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Silicon Purification by a New Type of Solar Furnace *

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We propose a new method to reveal a direct transformation from solar energy to solar electricity. Instead of using electricity in the process, we use concentrated solar rays with a crucibleless process to upgrade metallurgical silicon into solar-grade silicon feedstock.

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Silicon is the most important raw material used in photovoltaic devices in modern times. The demand for the high purity silicon has greatly increased recently due to the rapid development of the solar photovoltaic (PV) industry.[1] Classically, silicon is fabricated using the Siemens process.[2] In addition, metallurgical purification methods have begun to replace the Siemens process in some PV wafer fabrication. All these existing techniques suffer from the drawback of high electrical energy consumption. In this Letter, a new method described here reveals a direct transformation from solar energy to solar electricity. Instead of using electricity in the process, we use concentrated solar rays with a crucibleless process to upgrade metallurgical silicon into solar-grade (SoG) silicon feedstock.

Attempts have been made in the past[3] to use classical solar furnaces to purify metallurgical grade silicon. The processes described there are difficult for commercialization partly because of the high cost of the classical solar furnace and partly because of the issue of crucible contamination.

Recently, Chen and his colleagues[4,5] proposed a new type of solar furnace consisting of a heliostat with actively controlled facets and a very small secondary concentrator as shown in Fig. 1. They also published a spinning-elevation tracking formula that allows only for the movements of rows and columns, realizing the aberration corrections to first order, in order to reach concentration ratios of 10000 or higher. This new type of solar furnace can harness solar rays with high efficiency and high cost-effectiveness.

Experiments have been carried out to use the above described solar furnace for incident concentration of solar rays into a solid rod, which comprises a mixture of metallurgical-grade silicon powder and metal oxides such as CaO, Al_2O_3, SiO_2 and others. The impurities, including boron, inside the silicon oxidize instantly in the high ambient temperature (1700–2000°C) and are removed by some processes: they may transit into liquid metal oxides or become directly vaporized. Unlike the common thinking that such a process would take a long time to complete, we find that the purification speed is rather fast, i.e. on the order of a few seconds. This feature not only makes natural solar rays a practical energy source for industrial use but makes a crucibleless process possible.

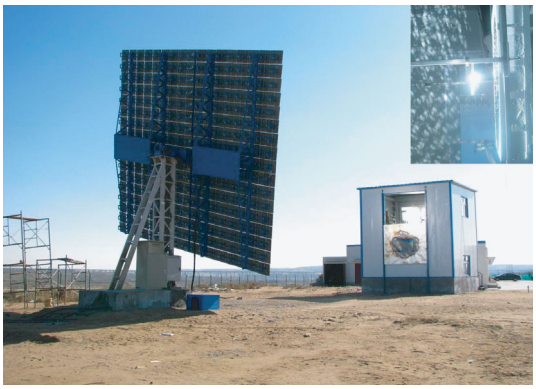


Fig. 1. A new type of solar furnace of aperture size 8 m × 8 m installed in the Inner Mongolia Autonomous Region of China. A non-imaging focusing heliostat consists of actively controlled facets impinging concentrated solar radiation onto a small secondary concentrator to achieve a 10000 times gain in intensity within the space of a tennis ball, as shown in the upper-right corner of the diagram.

There are three reasons for this ultra-fast purification speed. First, due to the instant heating effect of the photon radiation, the instantaneously melting part of the rod is rather small, so that the local temperature near the focusing point rises promptly (within a very short period of time, i.e., within seconds). The second reason is that, due to the poor thermal conductivity of the solid rod, the solar ray has to "peel

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off" the skin of the rod layer by layer, thus greatly increasing the reaction area. Thirdly, different from the classical refinement techniques where the process is conducted purely by thermal effects, in the process of the present work, the materials are exposed to very strong photonic radiation so that the chemical reaction speed becomes significantly faster than that of the normal reaction.

Even during daytime, solar radiation can often be interrupted (by clouds, etc.) with an uncertain time span, typically minutes or hours. That is the reason why most of people have historically believed that direct solar energy cannot be useful for this type of delicate process to purify silicon. However, a time span of a few seconds is much shorter than the usual period of interruption. During this short time frame, solar radiation can be considered to be continuous, thus making a solar process feasible. Moreover, as we are well aware, the nature of the intensity of solar radiation reaching the Earth's surface fluctuates. A successful chemical reaction using a non-constant power source must rely on a mechanism to maintain constant reaction temperature. In our research, we utilize a proper matching up of the composition of the metal oxides to maintain a phase changing-point of the mixture of the solid rod within a temperature range of 1700–2000°C. In this process, the temperature of the rod would not rise beyond this range because after the melting of the solid rod, the liquid will drip into a collector, draining out a portion of the absorbed energy and effectively keeping the temperature of the surface of the rod constant. On the other hand, when the sun's rays are unavailable for a period longer than the above reaction period, the temperature can fall naturally, and the purification process would stop but can be restarted within a few seconds when the sun's intensity is back to normal.

Table 1. Trace element analysis of the silicon feedstock refined by the solar process using a glow discharge mass spectrometer.

Element	Concentration (ppmW)	Element	Concentration (ppmW)
Li	< 0.005	Cr	< 0.005
B	1.1	Mn	< 0.005
Na	< 0.005	Fe	< 0.05
Mg	< 0.005	Co	< 0.005
Al	0.04	Ni	< 0.01
P	0.97	Cu	< 0.01
Sn	< 0.05	S	0.10
Sb	< 0.05	Ca	< 0.1
W	< 0.01	Ti	< 0.005
Pb	< 0.01	V	< 0.001
Bi	< 0.05	As	< 0.05
Zn	< 0.05	Mo	< 0.005

Indeed, without a container, the crucibleless process could not withstand long reaction times. We are still in favor of prolonging the reaction time if it is possible to ensure sufficient reaction. In our work, the free fall of the melt can extend the reaction time with an acceptable time span, e.g. 1.0 s to 1.2 s. The droplets consisting of both purified silicon and slag fall into a collector from a vertical distance of 5.5 m through a protection pipe. After that, the droplets are quenched by cold liquid. To ease the separation between the purified silicon shoots (mostly in the form of a spherical ball 1–10 mm in diameter) and the metal oxide slag, the liquid is prepared to be slightly acidic.

The process can be scaled up for mass production. A solar furnace consisting of a heliostat of 8 m × 8 m aperture size and a small secondary concentrator of aperture diameter 0.8 m has been installed in the Inner Mongolia Autonomous Region of China for industrialization testing. The above-described system can produce 3 kg of SoG silicon daily on average.

The collected silicon shoots would be acid leached prior to the directional solidification or Czochralski refinement process, to further remove other metal impurities. Trace element analysis using glow discharge mass spectrometry has been carried out a number of times, and the typical concentration of the remaining impurities of the silicon feedstock is shown in Table 1. Silicon feedstocks of 12 kg obtained by such processes have been employed to grow a monocrystal silicon ingot using the Czochralski method. Figure 2 shows the world's first monocrystalline silicon ingot grown using silicon feedstock prepared purely by the solar process. The ingot is 60% p-type and 40% n-type. The part that is p-type is cut and sliced to fabricate monocrystaline solar cells of 125 mm × 125 mm, using standard procedures. The efficiency of the cells ranges from 14.5% to 16.5%.



Fig. 2. The world's first monocrystaline silicon ingot grown using 100% silicon feedstock purified by the solar process.

The primary advantage of the new process is that its energy consumption can be as low as 30–40 kwh/kg with negligible emission of hazardous gases, unlike the traditional processes, which requires 200 kwh–400 kwh/kg and emit significant amounts of hazardous

gases that require considerable effort to deal with. Scaling up the process would thus have helpful global environmental impacts.

The second advantage of the new process is cost effectiveness. This could result in the present process becoming one of the major solar-grade silicon fabrication methodologies in the near future. At present, the fabrication cost of SoG silicon feedstock produced by the above method is in the range of 20–25 United States dollars per kg for small scale production. Analysis has shown that the major cost of the present process lies in the manufacturing of the solar furnace, but with mass production, cost reductions of up to half are feasible. Therefore, at a production rate of a few thousands of tons annually, the fabrication cost of SoG silicon feedstock by solar furnace using the crucibleless process illustrated here could be in the range of 10–15 United States dollars per kg.

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