COMMONWEALTH OF AUSTRALIA

Patents Act 1952

APPLICATION FOR A STANDARD PATENT OR A
STANDARD PATENT OF ADDITION

We, SKF PLASMA TECHNOLOGIES AB, a Swedish Company, of P. O. Box 202,
S-813 00 Hofors, Sweden,

hereby apply for the grant of a Standard Patent for an invention entitled:
"A METHOD AND APPARATUS FOR CONTINUOUSLY PRODUCING LIQUID SILICON FROM CARBON AND SILICA"

which is described in the accompanying specification.

We request that the Patent may be granted as a Patent of Addition to the Patent applied for on Application No. filed.

We request that the term of the Patent of Addition be the same as that of the patent for the main invention or so much of the term of the patent for the main invention as is unexpired.

This application is a Convention application and is based on an application/numbered 87 03 895-6 for a patent or similar protection made in Stockholm, Sweden, on 9th October 1987.

Our address for service is care of E. F. WELLINGTON & CO., Patent Attorneys, 457 St. Kilda Road, Melbourne, in the State of Victoria, Commonwealth of Australia.

Dated this 24th day of May, A.D. 1988

SKF PLASMA TECHNOLOGIES AB
COMMONWEALTH OF AUSTRALIA


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

In support of the Convention Application made by (SKF PLASMA TECHNOLOGIES AB)

(hereinafter referred to as the applicant) for a Patent for an invention entitled:

"A METHOD AND APPARATUS FOR CONTINUOUSLY PRODUCING LIQUID SILICON FROM CARBON AND SILICA"

do solemnly and sincerely declare as follows:

1. I am authorised by the applicant for the patent to make this declaration on its behalf.

2. The basic application as defined by Section 141 of the Act was made in Stockholm, Sweden on the 9th day of October, 1987, by SKF PLASMA TECHNOLOGIES AB.

3. JEROME FEINMAN, of 2654 Sperber Lane, Grand Junction, Colorado 81501, United States of America, who is an American citizen, is the actual inventor of the invention and the facts upon which the applicant is entitled to make the application are as follow:

The applicant is the assignee of the actual inventor.

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

DECLARED at Hofors, Sweden this 24th day of May, 1988.
Claim
1. A method of continuously producing liquid silicon from carbon and silica, wherein in a first step a first quantity of the silica is converted with carbon to form silicon carbide, and wherein in a second, consecutive step a second quantity of the silica is converted with the silicon carbide produced in the first step, to form liquid silicon, thermal energy being supplied during both steps of the process.

6. An apparatus for performing the method as claimed in claim 1 for continuously producing liquid silicon from carbon and silica with the addition of heat, comprising a reaction vessel containing coal in lump form, means arranged in the vessel for the supply of coal in lump form, a pipe connected to the reactor vessel for the removal of exhaust gases therefrom, a plasma generator arranged in the upper part of the vessel for introducing plasma gas into the upper part of the coal bed in the reactor vessel to form a first reaction zone in the bed, as well as means for injecting
silica into the first reaction zone, said apparatus further comprising a plasma generator located at a lower level in the reactor vessel, with means connected thereto for introducing silica into a second reaction zone formed by the plasma generator at the lower level, in a bed primarily of silicon carbide which has been produced in the first zone, and also an outlet in the lower part of the reaction vessel for tapping off the silicon produced.
Application Number:  
Lodged:  
Complete specification Lodged:  
Accepted:  
Published:  

Priority:  
Related Art: 

Name of Applicant: SKF PLASMA TECHNOLOGIES AB  
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Actual Inventor/x: JEROME FEINMAN.  

Address for Service: E. F. WELLINGTON & CO.,  
Patent and Trade Mark Attorneys',  
457 St. Kilda Road,  
Melbourne, 3004, Vic.  

Complete Specification for the invention entitled:  

"A METHOD AND APPARATUS FOR CONTINUOUSLY PRODUCING LIQUID SILICON FROM CARBON AND SILICA"  

The following statement is a full description of this invention including the best method of performing it known to me/us:
The present invention relates to a method of continuously producing liquid silicon from carbon and silica with the addition of thermal energy. The invention also relates to apparatus for performing the method.

In a conventional electrothermal process for producing silicon from silica, the quantity of silicon recovered is limited to about 80% due to the formation of silicon oxide in gaseous form, $\text{SiO}_g$, which leaves the reaction zone and the furnace before it can be reduced to liquid silicon $\text{Si}_l$, and also because SiC collects as a waste product in the cooler parts of the furnace. The mechanism and theory behind these phenomena are discussed in "Heat and Mass Transfer in the Fe-rosilicon Process", by M B Müller, S E Olsen and J K Tuset in The Scandinavian Journal of Metallurgy I (1972) 145 - 155.

An object of the present invention is thus to improve the silicon recovery percentage, thus reducing the consumption of material and energy.

This object is achieved according to the present invention in a method of continuously producing liquid silicon from carbon and silica, wherein in a first step a first quantity of the silica is converted with carbon to form silicon carbide, and wherein in a second, consecutive step the remaining, second quantity of the silica is converted with the silicon carbide produced in the first step, to form liquid silicon, thermal energy being supplied during both steps of the process. The first quantity of silica is chosen to be approximately two thirds and the second quantity is thus approximately one third of the whole.

The temperature during the first step is preferably at least 1900°C and the temperature during the second step
is preferably also at least 1900°C.

The method according to the invention thus offers a process wherein the above mentioned losses of SiO₂ are reduced or eliminated and the proportion of SiC remaining in the product is minimized.

The method according to the invention can be illustrated in the following way.

In the first step a carbothermic reduction of SiO₂ is performed in a first zone containing a surplus of carbon. SiC is thus produced as follows:

\[
\text{SiO}_2 + 2C \rightarrow 2\text{SiO}_g + 2\text{CO} \quad (1)
\]

\[
2\text{SiO}_g + 4C \rightarrow 2\text{SiC}_s + 2\text{CO} \quad (2) \text{ or }
\]

totally \[
2\text{SiO}_2 + 6C \rightarrow 2\text{SiC}_s + 4\text{CO} \quad (3).
\]

In the second step, the SiC produced in the first step is utilized to reduce additional SiO₂ as follows:

\[
\text{SiO}_2 + \text{SiC}_s \rightarrow \text{Si}_l + \text{SiO}_g + \text{CO} \quad (4), \text{ and }
\]

\[
\text{SiO}_g + \text{SiC}_s \rightarrow 2\text{Si}_l + \text{CO} \quad (5), \text{ or }
\]

totally \[
\text{SiO}_2 + 2\text{SiC}_s \rightarrow 3\text{Si}_l + 2\text{CO} \quad (6).
\]

The total reaction for the two combined steps (equation (3) plus equation (6)) is

\[
3\text{SiO}_2 + 6C \rightarrow 3\text{Si}_l + 6\text{CO} \quad (7).
\]
The method according to the invention is advantageously performed in a shaft furnace kept filled with carbonaceous material in lump form. This reactor is suitably provided with supply means for the carbonaceous material in lump form, and a pipe for withdrawing gases produced and introduced into the system. Means are provided at an upper level in the reactor for supplying silica and thermal energy in a first reaction zone. At a second, lower level in the reactor means are provided for the supply of silica and thermal energy for performing the second reaction step. The lower part of the reactor is designed in conventional manner to permit tapping off the silicon produced. It may advantageously be designed to permit temporary collection of liquid silicon which is then tapped off batchwise from the reactor.

The means for supplying thermal energy may preferably be means for the supply of energy independent of combustion, such as plasma generators, \( \text{SiO}_2 \) in particle form being injected at said means.

The process according to the invention, in accordance with the stoichiometric equations given above will now be described in more detail with reference to the single drawing showing schematically an apparatus for performing the method according to the invention.

The apparatus shown schematically in the drawing comprises a shaft furnace 1 provided with at least one plasma generator 2 at an upper level, and at least one plasma generator 3 at a lower level. The furnace is also provided with a conventional sluice 4 in the upper part 9 for the supply of coal in lump form, and a pipe 7 for the removal of gases produced and introduced into the furnace. Injection means 5 and 6, respectively, for injecting \( \text{SiO}_2 \)
in particle form at each plasma generator, are also shown. Tapping means 8 are also shown at the bottom of the furnace for periodic withdrawal of the product, liquid silicon.

The first step of the process is performed in the upper part of the shaft furnace 1 which comprises the plasma generators 2 at the upper level and the furnace volume above this upper level. This furnace volume is filled with coal in lump form which is fed through the upper part 9 of the shaft. Plasma gas, which may be recirculated exhaust gas or some other suitable gas, together with SiO\textsubscript{2} injected in particle form into the plasma gas, enters the carbon bed and reacts in accordance with equations (1) and (2) to form SiC and CO. The temperature in this reaction zone after substantially complete conversion will be at least 1900° C, preferably higher, and this temperature will decrease to about 1300° C at the top of the shaft furnace as the coal supplied in lump form moves down and is heated to reaction temperature by the rising gases. It is obvious that a considerable part of this pre-heating of the coal will be effected in the upper part of the reactor volume so that most of the volume above the level of the upper plasma generator 2 will be at or close to the reaction-zone temperature. This provides favourable conditions for complete reduction of all SiO\textsubscript{2} in accordance with reaction (2). The SiC produced in this first step will move downwardly, together with any unconverted coal, into the lower part of the shaft furnace 1, said part containing the plasma generators at the lower level 3 and means for collecting and withdrawing Si\textsubscript{1} and this lower part is utilized to perform the second step of the process. Additional SiO\textsubscript{2}, together with plasma gas from the plasma
generators at the lower level, is supplied here in the second step and will react with the downwardly moving SiC and unconverted coal to produce Si₁ and CO in accordance with reactions (4), (5) and possibly

\[ \text{SiO}_2 + C \rightarrow \text{Si} + \text{CO} \]  \hspace{1cm} (8).

Si₁ produced will be collected at the bottom of the furnace, and is periodically withdrawn by tapping in conventional manner. In a preferred embodiment of the invention, the furnace is started by filling its lower part with SiC in lump form and filling its upper part with carbon in lump form, which may be graphite or coke. The plasma generators at levels 2 and 3 are operated by a suitable inert plasma gas to heat the system to reaction temperature. Continuous production of Si₁ can then be started by injecting SiO₂ at the plasma generators 2 and 3 to obtain the reactions described earlier. An example of the process according to the invention, using relatively pure reactants, is given in the following table for a production flow of one ton silicon per hour.

<table>
<thead>
<tr>
<th></th>
<th>for the first step</th>
<th>for the second step</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbon consumption, kg</td>
<td>855</td>
<td>855</td>
</tr>
<tr>
<td>Silica, kg</td>
<td>2139</td>
<td>2139</td>
</tr>
<tr>
<td>Electricity, kWh</td>
<td>7924</td>
<td>12915</td>
</tr>
<tr>
<td></td>
<td>4546</td>
<td>7590</td>
</tr>
<tr>
<td></td>
<td>3378</td>
<td>5325</td>
</tr>
</tbody>
</table>

The reaction-zone temperatures were maintained at about 1900°C in the example.

Examples of operation at a production rate of 1 ton Si per hour, per ton Si

cont'd
Plasma gas, kmol
for the first step  49  58
for the second step  34  34

Off-gas, kmol*
125  134

CO, mol fraction  0.938  0.938
N₂, mol fraction  0.062  0.062

Off-gas temperature, °C  \(\sim\)1250  \(\sim\)1210

* The plasma gas in these operating examples is recycled off-gas.

The matter contained in each of the following claims is to be read as part of the general description of the present invention.
CLAIMS
The claims defining the invention are as follows:

1. A method of continuously producing liquid silicon from carbon and silica, wherein in a first step a first quantity of the silica is converted with carbon to form silicon carbide, and wherein in a second, consecutive step a second quantity of the silica is converted with the silicon carbide produced in the first step, to form liquid silicon, thermal energy being supplied during both steps of the process.

2. A method as claimed in claim 1, wherein the first quantity of silica is chosen to be approximately 2/3 and the second quantity of silica is thus chosen to be approximately 1/3 of the whole.

3. A method as claimed in claim 1 or 2, wherein the temperature during the first step is maintained at at least 1900°C.

4. A method as claimed in any one of claims 1-3, wherein the temperature during the second step is maintained at at least 1900°C.

5. A method as claimed in any one of claims 1-4, wherein the first step is performed by introducing plasma gas with SiO₂ injected therein, into a bed to form a first reaction zone located at a level above a second reaction zone in the reactor containing primarily silicon carbide which has been produced in the first zone, the second step being performed in the second zone, plasma gas with silica injected therein being simultaneously introduced to said second zone.
6. An apparatus for performing the method as claimed in claim 1 for continuously producing liquid silicon from carbon and silica with the addition of heat, comprising a reaction vessel containing coal in lump form, means arranged in the vessel for the supply of coal in lump form, a pipe connected to the reactor vessel for the removal of exhaust gases therefrom, a plasma generator arranged in the upper part of the vessel for introducing plasma gas into the upper part of the coal bed in the reactor vessel to form a first reaction zone in the bed, as well as means for injecting silica into the first reaction zone, said apparatus further comprising a plasma generator located at a lower level in the reactor vessel, with means connected thereto for introducing silica into a second reaction zone formed by the plasma generator at the lower level, in a bed primarily of silicon carbide which has been produced in the first zone, and also an outlet in the lower part of the reaction vessel for tapping off the silicon produced.

7. An apparatus as claimed in claim 6, substantially as described herein with reference to the accompanying drawing.

8. Silicon when obtained by the method as claimed in any one of claims 1 to 5, or when obtained by use of the apparatus as claimed in claim 6 or 7.

DATED this 2nd day of June, A.D. 1988

SKF PLASMA TECHNOLOGIES AB,
By its Patent Attorneys,
E. F. WELLINGTON & CO.,
By:

BRUCE S. WELLINGTON
DRAWINGS