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Fabrication of thin film silicon solar cells on plastic substrate by very high frequency PECVD

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ABSTRACT

The paper describes the way to transfer process technology of state-of-the-art high efficiency thin film silicon solar cells fabrication on cheap plastic (such as PET or PEN) substrates, by two completely different approaches: (i) by transfer process (Helianthos concept) of thin film silicon cells deposited at high substrate temperature, T_s (~200 °C) and (ii) direct deposition on temperature sensitive substrates at low T_s (~100 °C). Adaptation of the process parameters and cell processing to the requirement of the flexible/plastic substrate is the most crucial step. In-situ diagnosis of the plasma has been done to understand the effect of inter-electrode distance, substrate temperature and hydrogen dilution on the gas phase conditions. Whereas, for the transfer process, the inter-electrode distance is a critical deposition condition that needs to be adapted for the flexible substrates, the direct deposition on plastic substrates has an added issue of loss in material quality and the deposition rate due to depositions at low T_s. Our studies indicate that ion energy is crucial for obtaining compact films at low temperature and high hydrogen dilution helps to compensate the loss of ion energy at low substrate temperatures. Efficiencies of ~5.9% and 6.2% have been obtained for n–i–p type a-Si cells on PET and PEN substrates, respectively, using direct deposition. Using an adapted inter-electrode distance, an a-Si/nc-Si tandem cell on plastic (polyester) substrate with an efficiency of 8.1% has been made by Helianthos cell transfer process.

1. Introduction

Flexible electronics is getting momentum due to its advantages of flexible and unbreakable product, cheap substrate and its ease of handling and transportation. Various types of devices ranging from displays, sensors, markers to solar cells are being made on flexible substrates. Each device has an optimum process temperature that depends on the various constituents of the device and most crucially the optoelectronic material that is the central piece of such devices. For thin film silicon based solar cells, the silicon layers, i.e., amorphous silicon (a-Si) and nanocrystalline silicon (nc-Si) materials, are generally made at 200 °C, the optimum temperature at which the best electronic qualities (low defect density, higher structure order, high photosensitivity, etc) are obtained. This is related to optimum diffusion length of growth precursors such as SiH_3 on the growing surface [1]. Silicon films deposited at lower temperatures, for example, at 100 °C, are more porous and disordered; however, the structural quality of the films can be improved by providing extra energy through flux of atomic hydrogen and ions to the growing surface [2]. In this sense, plasma mediated growth processes, such as plasma enhanced chemical vapour deposition (PECVD) [3] has a definite advantage and very high frequency (VHF) PECVD [4] in combination with hydrogen dilution can deliver device quality materials at substrate temperature as low as 100 °C [5,6]. Deposition rate of nanocrystalline silicon has also received much attention in recent years due to its importance as a rate limiting step in the fabrication of multi-junction solar cells using nc-Si as one of the bottom cells in combination with a-Si top cell. It is a scientific as well as technological challenge to obtain high efficiency nc-Si cells at deposition rate ≥5 nm/s. Nanocrystalline silicon cells with efficiencies of 10% at 0.5 nm/s and 6.7% at ~5 nm/s [7] have been obtained on glass substrates at a substrate temperature (T_s) of 200 °C. At low temperature, the deposition rate becomes even more critical, both for a-Si and for nc-Si, due to slow diffusion of physisorbed species on the growing surface. Hence, the deposition process has to be adapted to meet the temperature limit that the substrate imposes. For flexible substrates, this is also the case, though the processing temperature and other conditions, mostly related to stress, depend on whether the processing is directly on the permanent substrate or on a temporary substrate and later transferred to the permanent substrate. In the first case, the glass transition temperature (or the deformation temperature) of the substrate is the deciding factor. Whereas cell processing directly on polyimide (also called Kapton) plastic substrates or metal foils such as stainless steel foil type of substrates can be made at the

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Table 1
Reported efficiencies at AM1.5 light condition of thin film silicon solar cells on plastic substrates.

Cell type	Substrate	Source	Eff. (%)	Ref.
n–i–p (a-Si/a-SiGe/nc-Si) D	SS	United Solar Ovonics, USA	15.4	[23]
n–i–p (a-Si/a-SiGe) D	Kapton	Fuji Electric, Japan	10.1	[24]
n–i–p (a-Si) D	PEN	IMT, Switzerland	8.7	[25]
n–i–p (a-Si) D	E/TD	AIST, Japan	6	[26]
n–i–p (nc-Si) D	LCP	AIST, Japan	8.1	[27]
n–i–p (a-Si/a-Si) D	PET	Univ. Stuttgart, Germany	4.9	[28]
n–i–p (a-Si) D	PET	Uinv. Utrecht, Netherlands	5.9	This work
n–p (mono-Si) T	Plastics	Univ. Stuttgart, Germany	14.6	[29]
p–i–n (a-Si) T	Polyester	Univ. Utrecht/Nuon, Netherlands	7.7	[22]
p–i–n (a-Si/nc-Si) T	Polyester	Univ. Utrecht/Nuon, Netherlands	8.1	This work
n–i–p (a-Si/a-SiGe/a-SiGe) DT	Polymer	United Solar Ovonics, USA	9.7%*	[30]

SS: stainless steel; E/DT: tetracyclododecene co-polymer; LCP: liquid crystal polymer; PEN: polyethylene naphtalate; PET: polyethylene terephtalate; Kapton: polyimide; *AM0 light condition; D: direct deposition; T: transfer method; DT: direct deposition+tranfer.

standard processing temperature ($\sim$200 °C) of thin film silicon cells, the deposition on cheap plastic substrates such as polyethylene terephtalate (PET) needs much lower processing temperature due to its glass transition temperature of 70 °C. There are, however, a number of plastic materials to choose, which can withstand intermediate temperatures: polyester sulphonate (PES), polyethylene naphtalate (PEN), thermoplastic ethylene–tetracyclododecene co-polymer (E/TD), liquid crystal polymer, etc. (see Table 1 and references therein). In the second case, the processing temperature is not a critical issue as the cell processing takes place on a substrate that can resist heat above the standard processing temperature of the cell, i.e., 200 °C. The transfer of the cell from a temporary to permanent substrate is the critical issue here. The advantage of this processing is that almost any permanent substrate can be employed, as the transfer processing takes place at room temperature. Examples of such processes (see Table 1 and references therein) are (i) the fabrication of monocrystalline n–p junction on c-Si wafer followed by transfer to plastic substrate and (ii) deposition on metal foil (Al) followed by transfer to plastic substrate (Helianthos concept). In this paper we will describe the processing of high efficiency solar cells, with the aim of fabricating a-Si/nc-Si tandem solar cell on plastic substrates. This type of solar cells on TCO coated glass gives initial and stabilized efficiencies of 12% and 11.4%, respectively [7]. We will show what sorts of adaptation to cell processing are needed to make such cells on plastic substrates by direct and transfer methods.

2. Experimental

The silicon films were made in an ultra high vacuum (UHV) multi-chamber deposition system called ASTER [8], by very high frequency plasma enhanced chemical vapour deposition (VHF PECVD). In-situ monitoring tools, such as optical emission spectroscopy (OES) and voltage–current impedance (V–I) probe, were employed to record and monitor the gas phase processes and the delivered power to the reactor, respectively.

For fabrication of a-Si solar cells directly on plastic substrates, the i-layer was amorphous silicon made at 100 °C, high hydrogen dilution (H_2/SiH_4) of 15 at the edge of transition to nanocrystalline material (the so-called "proto-crystalline silicon regime"), whereas the doped layers were either amorphous or nanocrystalline. The VHF plasma frequency was 50 MHz and the inter-electrode distance was 27 mm. The amorphous silicon doped layers were made with a high dopant concentration in the gas phase. Doped layers were developed at substrate temperature, T_s=100 °C, taking the i-layer recipe as a basis. Band gaps of the doped layers E_{04} were around 2.07 eV. Solar cells were made in the substrate configuration, substrate/Ag/ZnO:Al/n/i/p/ITO/Au grid-lines. Three types of cells were made:

(1) substrate/Ag/ZnO:Al/n-nc-Si/i-Si/buffer layer/p-nc-Si/ITO/Au (*single doped n-layers of nanocrystalline type*),
(2) substrate/Ag/ZnO:Al/n-nc-Si/n-a-Si/*i*-Si/buffer layer/p-nc-Si/ITO/Au (*double n-layer*) and
(3) substrate/Ag/ZnO:Al/n-nc-S/n-a-Si/*i*-Si/p-a-Si/ITO/Au (*double n-layer+amorphous p-layer*).

For the fabrication of a-Si/nc-Si solar cells by transfer process, we used the Helianthos concept [9]. The module fabrication by the Helianthos concept goes through various steps starting with (1) the transparent conducting oxide (TCO) layer deposition on Al foil by a roll-to-roll process, followed by (2) p, i and n silicon layers by radio frequency plasma enhanced chemical vapour deposition (RF PECVD) roll-to-roll deposition, (3) deposition of a ZnO:Al/Al back reflector, (4) roll-to-roll monolithic series integration, (5) transfer to permanent polymer substrate through roll-to-roll lamination, (6) removal of the temporary superstrate by wet etching in a roll-to-roll process and (7) roll-to-roll encapsulation. For the laboratory cell-on-foil studied here, only step (1) has been done in a roll-to-roll process to deposit SnO_2:F layer by atmospheric pressure chemical vapour deposition (APCVD) on Al foil at Helianthos b.v. [10]. All the silicon layers were deposited in laboratory batch type reactors in ASTER at Utrecht University, on 10×10 cm^2 area substrates, cut out from the 35 cm wide TCO coated foil roll. Whereas a-Si for the top cell is made at 50 MHz at a substrate-to-electrode distance of 27 mm, nc-Si for the bottom cell is made at 60 MHz at variable inter-electrode distances of 6–12 mm. The single junction a-Si cell as well as the tandem a-Si/nc-Si cell was deposited in a superstrate structure on the TCO coated Al foils.

The solar cells were characterized by current density–voltage (J–V) measurement in dark and under AM1.5 100 mW/cm^2 white light condition by a dual beam solar simulator (Wacom, Japan) and spectral response measurement from which external collection efficiency (ECE) was obtained. The cells were degraded under approximate AM1.5 light condition at 50 °C.

3. Results and discussion

3.1. Fabrication of thin films silicon cell directly on plastic substrates

In order to make a-Si/nc-Si type tandem cell directly on plastic substrates, it is not just enough to take the recipe of our state-of-the-art a-Si/nc-Si cell, which gives 12% initial efficiency