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For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

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(54) Title: THE METALLURGICAL METHOD OF RECEIVING THE HIGH PURITY SILICON POWDER BY CHEMICAL PROCESSING

(57) Abstract: The invention relates to the metallurgical method of receiving the high purity silicon powder by chemical refinement and allows using it for production of solar cell, and silicon hydride in case of a need. To achieve the object, this experiment of quartzite carbothermical reduction and processing of the obtained silicon by inorganic acid water solution, differs by the fact, that the initial material - quartzite is processed with inorganic acid and the reduction process includes silicon fining with the mixture of SiOz x CaFz in the ratio of 2:1 and 5-50% of the reducing agent's charge in a number. Aluminum is used as reducing agent and furnace is the base material of SiOz x CaO in the mass ratio of 1:3-6. This allows increasing the range of target product's purity.

The metallurgical method of receiving the high purity silicon powder by chemical processing.

An invention relates to the metallurgical method of receiving the high purity silicon powder by chemical processing, which can be used for production of solar cell.

There is a known method to receive the silicon powder, that includes processing of grinded technical silicon by 4-20 mass shares of HC1 solution, ablution and processing at the second stage by the solution of HC1 x HF in the following ratio of 10-24:1 (the patent RU #2049056 dated 27.11.1995, bulletin #33). Disadvantage of the above method is low range of silicon powder purity. The task of the invention is to increase the range of silicon powder purity and it's activity during the period of reduction and chemical processing. The metallurgical method includes the following stages of carbothermical reduction of quartzite and processing with inorganic acid water solution that help to achieve technical results. During this, quartzite is processed by inorganic acid and the reduction process includes silicon fining with the mixture of SiOs x CaFz in the ratio of 2:1 and 5-50% of the reducing agent's charge in number. Aluminum is used as reducing agent and furnace is the base material of Si02 x CaO in the mass ratio of 1:3-6.

For the purpose of silicon activity increase the alkali and alkaline-earth materials are additionally put into furnace materials at the amount of 5-60% of the reducing agent's charge.

Example 1. Implementation of the method.

1 kg of aluminum to pour in cmcible and turn with the heat. The furnace material, that consists mostly of CaO and preliminarily purified by Si02 inorganic acid is equal to ratio 1:1 at the quantity of 4.5 kg should be mixed with 400 gram of SiOa x CaFz in 2:1 ratio. Pour the received mixture on molten aluminum. Reducing agent's reaction is exothermic and is followed by temperature rise till 2000°C. On the completion of reducing reaction, when temperature falls, time for cooling. The obtained silicon is pounded, and this powder might be used for receiving silane gas (SFLt) in big numbers. The gas itself is a product to get the silicon of electronic quality. The last step is processing with the water solution of inorganic acid.

Table 1

№	Ma. ss shares		Si %	The ratio of refined mix and reducing agent		Si %
	Furnace	Reducing agent		SiO_2 : CaF_2	Reduc, agent Al	
1	3	1	98,6	5		99,9
2	4	1	99,2	20		99,99
3	4.5	1	99,4	40		99,996
4	5	1	99,5	50		99,996
5	6,5	1	99,2	60		99,992
the prototype						99,8

Example 2. Implementation of the method.

The proportion of furnace materials in Table 1 was used for receiving the silicon of high activity extent.

1 kg of aluminum to pour in cmcible and turn with the heat. The furnace material, that consists mostly of CaO and preliminarily purified by Si02 inorganic acid is equal to ratio 1:1 at the quantity of 4.5 kg should be mixed with 350 gr. of alkali and alkaline-earth elements and 400 gr. of StOi x CaP3 in 2:1 ratio. Pour the received mixture on molten aluminum. Reducing agent's reaction is also exothermic and is followed by temperature rise till 2000°C. On the completion of reduction reaction it's time for cooling. The obtained silicon is pounded and the powder's activity to be conducted by the following. 10 gr. of the product put into thermostable beaker, which is then to be filled with inorganic acid. An interaction of acid and silicon powder takes place by isolation of blown silicon hydride in the form of white soot SiOs. On the reaction completion the silicon powder residuum comes to 2.8 gr. that forms 28% of initial weight, and means that 72% of the product enters into reaction. The experiment results are shown in Table 2.

Table 2

№	The proportion of activating addictive and reducing agent		Silicon hydride output, %
	Alkali and alkaline-earth additives %	Redusing agent Al	
1	5		35
2	20		64
3	35		72
4	50		70
5	60		65

It is easy to get high purity silane from metallurgically received silicon powder by acid hydrolysis with some use low-temperature rectification.

The alloy activity preliminary appraisal illustrated that under the room temperature and standard atmosphere pressure conditions an output of silane, identified with mass-spectrums as monosilane (SiH^) amounts no less then 0.5 kg from the initial 1 kg of silicon powder. Synthesized silane cob contained an admixture of<10"^{4} mass shares, the phosphine and dibonrane were not detected in mass-spectrums.

CLAIMS

The metallurgical method of receiving the high purity silicon powder by chemical processing includes the carbothermical reduction of quartzite and processing with the inorganic acid water solution. It differs by the fact, that the initial material -quartzite is processed with inorganic acid and the reduction process includes silicon fining with the mixture ofSi02 x CaFs in the ratio of 2:1 and 5-50% of the reducing agent's charge in number. Aluminum is used as the reducing agent and furnace is the base material of SiOz x CaO at the mass ratio of 1:3-6. This method is also new, because in case of need to increase the range of silicon activity alkali and alkaline-earth materials are additionally put into furnace materials during the reduction process by 5-60% of the reducing agent's charge.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/KZ 03/00002

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C01B33/023 C22B34/20

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C01B C22B

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

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

EPO-Internal, WPI Data, INSPEC, CHEM ABS Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 089 010 A (HELIOTRONIC GMBH) 21 September 1983 (1983-09-21) page 3, last paragraph; example 1 ---	1
X	DATABASE CA 'Online! CHEMICAL ABSTRACTS SERVICE, COLUMBUS, OHIO, US; OSPANOV, ZHUMAGALI O. ET AL: "Method for silicon production" retrieved from STN Database accession no. 122:137493 CA XP002265236 abstract & SU 1 808 812 A (KAZAKHSKIJ KHIMIKO-TEKHNOLOGICHESKIJ INSTITUT, USSR) 15 April 1993 (1993-04-15) --- -/--	1

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
16 December 2003	04/02/2004

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31–70) 340–2040, Tx. 31 651 epo nl, Fax: (+31–70) 340–3016	Reedijk, A

INTERNATIONAL SEARCH REPORT

International Application No
PCT/KZ 03/00002

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
Y	DATABASE INSPEC 'Online! INSTITUTE OF ELECTRICAL ENGINEERS, STEVENAGE, GB; SHARMA I G ET AL: "Aluminothermic preparation of Hf-Ta and Nb-10Hf-1Ti alloys and their characterization" Database accession no. 7634810 XP002264730 abstract & JOURNAL OF ALLOYS AND COMPOUNDS, 17 FEB. 2003, ELSEVIER, SWITZERLAND, vol. 350, pages 184-190, ISSN: 0925-8388	1
Y	KELLER R: "PROCESS MAKES HIGH GRADE SILICON" , APPLIED OPTICS, OPTICAL SOCIETY OF AMERICA,WASHINGTON, US, VOL. 27, NR. 20, PAGE(S) 4346 XP000099063 ISSN: 0003-6935 the whole document	1
A	EP 0 024 614 A (CONSORTIUM ELEKTROCHEM IND) 11 March 1981 (1981-03-11) page 7, paragraph 1; claims	1
A	DATABASE WPI Section Ch, Week 199712 Derwent Publications Ltd., London, GB; Class E36, AN 1997-130645 XP002264731 & SU 835 063 A (IRKUT SECT ALUMINIUM MAGNESIUM ELECTRODE), 27 July 1996 (1996-07-27) abstract	1
A	DATABASE WPI Section Ch, Week 198524 Derwent Publications Ltd., London, GB; Class M25, AN 1985-143788 XP002264732 & JP 60 077942 A (NIPPON DENKO KK), 2 May 1985 (1985-05-02) abstract	1

INTERNATIONAL SEARCH REPORT

ational application No.
PCT/KZ 03/00002

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☒ Claims Nos.: -
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:

see FURTHER INFORMATION sheet PCT/ISA/210

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest.

☐ No protest accompanied the payment of additional search fees.

Continuation of Box I.2

Present claim 1 relates to an extremely unclear method. In fact, the claim contains so many spelling/language mistakes and unclarities, that a lack of clarity (and/or conciseness) within the meaning of Article 6 PCT arises to such an extent as to render a meaningful search of the claims impossible.
Consequently, the search has been carried out for those parts of the application which do appear to be clear (and/or concise), namely a method recited in the example 1 and closely related homologous compounds etc. The method searched is the aluminothermal reduction of silica in the presence of Calciumoxide and calciumfluoride in order to obtain a high purity silicon.

The applicant's attention is drawn to the fact that claims, or parts of claims, relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure.

INTERNATIONAL SEARCH REPORT

nformation on patent family members

Inte[redacted]onal Application No
PCT/KZ 03/00002

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