. However, it is unexpectedly difficult to accurate control the liquid fraction, and hence further improvement has been desired in terms of operation performance.

Through intensive research carried out by the inventors of the present invention, it was discovered that without performing a relatively difficult control, such as the liquid fraction control of slag, when a temperature for producing initial molten slag, which is produced in a reducing and melting process for a raw material mixture and which can be obtained from gangue components, unreduced iron oxide (FeO), and an ash component of a carbonaceous reducing agent, is properly controlled, metallic iron is not only efficiently manufactured, but the carbon concentration of the metallic iron can also be controlled; hence, as a result, the present invention was made.

After an unreduced molten iron oxide (FeO) derived from a raw iron oxide contained in molten slag which is produced in a heating and reducing process for a raw material mixture is brought into contact with carbon (or carbon monoxide) functioning as a reducing agent to perform melting reduction reaction, produced molten iron is further carburized to increase the carbon concentration, and as a result, the melting point thereof is further decreased. By this reaction, since the FeO concentration in slag in contact with a carbonaceous material is decreased and is different from the FeO concentration in slag in contact with reduced iron, the molten iron tends to move in a direction apart from the carbonaceous material. In addition, as is the case of the above Non-Patent Document 1, also in a direct iron-making method, molten iron which is carburized may be rapidly moved in a molten slag phase in a direction towards solid reduced iron and may then be unified therewith. Accordingly, after the unification with the solid reduced iron, a carbon component at a high concentration which enters the molten iron by carburization rapidly diffuses into the solid reduced iron, and as a result, the carbon concentration of the entire solid reduced iron is increased. That is, also in a direct iron-making method, when molten slag is produced at an early stage, metallic iron produced in the molten slag by reduction of molten iron oxide functions as a carrier which carries a carbon component in the molten slag in a direction toward solid reduced iron, and as a result, the carburization of the solid reduced iron is accelerated.

As apparent from the thermodynamic phase diagram of a Fe-C system, the carbon concentration of molten iron can be increased to that at 1,147°C, which is a eutectic point in terms of equilibrium, that is, can be increased to 4.3 mass percent, and also as apparent from the phase diagram described above, up to the eutectic temperature, the carbon concentration is increased as the temperature of the system is decreased.

Hence, when metallic iron in a molten state is manufactured by heating and reducing a raw material mixture, in order to increase the carbon concentration of molten iron to be produced, it is preferable that a melt of a slag-forming component containing unreduced iron oxide derived from an iron source be produced at a temperature as low as possible to advance reduction of the iron oxide (formation of reduced iron) and to promote the movement thereof in a direction towards solid metallic iron, and that production of molten iron by carburization be further accelerated. That is, it is preferable that a temperature producing a melt (that is, the initial molten slag) derived from a slag-forming component including iron oxide, which is first produced in a raw material mixture in a heating and reducing step, be decreased. In other words, when the raw material mixture is heated, as the reduction proceeds from the exterior of this raw material mixture, since slag is produced therein, this slag forming temperature is preferably decreased. In this reduction process, even when the exterior is partly reduced, the above initial molten slag may be produced in some cases.

In addition, as the forming temperature of initial molten slag produced in a heating and reducing process for a raw material mixture is decreased, the carburization rate of solid reduced iron is accelerated, and hence molten iron having a high carbon concentration can be obtained. Hence, when this phenomenon is effectively used, by controlling the forming temperature of initial molten slag, the carbon concentration of metallic iron to be obtained can be controlled. Furthermore, when the carburization of solid reduced iron is promoted as described above, the melting temperature (meltdown temperature) thereof is also decreased, and as a result, the operation temperature for metallic iron manufacturing can be decreased.

Incidentally, in order to determine the forming temperature of initial molten slag, which triggers off carburization and melting (meltdown) of solid reduced iron at a final stage of reduction of the raw material mixture, it is fundamental to set unreduced iron oxide derived from an iron source, gangue components, and an ash component in a carbonaceous material, which are components in molten slag (that is, slag components). The forming temperature of multi-component initial molten slag can be obtained from a multi-component thermodynamic phase diagram containing SiO_2, Al_2O_3, CaO, MgO, FeO, and the like. In recent years, it could be obtained from a computer-programmed phase diagram of multi-component slag.

For example, Fig. 2 is a phase diagram obtained by synthesizing a SiO_2-Al_2O_3-CaO system and a SiO_2-Al_2O_3-FeO system. According to this figure, in the case in which the composition of molten slag is a SiO_2-Al_2O_3-CaO system, when the composition is as shown by a dotted circle A in which Al_2O_3 is approximately 20% and a CaO/SiO_2 ratio is approximately 5/5, or when the composition is as shown by a dotted circle B in which Al_2O_3 is approximately 15% and a CaO/SiO_2 ratio is approximately 30/70, the melting temperature of the above four-component system slag shows the lowest value. In addition, in the case in which the composition of molten slag is a SiO_2-Al_2O_3-FeO system, when the composition is as shown by a thick line C in which a FeO content is in the range of approximately 35% to 50% (more preferably approximately 40%) and a SiO_2/Al_2O_3 ratio is approximately 45/55 or 40/60, the melting temperature of the above four-component system slag shows the lowest value.

Accordingly, with reference to those phase diagrams, when the individual component compositions of slag-forming components are adjusted in target ranges so that a slag composition produced in a heating and reducing process has the lowest temperature as described above, the forming temperature of initial molten slag can be decreased to the lowest temperature.

As a particular method for adjusting the slag-forming components, a method may be used in which in accordance with slag-forming components in raw material components (including gangue components in an iron source, an ash component of a carbonaceous material, an inorganic binder component, and the like), additional materials, such as CaO, SiO_2, or Al_2O_3, may be added. In this case, the addition is most generally performed when a raw material mixture is prepared; however, component adjustment may also be performed by additional supply at an early stage of heating and reducing. In addition, as for an iron oxide (FeO) component in slag, the amount of iron oxide remaining in unreduced state, which can be controlled by the metallization degree, may be used for the adjustment, the above iron oxide being one of iron oxide sources contained in a raw material mixture.

When a direct iron-making method as is the present invention is performed, the ratio of metallic iron recovered from iron oxide in a raw material is represented by the metallization degree, and it is judged that as the metallization degree is high, the productivity is superior. Hence, much energy was spent in the past to find a way of increasing the metallization degree. However, it is very difficult to increase the metallization degree to 100% by reducing all of an iron oxide source, the metallization degree obtained under general conditions is approximately up to 90% to 95%, and unreduced iron oxide in an amount of several percent remains.

However, on the other hand, the present invention positively uses unreduced iron oxide which remains in a heating and reducing process. That is, in addition to decrease in forming temperature of initial molten slag by mixing unreduced iron oxide in slag, unreduced iron oxide in a molten state mixed in the slag is reduced and carburized, so as to enable carburized iron thus obtained to function as a carrier carrying carbon in a direction towards solid reduced iron. As a result, the production efficiency of metallic iron can be improved as a whole.

Accordingly, in the present invention, in accordance with the types and/or the amounts of slag-forming components in a raw material mixture and additives for slag component adjustment, the metallization degree (reduction degree of an iron oxide-containing material) of an iron oxide source may be controlled so as to obtain a remaining amount of unreduced iron oxide corresponding to an optimum FeO content. In particular, until a target metallization degree is obtained, a heating temperature pattern or a reduction potential is adjusted, and in addition, the raw material mixture may be heated. As adjustment of the heating temperature pattern, for example, control of temperature, time, or temperature rise rate in heating and reducing may be mentioned. In addition, as adjustment of the reduction potential, for example, control of the amount of a carbonaceous reducing agent, the amount of a reducing agent used as a hearth protection material, or an in-furnace atmospheric gas may be mentioned.

Fig. 3 is a graph showing the result of research on the relationship between the metallization degree and the remaining FeO amount in an iron oxide source, which was obtained when a material produced in South America and supplied by MBR was used as iron oxide source (iron ore), Oak Grove coal produced in North America was used as a carbonaceous material, and heating and reducing were performed at 1,250 to 1,350°C (in which the content ratio of slag-forming components on a mass basis was set so that $CaO/SiO_2/Al_2O_3=4/68/28$ was satisfied, and the amount of the carbonaceous material was set to 0.95 times the theoretical carbon amount which is necessary for reducing all iron oxide by the carbonaceous material itself), and it is understood that there is a certain relationship between the metallization degree and the remaining FeO. The relationship described above may vary to a certain extent by the types of iron oxide source and/or carbonaceous material, mixing ratios thereof, heating and reducing conditions, and the like; however, when the relationship therebetween is obtained beforehand by a preliminary experiment, the remaining FeO amount can be adjusted by controlling the metallization degree of a raw material mixture, and as a result, the FeO content in produced slag can be properly obtained.

In addition, Fig. 4 is a graph showing the changes in temperature and metallization degree (that is, reduction degree) with time, which were obtained in the case in which a target temperature of a heating and reducing furnace was set to 1,400°C, a raw material mixture similar to that described above was supplied thereto, and heating and reducing were then performed. As can be seen from this graph, although being slightly changed by characteristic properties of a heating and reducing furnace, the metallization degree is increased as the heating time passes and is rapidly increased 4 to 5 minutes after the start of heating at a heating temperature used in this case, and the degree of increase in metallization degree is rapidly decreased approximately 9 minutes after the start. However, after all, the metallization degree reaches approximately 90 mass percent about 8 minutes after the start of heating, and at this stage, the amount of iron oxide remaining in an unreduced state is approximately 10 mass percent.

Hence, in accordance with the component compositions and the contents of slag-forming components in raw materials used in this operation process, when the proceeding of reduction is controlled to obtain the metallization degree of an iron oxide source, that is, the remaining iron oxide amount, so that the above remaining iron oxide amount corresponds to the above target ratio of content (that is, the ratio of content at which a slag composition corresponding to a target carbon concentration is obtained), the initial molten slag forming temperature can be controlled to the lowest temperature.

Fig. 5 is a graph showing the relationship between the iron oxide (FeO) concentration in slag and the initial molten slag forming temperature (and CO gas generation start temperature), which was obtained when a CaO/SiO$_2$ mass ratio in slag was maintained constant at 0.38. As shown in the figure, as the FeO content in slag is increased, the initial molten slag forming temperature (meltdown temperature of slag) is decreased, and in addition, the CO gas generation start temperature is also decreased, the CO gas generation being caused by reduction of molten iron oxide (FeO).

Fig. 6 is a graph showing the result of an experiment similar to that described above except that the CaO/SiO$_2$ mass ratio in slag was changed to 0.92, and although the slag composition was changed, the tendency in which the initial molten slag forming temperature and the CO gas generation start temperature are decreased as the FeO content is increased is the same as that shown in Fig. 5.

Fig. 7 is a graph showing the result obtained when the influence of the initial molten slag forming temperature (slag meltdown temperature) on the carbon concentration (C concentration) in produced molten metallic iron (meltdown metallic iron) was investigated, and from this graph, the tendency can be confirmed in which as the initial molten slag forming temperature (slag meltdown temperature) is decreased, the carbon content in metallic iron, which is a reduced product, is increased.

From the results described above, it was understood that the forming temperature of the initial molten slag composed of a mixture containing slag-forming components (CaO, SiO_2) and unreduced iron oxide (FeO), which is produced in a reducing and melting process for a raw material mixture, and the carbon content in molten metallic iron to be produced has a certain relationship, and that when the forming temperature of the initial molten slag is controlled, the carbon content of obtained metallic iron can be controlled. In addition, the tendency in which as the forming temperature of the initial molten slag is decreased, the carbon content of meltdown metallic iron is increased indicates that metallic iron having a high carbon content can be efficiently obtained by positively decreasing an operation temperature for metallic iron production, and this tendency is also significantly effective in view of decrease in thermal consumption.

In addition, as for the forming temperature of the initial molten slag, as described above, based on a multi-component phase diagram (such as that shown in Fig. 2), which includes unreduced iron oxide remaining in reduction besides slag-forming components in raw materials (gangue components in an iron source and/or an ash component contained in a carbonaceous material) which are first used to form a raw material mixture, an appropriate amount of a slag-forming component other than the gangue components in the above iron source may be added (hereinafter referred to as "addition of a third slag-forming component" in some cases) at the stage when a raw material mixture is prepared, is charged, or is heated so that a melting temperature of slag having the above mixed composition is further decreased. In addition, as for the iron oxide content in slag, as described above, the metallization degree of an iron source in a raw material mixture and the heating temperature pattern may be properly controlled in a metal iron-production process so that a necessary iron oxide content in slag is ensured by iron oxide remaining in an unreduced state.

In addition, when a target carbon concentration of a metallic iron product is determined beforehand, corresponding to the target carbon concentration, the optimum initial molten slag forming temperature is obtained from the relationship shown in Fig. 7, and based on a multi-component phase diagram as shown in Fig. 2, compositions of slag-forming components and an unreduced iron oxide may be adjusted so as to obtain the above optimum initial molten slag forming temperature. The adjustment of the initial molten slag composition in this case may also be performed by addition of a third slag-forming component or by the metallization degree of an ion source in a raw material and the heating temperature pattern.

The addition of a third slag-forming component at the stage when a raw material mixture is prepared will be described in detail. When the present invention is carried out, the control of the initial molten slag forming temperature can be adjusted by mixing use of another iron ore in accordance with the composition of gangue components . contained in an iron ore and the like used as an iron oxide source so as to obtain an appropriate slag-forming component composition; however, in accordance with a gangue component composition contained in a raw material ore, an additional raw material which can change the initial molten slag forming temperature is preferably added. As the additional raw materials, for example, calcined lime (CaO), lime stone ($CaCO_3$), silica (SiO_2), serpentine rock ($MgO + SiO_2$), Mn ore ($MnO + FeO$), and bauxite (Al_2O_3) may be mentioned, and those materials may be added alone or in combination. In particular, when a raw material mixture is prepared by mixing an iron oxide source and a carbonaceous reducing agent, plus a binder component whenever necessary, after a melting temperature is obtained from the composition of gangue components contained in raw materials based on a multi-component phase diagram, an appropriate amount of oxide as mentioned above may be mixed as an additional raw material with the raw material mixture so as to obtain a target initial molten slag forming temperature.

In order to effectively and practically achieve reduction and carburization of molten iron oxide as described above, rapid movement of the molten iron oxide to a direction towards solid reduced iron and concomitant promotion of carburization thereof by a carbon carrier effect, and decrease in meltdown temperature, it is necessary to cause sufficient lowering of melting point by carburization of solid reduced iron. Hence, it is preferable that the carbon concentration of metallic iron after carburization be controlled in the range of 0.5 to 4.3 mass percent, and that, in addition, the initial molten slag forming temperature be controlled in the range of 1,147 to 1,500°C. In addition, it is more preferable that the carbon concentration of metallic iron after carburization be controlled in the range of 1.5 to 3.5 mass percent, and that, in addition, the initial molten slag forming temperature be controlled in the range of 1,200 to 1,450°C. The carbon concentration of metallic iron after carburization may be adjusted by the amount of a carbonaceous reducing agent to be mixed at a raw material preparation stage, and in particular, as the amount of a carbonaceous reducing agent, the total of a necessary theoretical amount required for reducing an iron oxide source and the above amount necessary for carburization may be used. However, under general operation conditions, since a carbonaceous reducing agent is partly consumed by an oxidizing gas generated, for example, by burner heating in reduction, when the actual carbonaceous material amount is determined, the amount must be adjusted in consideration of the consumption amount described above.

In addition, when the amount of a carbonaceous reducing agent is adjusted when raw material mixing is performed as described above, a carburization amount for metallic iron in carburization and melting can be adjusted, and hence a carbon content of metallic iron which is finally obtained can be optionally adjusted whenever necessary.

As a raw material mixture used in the present invention, in order to enable heating and reducing of iron oxide by a gas derived from a carbonaceous reducing agent to efficiently proceed, an iron oxide source and a carbonaceous reducing agent are both preferably in the form of powder and are preferably placed in a mixed state for the use. This raw material mixture may be lightly solidified on a hearth with pressure and then be supplied; however, as disclosed in the above Patent Document 1, when the mixture is formed into an agglomerated material having an optional shape such as approximately spheres, briquettes, or pellets and is then supplied, a metallic shell of solid reduced iron is formed around the periphery of the agglomerated material in solid reduction by heating, and a high reduction potential can be maintained inside; hence, the metallization degree can be further efficiently improved, which is preferable.

Particular apparatus, fundamental operation conditions, and the like for performing the present invention are basically equivalent to the apparatus, operation conditions, and the like disclosed in the above Patent Document 1 or the like. In particular, an apparatus may be used in which a circular or a doughnut-shaped rotary hearth is provided; a heating and reducing furnace is used which has a raw material mixture supply zone, a preheating zone, a heating and reducing zone, a metallic iron melting zone, a cooling zone (metallic iron solidification zone), and a discharge zone provided in that order in a rotary direction; and a series of operations including supply of raw materials, heating and reducing, cooling and solidification of produced metallic iron, and recovery can be continuously performed. When continuous operation is performed by the apparatus as described above, the composition of slag-forming components in raw materials and the carbon content thereof are determined corresponding to a target carbon concentration by a preliminary experiment, and in addition, the metallization degree is also adjusted to obtain the optimum initial molten slag forming temperature in heating and reducing, so that the content of unreduced iron oxide in the initial molten slag may be ensured.

When metallic iron, which is coalesced into particles by heating and reducing, followed by carburization and melting, is cooled, solidified metallic iron coalesced into particles can be obtained. Subsequently, the metallic iron can be easily separated by sieving or magnetic separation from produced slag which is simultaneously cooled and solidified.

As described above, according to the present invention, when metallic iron is manufactured by heating a raw material mixture, followed by reduction and melting, by properly controlling a forming temperature of initial molten slag composed of unreduced iron oxide and gangue components, which is produced in a reducing and melting process for a raw material mixture, base on a multi-component phase diagram, the carbon concentration in metallic iron to be obtained can be optionally controlled. In addition, concomitantly, the following secondary effects can also be obtained.

That is, since the carbon content of metallic iron obtained by the method according to the present invention is increased as the initial molten slag forming temperature is decreased, that is, in other words, since when an operation temperature is decreased, metallic iron having a high carbon content can be obtained, a thermal consumption for heating and reducing can be decreased. In addition, molten iron oxide contained in initial molten slag functions as a carbon carrier for solid reduced iron produced by gas reduction, rapidly performs carburization of solid reduced iron (solid metallic iron), and promotes its meltdown; hence, melting of solid reduced iron is significantly accelerated, and as a result, the production efficiently can be significantly improved as a whole.

In the initial molten slag, unreduced iron oxide is contained as described above.. In the present invention, this iron oxide is reduced to metallic iron, and metallic iron functioning as a carbon carrier moves in molten slag to a solid reduced iron side. In this process, since the amount of iron oxide in molten slag is decreased, the composition of molten slag is changed with time. Hence, in order to efficiently manufacture metallic iron, even when the amount of iron oxide in molten slag is decreased, it is necessary for metallic iron to rapidly move. From this point of view, instead of generating temperature of initial molten slag, it is preferable to control a melting point of slag even after metallic iron is produced. The reason for this is that since slag is in a liquid state even after metallic iron is produced, the movement of metallic iron is not inhibited. As one example, base on an average composition of slag which is finally produced, a melting temperature (that is, slag meltdown temperature) of final slag, which should be controlled, may be determined from a phase diagram. Accordingly, even when reduction reaction is advanced, since final slag in a molten state is produced at a low temperature, metallic iron can be efficiently manufactured.

[Examples]

Hereinafter, the present invention will be described in detail with reference to examples; however, of course, the present invention is not limited to the following examples and may be performed after optionally making modifications within the scope which was described above and which will be described below, and any modifications mentioned above are also included in the technical scope of the present invention. In the following examples, "%" and "part(s)" indicate "percent by mass" and "part(s) by mass", respectively.

Example 1

An iron ore (67.74% of iron component content, 0.05% of CaO, 0.99% of SiO_2, 0.56% of Al_2O_3, and 1.55% of others) produced in South America as an iron source and materials shown in Table 1 as a carbonaceous material were used, and by using raw material pellets of metallic iron produced by heating and reducing the above materials which were mixed together to have a carbon content of approximately 1%, 2%, 3% or 4%, heating and reducing experiments were performed under the following conditions.

Table 1

CARBONACEOUS MATERIAL

	PRODUCING DISTRICT	BRAND	ASH COMPONENT (mass%)	VOLATILE COMPONENT (mass%)	FIXED CARBON (mass%)	S CONTENT (mass%)
CARBONACEOUS MATERIAL 1	CANADA PRODUCT	GreggRiver	9.36	21.25	69.39	0.334
CARBONACEOUS MATERIAL 2	USA PRODUCT	Oak Grove	8.8	19.6	71.6	0.53
CARBONACEOUS MATERIAL 3	SOUTH AFRICA PRODUCT	Optimum	9.95	36.41	53.63	0.32
CARBONACEOUS MATERIAL 4	GRAPHITE	REAGENT	-	-	100	-
CARBONACEOUS MATERIAL 5	CHINA PRODUCT	SHANXI	14.4	6.8	77.8	0.22

For the experiments, an apparatus composed of an infrared heating furnace in combination with a laser microscope was used, individual raw material pellets were continuously heated to 1,450°C at a temperature elevating rate of 200°C/min, behavior from reduction to melting in a temperature elevating process and behavior of bubble generation were directly observed, and a gas generation temperature and an initial molten slag forming temperature were obtained.

In this case, in order to finally obtain metallic iron having the above carbon content, in consideration of the forming temperature of the initial molten slag which was generated in heating and reducing, the metallization degree in heating and reducing was adjusted, so that appropriate individual initial molten slag forming temperatures (or slag meltdown temperatures at which slag was placed in one phase state which was composed only of liquid) were obtained based on a multi-component phase diagram.

The results are shown in Table 2. Although the initial molten slag forming temperatures corresponding to the respective carbon contents vary to a certain extent by carbonaceous materials which are used, when the individual initial molten slag forming temperatures are controlled to their approximate temperatures, metallic iron having an approximately target carbon content can be obtained.

Table 2

RELATIONSHIP BETWEEN CARBON CONTENT OF REDUCED IRON AND INITIAL MOLTEN SLAG FORMING TEMPERATURE

CARBON CONTENT OF REDUCED IRON PRODUCT (mass%)		1.1	2.04	2.98	4.26
INITIAL MOLTEN SLAG FORMING TEMPERATURE (°C)	CANADA COAL	1527	1423	1333	1259
	USA COAL	1538	1452	1342	1300
	SOUTH AFRICA COAL	1544	1413	1308	1294
	CHINA COAL	-	1465	1360	1278
	GRAPHITE	1516	1407	1258	1205
	AVERAGE	1531	1432	1320	1266

Example 2

The initial molten slag forming temperature is determined by slag-forming components contained in raw materials and a remaining unreduced iron oxide amount in heating and reducing; however, when the slag-forming components are not appropriate, by additionally adding a material containing a slag-forming component, the initial molten slag forming temperature can be decreased.

For example, Table 3 is a table showing the result of research on the influence of a CaO addition amount on the initial molten slag forming temperature. In the above research, when the target carbon concentration of metallic iron to be obtained was set to approximately 3% in Example 1, in order to ensure the initial molten slag forming temperature corresponding to that concentration, lime stone as CaO source was additionally added as a material containing a slab-forming component when a raw material mixture was prepared. Although two of the materials used in Example 1 were used as the carbonaceous material, in both cases, it was understood that when an appropriate amount of CaO was added, the initial molten slag forming temperature could be decreased. In the individual cases, the carbon contents of obtained metallic iron were 1.8%, 1.7%, 2.9%, and 3.5% when the CaO addition amounts in Table 3 were 0.3%, 0.4%, 2.0%, and 4.0%, respectively.

That is, in view of ensuring process productivity of heating and reducing operation, for example, when an operation temperature pattern and/or residence times in respective zones for heating, reducing, and the like are determined, and when it is desired to further decrease the initial molten slag forming temperature, by additionally adding a slag-forming component such as CaO as an additive at the stage when a raw material mixture is charged or as a slag-forming agent at the stage when a raw material mixture is heated, the initial molten slag forming temperature can be effectively controlled.

Table 3

INITIAL MOLTEN SLAG FORMING TEMPERATURE IN THE CASE OF ADDITION OF CaO

CaO ADDITION AMOUNT (mass%)		0.3	0.4	2.0	4.0
INITIAL MOLTEN SLAG FORMING TEMPERATURE (°C)	CANADA COAL 1	1318	-	1224	1262
	CANADA COAL 2	-	-	1206	1226
	CANADA COAL 3	-	-	-	1206
	AVERAGE	1318	-	1215	1231
	SOUTH AFRICA COAL 1	-	-	1282	1194
	SOUTH AFRICA COAL 2	-	1350	1244	1188
	SOUTH AFRICA COAL 3	-	1320	1232	1178
	AVERAGE	-	1335	1253	1187

Example 3

According to the present invention, by changing properties of a carbonaceous reducing agent (carbonaceous material) used as a reducing agent, the initial molten slag forming temperature and/or the remaining unreduced iron oxide amount in heating and reducing can also be controlled. In this example, the case is shown in which a CaO content is intentionally increased by adding CaO to a carbonaceous material, so that the initial molten slag forming temperature is changed.

That is, Table 4 below shows the results of research on the change in initial molten slag forming temperature, the results being obtained by adding CaO in amounts shown in Table 4 to the aforementioned 3 types of carbonaceous materials to change the ash content. As apparent from this table, when an appropriate amount of CaO is added to a carbonaceous material, the initial molten slag forming temperature is apparently decreased. In addition, since it has been known that Ca ions in CaO has a catalytic effect as an alkali to enhance reducing capability of a carbonaceous material and also contributes to improve reactivity thereof, it is believed that CaO can also be used to control a remaining unreduced iron oxide amount in heating and reducing.

Table 4

CaO AMOUNT IN LIME STONE AND INITIAL MOLTEN SLAG FORMING TEMPERATURE

CaO AMOUNT IN COAL ASH (mass%)		3	5	10	20	30	40	60
INITIAL MOLTEN SLAG FORMING TEMPERATURE (°C)	SOUTH AFRICA COAL	-	1294	-	1269	1263	1237	1238
	CANADA COAL	1259	-	-	1269	1252	1234	1212
	USA COAL	1299	-	-	1301	1253	1240	1227

As has thus been described, the present invention relates to a method for manufacturing metallic iron from a raw material mixture containing a carbonaceous reducing agent and an iron oxide-containing material, and this method has a step of determining a target temperature of initial molten slag formation corresponding to a predetermined target carbon concentration in the metallic iron, the initial molten slag containing a gangue component, an unreduced iron oxide, and an ash component of the carbonaceous reducing agent, and being first produced in the raw material mixture by heating thereof; a step of preparing the raw material mixture producing a composition of the initial molten slag corresponding to the target temperature; and a step of heating the raw material mixture to reduce and melt the raw material mixture and to produce the initial molten slag. The target temperature of the initial molten slag formation may be a specific temperature or a temperature range having a specific upper and lower limit. Furthermore, in accordance with operational restriction caused by apparatuses and/or steps, the above "specific temperature" may be a "temperature higher than the lowest temperature" in a changeable range of each component composition of a slag-forming component. The same thing can be said for the "upper limit" and the "lower limit" in the above temperature range. Accordingly, metallic iron having a predetermined carbon concentration can be efficiently manufactured.

When the method described above is performed, in the preparation step, a slag-forming component may be mixed with the iron oxide-containing material. In the preparation step, a slag-forming component may be mixed with the carbonaceous reducing agent. The raw material mixture may further contain an additional raw material, and in the preparation step, a slag-forming component may be mixed with the additional raw material. Before the heating step, a step of charging an additive containing a slag-forming component may be performed. In addition, in the heating step, a slag-forming agent containing a slag-forming component may be added. In addition, instead of a slag-forming agent, a flux containing a slag-forming agent may be added, or a slag-forming agent containing a slag-forming component and a flux containing a slag-forming component may be added. In other words, in any of the above steps, by adding a third slag-forming component, the target temperature may be controlled.

By performing at least one of the operations described above, a necessary slag-forming component can be optionally added into initial molten slag at the stage when a raw material mixture is prepared, is charged, or is heated, and as a result, the initial molten slag can be produced at a target temperature.

The target temperature can be determined by a multi-component phase diagram composed of gangue components, unreduced iron oxide remaining in reduction, and an ash component of a carbonaceous reducing agent. Accordingly, when individual component compositions of slag-forming components are adjusted within their changeable ranges, the target temperature which corresponds to the lowest temperature of initial molten slag formation in a target composition range can be easily determined. Alternatively, when the relationship between the forming temperature of initial molten slag and the carbon concentration in metallic iron is investigated beforehand, the target temperature can also be determined by the target carbon concentration in accordance with the above relationship. Accordingly, metallic iron having a target carbon concentration can be stably manufactured.

In addition, before the heating step, there may further be provided a step of setting a target content of the unreduced iron oxide in the initial molten slag and calculating a target reduction degree of the iron oxide-containing material corresponding to the target content, and in the heating step, until a reduction degree of the iron oxide-containing material reaches the target reduction degree, a heating temperature pattern or a reduction potential may be adjusted, and simultaneously heating may be performed. Accordingly, unreduced iron oxide, carburized iron, which remains in heating and reducing can be positively used as a carrier carrying carbon in a direction towards solid reduced iron, and as a result, the production efficiency of metallic iron can be improved as a whole.

In addition, in the heating step, heating may be performed based on a melting point of slag containing unreduced iron oxide in reduction. Accordingly, even when the amount of unreduced iron oxide in molten slag is decreased, since rapid movement of metallic iron in the molten slag can be ensured, even at the stage at which the reduction reaction has proceeded, metallic iron can be effectively manufactured.

According to the present invention, compared to the method disclosed in Patent Document 4 developed by the inventor, by controlling the amounts of gangue components in a raw material mixture, an ash component in a carbonaceous reducing agent, and reduced iron oxide remaining in heating and reducing, which is not a small amount, the temperature of initial molten slag formation can be controlled. Accordingly, the carbon concentration in obtained metallic iron can be adjusted, and metallic iron having a desired carbon concentration can be efficiently obtained. In addition, according to the present invention, as has thus been described in detail, by using molten iron derived from an iron oxide material (iron oxide-containing material), which enters molten slag from a raw mineral ore, as a carrier of carbon used for carburizing solid reduced iron, the carburization thereof is rapidly advanced, meltdown can be performed at a low temperature, and as a result, the productivity can be improved while consumption of heat energy for metallic iron manufacturing is decreased.

The present invention contains subject matter related to Japanese Patent Application No. 2006-008743 filed on January 17, 2006.

We claim:

1. A method for manufacturing metallic iron from a raw material mixture containing a carbonaceous reducing agent and an iron oxide-containing material, comprising:

a step of determining a target temperature of initial molten slag formation corresponding to a predetermined target carbon concentration in the metallic iron, the initial molten slag containing a gangue component, an unreduced iron oxide, and an ash component of the carbonaceous reducing agent, and being first produced in the raw material mixture by heating thereof;

a step of preparing the raw material mixture producing a composition of the initial molten slag corresponding to the target temperature, wherein in the preparation step, a slag-forming component is mixed with the iron oxide-containing material and the carbonaceous reducing agent; and

a step of heating the raw material mixture to reduce and melt the raw material mixture and to produce the initial molten slag, wherein a FeO content of the initial molten slag is in the range of 35% to 50% in the SiO_2-Al_2O_3-FeO system.

2. The method according to Claim 1,

wherein the raw material mixture further contains an additional raw material, and in the preparation step, the slag-forming component is mixed with the additional raw material.

3. The method according to any one of Claims 1 or 2, further comprising a step of charging an additive containing the slag-forming component before the heating step.

4. The method according to any one of Claims 1 to 3, wherein in the heating step, a slag-forming agent containing the slag-forming component is added.

5. The method according to any one of Claims 1 to 4, wherein the target temperature is determined by a multi-component phase diagram composed of the gangue component, the unreduced iron oxide remaining in reduction, and the ash component of the carbonaceous reducing agent.

6. The method according to any one of Claims 1 to 5, wherein a relationship between a temperature forming the initial molten slag and a carbon concentration in the metallic iron is evaluated beforehand, and the target temperature is determined by the target carbon concentration in accordance with the relationship.

7. The method according to any one of Claims 1 to 6,

further comprising, before the heating step, a step of setting a target content of the unreduced iron oxide in the initial molten slag and calculating a target reduction degree of the iron oxide-containing material corresponding to the target content,

wherein in the heating step, until a reduction degree of the iron oxide-containing material reaches the target reduction degree, a heating temperature pattern or a reduction potential is adjusted, and simultaneously heating is performed.

8. The method according to any one of Claims 1 to 7,

wherein in the heating step, heating is performed based on a melting point of slag containing unreduced iron oxide in reduction.

FIG. 1

Prior Art

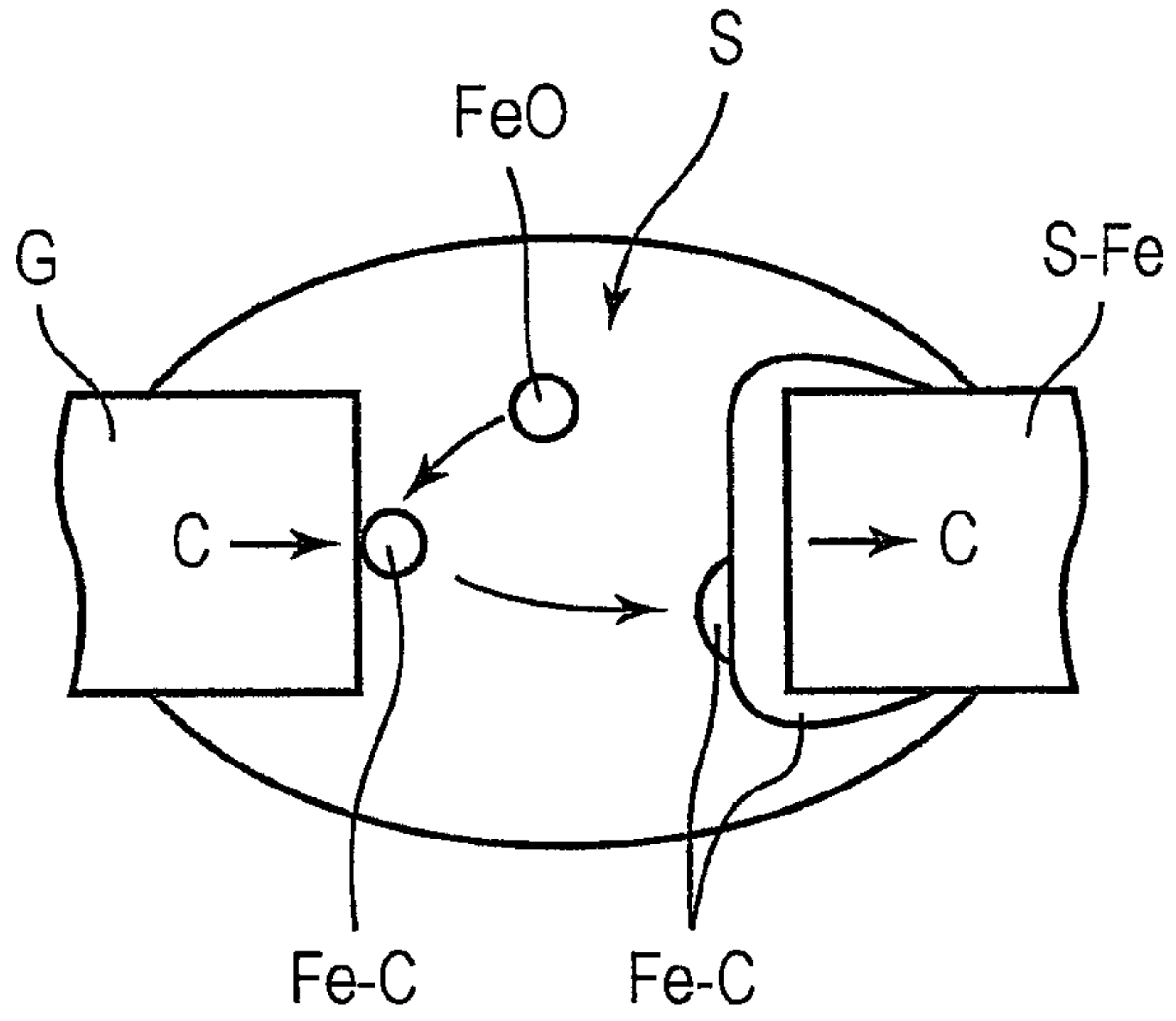
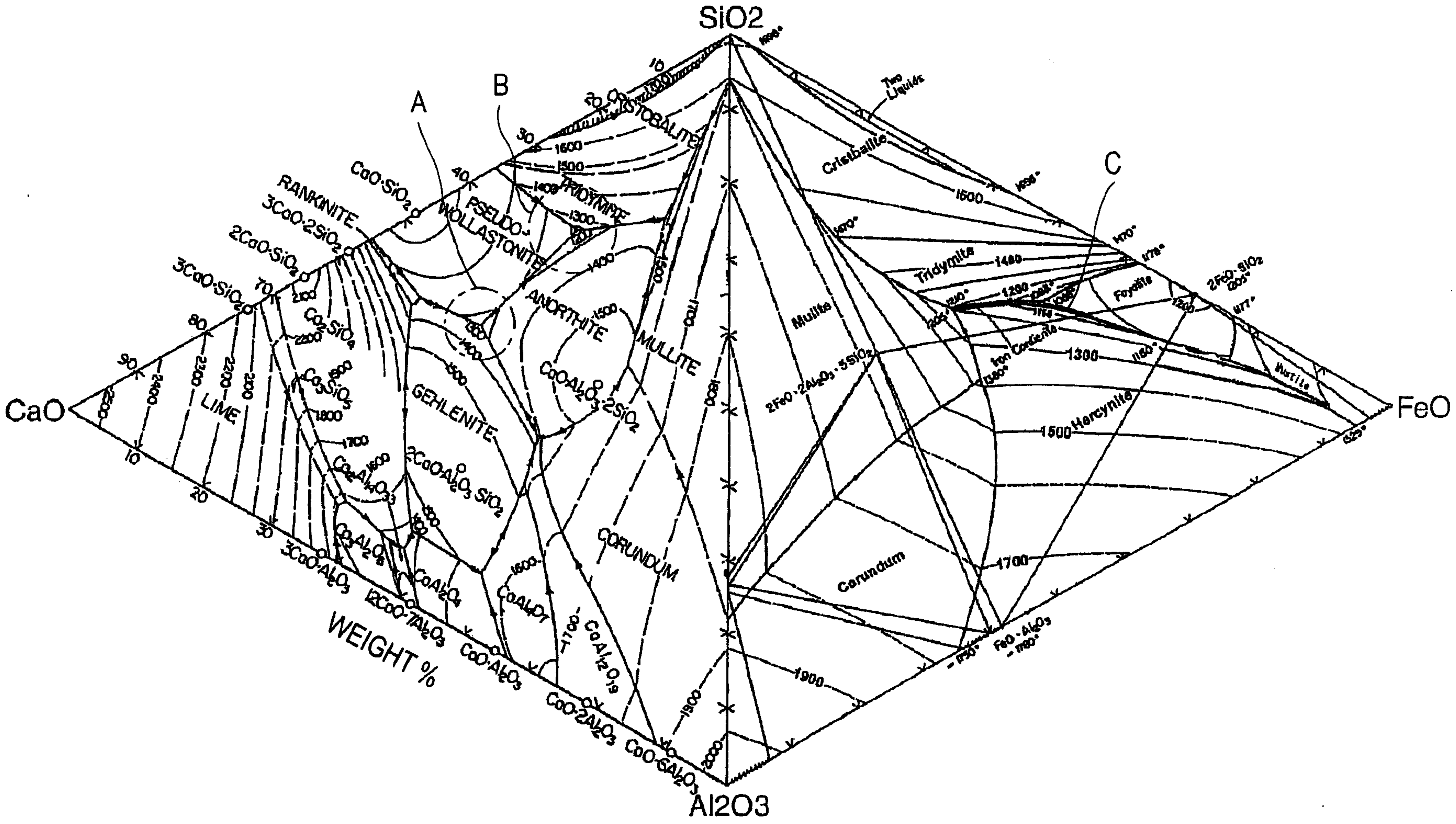


FIG. 2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CaO
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C
WEIGHT %
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RANKINITE
ANORTHITE
MULLITE
GEHLENITE
CORUNDUM
LIME
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3CaO·2SiO2
2CaO·SiO2
3CaO·SiO2
Ca2SiO4
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30
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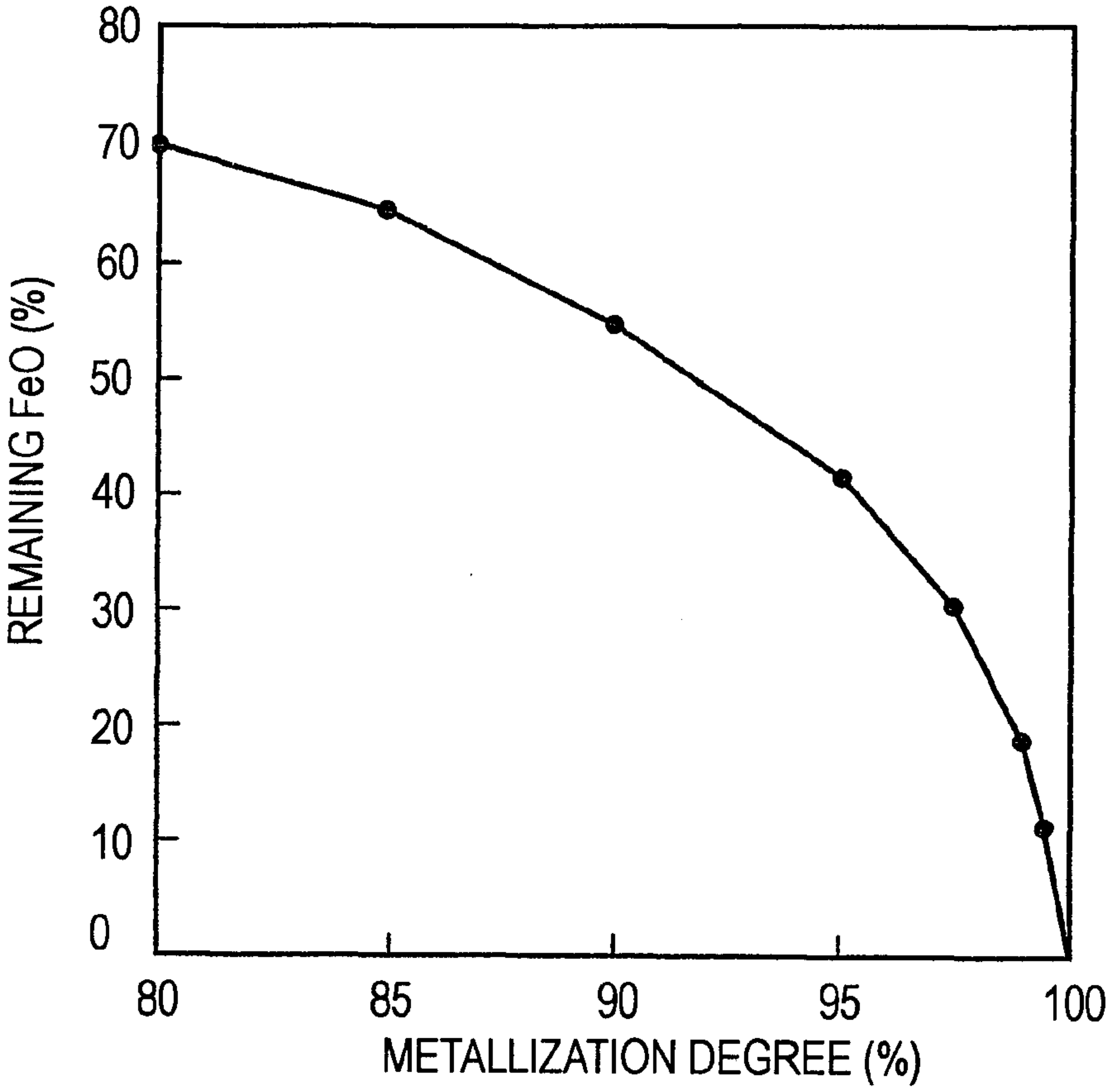
2400
2500

FIG. 3


80
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REMAINING FeO (%)
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95
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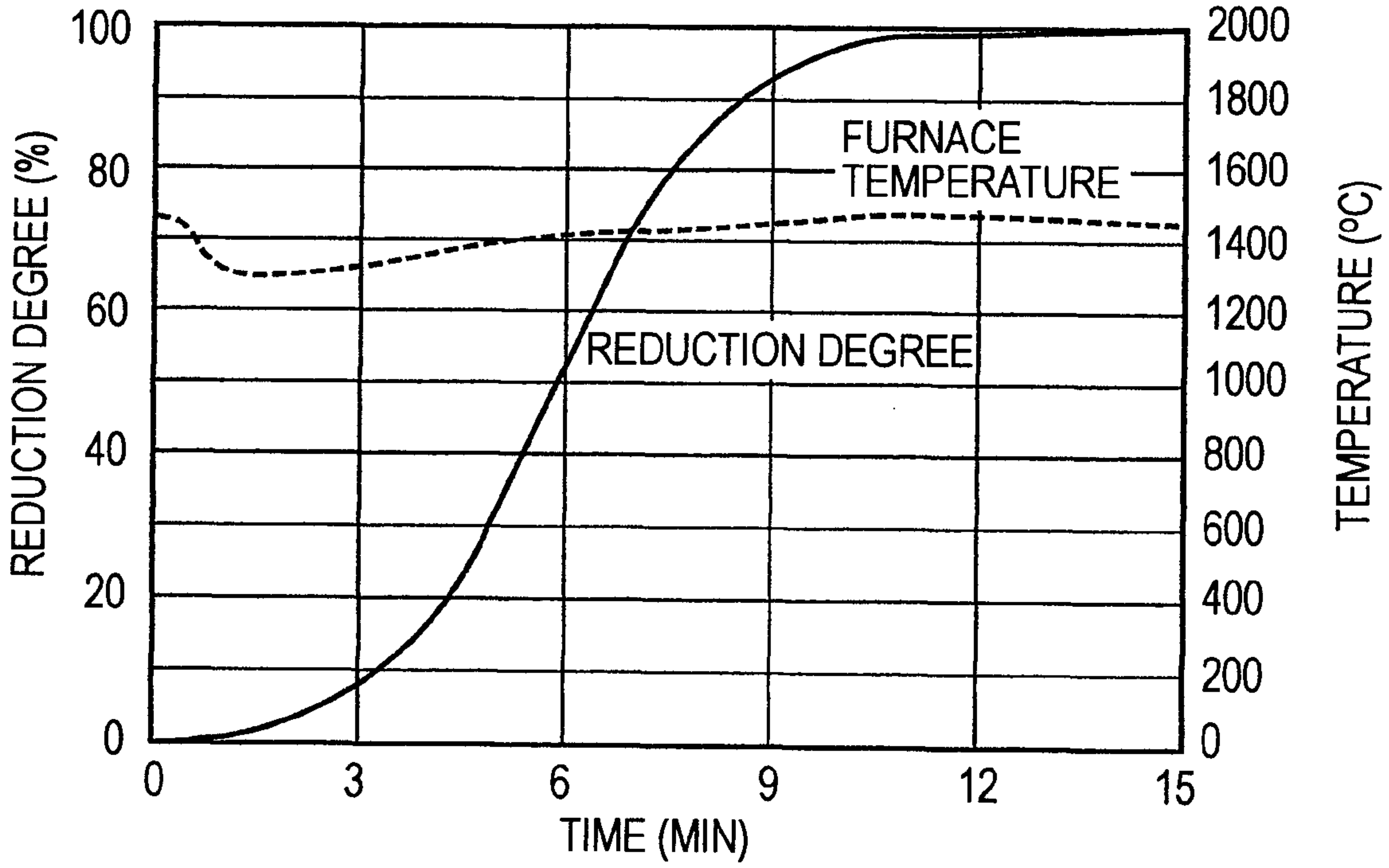
METALLIZATION DEGREE (%)

FIG. 4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20
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1000
800
600
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FURNACE TEMPERATURE
REDUCTION DEGREE
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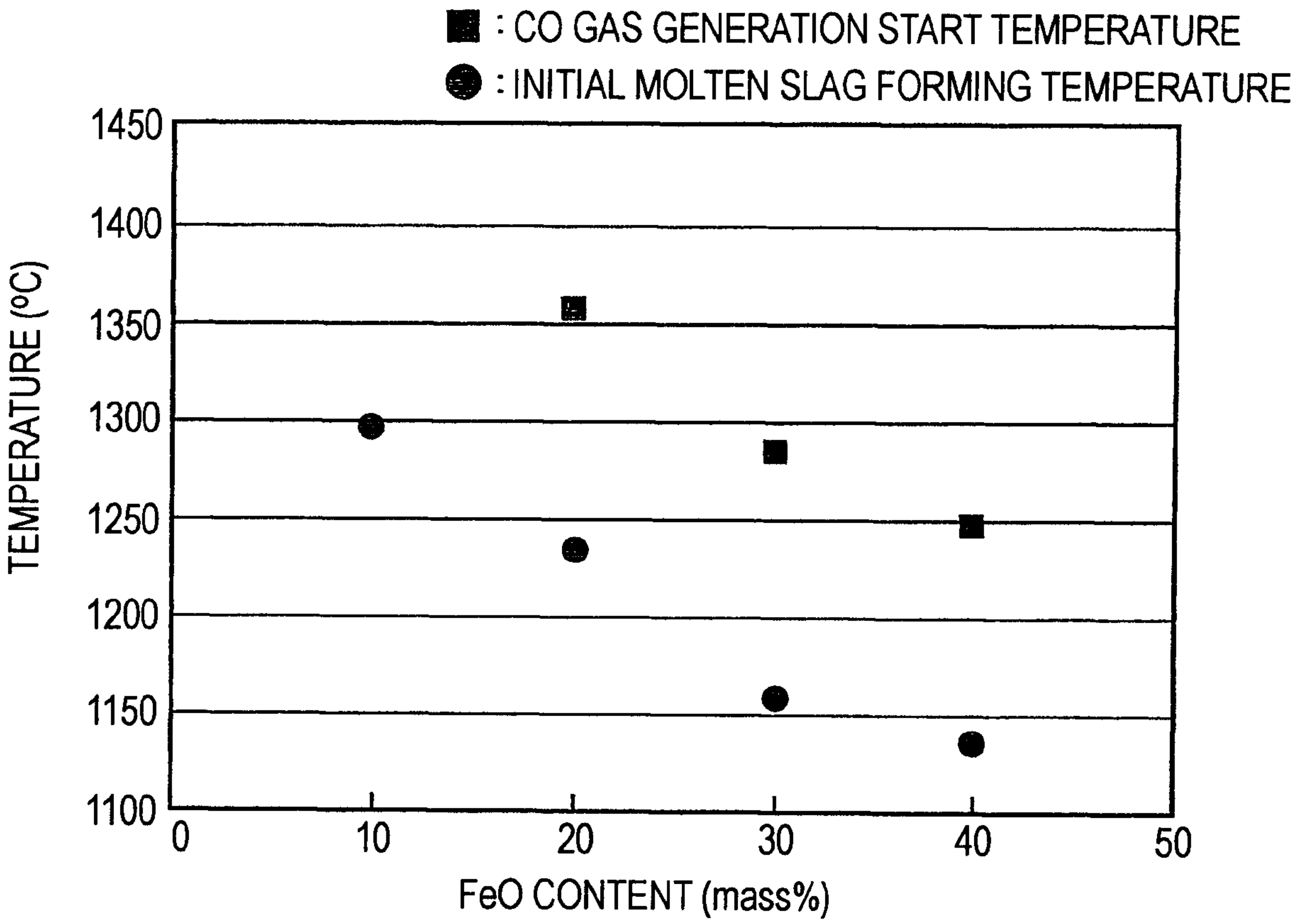
3
6
9
12
15
TIME (MIN)

FIG. 5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● : INITIAL MOLTEN SLAG FORMING TEMPERATURE
TEMPERATURE (ºC)
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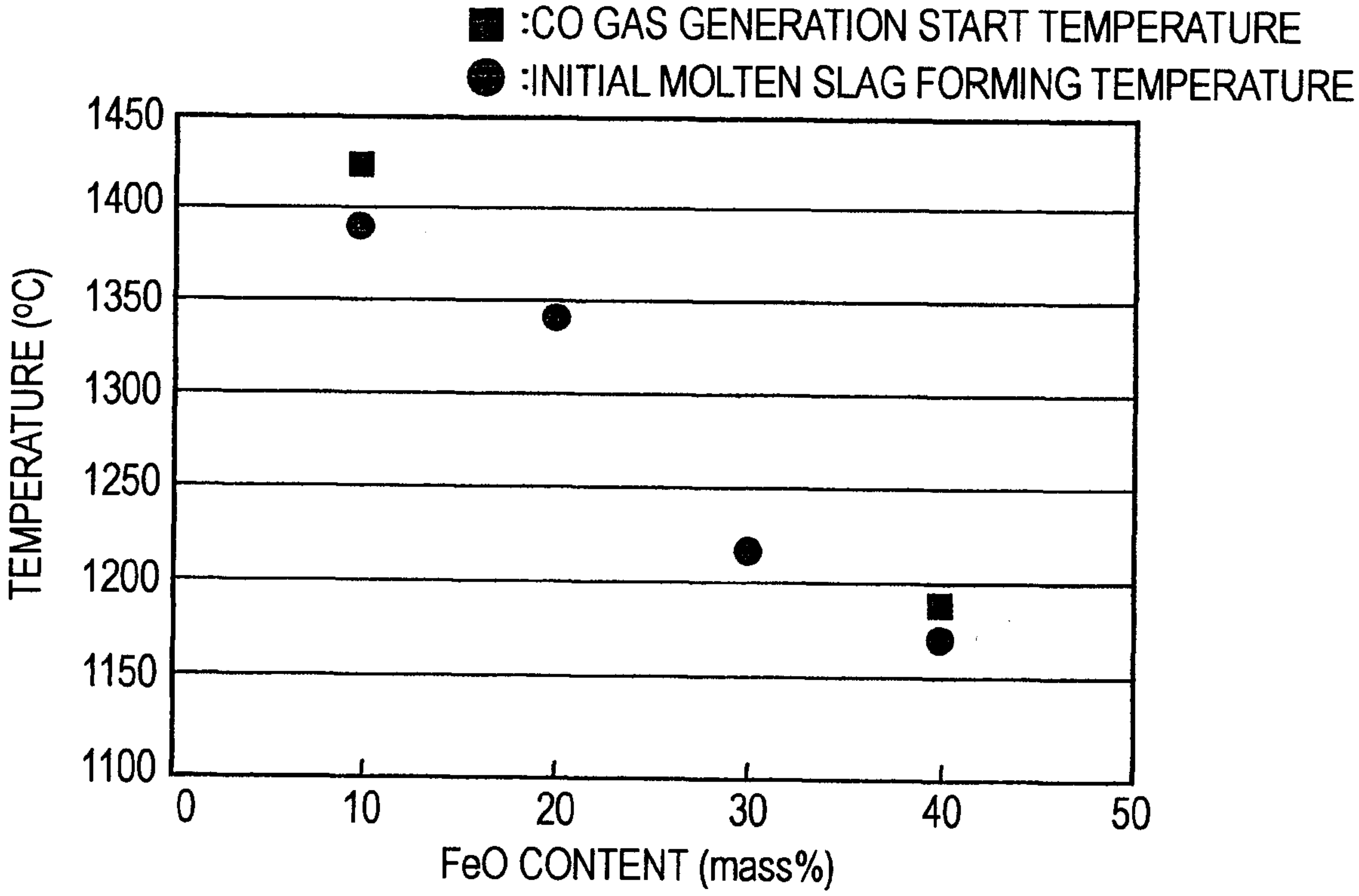
1300
1250
1200
1150
1100
0
10
20
30
40
50
FeO CONTENT (mass%)

FIG. 6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● :INITIAL MOLTEN SLAG FORMING TEMPERATURE
TEMPERATURE (ºC)
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1300
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1150
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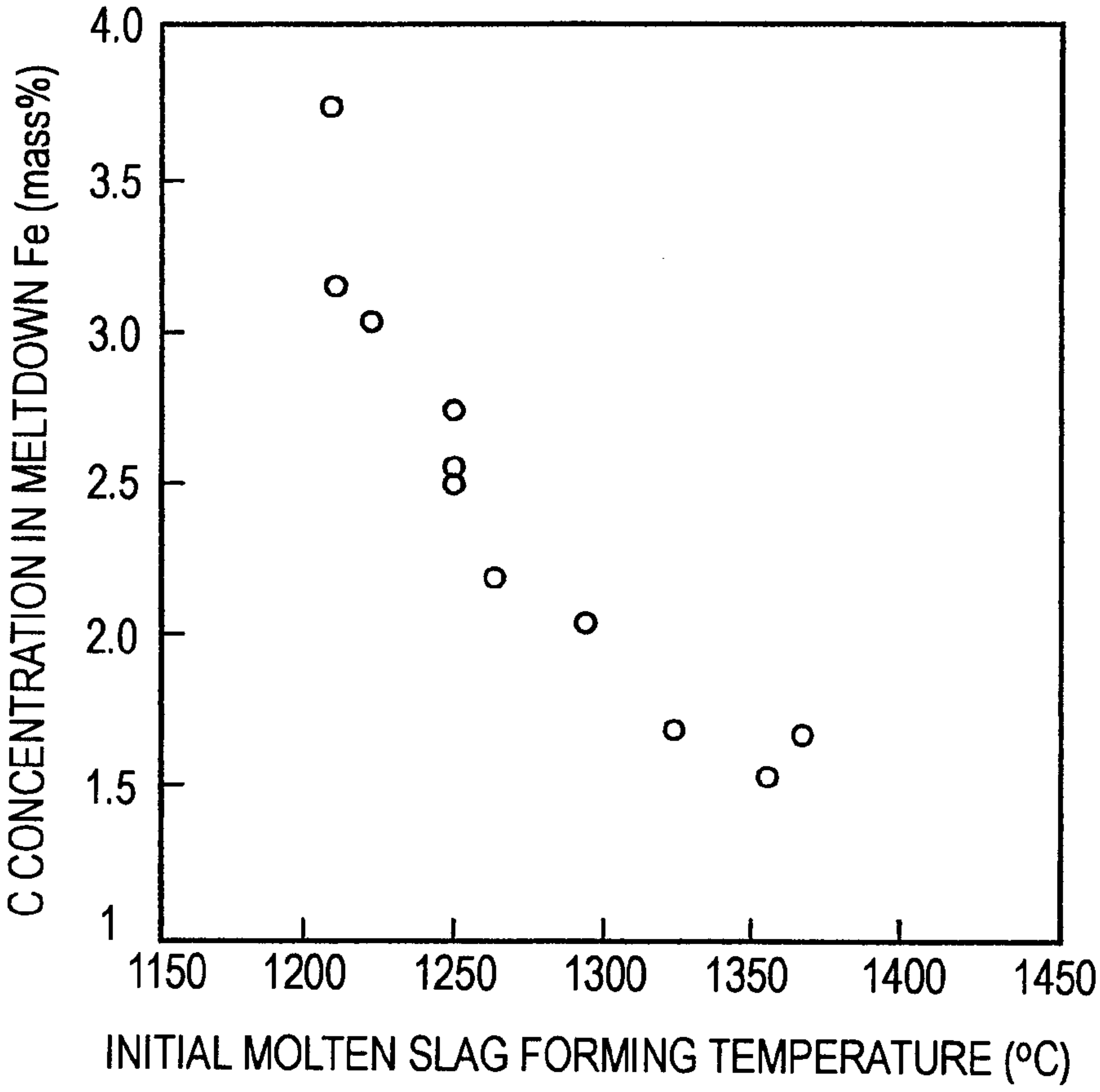
40
50
FeO CONTENT (mass%)

FIG. 7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3.0
2.5
2.0
1.5
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C CONCENTRATION IN MELTDOWN Fe (mass%)
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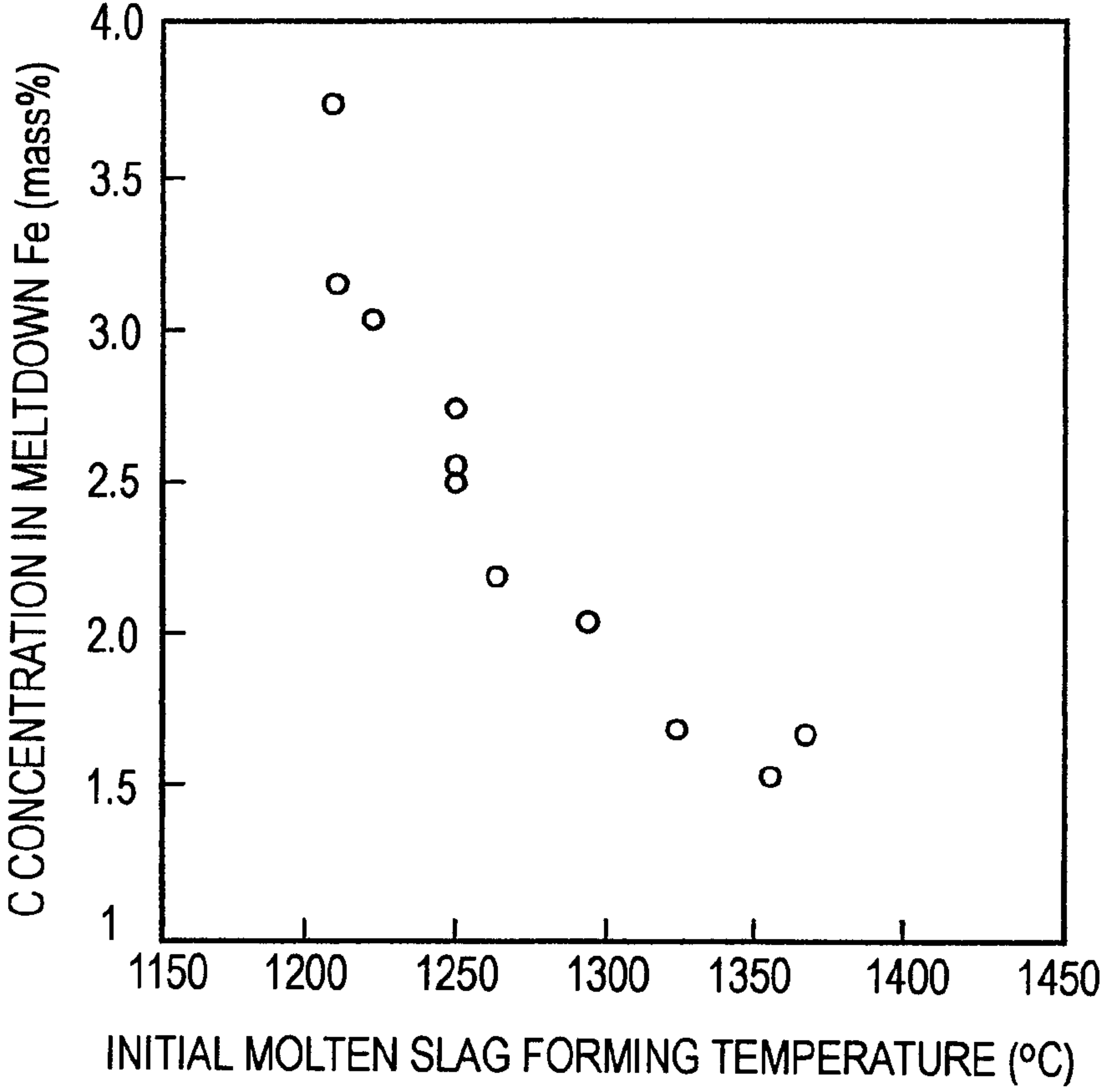
1300
1350
1400
1450
INITIAL MOLTEN SLAG FORMING TEMPERATURE (°C)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3.5
3.0
2.5
2.0
1.5
1
C CONCENTRATION IN MELTDOWN Fe (mass%)
1150
1200
1250
1300
1350
1400
1450
INITIAL MOLTEN SLAG FORMING TEMPERATURE (°C)