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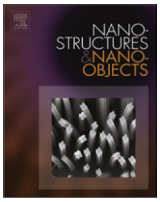

Nanowire polymer transfer for enhanced solar cell performance and lower cost

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HIGHLIGHTS

- An easy approach to substrate reuse for nanowire solar cells is demonstrated.
- Lift-off of nanowires embedded in a polymer layer is used to enable reuse.
- By adding a mirror to the nanowires in the polymer, photoluminescence is enhanced.
- A path towards lower cost and higher performance is simultaneously shown.

GRAPHICAL ABSTRACT

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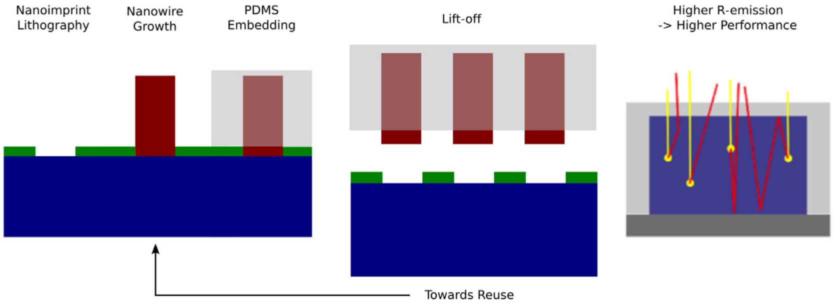

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ABSTRACT

The diffusion of photovoltaic systems is deterred by the struggle in combining high efficiency and low cost. Nanowire devices have been demonstrated to hold great promise to solve this predicament, but the substrate cost is still an unavoidable obstacle. Repeated nanowire growth on a single substrate is demonstrated by embedding InP nanowires in a polymer layer and removing them from the substrate. Our approach promises cost reduction by using the substrate multiple times. In addition, it provides a pathway to increase the open-circuit voltage by placing a mirror at the backside of the cell.

1. Introduction

Solar cell market acceptance is limited by issues in overcoming the necessity to combine low cost and high efficiency. Nanostructures can improve performance in several aspects of this topic [1–3], and in particular nanowires are great candidates to solve this problem [4], thanks to intrinsic higher performance [5–7], extraordinary absorption-to-volume ratio [8,9] leading to reduction of the amount of material consumed, and tolerance to lattice-mismatched structures [10–12]. InP nanowires, in particular, have demonstrated an exceptional potential for high efficiency [13–16]. Since the cost of the substrate is still a core issue though [17], a promising pathway to reduce the fabrication costs is the reuse of the substrate by repeated growth on a single wafer. In thin film growth, several techniques are used, such as epitaxial lift-off (ELO), substrate spalling, and the employment of porous release layers [18]. All these methods share one common trait: they all rely on the presence of a thin (as much as possible) sacrificial layer, either epitaxially grown or modified *ex-situ*, which is removed by etching or by mechanical means, and thus result in material waste. Nanowires instead allow a more elegant solution, as they can be

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easily embedded in a polymer layer such as Polydimethylsiloxane (PDMS). The layer of nanowires embedded into PDMS can subsequently be mechanically removed by lift-off [19,20], allowing a new nanowire growth on the same substrate. This approach has been shown previously [21] for microwires. Due to their larger dimensions, though, they are more easily removed from the substrate. Furthermore, since the microwire/substrate surface is much larger, tolerance to defects introduced by the lift-off and cleaning procedure is higher. In addition, microwires were grown by a metal-catalyzed vapor–liquid–solid (VLS) mechanism, which results in additional processing complexity, since the catalyst has to be redefined every time after wire removal. Selective area (SA) growth of nanowires [22–24] is instead particularly suitable for reuse, as after the substrate is cleaned from polymer residues and the bases of the nanowires are removed, the growth mask is promptly ready for another growth cycle. Moreover, embedding the nanowires in a polymer allows the placement of a back-mirror, which is known to increase the optical length of the absorber layer in solar cells [25]. At the same time, the optical emission can be expected to increase thanks to increased reflection of re-emitted photons, leading to an increased implied solar cell performance [7]. Here, we show the reuse of the substrate for SA-grown nanowires and an improvement of the optical performance of transferred wires.

2. Results and discussion

2.1. Substrate reuse procedure

The procedure for repeated nanowire growth is presented in Fig. 1a. After patterning a (111)A InP substrate with nanoimprint [26,27], wurtzite InP nanowires are grown by the selective area method in the openings in a SiN_x mask [6]. Although these specific nanowires are wurtzite, our conclusions apply to any kind of nanowires. The nanowires are then embedded in PDMS and the whole layer is removed from the substrate [19,28]. After lift-off, a residue persists on the substrate, consisting of the remains of PDMS and InP NW bases, which must be cleaned. In the case of selective area InP growth, special care must be taken to avoid dissolution of the SiN mask and to prevent etching the underlying InP (111)A substrate that could result in uneven growth. Favorably, PDMS can be etched at room temperature by a fluoride ion-based solution, tetra-butyl-ammonium fluoride (TBAF) [29]. TBAF must be diluted with a solvent that can dissolve the etched products, such as dimethyl-formamide or N -methylpyrrolidinone (NMP): in our case, the latter is used. This solution does not etch either SiN or InP, as required. To remove the base parts of the nanowires, a solution of HCl: H_2O 1:1 was used, which is known to etch the [111]B facet, as well as {100} and {110} facet families of cubic InP, but etches the [111]A facets extremely slowly, effectively leaving them untouched [30]. InP SA nanowires have {1120} lateral facets, corresponding to {110} facets in cubic notation, which are quickly eliminated, so that the nanowires are removed but the [111]A top surface of the substrate is preserved. At the same time, HCl does not attack the SiN_x mask, which remains intact. After the cleaning procedure (see SI for more information), the substrate is ready for another growth, in which the growth yield is unaffected by the cycle (Fig. 1b). The whole procedure leaves the substrate and the growth mask effectively untouched, so that its properties are unchanged and regrowth can occur again. The cleaning process is very gentle, but can be optimized further, for instance by introducing a sacrificial resist layer at the bases of the nanowires to facilitate lift-off, or by using a different polymer that is easier to remove and clean.

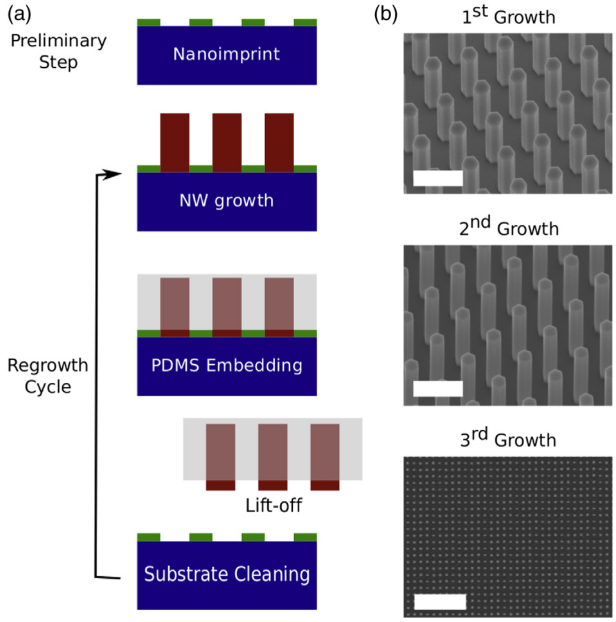

Fig. 1. Regrowth cycle. (a) After patterning the substrate by nanoimprint, growth takes place in the openings of the mask. Nanowires are then embedded in PDMS and removed by mechanical lift-off. Nanowires embedded in PDMS can also be processed towards flexible solar cells or other devices. After cleaning the substrate from residues of the polymer and the nanowire bases, regrowth is possible again on the same substrate. (b) SEM images of the sample after the first, second, and third growth. First and second image are taken at a 30° tilt. In the third case, a top-view SEM is shown to highlight the homogeneity and 100% yield of the sample. Scale-bars are 500 nm, 500 nm, and 5 μm respectively.

2.2. Enhanced photoluminescence

Using this procedure, the repeated reuse of expensive substrates is enabled. In addition, the development of flexible nanowire solar cells with a highly reflecting back mirror is facilitated with this approach [31]. The simplest way to obtain a flexible nanowire solar cell is to lay a transparent conductive oxide on the front side of the device facing the sun, and to deposit a reflective electrical contact on the backside, which will additionally act as a back-mirror. The first advantage of a back-mirror is an effective doubling of the absorption length for solar photons, which allows making the nanowires half as long for the same photocurrent generation. The second advantage is an enhanced luminescent emission at the front side of the cell, which results in a positive impact on the open circuit voltage of a solar cell. In the presence of a substrate, downward emitted photons can be reabsorbed by the underlying bulk substrate, in which they non-radiatively recombine or are re-emitted in a random direction (Fig. 2a). The reflectivity at the NW/substrate interface is in fact almost negligible as can be estimated using the complex Fresnel equations (see SI). Effectively most of the photons that travel from the wires into the substrate will be lost thus, because both the internal and external radiative efficiency of the substrate are much lower than that of the nanowires, due to both the lower crystal quality of the semiconductor substrate, and the geometry of the system that reduces escape probability [7]. If a (Ag) mirror is positioned below the nanowires instead of the substrate, most of the photons resulting from radiative recombination within the nanowires are reflected back, as the calculated NW/Ag reflectance is ∼95%. In this configuration, the solid angle of the emission is halved, as no photons escape from the backside, resulting in an increase of the front-side external radiative emission efficiency. As the radiative emission wavelength is very close to the bandgap, reabsorption within the nanowires is rather low, while in the case of the substrate nearly all emitted light is lost because the wafer is very thick compared to the nanowires and the band gap energy of the cubic substrate is lower than that of the wurtzite nanowires [6]. To experimentally prove this enhancement, two samples grown under identical conditions were fabricated: a first sample in which the nanowires are embedded in PDMS and are still on the substrate, and a second sample in which the nanowires/PDMS layer is lifted-off, and a 300 nm thick Ag mirror is deposited on the back by evaporation. The nanowires are 4 μm long and have a diameter of 180 nm. These dimensions have been chosen on purpose to exclude any effect of the absorptance on this test (discussed later), and allow a straightforward explanation of this phenomenon. The photoluminescence (PL) is measured by using a 100x objective focusing a 532 nm laser to a spot with a diameter of approximately 2 μm. In Fig. 2b the PL intensity of the two samples is then compared. A clear increase in the measured front-side emitted PL is observed. The back-mirror increases the collected emission by 60% at the front-side as compared to the sample without a back-mirror, which corresponds to an enhancement in the V_{oc} of ∼12 mV. This increase also guarantees that the high optical quality of the nanowires is maintained by the procedure. As presented before in more detail [31], the front-side emission, measured as the intensity of photoluminescence, is in fact directly correlated with the open circuit voltage of a solar cell by the relation

$$V_{oc} = V_{oc,ultimate} - \left| \frac{k_B T}{q} \ln \left(\frac{\eta_{ext} \varepsilon_{sun}}{\varepsilon_{emission}} \right) \right|$$

in which ε_{sun} is the solid angle of the sunlight falling on the solar cell ($\varepsilon_{sun} = 6.85\ 10^{-5}$) and $\varepsilon_{emission}$ is the solid angle of the light being emitted from the solar cell [32]. The solid angle of the emitted light is 4π for the nanowires embedded in PDMS without a back-mirror, while it is 2π for the sample with a back-mirror. As can be seen from the equation, the open circuit voltage increases when all emission in the backward direction is reflected back to the top. As demonstrated by several groups, such an enhancement in the optical emission is directly correlated to an increase in performance [33,34]. While we do not demonstrate experimentally this increase, the published literature strongly supports our conclusion. Furthermore, no degradation of photoluminescence has been observed after a complete regrowth cycle. Finally, to exclude any influence of the mirror on the absorption of the incoming illumination, the absorptance has been measured. According to simulation data [9], all light is expected to be absorbed in both samples since the NW length and diameter are ∼ 4 μm and 180 nm respectively. No observable effect of the silver mirror can be thus expected on the absorptance. This is verified by using an integrating sphere with a white light source. The reflectance is collected by the integrating sphere and is measured by a spectrometer, allowing to calculate the absorptance. In accordance with the simulations, the measured absorptance is very high in the visible range, and very similar between the two samples (inset of Fig. 2), with a value of ∼ 95% at 532 nm, which is the wavelength of the laser used for the PL measurements. As intended thus, while no influence of the back-mirror is expected on the absorptance, its effect on photon emission is clear.

3. Conclusion

To conclude, a procedure has been developed to enable repeated nanowire growth on a single substrate. By reusing the substrate, its influence on the total cost of the device is reduced. The nanowires are embedded in a PDMS layer and detached from the substrate: after cleaning, the substrate is ready for another growth and embedding cycle. We demonstrated a high yield for nanowire regrowth by repeating the complete cycle twice. Removing the nanowires from the substrate also allows adding a back mirror, which not only doubles the effective absorption length, but in addition allows increasing the open-circuit voltage. This process results thus in three advantages: it allows substrate reuse, opens a route towards nanowire-based flexible photovoltaic devices, and enhances the performance of solar cell devices.

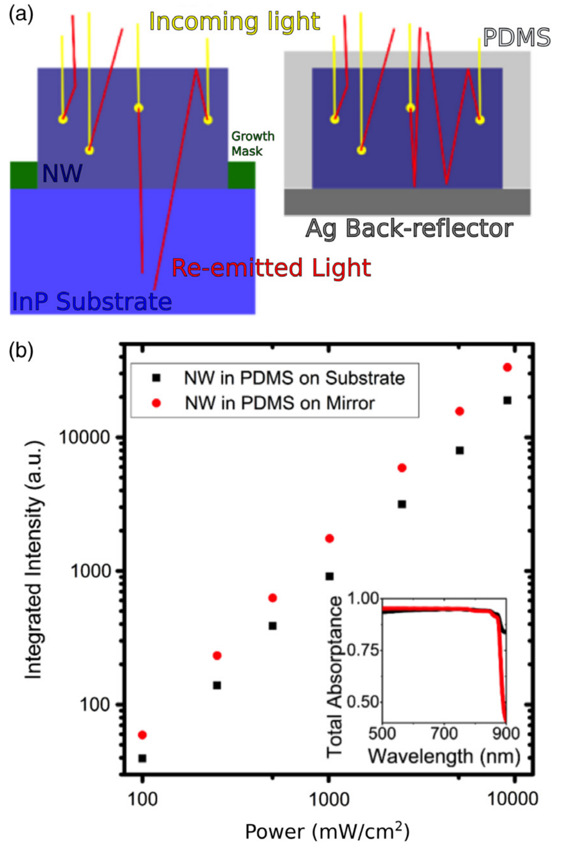

Fig. 2. Influence on the optical properties of the nanowire sample with either a mirror or the substrate placed at the backside of the nanowires. (a) Effect of the presence of the substrate on the front side emission. The emission should be doubled, in a lossless cell. (b) Integrated photoluminescence intensity of nanowires embedded in PDMS on the native growth substrate, and after polymer lift-off with a 300 nm Ag mirror on the backside, as a function of incident laser intensity. Inset: Absorptance of nanowires embedded in PDMS on the native growth substrate, and after polymer lift-off.

Notes

The authors declare no competing financial interest.

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Author Contributions

A. C. conceived and designed the experiment, as well as fabricated and measured the samples, and wrote the manuscript. A.D. designed and performed the integrated sphere measurement. J. E.

M. H. and E. P. A. M. B. provided the theoretical background to the study and the result interpretation, as well as supervised the study and strongly supported and encouraged its development. All authors reviewed, discussed and approved the results and conclusions of this article.

Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.nanoso.2018.03.014.

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