ISSN 0003-701X, Applied Solar Energy, 2018, Vol. 54, No. 2, pp. 85–90. © Allerton Press, Inc., 2018.
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DIRECT CONVERSION OF SOLAR ENERGY TO ELECTRICITY

Prospects for the Production of Silicon and Solar Energy Products in the Republic of Uzbekistan

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Received January 5, 2018

Abstract—The paper presents the results of a comparative analysis of the technical and economic parameters of the trichlorosilane and monosilane processes of silicon production. It is shown that Russia's available gangue quartz, quartz sands, natural gas, and new methods of smelting technical silicon, as well as new technologies for its conversion into monosilane, makes it possible to organize environmentally friendly industrial production of raw silicon and competitive, highly efficient photovoltaic energy products based on monosilane technology.

DOI: 10.3103/S0003701X1802007X

INTRODUCTION

The creation of Uzbekistan's own production of technical silicon (TS) based on domestic silicon-containing raw materials (SCRMs) has opened up fundamentally new possibilities for the production of silumin, rubber, high-quality steels, bronzes, etc., as well as for the organization of a chain of enterprises for chemical–metallurgical conversion of TS into electronic-grade raw silicon and then into monocrystalline (MCS) or large-block polycrystalline silicon (PCS), used directly for the manufacture of solar cells (SCs) [1]. One of the variants that in general coincides with those discussed in [2, 3] is given in [4]. It is important that in [4], the question of the competitiveness of solar energy was raised simultaneously raised in comparison with traditional methods of obtaining electric power and in the harshest operating conditions for its objects, i.e., photovoltaic modules (PVMs) and solar photovoltaic stations (SPVSs). It should be noted that in comparing competitiveness, costs were taken into account not only for the creation of solar cells, but also for the manufacture of the actual final products in the form of PVMs and SPVSs, i.e., load-bearing structures with modules or panels with SCs equipped with protective glass, and an inverter, and a battery housing. Estimates given in [1, 5] show that along with ~10 g of MCS used to manufacture a 1-W SC, the creation of an SPVS with all the infrastructure mentioned above requires specific costs of other materials: glass, duraluminum, steel, copper, tin, etc., with a total mass of 217 g, excluding the specific costs for concrete spent on making the foundations for the station's support devices, which are about 100 g per 1 W of the installation's power. The competitiveness of solar energy products increased sharply by 2010 due to the almost tenfold drop in raw silicon prices, which resulted when its producers switched en masse to new recycling technologies. This made it possible to significantly increase the yield of silicon obtained in the trichlorosilane (TCS) process, which resulted in a multiple reduction in prices for MCS and PCS SCs, as well as PVMs and SPVSs [1, 6]; it also led to a reduction in the severity of environmental problems associated with it.

Thus, under conditions associated with the drop in world prices for silicon, the competitiveness of newly created silicon plants in Uzbekistan can be ensured only by selection of the most productive and least energy-intensive technologies and reliance entirely on Uzbekistan's own raw materials. Note that for Uzbekistan to obtain its own end products, i.e., PVMs and SPVSs, in parallel to silicon production, it is necessary to expand factories that manufacture both SCs and other components—primarily, protective impact-resistant glass for PVMs at a unit cost of 9.6 kg per 1 m^2 of PVM area, as well as modern types of batteries and inverters with a high performance index.

Let us assess the preconditions existing in Uzbekistan for work in this direction and the already existing results.

RAW MATERIALS BASE AND THE FIRST METALLURGICAL PROCESSING

Uzbekistan has seen in its territory the discovery of a large number of deposits and occurrences of SCRMs in the form of gangue quartz (GQ) with a SiO_2 content above 95% with total reserves of different categories of more than 20 mln t [7].

Together with geologists, in addition to standard indirect assessments of the suitability of silica available there, we carried out direct assays by melting TS in an electric arc furnace (EAF) using samples from these deposits and standard carbonaceous reductants (CRs) [1, 8, 9]. The possibility of obtaining high-grade TS, i.e., Kp1 and Kp0 with a 97 and 98% Si content, respectively, using standard technologies [1, 10] was shown. In the same way, we evaluated the suitability of domestic quartz sands (QSs) for the production of TS of Kp00 grade with 98.8% Si content [11]. In [8, 9] and [11], respectively, detailed information is given on the content of all possible impurities, and not only those regulated by GOST, in TS manufactured by us on the basis of GQ. The particular value of these results and, accordingly, of raw materials is that both our measurements and independent expertise found either no boron impurity or only a trace level in the melted TS, which is important, since purification of TS from this impurity by refining and then by remelting raw silicon is a costly and inefficient operation.

We have also experimentally substantiated ways to solve a problem which is extremely acute for Uzbekistan's production of TS, FS and other metallurgical processes: replacing CRs in the form of coke, which is imported for hard currency. Studies [1, 12] present results illustrating the purity of TS at the level of Kp0 and Kp00 grades and minimum harmful impurities in ferrosilicon (FS) when the coke is replaced by gaseous CR in the form of natural methane gas, and it is concluded that the supply of CH_4 directly to the arc combustion zone allows TS and FS of any given grades to be obtained. Other variants of mixed feed are gaseous CR in the form of CH_4 and solid CR in the form of briquettes formed from SCRMs in a mixture with preheat-treated local coal, as well as from their mixtures with soot waste formed in fuel power production, e.g., at Novoangrenskaya GRES, which makes it possible to produce regular grades, conserve energy, and smelt any grade of FS without any restriction. The novelty of the developed technical solution is the fundamental change in the course of physicochemical reactions of SiO_2 reduction in EAF related to gaseous CR supplied directly to the electric arc combustion zone. In addition, methane on the feed is intentionally ionized, and water vapor can be fed along with it into the reaction zone. This makes it possible to regulate and significantly increase the reactivity of carbon and hydrogen atoms making up this CR, as well as to thereby increase the silicon yield and reduce the specific electricity consumption by 7–10% [9, 12].

In order to increase the profitability of the energy-consuming and ecologically unsuccessful process of first metallurgical processing, we also propose schemes for utilizing waste from the production of TS and FS in the form of microsilicon earth (MSE), which are conglomerates of nanosized particles on the order of 50–100 nm, and SiO_2 particles formed in the working zone of the EAF, the amount of which can reach up to 30% of the mass of the desired product.

The largest-scale effect can be obtained via MSE for the production of SiC micro- and, especially, nanopowders, based on plasmetallurgical technology (PMT) [16] and furnace synthesis (FSyn) [17]. PMT makes it possible to obtain SiC nanopowders with a purity of 91–94% and a size of cut particles of 60–70 nm, which have a minimum number of surface defects with a specific surface area of 36000–38000 m^2/kg, and also to impart to them specific electrophysical properties. At the same time, with the help of a FSyn, achieved with a particular MSE, i.e., under conditions where the synthesis rate is determined by the degree of grinding of the raw materials involved in the reaction, SiC can be obtained as a micropowder with a particle size of 200 to 900 nm or larger and a specific surface area of 3000–4000 m^2/kg. Due to the mentioned size effect, the temperature for synthesis of SiC from MSE is noticeably lower than in the traditional production of this material, as well as the duration of the process, which makes it possible to reduce the unit cost of electric power by almost a factor or 2. Note that SiC powders with a particle size of less than 1 μm, so-called micronized silicon carbide, are consumed mainly by the production of ceramics; nanosized SiC, i.e., with a particle size less than 100 nm, is in demand for the manufacture of high-quality structural ceramics and electroplating: it is used in a number of new, relevant, and promising fields of science and technology and is a very liquid commodity.

In summary, we believe that, based on local SCRMs, both in the form of GQ and briquettes made of QSs, it is possible for Uzbekistan to produce high-quality TS for subsequent conversion into electronic-grade silicon. At the same time, due to the replacement of coke, methanol increases the environmental purity of the process and reduces the specific energy consumption and, hence, the competitiveness of domestic enterprises. When mastering the synthesis of silicon carbide powders based on MSE, smelting of TS from the most cost-effective link of the technological chain [4] of solar products can become the most cost-effective.

PROCESSING OF TS INTO ELECTRONIC-GRADE RAW SILICON

The world production of raw silicon is based mainly on the TCS process, which, as already mentioned, was radically modernized at the beginning of the 21st century. It resulted in a sharp increase in production to 250000 t in 2014, a reduction in harmful emissions, and, as already mentioned, a tenfold decrease in prices, which entailed not only a reduction in the cost of SCs, PVMs and SPVSs, but also outstripping growth in the world production of these solar energy products [1, 6, 18–20].

Over 60% of the world's total silicon content is produced by the TCS process [1, 18–20]. This requires an answer to the question of the expediency of deploying this link of the technological chain [4] in Uzbekistan, and with a positive response to it, the competitiveness of domestic production. In favor of the decision to master the industrial technology for obtaining raw silicon in Uzbekistan is the above-described availability of high-quality SCRMs, new technologies for the metallurgical processing of silica into TS, as well as the experience of obtaining 60 t of it per year, which that had existed in Uzbekistan before 2000, and based on a slightly modified version of the Siemens–TCS process [1, 4], in which not trichlorosilane, but rectified silicon tetrachloride was used as the initial regent.

Reduction of harmful, chlorine-containing emissions into the atmosphere in the most modernized TCS process does not in any way reduce the danger to the environment of chlorine and hydrogen chloride production, which are the initial reagents for the manufacture of TCS. These are an integral part of the TCS process, and they will have to be created in the Republic of Uzbekistan, because for large-scale production of raw silicon, it is extremely dangerous to import and transport these reagents in significant quantities. In addition, the energy intensity of the modernized TCS process has remained at the previous relatively high level, since the production of raw Si by Siemens technology, as well as all processes used in the technology of silicon and silicon structures associated with hydrogen reduction of TCS, and particularly $SiCl_4$, are invariably carried out at temperatures of ~1500 K. The competitiveness of such domestic production will be extremely difficult, not to mention the export of products, the cost of which did not exceed US $12 per kg in 2010 [1, 19, 20]. Thus, the production volume for raw Si in China in 2013 reached ~68 400 t, which at that time was 30% of world production. Note that at the same time China imported 65 000 t of silicon per year for the needs of its photovoltaic industry, which at that time produced 79% of the world production of all types of silicon SCs and PVMs. In addition, in 2012–2013, in China many raw silicon producers reduced the production volume, stopped production, or even went bankrupt, unable to withstand competition due to high production costs against the background of a decline in world prices.

A significant share of production falls on South Korea with ~47 800 t of polysilicon per year, and Japan, in which raw silicon is produced by the following companies: Tokuyama, Mitsubishi, Osaka Titanium Technologies and M. Setek [6, 19, 20] with the use of the most advanced innovations in TCS technology.

Obviously, it is necessary to adopt an alternative solution that would make it possible to guarantee a reduction in the cost of raw silicon: in our opinion, it is the industrial development of monosilane (MS) production in Uzbekistan and then the MS process for obtaining raw silicon [1] with emphasis on the primary introduction of a technology option in which silicon is produced in the form of pellets in fluidized-bed reactors (FB-MS). The following arguments can be stated in favor of such a solution:

—All MS processes are ecologically flawless in comparison with TCS technology.

—The technological temperatures of any operations related to MS do not exceed 1000 K with speeds of obtaining desired products comparable with the TCS process, as a result of which their specific energy intensity is almost two times lower. At the same time, the minimum specific energy intensity, namely, ten times lower than in the TCS process with the deposition of silicon on target rods, is characteristic of the FB-MS process of obtaining silicon beads, which is carried out at a record low temperature for silicon technologies, 900 K, which makes it possible to obtain raw Si with a purity of 99.99999%, which is quite acceptable for creating the working body of an SC [6, 18–20].

—In Uzbekistan, fundamentally new technologies have been created for preparing the precursors of MS in the form of alkoxysilanes, as well as MS itself and a novel instrumental design for the most important operations.

Despite the clear advantages of the MS process in terms of environmental purity and lower energy intensity, the world production of raw silicon with its help in 2013 did not exceed 18%, although experts on the problem, e.g., from the International Technology Roadmap for Photovoltaics [21] predicted back in 2012 that the share of polycrystalline semiconductor silicon produced by the FB-MS will monotonically but steadily increase to 40% in the total production volume by 2023, while the share of the Siemens–TCS process, on the contrary, will decrease to 55% and apparently stabilize at this level in subsequent years.

The reason for the relatively slow rivalry, which stretched over a whole decade, of advanced MS technology with the obviously failing TCS process was not only the need to break established tradition and the associated material costs for re-equipping the existing polycrystalline semiconductor silicon production, the annual volume of which is 300 000 t, but also the difficulty of mastering, e.g., one of the most advanced low-energy MCS technologies [22, 23]. This technology entails the production of MCS precursors via the reaction of TS interaction with ethanol or methanol and subsequent isolation of MS itself, respectively, from ethoxysilane or methysiloxane, which in our designs was adopted as a basis and radically modernized.

As a result, we revealed the physicochemical nature of the so-called induction period of the reaction, the unpredictability of the course and duration of which reduced to nothing all known attempts to make the synthesis of alkoxysilanes manageable and continuous with this technology. The technical solution [24] in the synthesis of triethoxysilane developed by us generally eliminates the induction period and develops continuous selection of the reaction by-products from the reactor formed due to impurities in the initial TS. The novel synthesis technology of the MS itself [25], first, provides for the removal from the reaction products of unreacting ethanol and its azeotropes with triethoxysilane at atmospheric pressure and directly during condensation of the desired reaction products. Second, the disproportionation of ethoxy silane is carried out using freshly prepared sodium ethoxide as a catalyst at temperatures from 0 to 50°C. Finally, purification of the resulting MS is carried out by absorbing impurities in triethoxysilane at temperatures below −140° C, where the MS is fed to the absorber in the liquid state; the purification process itself proceeds in a straight-through mode. The entirety of these technical solutions, which, in addition to Uzbekistan's patents, are also protected by patents in China, Japan, the United States, South Korea, and a number of other countries, makes it possible to obtain MS in a continuous mode with ensuring high purity by impurities. Thus, measurements of mono- and large-block polycrystalline films grown from MS synthesized by us show that the total content of impurities in them does not exceed 10^{13} cm^{-3}, and this level is apparently limited by the purity used in the growth of films and hydrogen. In this regard, we note that when assembling the furnace load for processing raw silicon of various technological origin in MC for electronic engineering, the pellets obtained by the FB-MS method are usually mixed with raw silicon from the Siemens rods from the core, because the FB-MS contains more impurities than Si from the Siemens rods, which apparently is caused by the developed surface of granules, which sorbs impurities easier. Raw silicon of such technological origin is used directly for metallurgical processing into solar-cell-quality silicon. Taking into account the possibility of organizing in the Republic of Uzbekistan gross production of high-grade TS using new technologies [8, 11, 12, 14] and new technologies for synthesis of MS [24–26] using domestic TS and specially purified ethanol [27], it becomes possible to form a furnace load for smelting raw silicon into MCS for microelectronics based only on the FB-MS product.

In contrast to the TSC-based process, the production of MS-based raw silicon based on a lower-energy-intensity, higher-production culture, higher environmental purity, and a number of other advantages is a priori more competitive and can ensure the production of a quality product that has a much cheaper unit cost. This makes it possible not only to meet the raw material needs of Uzbekistan's solar energy industry, but also to organize the export of raw silicon. It should not be forgotten that the MS itself is a highly liquid product that may also be profitably exported. The availability of MA production in Uzbekistan opens essentially new possibilities in the manufacture of SCs.

EXISTING AND PROMISING DESIGNS FOR SOLAR CELLS BASED ON THE USE OF MONOSILANE

Modern designs [28–30] of voluminous SCs are concentrated in three alternative variants almost equal in efficiency, originality, and popularity [31–34]; they are realized from structures manufactured by MS technologies with a base made of MCS or PCS:

PERL structure (passivated emitter rear localized cell);

HIT structure (heterojunction with intrinsic thin layer structure);

BC–BJ structure (back contact–back junction structure).

An SC of the first modification, i.e., with the PERL design [28], is equipped with a thick MCS or PCS *p*-type base and a thin (0.2–0.3 μm) n^+-type layer made of PCS manufactured using the MS technology, as well as passivated front and back surfaces. In this type of SC, surface texturing is used, as well as an antireflection coating and a rear potential barrier. In practice, the entire photocurrent is generated in the MCS base of this SC and the record value of its efficiency is 25% [31, 32], which naturally depends strongly on the quality of MCS.

Reduction in recombination in the HIT structure is ensured by replacement of the *p*–*n*-junction with a (*p*–*i*–*n*)-heterojunction with a high-resistance *i*-interlayer. Instead of PCS, thin layers of amorphous silicon (a-Si–H) of *p*-, *i*- and *n*-conductivity types, grown on both sides of the MCS base, are used here [35]. The main role of the layers (a-Si–H) is an increase in the voltage U_{xx}, due to the large width (1.7–1.8 eV) of the forbidden a-Si–H band. An *i*-type layer compensates the effect of defects when films grow at the interface between the MCS substrate and *p*- and *n*-layers of a-Si–H. The record value of efficiency for a SCt of the HIT type was demonstrated by Panasonic (Japan): 25.6% [37].

SEs representing the so-called BC–BJ structure [38] is characterized by passivation of both the front and back surfaces; the presence of a broadband, textured, antireflection coating; a face potential barrier based on a thin n^+-layer; the arrangement of regions with *n*- and *p*-type doping on the rear surface. In the BC–BJ elements, the thickness of the photovoltaic layer should be comparable with the diffusion length of the minority charge carriers in the silicon used for their production [38]. Whereas in MCS this value is about 100 μm, in amorphous silicon it is only 150 nm. For this thickness, the ohmic losses of SCs decrease and U_{xx} decreases insignificantly; conversely, the duty cycle and, consequently, the efficiency increase.

Obviously, it is in these directions [31–38] that further improvement of SC designs will take place, and they themselves will continue to displace the types of SC most prevalent on the market with an efficiency of less than 20%, with AM 1.5 making up the current basis of "large" solar energy.

In Uzbekistan, it is equally important to develop MS technologies for so-called thin-film SC designs, used as accessories in household appliances, portable radio equipment, and in short-term or one-use power supply systems deployed in the event of natural disasters, etc. Only based on MS technology is it possible to fabricate an SC with an a-Si–H base that forms in thin films deposited on flexible steel or transparent bearing surfaces. Despite the small efficiency, ~ 10%, SCs and PVMs of this type have a quite extensive area of application and will be in demand in Uzbekistan's developing economy.

Finally, only on the basis of MS technologies it is possible to conduct promising scientific research on the creation of fundamentally new types of solar radiation converters like tandem SCs, SCs combining amorphous and MCS layers, structures containing nanoscale inclusions of the crystalline phase in an amorphous material, and composite a-Si–Hc luminescent coatings [39, 40]. An illustration of the real possibilities for mastering these technologies in Uzbekistan can be found in [41], which describes the preparation of silicon films with nanoscale inclusions made of domestic MS, also obtained from domestic TS, and shows ways to control the crystallinity and size of Si nanoparticles during their synthesis in a glow-discharge plasma.

CONCLUSIONS

Uzbekistan has all the conditions necessary for the industrial development of new domestic technologies for the production of TS, which are characterized by low energy intensity, ecological purity, and the possibility of obtaining a high-quality product based on Uzbekistan's own SCRMs and CRs. The new technological methods can also be successfully used in electric arc production of FS, silicocalcium, or silicomanganese.

In the Republic of Uzbekistan, new technologies for the synthesis of high-purity alkoxysilanes and MS based on the use of TS produced by Uzbekistan, are being developed, which may be used as a basis for the newly created industrial production of electronic-grade polycrystalline raw silicon, which will find application both in the manufacture of electronic products and the production of highly efficient SCs.

It is expedient to organize the manufacture of products for large solar energy in Uzbekistan based on MS technologies, which will make it possible to master the output of the most promising and highly efficient SCs with an efficiency of at least 20% with AM 1.5, simultaneously reducing the unit cost of materials and energy in comparison with other technologies.

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Translated by S. Avodkova