



Polycrystalline silicon thin-film solar cells: Status and perspectives

C. Becker^{a,b,*}, D. Amkreutz^a, T. Sontheimer^a, V. Preidel^{a,b}, D. Lockau^a, J. Haschke^a,
L. Jogschies^a, C. Klimm^a, J.J. Merkel^{a,b}, P. Plocica^a, S. Steffens^a, B. Rech^a

^a Helmholtz-Zentrum Berlin für Materialien und Energie, Institut Silizium Photovoltaik, Kekuléstr. 5, 12489 Berlin, Germany

^b Helmholtz-Zentrum Berlin für Materialien und Energie, Young Investigator Group Si-Nanoarchitectures for Photovoltaics and Photonics, Kekuléstr. 5, 12489 Berlin, Germany

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ABSTRACT

The present article gives a summary of recent technological and scientific developments in the field of polycrystalline silicon (poly-Si) thin-film solar cells on foreign substrates. Cost-effective fabrication methods and cheap substrate materials make poly-Si thin-film solar cells promising candidates for photovoltaics. However, it is still the challenge for research and development to achieve the necessary high electrical material quality known from crystalline Si wafers on glass as a prerequisite to harvest the advantages of thin-film technologies. A wide variety of poly-Si thin-film solar cell approaches has been investigated in the past years, such as thermal solid phase crystallization – the only technology that had already been matured to industrial production so far – the seed layer concept where a large-grained seed layer is epitaxially thickened, direct growth of fine grained material, and liquid phase crystallization methods by laser or electron beam. In the first part of this paper, the status of these four different poly-Si thin-film solar cell concepts is summarized, by comparing the technological fabrication methods, as well as the structural and electrical properties and solar cell performances of the respective materials. In the second part, three promising technologies are described in more detail due to their highly auspicious properties regarding material quality and throughput aspects during fabrication: (1) High-rate electron-beam evaporation of silicon for the low-cost deposition of high-quality material, (2) large-area periodic nano- and micro-structuring of poly-Si by the use of imprinted substrates providing a large absorption enhancement by a factor of six at a wavelength of 900 nm, (3) liquid-phase crystallization of silicon thin-film solar cells by electron-beam, yielding an excellent poly-Si material quality reflected by an open-circuit voltage of 582 mV which has been achieved only very recently. A successful combination of these three complementary technologies is envisaged to be the basis for a prospective low-cost and highly efficient poly-Si solar cell device.

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1. Introduction

Currently, the photovoltaic sector is dominated by wafer-based crystalline silicon solar cells with a market share of almost 90%. Thin-film solar cell technologies which only represent the residual part employ large-area and cost-effective manufacturing processes at significantly reduced material costs and are therefore a promising alternative considering a massive global deployment of photovoltaics in the coming decades. So far, polycrystalline silicon (poly-Si) thin films have not reached the maturity and performance required for market entrance. However, poly-Si layers exhibit distinct advantages since they could benefit from the advantages of the crystalline silicon wafer approach while maintaining the advantages of thin-film

technologies [1–6]: Crystalline silicon is a non-toxic material with high stability and durability, and possesses an energy gap of 1.12 eV which is nearly perfectly suited for maximum single-junction solar cell efficiencies. Furthermore, silicon is an abundant material and therefore not subject to natural resource limitations in case of a strong rise of the solar electricity demands in the future [7]. Fabricated as thin layers, polycrystalline silicon also features all advantages of thin-film technologies, namely low costs due to low material wastage with up to factor 100 less material compared to wafer-based solar cells, and the technically feasible monolithic fabrication of large area devices. With an appropriate light trapping concept crystalline silicon thin-film solar cells can principally reach single-junction efficiencies of more than 17% close to that of silicon wafer-based solar cells, as calculated by Brendel in 1999 [8]. Despite all those promising properties, poly-Si thin films did not establish itself on the photovoltaic market so far. CSG Solar, the only company that produced poly-Si thin-film solar cells on glass on industrial scale, fell victim to the crisis in the photovoltaic sector in the year 2011. The technology of CSG Solar relied on thermally solid phase crystallized silicon thin films on glass substrate [9] with a module

* Corresponding author at: Helmholtz-Zentrum Berlin für Materialien und Energie, Young Investigator Group Si-Nanoarchitectures for Photovoltaics and Photonics Kekuléstr. 5, 12489 Berlin, Germany.
Tel.: +49 30 8062 41392.

E-mail address: christiane.becker@helmholtz-berlin.de (C. Becker).

efficiency of 10.4% demonstrated on an area of $10 \times 10 \text{ cm}^2$ [10]—being up to now the world record of poly-Si thin-film submodules [11]. In the past years, research in poly-Si thin-film solar cells has considerably moved forward, providing a roadmap to higher efficiencies at which poly-Si will compete with incumbent technologies.

This paper reviews four technological methods for the fabrication of poly-Si thin-film solar cells on foreign substrates that have been subject of intensive research activities in the past years: The above mentioned solid phase crystallization of amorphous silicon layers by thermal annealing (Section 2.1), the so called “seed layer approach” based on epitaxial thickening of large-grained poly-Si seed layers (Section 2.2), the direct growth of fine-crystalline silicon layers at elevated deposition temperatures (Section 2.3), and the newly emerging liquid phase crystallization processes inducing the recrystallization of fine-crystalline silicon by a rapid heat-up above the silicon melting point at 1414°C (Section 2.4). These four poly-Si material types are compared in terms of structural and electrical properties, solar cell performance and technological complexity with regard to industrial production. We limit the scope on entirely crystalline silicon thin films prepared on foreign substrates by large-area thin-film deposition techniques. So called “microcrystalline” or “micromorph” silicon solar cell materials consisting of nanocrystallites embedded in an amorphous matrix [12–14] and silicon transfer techniques from wafers [15–17] are therefore excluded from this review.

The second part of the paper covers three technologies in poly-Si thin-film photovoltaics which are explored at Helmholtz-Zentrum Berlin and which exemplify emerging trends that have the potential to boost solar cell performance and throughput towards cost-effective high-efficiency devices. Section 3.1 describes electron-beam evaporation of silicon as an alternative high-rate deposition technique for poly-Si thin-film solar cells allowing for roughly 30-fold larger rates than conventional plasma enhanced chemical vapor deposition methods. The development of tailored light trapping structures is of great importance due to the inherent low absorption coefficient of thin crystalline silicon films. In Section 3.2 we will address highly absorbing large-area poly-Si microarchitectures by combining electron-beam evaporation of silicon with nanoimprint-lithographically structured substrates. In Section 3.3 electron-beam crystallization is discussed as a representative technique for liquid phase crystallization as a promising high-throughput fabrication method for high-quality poly-Si thin-film solar cells.

2. Status of poly-Si thin-film solar cells

This section summarizes four important approaches for the fabrication of poly-Si thin-film solar cells on foreign substrates, which have been subject to intensive research activities in the past years. In all poly-Si fabrication approaches described here, silicon films are grown by vacuum deposition techniques onto a foreign substrate, such as glass, and are aiming at an electrical material quality as close as possible to crystalline silicon wafer material. We do not give a complete overview of the entire literature, but highlight fundamental technological aspects, characteristic structural and electrical material properties as well as current record solar cell results in terms of efficiency and open circuit voltage.

2.1. Solid phase crystallization

2.1.1. Technological aspects

Solid phase crystallization (SPC) of amorphous silicon thin films by thermal annealing is a popular technique for the fabrication of poly-Si layers due to its technological simplicity. As only moderate temperatures around 600°C are required for the crystallization

process in a time span of several hours, cheap substrates such as glass or metal can principally be used. According to Bergmann [20] the substrate has just to fulfill three requirements: (1) It has to be stable upon poly-Si processing temperatures. (2) Diffusion of impurities into the silicon bulk has to be prohibited either by the inclusion of barrier layers or by the use of high-quality substrates. Usually this requirement results in the integration of barrier layers as pure substrates are often very expensive. (3) The thermal expansion coefficient has to be adapted to that of crystalline silicon in order to avoid cracking and flaking during thermal treatment.

2.1.2. Structural and electrical material quality

The typical microstructure of poly-Si thin films prepared by SPC is shown in Fig. 1a, exhibiting randomly oriented grains with a size of $1\text{--}3 \mu\text{m}$ if an amorphous substrate like glass is used. Nucleation, growth and resulting grain size can be influenced by experimental parameters, such as annealing time, temperature and substrate texture [20–22]. Raman spectroscopy has been used to evaluate the crystallinity of this poly-Si material [23] as shown in Fig. 2. The inset shows the respective Raman spectra for directly grown poly-Si (solid line) and e-beam crystallized material (dotted line). The full width at half maximum (FWHM) of the transverse optical phonon mode of solid phase crystallized silicon at 520 cm^{-1} as a measure for structural order is about 4.8 cm^{-1} in the as-crystallized state and 4.1 cm^{-1} after rapid thermal annealing (RTA), which is considerable larger than the respective value of a monocrystalline silicon wafer with 3.2 cm^{-1} . In the as-crystallized state, the quality of such SPC poly-Si material is not sufficient for photovoltaic applications as it contains many deep level intra-grain defects and grain-boundary defects, limiting the electrical performance. Post-crystallization treatments are necessary such as rapid thermal annealing (RTA) at temperatures above 900°C for the activation of dopants and to heal out extended defects [24–26], and hydrogen passivation in order to saturate silicon dangling bonds [27,28]. By a combination of both treatments the electrical material quality can be considerably enhanced. The open circuit voltage increases from about 150 mV up to about 500 mV and accordingly the density of paramagnetic defects which can be attributed to silicon dangling bonds decreases from about $1 \times 10^{18} \text{ cm}^{-3}$ to below $1 \times 10^{16} \text{ cm}^{-3}$ [29]. It has to be noted that these post-crystallization treatments also affect the FWHM of the TO phonon peak in the Raman spectrum, underlining their clear impact on the microstructural properties of the poly-Si thin-film material (Fig. 2).

2.1.3. Solar cells

Matsuyama et al. from Sanyo Electric Co. could achieve impressive solar cell results by using solid phase crystallized thin silicon films on metal substrates [30–32]. With an efficiency of 9.7% (active area) and an open-circuit voltage of 553 mV they could set a remarkable benchmark in the 1990s [31]. It was not possible to reproduce such high open-circuit voltages in SPC grown poly-Si thin-film solar cells down to the present day (Table 1). In their unique approach based on plasma-enhanced chemical vapor deposition (PECVD) they were able to grow $5 \mu\text{m}$ long columnar grains inside a thick ‘crystal growth layer’ by using a thin so called ‘nucleation layer’.

The record SPC poly-Si thin-film solar cell device has been developed by CSG Solar (formerly Pacific Solar) in Sydney [9] with an efficiency of 10.4% demonstrated in the year 2007 [10]. Here, a textured borosilicate glass with a thermal expansion coefficient very close to that of silicon is used as a substrate, on which a silicon (oxi) nitride diffusion barrier and an $1\text{--}2 \mu\text{m}$ thick n^+pp^+ -doped a-Si:H layer

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