LOWERING THE ENERGY CONSUMPTION OF THE CVD POLYSILICON PRODUCTION PROCESS FOR PHOTOVOLTAICS

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ABSTRACT

In this work, we analyze the energy consumption during the chemical vapor deposition (CVD) of polysilicon for Photovoltaic (PV) applications. Through theoretical models describing heat transfer and thermodynamics, computational fluid-dynamics modeling (CFD) and experimental research in a laboratory scale prototype reactor, possible energy savings strategies are identified.

Models for radiation, conduction and convection heat losses in Siemens-type reactors are developed, shaping a comprehensive model for heat loss. These models take into account the changing conditions over a deposition process and have been validated through experiments carried out in a laboratory scale reactor. Moreover, CFD modeling results for the laboratory reactor are in agreement with the previous ones. From the above, radiation heat loss is shown to be the greatest responsible for the high energy consumption of these type of reactors (~50% for industrial scale reactors). Also, the use of thermal shields and alternative deposition surfaces to maximize the volumetric deposition rate, such as hollow cylinders, are identified as important energy saving measures; and these are explored both theoretically and experimentally. We have demonstrated that energy consumption reductions per kilogram of silicon in the range of 30% are achievable.

INTRODUCTION

Solar energy is called to play a leading role in the energy mix for a high-efficiency sustainable energy future. Photovoltaic solar energy (PV) is the dominating technology converting solar energy into electricity; it has experienced a geometric growth in the last decade, and it is responsible for about 90% of the global solar power capacity installed. Furthermore, silicon-based PV technology accounts for the majority of the total world production: about 92% in 2014 [1]. The price of the silicon-based PV modules has decreased sevenfold over the last five years, and the cost of the installed silicon PV system is reaching the level of 0.7-0.9 \$/Wp [2].

One of the areas of research that can have a great impact on further improving silicon PV is the process to obtain polysilicon (ultrapure silicon feedstock, reaching purity grades higher than 99.999999%, 8N). Polysilicon accounts for between one fourth and one third of the total energy consumption for silicon-based technologies [3,4], greatly impacting on PV cost.

PV polysilicon ultimately refers to any grade of silicon usable in manufacturing solar cells. To obtain PV polysilicon, there was a vast effort on research and development (R&D) after the oil crisis in the 1970s, with many alternative paths being explored. Most of them did not reach an industrial scale, and PV kept relying on the traditional chemical route, the so called Siemens process [5,6,7]. Nowadays, the vast majority of polysilicon is still produced by the Siemens process -more than the 90%-; as this class of process is currently the only one available from technology suppliers and engineering firms [8].

In this paper, major milestones in our research in this field are described, the theoretical and the experimental work developed are summarized, and the most important results are exhibited.

NOMENCLATURE

c_p	[J /kg K]	Heat capacity at constant pressure
E_b	[W/m^2]	Emitted radiation of a black body
F_{i-j}	[-]	Coefficient dependent on geometric property values
h	[W/m^{2}K]	Convection heat transfer coefficient
J	[W/m^2]	Radiosity
k	[W/mK]	Thermal conductivity
Q	[W]	Heat power
r, θ, z	[m]	Cylindrical coordinates
S	[m^2]	Surface area
T	[K]	Temperature
T_∞	[K]	Free stream temperature
Special characters		
ε	[-]	Emissivity
λ	[m]	Wavelength
ρ	[kg/m^3]	Density of the gas mixture

POLYSILICON PRODUCTION PROCESS

Polysilicon production through the Siemens process consists of chemical vapor deposition (CVD) of high purity trichlorosilane (TCS) on a hot filament, leading to high quality polysilicon at the expense of high energy consumption [9,10]. TCS decomposition to polysilicon occurs on the surface of a number of U-shaped pure silicon rods at temperatures higher than 1000 ºC, while the reactor wall is kept cooled at temperatures below 400 ºC. This large temperature difference between the deposition surfaces and the reactor wall is required to avoid certain chemical reactions in the bulk of the gas that would lead to lower quality of the resulting material. The initially slim U-shaped rods are heated up by Joule's effect in a H_2 atmosphere until decomposition temperature, then TCS starts to be introduced in the reactor chamber (typically in a low percentage ~10% mol) and due to the reduction reaction the thickening of the rods begins. In Figure 1 a simplified sketch of a Siemens-type CVD reactor is presented.

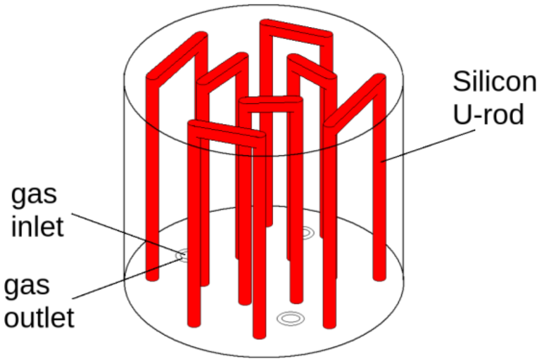

Figure 1 Simplified sketch of a Siemens-type CVD reactor. State-of-the-art reactors have 18 to 24 U-rods.

CVD processes are complex; heat transfer mechanisms, fluid-dynamics, kinetics and chemistry are involved [11,12,13]. The understanding of the fundamental reactions and how they influence the product quality is key, along with the comprehension of the phenomena responsible for the energy consumption, to aid further development in polysilicon CVD and seeking to reduce energy costs of the Siemens process.

RESEARCH STAGES

Research on PV polysilicon production through the chemical route using an industrial scale CVD reactor is very costly in time and money. Thus, laboratory scale reactors allow this research to be carried out in a more affordable way, provided extrapolation of results to the industrial scale can be clearly justified. At the Solar Energy Institute (IES) there is an in-house made laboratory Siemens reactor at our disposal. All working conditions of the CVD industrial process -except the pressure inside the reactor chamber- can be reproduced in the laboratory reactor; which is described in [14], together with the main aspects of the experimental procedure.

For several years, we have conducted experimental and theoretical research on PV polysilicon. On the one hand, more than 40 experiments have been conducted in the laboratory Siemens reactor. On the other hand, the problem of the energy consumption in a Siemens reactor is addressed from the theoretical point of view. Theoretical models for radiation, conduction and convection heat loss are developed; shaping a comprehensive theoretical model for heat loss in Siemens reactors. These models take into account the changing conditions over a deposition process and have been validated with the laboratory scale reactor. Moreover, computational fluid-dynamics (CFD) modeling of the laboratory reactor is developed, and results obtained are in agreement with the previous ones.

The comprehensive model for heat loss in a Siemens reactor becomes an important tool for PV polysilicon research. It allows to study the effect of different configurations, deposition conditions or some reactor modifications on the energy consumption per kilogram of silicon obtained (kWh/kg). Those theoretical proposals that result in appreciable energy consumption savings are tested in the laboratory reactor.

EXPERIMENTAL RESEARCH

Experiments conducted in the laboratory scale reactor are grouped together in five different batches. In all these experiments temperature measurements of the gas and the reactor structure are conducted with thermocouples, the rods temperature is monitored by means of a two-color pyrometer, the pressure is controlled and the inlet and outlet gas composition is analyzed using a mass spectrometer [14].

The first batch of experiments comprises some heating-up test conducted without deposition to cover different aspects related to the process control, electrical behavior, operation parameters or accuracy of temperature measurements, among others.

The second batch comprises a number of deposition experiments with different rod configurations. This batch of experiments is the first approach to the understanding of the polysilicon CVD from TCS under different deposition conditions. A proper heating of the rods and the adequate control of the temperatures of the whole system allowing polysilicon deposition are achieved. In addition, deposition yield dependence is studied by means of a mass spectrometer. In Figure 2, polysilicon rods obtained from some of these experiments are shown.

Figure 2 Polysilicon rods after deposition processes conducted in the laboratory Siemens reactor at 1000 ºC (bottom), 1100 ºC (middle) and 1150 ºC (top)

The third and fourth batches of deposition experiments consider an extra surface -thermal shield-, and a single rod and 7-rod configuration, respectively. Different thermal shields materials (silicon, alumina, stainless steel…) are tested in the laboratory Siemens reactor during polysilicon deposition experiments. Experiments with each configuration and the same deposition conditions without any shielding are also conducted; allowing to analyze the effect of the thermal shields. The concept of a thermal shield and the most important results obtained will be presented later on in this article.

Finally, the fifth batch comprises a number of experiments to study polysilicon deposition on different geometries more favorable than that of the state-of-the-art slim rod; this concept will be also presented later.

THEORETICAL MODELING

To address the challenge of energy consumption reduction of polysilicon CVD reactors, first, the heat loss problem associated with polysilicon CVD is studied. The energy consumption is obtained applying the law of conservation of (either the internal or total) energy to a volume element, and the relevant heat loss contributions to the Siemens reactor are identified. These phenomena are radiation, conduction and convection via gases, as reaction heat loss is not significant. In a Siemens reactor the major contributor to the energy consumption is radiation heat loss [15].

Models for radiation, conduction and convection heat loss in Siemens reactors are developed, which shape a comprehensive theoretical model for heat loss. These models are briefly described below, and they have been presented in detail in [16,17].

From the energy balance equation the temperature distribution in the gas surrounding a polysilicon rod is calculated. In particular, the internal energy equation is applied to a stationary volume element (VE), through which a moving fluid can be defined [18]. This volume element is a cylinder whose radius is defined by half the distance between either two seed rods; the center of the silicon rod is situated along the longitudinal axis of the cylinder and the reaction gases are flowing through the space between the rod surface and the cylinder limits (see Figure 3). For the present case, the internal energy equation for molecular and mass transport mechanisms applied to the gas region of the VE -expressed in terms of temperature- leads to Equation 1.

$$\rho c_P \frac{DT}{Dt} = -\nabla(k \nabla T) \qquad (1)$$

From Equation 1, convection and conduction heat loss phenomena must be studied together and none can be disregarded [19]. Radiation heat loss is calculated separately.

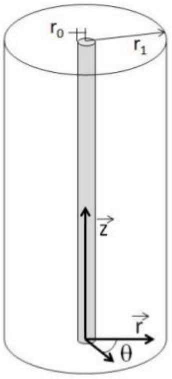

Figure 3 VE for a single rod in a Siemens reactor

Conduction heat loss modeling

Once the temperature distribution is obtained, conductive heat loss through the gases is calculated from the Fourier's law. Equation 2 shows the general expression for conductive heat loss in Watts per unit area at steady-state without internal heat generation. The boundary conditions needed to solve this equation are the temperature at the rod surface, $T(r = r_0) = T_0$, and at the volume boundaries, $T(r = r_1) = T_1$.

$$Q_{conduction} = -k \nabla T \qquad (2)$$

Convection heat loss modeling

Convective heat loss is also calculated starting from the gas temperature distribution; but first the convection coefficient needs to be calculated, for which the thermal conductivity of the solid surface and the thermal boundary layer (BL) need to be known. Thermal boundary layer calculations are not straightforward [20,21]. Heat transfer by convection, in Watts per unit area, can be calculated if the convection coefficient (h), the temperature at the rod surface (T_s) and the average temperature of the free stream (T_∞) are known, as expressed in Equation 3.

$$Q_{convection} = h \cdot (T_S - T_\infty) \qquad (3)$$

Different procedures exist to establish the heat transfer convection coefficient *h*, but exact solutions only exist for a few geometries and flow conditions [22]. This model for convection heat loss has found a known analytical solution, 'cylinder in parallel flow', acceptable to approximate the convection coefficient for the geometry and flow conditions under investigation. To the author's knowledge this is the first time such a model has been applied to convective heat loss in a Siemens reactor.

Moreover, flow conditions must be analyzed: in a Siemens reactor natural and forced convection coexist.

Radiation heat loss modeling

Radiation heat loss is calculated separately. It is strongly dependent on the surface temperature, as well as on the rods arrangement and the properties of all surfaces involved. The model for radiative heat loss calculations is described in detail and applied to different reactor configurations in [23,15]. Radiation heat loss is calculated through Equation 4, where *J* is the radiosity (the rate of outgoing radiant heat per unit area from a surface), *S* is the surface and F_{j-i} is called the configuration factor (the fraction of energy leaving a certain surface *i* that arrives at another surface *j*).

$$Q_i = S_i \cdot J_i - \sum_{j=1}^{n} S_j \cdot F_{j-i} \cdot J_i \qquad (4)$$

The calculation of the configuration factor is done using a geometric method called Hottel's crossed-string method [24], and the radiosity is obtained from Equation 5: *n*-equation systems being *n* the number of surfaces involved (*i= 1,…, n*).

$$S_i \frac{1}{1-\varepsilon_i} J_i - \sum_{j=1}^{n} S_i \cdot F_{i-j} \cdot J_j = S_i \frac{\varepsilon_i}{1-\varepsilon_i} \sigma T_i^4 \qquad (5)$$

This radiative model allows considering extra surfaces or different materials (defined by their optical properties) in the Siemens reactor, and studying the positive or negative effect on heat savings of such modifications. That can be the case of a thermal shield. If extra surfaces are considered, Equation 5 is substituted by Equation 6 and an extra equation per each extra surface needs to be added to the n-equation system (Equation 7, where *i = 1, ..., m-1*).

$$S_i \frac{1}{1-\varepsilon_i} J_i - \sum_{j=1}^{m} S_i \cdot F_{i-j} \cdot J_j = S_i \frac{\varepsilon_i}{1-\varepsilon_i} \sigma T_i^4 \qquad (6)$$

$$S_m \frac{1}{1-\varepsilon_m} J_m - \sum_{j=1}^{m} S_m \cdot F_{m-j} \cdot J_j = S_m \frac{\varepsilon_m}{1-\varepsilon_m} \sigma T_m^4 \qquad (7)$$

Further, a novel contribution to the radiation heat loss model has been developed; the emissivity variation with the wavelength is introduced. The new equations are similar to the ones presented above, but the parameter emissivity is going to be wavelength dependent, as well as the radiosity of each surface. Thus, radiation heat loss is now calculated integrating heat loss obtained for each wavelength along all the radiation spectrum, Equation 8 [14].

$$Q_i = \frac{\int_0^\infty Q_i(\lambda) E_b(\lambda) d\lambda}{\int_0^\infty E_b(\lambda) d\lambda} \qquad (8)$$

These theoretical models are validated experimentally, through the comparison of the power consumption calculated theoretically and the experimental data measured at the laboratory [19]. From the experiments conducted in the laboratory reactor, a number of them are selected; then the theoretical models are employed under the same set of conditions. The average measured radiation heat loss per unit area along a number of deposition processes compared to that for the theoretical model results in 3.6% difference, for the worst case. The difference between the average of the total power consumption measured and the predicted one by the theoretical models is below 4.0%.

CFD modeling

Taking advantage of both theoretical and experimental experience, a CFD model for the laboratory Siemens reactor is developed. Due to the cylindrical configuration of the laboratory Siemens reactor, the CFD model developed is well represented by an axisymmetric model. The geometry and the mesh have been developed by means of the commercial software ‘PATRAN’. The different domains defined are indicated: the rod (silicon), the casing (stainless steel), the fluid flow domain (TCS and H_2) and the cooling (water). The global CFD model accounting for all important aspects of heat transfer and gas flow has been developed using SiSim [25,26,27], a software dedicated to silicon production processes developed in the frame of the Norwegian center Solar United, and details of this model can be found in [28]. From this model, information about the fluid flow, the thermal fields and the energy consumption, is obtained. In Figure 4, the main domains of the laboratory Siemens reactor are indicated and the temperature distribution and fluid flow field resultant for a certain deposition conditions are presented.

Good agreement is found between experimental and simulation data (power consumption and temperature), the maximum differences found in the rod temperature, the inlet temperature and the outlet temperature are below 2, 0.5 and 12%, respectively.

From the laboratory to the industrial scale

Once the theoretical models are empirically validated, the radiation and convection heat loss models are extended to the industrial scale. In the laboratory scale Siemens reactor the radiation heat loss is responsible for 63.6-70.3% of the total energy consumption and conductive and convective phenomena are responsible for 13.7-16.6% and 7.2-9.3%, respectively. In industry, this numbers vary due to the improvement in compactness; e.g., while the relation between radiative and conductive heat loss for the laboratory reactor is 7.5-8.8, for the industrial scale Siemens reactors is 0.88-1.20.

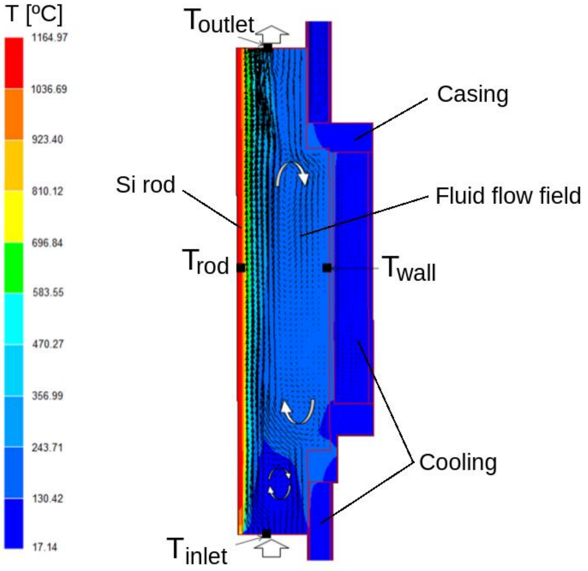

Figure 4 Axisymmetric temperature distribution and fluid flow field of the laboratory Siemens reactor (T_{rod} = 1150 ºC)

TOWARDS ENERGY CONSUMPTION REDUCTION IN CVD POLYSILICON

Among all the relevant results and knowledge gained during this research, we want to stand out here those that are translated into significant energy savings in the CVD polysilicon production process. Advantage is taken of the comprehensive model and several approaches to reduce heat loss in Siemens-type CVD reactors are proposed: thermal shields and new deposition surface concepts. These novel concepts are explored by means of theoretical calculations and tests in the laboratory Siemens reactor. Moreover, the relevance of the novel contribution to the radiation heat loss model described above is exposed.

Thermal shields

A thermal shield is a cylinder surrounding the polysilicon rods and placed between them and the reactor wall. The presence of this shield may block an important part of the radiated heat that otherwise is lost through the reactor wall (see Figure 5).

The potential of different thermal shields in an industrial Siemens reactor is first studied; the key parameter for the selection of the thermal shields material is the emissivity (ε). The potential savings are explored using the radiation heat loss model that takes emissivity variations with wavelength into account, which is important for materials that do not behave as grey bodies. The theoretical calculations confirm that materials with lower surface emissivity lead to higher radiation savings. Assuming that radiation heat loss is responsible for around 50% of the total power consumption, a reduction of 32.9% and 15.5% is obtained if thermal shields with constant emissivities of 0.3 and 0.7 are considered, respectively [29]. It is worth mentioning that the temperature reached by the different thermal shields is in all cases -and from the beginning of the process- above 850 ºC, which will result in polysilicon deposition on these surfaces. Thus, after a few minutes of deposition process the thermal shields surface emissivity will be 0.7, e.g., that of silicon.

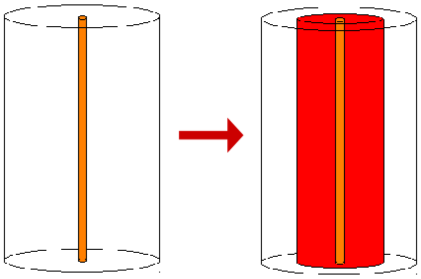

Figure 5 Sketch of the concept of a thermal shield

Since the temperature of the thermal shields in the laboratory Siemens reactor is lower than in the industrial case (no polysilicon deposition is expected onto the thermal shields), the laboratory reactor allows us to test the effect of thermal shields with different emissivities. Experiments considering different thermal shields conducted in the laboratory reactor confirm that the real materials do not behave as grey bodies, and prove that significant energy savings in the polysilicon deposition process are obtained. Using silicon as a thermal shield leads to energy savings of between 26.5-28.5%; very close to the theoretical calculations for the laboratory reactor [29].

Finally, for wavelength-dependent emissivities, the model shows that there are significant differences in radiation heat loss, of around 25%, when compared to that of constant emissivity [14]. The results of the model highlight the importance of having reliable data on the emissivities within the relevant range of wavelengths, and at deposition temperatures, which remains a pending issue.

New deposition surfaces

Polysilicon deposition onto different kind of surfaces and geometries has been proposed as a means to reduce the high energy consumption of the Siemens reactors [30,31]. As the energy consumption per kilogram of silicon produced is not constant along a conventional deposition process, the idea is to obtain a kWh/kg ratio as low as possible along all the deposition process by means of considering deposition surface geometries more favorable than that of the state-of-the-art slim rod; that is, taking advantage of the higher volume deposited per time unit when higher deposition area is available.

Here, the concept of the hollow rods is explored. This concept consists of replacing the characteristic slim seed rods of a Siemens reactor with hollow rods with an initial outer diameter larger than the one of the slim rods (see Figure 5).

Calculations carried out with the comprehensive model are applied to an industrial 36-rod Siemens reactor. A constant growth rate of 12 µm/min is considered, the rods height is 2 m, and initial and final diameters in scenario (a) are 0.7 and 15 cm, and 5 and 15 cm in scenarios (b.1) and (b.2), respectively. The thickness of the hollow rods is 0.5 cm.

The averaged kWh/kg ratio along a deposition process for the scenarios (a), (b.1) and (b.2) are 80, 53 and 48 kWh/kg, and the kilograms of silicon deposited are 62, 53 and 59, respectively. Scenario (b.1) presents ~34% kWh/kg savings and a productivity of 85%, and scenario (b.2) ~40% kWh/kg savings and a productivity of 95%. The lower deposition time and the larger volume deposited per time unit are responsible for the greatest energy consumption reduction in cases (b.1) and (b.2). However, the power consumed instantly -in Watts- in these cases is higher than in case (a); since heat loss is directly dependent on the surface area, that is bigger in cases (b.1) and (b.2).

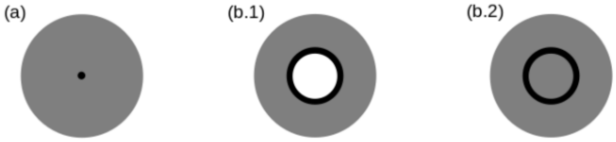

Figure 6 Sketch of the scenarios conventional slim rods (a), hollow rods with polysilicon deposition on the outer surface (b.1), and hollow rods with polysilicon deposition in both - inner and outer- surfaces (b.2)

A sensitivity analysis of the effect of the main parameters affecting the polysilicon production and energy consumption is performed. Increasing the working temperature and the growth rate -if possible-, the kWh/kg ratio decreases for the same productivity in all scenarios (the deposition process duration shortens).

Experiments to test the viability of new deposition surface concepts as the ones described previously were developed. These experiments still continue today due to the promising results so far; in short, homogeneous heating of a hollow rod until deposition temperature and polysilicon deposition onto different surface geometries has been successfully obtained.

CONCLUSION

Experiments conducted in the laboratory Siemens reactor at the IES have proven its satisfactory operation, and have allowed to acquire an important knowledge. Contributions to the energy consumption of the polysilicon CVD process are put forward. Phenomena responsible for the energy consumption are radiation, conduction and convection via gases heat loss. Of these, the major contributor in a Siemens reactor is radiation heat loss. Theoretical models for radiation, conduction and convection heat loss in Siemens reactors have been developed, shaping a comprehensive theoretical model for heat loss. This model has been empirically validated. A CFD model for the Siemens-type laboratory prototype has also been developed. Simulated results are in agreement with the previous ones, both theoretical and experimental ones.

Theoretical models allow to identify the parameters responsible for the greatest power consumption and, thus, suggest some modifications that could decrease the kWh/kg of silicon produced. An important potential to reduce radiation heat loss has been identified, by using thermal shields. We have demonstrated that energy consumption reductions per kilogram of silicon in the range of 30% are achievable. Also, novel surface deposition concepts have been explored -polysilicon deposition onto hollow rods and flat surfaces- that also can succeed in reducing the high energy consumption. There are intriguing advantages regarding to polysilicon productivity and energy consumption if considering polysilicon deposition onto these new surface concepts, which could lead to energy savings per kilogram of silicon produced up to 40%.

Summarizing, silicon PV is a moving target for competing technologies, as it continues to evolve, improve and reduce its costs. And the use of thermal shields and the deposition onto new surface concepts are promising paths to reduce the energy consumption in the polysilicon CVD process.

ACKNOWLEDGEMENTS

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