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REVIEW

Transfer Printing Methods for Flexible Thin Film Solar Cells: Basic Concepts and Working Principles

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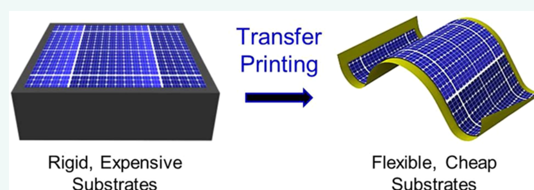
ABSTRACT Fabricating thin film solar cells (TFSCs) on flexible substrates will not only broaden the applications of solar cells, but also potentially reduce the installation cost. However, a critical challenge for fabricating flexible TFSCs on flexible substrates is the incompatibility issues between the thermal, mechanical, and chemical properties of these substrates and the fabrication conditions. Transfer printing methods, which use conventional substrates for the fabrication and then deliver the TFSCs onto flexible substrates, play a key role to overcome these challenges. In this review, we discuss the basic concepts and working principles of four major transfer printing methods associated with (1) transfer by sacrificial layers, (2) transfer by porous Si layer, (3) transfer by controlled crack, and (4) transfer by water-assisted thin film delamination. We also discuss the challenges and opportunities for implementing these methods for practical solar cell manufacture.



KEYWORDS: transfer printing methods · thin film solar cells · flexible solar cells · epitaxial lift-off method · epitaxial layer transfer method · controlled spalling method · peel-and-stick method

Thin film solar cells (TFSCs) that are fabricated on flexible substrates, such as plastics, paper, and fabrics, will significantly broaden the applications of solar cells, ranging from wearable solar chargers for portable electronics, building-integrated photovoltaics on sidewalls and curved rooftops, to lightweight solar cells for aerospace and space applications.[1−6] In addition, flexible TFSCs will greatly reduce the material cost and potentially installation cost.[7] Despite these benefits, it remains challenging to find efficient and economical methods for fabricating TFSCs on flexible substrates because the thermal, mechanical and chemical properties of these substrates typically are not compatible with the processes used for manufacture of the high efficiency TFSCs. For example, polyethylene terephthalate (PET) is one of the most widely used flexible substrates, yet it has a melting temperature of about 250 °C, which is close to or well below the deposition or recrystallization temperatures for absorber materials of TFSCs (*e.g.,* 250 °C for a-Si (amorphous Si), 600 °C for CIGS (copper−indium-gallium−selenide), and 620 °C or higher for poly-Si (polycrystalline Si)). As such, higher temperature tolerant substrates, instead of PET, have to be used for the fabrication of TFSCs. Alternatively, the process temperatures have to be lowered to accommodate the substrate temperature constraint, which frequently leads to the TFSCs of lower efficiency.[8−10]

The effect of process temperature on the efficiency of several common TFSCs is illustrated in Figure 1.[11−27] In the lower right half region of Figure 1, TFSCs are deposited directly onto various substrates and it is clear that the maximum process temperature of TFSCs is strictly limited by the maximum temperature that the substrate materials can tolerate. For instance, amorphous Si thin films, due to its relative low deposition temperature of 300 °C or below, can be deposited directly on many flexible substrates, including plastics[28] and polyimide.[20] In comparison, polycrystalline Si or CIGS thin films need to be deposited/recrystallized at temperature at least above 400 °C, so they can be only deposited onto high temperature resistant glass,[27] metal,[29,30] and graphite substrates.[17,18] As the substrates with higher temperature tolerance are used for TFSCs of the same type, the efficiency normally

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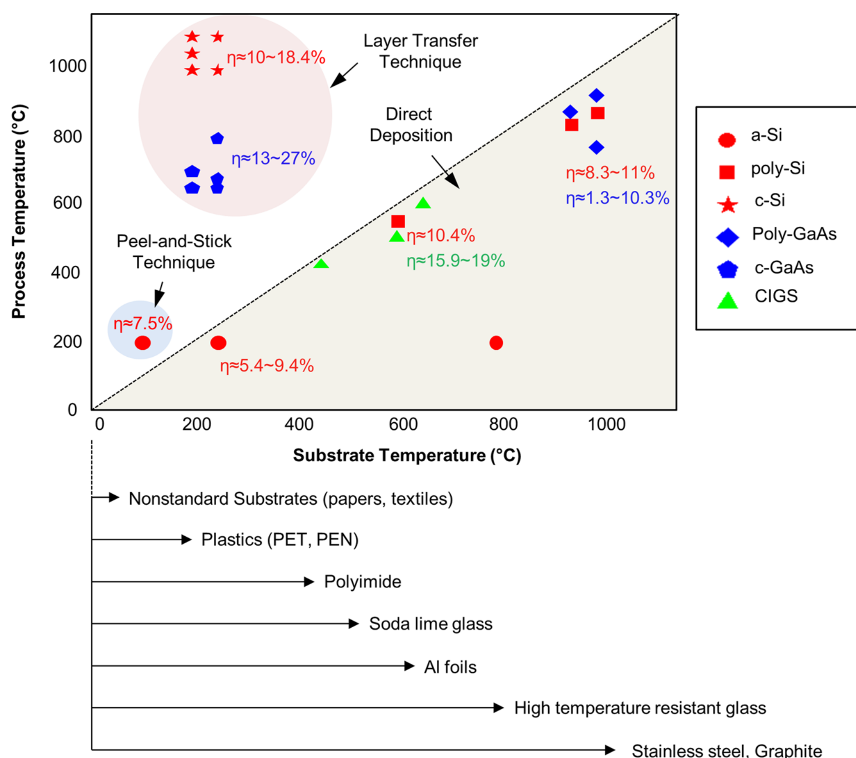

Figure 1. An illustration showing the effect of process temperature and substrates on the efficiency of several common thin film solar cells (TFSCs). The TFSCs in the lower right half region (blue highlight color) are directly deposited onto the substrates, so the maximum process temperature of the TFSCs is strictly limited by the maximum temperature that the substrate materials can tolerate. The TFSCs in the upper left half region (red highlight color) are first deposited on a donor substrate, and then transferred to other substrates using transfer printing methods so that high temperature processed TFSCs are integrated with low temperature tolerant substrates that are cheap, flexible, and lightweight (Si, refs 11−21; **GaAs, refs** 29, 30, 34, 41, 44, 68−71, **CIGS, refs** 25−27). **The bottom half shows the maximum temperature that various flexible substrates can tolerate.**

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increases with the process temperature. Hence, the major research efforts in this region focus on developing high temperature sustainable substrates or low temperature deposition techniques, such as plasma-assisted chemical vapor deposition, to achieve higher quality materials.[26,27]

On the other hand, in the upper left half region of Figure 1, TFSCs are first deposited or fabricated on a donor substrate that is compatible with the fabrication conditions of TFSCs, then separated from the donor substrate and finally transferred to another receiver substrate. Such approaches are generally called transfer printing methods and enable the integration of high temperature processed TFSCs onto low temperature tolerant substrates. Comprehensive review about flexible TFSCs is reported in ref 31, and the basic approaches and procedures for general transfer printing methods are reviewed in ref 32 for broad application areas of nanoelectronics,[33] optoelectronics,[34] metamaterials[35] and solar cells.[24,36,37] Herein, this review focuses on the transfer printing methods used for flexible TFSCs (upper left half region in Figure 1). We will focus on four major transfer printing methods that are categorized on the basis of their fundamental working principles: (1) transfer by sacrificial layers, (2) transfer by porous Si layer, (3) transfer by controlled crack, and (4) transfer by water-assisted thin film delamination. For each transfer printing method, we will briefly discuss its basic concepts, working principles, and applications for TFSCs, followed by discussion on their opportunities and challenges for practical applications.

RESULTS AND DISCUSSION

Transfer by Sacrificial Layers. In 1978, Konagai *et al.*[38] pioneered the "Peeled Film Technology (PFT)", which also referred to as "Epitaxial Lift-Off (ELO) method", and demonstrated the transfer by using a sacrificial layer to deliver monocrystalline thin films onto flexible substrates. The basic procedures of this approach are described in Figure 2a. A sacrificial layer (*e.g.,* AlGaAs) and the desired monocrystalline device film (*e.g.,* GaAs) are epitaxially grown in sequence on a host substrate (*e.g.,* GaAs wafers) by using metal organic chemical vapor deposition at the temperature range of 700−800 °C (Figure 2a, left). Subsequently, the whole structure is immersed in an etchant solution that etches the sacrificial layer material significantly faster than the monocrystalline device film (Figure 2a, middle). After completely etching away the sacrificial layer, the top monocrystalline device film is released from the host substrate (Figure 2a, right) and subsequently attached onto other receiver substrates. Additional fabrication

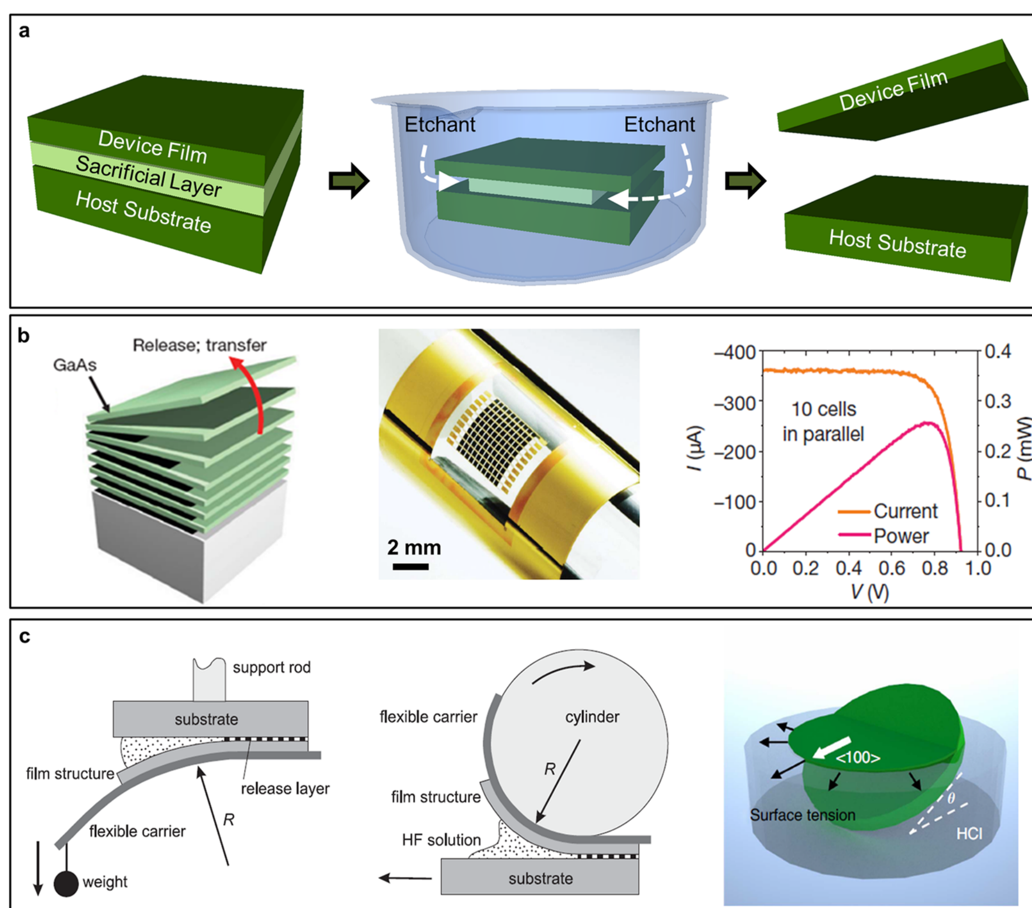

Figure 2. Working principles and solar cell applications of the Epitaxial Lift-Off (ELO) method. (a) Schematic illustration of the general ELO process. Reprinted with permission from ref 41. Copyright 2013 Nature Publishing Group. (b) Schematic illustration of simultaneous release of multiple device thin films with interweaving sacrificial layers (left); a photograph of a 10 × 10 solar cell array that is assembled using a deterministic technique in a repeat fashion onto a plastic substrate (middle); a representative light current−voltage (*I*−*V*) and power−voltage (*P*−*V*) curves for 10 parallel solar cells (right). Reprinted with permission from ref 34. Copyright 2010 Nature Publishing Group. (c) Schematic illustration of diverse modified ELO methods to facilitate the lateral etching process, including weight-assisted (left), roller-assisted (middle) and surface tension-assisted (right) methods. Reprinted with permission from refs 41, 42, 44. Copyright 2013 Nature Publishing Group, 2000 American Institute of Physics, and 2005 Wiley-VCH Verlag, respectively.

steps, such as contact metal deposition, can be carried out to finish the TFSC.

The first successful implementation in TFSCs was carried on a GaAs wafer.[38] Both the GaAs solar absorber layer and the sacrificial AlGaAs layer (5 μm thick) were epitaxially grown on the GaAs wafer. Afterward, the sacrificial AlGaAs layer was selectively etched away by using hydrofluoric acid (HF) solution that etches AlGaAs about 10^6 times faster than GaAs.[39] Hence, both the top GaAs solar absorber layer and the original GaAs host wafer were not affected during the HF etching process. As such, monocrystalline GaAs single junction solar cells (30 μm thick) were successfully transferred onto Al plates with an efficiency around 9−11%.[38] This approach has been used to transfer print other types of semiconducting thin films, including InP[40] and Si,[24] with appropriate sacrificial layers (*i.e.*, AlAs for releasing InP and SiO_2 for releasing Si, respectively) for diverse applications ranging from photodetectors,[34] field effect transistors[34] to light emitting diodes.[41]

This method enables to simultaneously transfer multiple individual device thin films by growing the sacrificial and device films alternatively.[34,38] For example, when AlGaAs and GaAs layers were alternatively grown on a GaAs substrate (Figure 2b, left),[34] all the intermediate AlGaAs layers were selectively dissolved by HF solution, leading to the detachment of multiple GaAs thin films simultaneously. The lifted individual GaAs thin film was subsequently assembled onto a flexible polyimide substrate using a deterministic assembly technique in a step and repeat fashion, followed by postfabrication processes to employ metal contacts, encapsulations and other components for completing the devices (Figure 2b, middle).[34] With this approach, Yoon *et al.*[34] demonstrated the 10 × 10 arrays of GaAs single-junction solar cells (0.5 mm × 0.5 mm) on a PET substrate with a conversion efficiency of 20.5%. A representative light current−voltage (*I*−*V*) characteristics of the 10 solar cells connected in parallel appear in Figure 2b (right)[34] with the maximum

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output voltage and power of 0.93 V and 0.23 mW, respectively.

The main advantage of this transfer printing method is its capability to yield extremely high efficiencies over 20% because it allows using monocrystalline thin film as an absorber material. However, the key challenge lies on its low throughput because of the slow etch rate of the sacrificial layer. Although the etch rate of the sacrificial layer is much faster than that of the device film, it is still slow because the etchant etches the sacrificial layer laterally through the small thickness of around a few hundred nanometers or less. For instance, the lateral etching rate for a 10 nm thick AlGaAs is about 1 mm/h,[39] which is significantly slow for practical manufacturing. To facilitate the lateral etching rate, diverse modified methods, such as weight-assisted (Figure 2c, left),[42,43] roller-assisted (Figure 2c, middle)[44,45] and surface tension-assisted (Figure 2c, right)[41] methods, are developed to facilitate the transport of etchant into the sacrificial layer. Comprehensive review of their recent advances appears in ref 44. Briefly herein, the weight-assisted and roller-assisted methods applied external force during the separation of the epitaxial thin film from its host substrate, in which the lateral etching rate was accelerated. With these approaches, the lateral etching rate was almost 40 times increased up to 11.2 mm/h comparing to that of 0.3 mm/h when using standard peel off configuration.[42] In addition, since the etchant solution etched the sacrificial layer simultaneously from multiple sides (Figure 2c, left and middle), the practical lateral etching rate was even faster and an epitaxial thin film of 15 $\times$ 15 mm^2 was released within an hour.[42] However, both weight-assisted and roller-assisted methods required single-wafer set up with consequent low throughput and the flexible carrier needed carefully handling to prevent the thin film from cracking. In 2013, Cheng *et al.*[41] reported a surface tension-assisted method to enhance the lateral etching rate (Figure 2c, right). The sample was placed obliquely with an angle of 1−20 degree from the etchant solution surface, and the etchant solution was added to the level of the etching front. During this step, surface tension pulled the thin film away from the substrate and flattened on the surface of the etchant solution. With this method, the achieved lateral etching rate was around 5.9 mm/h and a 2 in. GaAs thin film was completely released within 8.5 h. Nevertheless, the etching rate is still slow for practical large throughput manufacture of TFSCs. In addition, the exposure to the HF solution severely affects the surface quality of the host GaAs wafer,[46] and therefore, additional surface treatments such as chemical/mechanical polishing are required to recover the GaAs surface for subsequent epitaxial growths,[47] which would increase the manufacture cost. Though these methods have made continuous progress over the past 35 years, commercial implementation is yet to be seen.

Transfer by Porous Silicon Layer. Similar to the above method, a porous Si layer can be also used as a sacrificial layer for transfer printing. This approach, referred to as "Epitaxial Layer TRANsfer (ELTRAN)", was first demonstrated in 1994 by Yonehara *et al.*,[48] for which a monocrystalline Si thin film was transferred by using an underneath porous Si layer as a sacrificial layer. This method was originated and developed by Canon, Inc. in Japan, and it is the first manufacturable and commercially available method of using porous Si. The basic procedures of this method involve four steps (Figure 3a). First, the top layer of a monocrystalline Si wafer is converted into porous Si by electrochemical etching.[49,50] Specifically, the Si wafer is immersed in HF solution while passing through an electric current across the wafer, and the Si is oxidized by the current and subsequently etched by HF. The applied current density is typically tuned during the etching process to create a double porosity structure: low-porosity (10−20%) layer at the top and high-porosity (50−70%) one at the bottom (Figure 3a, left). Second, since the Si surface after the porous Si formation has a roughness about 10 nm, the Si surface is subsequently smoothened by a high temperature annealing step in hydrogen ($\geq$1050 °C).[49,50] After annealing (Figure 3a, middle), the low porosity Si layer is transformed to a smooth and quasi-monocrystalline Si layer that is then used for the epitaxial growth of monocrystalline Si on top. The high porosity Si layer is turned into a separation layer with bigger pores for easy detachment. Figure 3b shows a representative scanning electron microscopy (SEM) image of the epitaxially grown monocrystalline Si film and the underneath low porous Si layer and separation layer.[49] Third, the monocrystalline Si layer grown on the porous silicon layer is then separated from the host Si wafer by applying mechanical forces at the high porosity separation layer with water jets,[51] or ultrasonic vibration[50] or mechanical tensile force[52] (Figure 3a, right). Finally, the monocrystalline Si film is bonded onto a receiver substrate and additional fabrications are performed to finish the final devices. The remaining porous silicon on the monocrystalline Si film is removed by selective chemical etching or can be kept as light-trapping structures at the bottom of TFSCs. As such, monocrystalline Si films/devices are successfully integrated with other substrates. To complete the TFSCs, additional fabrication steps, such as metal depositions for electrical interconnections, are carried out. In 2009, Reuter *et al.*[22] utilized this method to manufacture a free-standing 47 μm thick monocrystalline Si TFSC with a conversion efficiency of 17.0%. Subsequently in 2013, Dross *et al.*[23] further optimized the process and improved the efficiency of the monocrystalline Si TFSC to 18.4%.

The main advantage of this method is its scalability because the mechanical separation process of the porous Si layer does not significantly depend on the size. As shown in Figure 3c,[50] Canon, Inc. successfully

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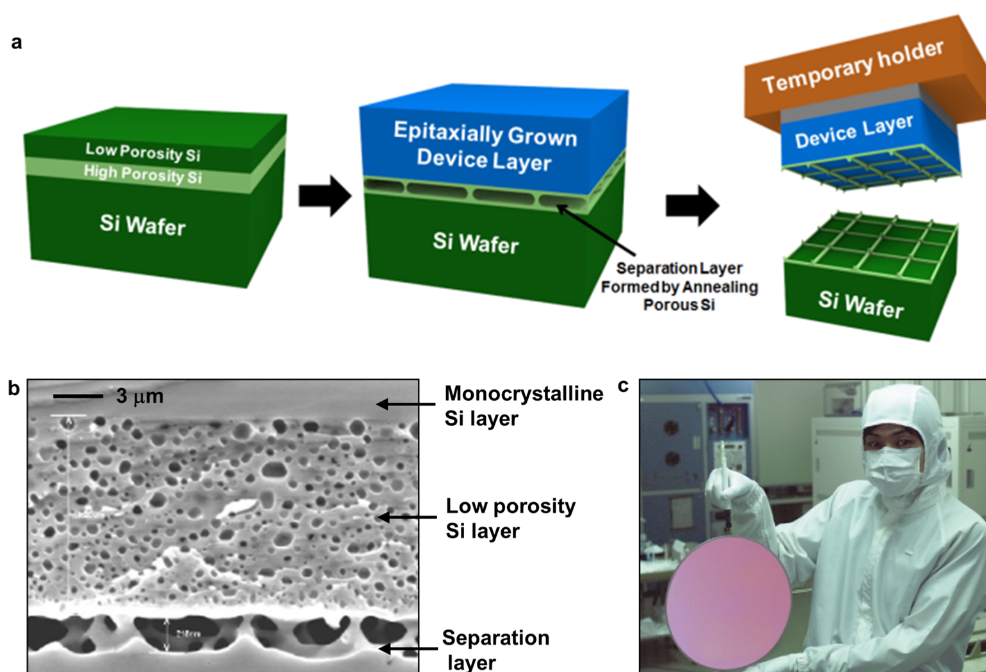

Figure 3. Working principles and solar cell applications of the Epitaxial Layer TRANsfer (ELTRAN) method. (a) Schematic illustration of using the ELTRAN process for transferring a monocrystalline Si device layer onto a foreign substrate. (b) Scanning electron microscopy images of a monocrystalline Si layer epitaxially grown on the porous Si surface. Reprinted with permission from ref 49. Copyright 2013 Elsevier. (c) A photograph of an 8 in. wafer scale transferred monocrystalline Si layer with the ELTRAN method by Canon, Inc. Reprinted with permission from ref 50. Copyright 2002 Kluwer Academic Publishers.

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transferred an 8 in. wafer scale of monocrystalline Si layer with the ELTRAN method. However, the challenges arise from the high cost for solar cell applications, because the thickness of the monocrystalline Si needs to be at least tens of micrometers, which significantly increases the cost and time for the epitaxial growth step. One cost saving strategy is to reuse the host Si wafer after additional surface treatments, such as chemical/mechanical polishing to remove the high porosity Si layer and smooth the surface. This method works only for Si; it is unlikely that the method in the previous section works for GaAs,[34,41] InP[40] as well as Si.[24] Commercial implementation of this method for TFSCs is yet under development, and to date, the most successful application is on the fabrication of silicon-on-insulator (SOI) wafers,[50] which was commercialized in 1997.

Transfer by Controlled Crack. For both above methods, the monocrystalline thin film used for the TFSCs needs to be grown epitaxially on top of a sacrificial layer, which is time-consuming and expensive. To reduce the needs of the sacrificial layer and potentially the epitaxial growth step, in 2012, Bedell *et al.*[53,54] reported a new concept of transfer printing by utilizing cracks, which is named as the controlled spalling method. The basic idea of this method is to use mechanical stress to separate thin films from a donor substrate by propagating a crack inside the donor substrate parallel to the surface. Specifically, first, a thick metal layer (*e.g.,* 6 μm thick Ni) is deposited on a donor substrate (*e.g.,* a Si wafer). Second, another flexible handling layer (*e.g.,* thin polyimide tape) is attached to the top of the Ni film, and the flexible handling layer is mechanically pulled back. As a result, a crack is initiated at the edge of the donor wafer. Since the stress field at the crack tip is composed of both the pure opening stress mode and the shear stress mode (Figure 4a), the crack tends to follow a trajectory where the shear stress component is minimized.[53] Consequently, the equilibrium crack depth is at a position a few microns below the metal film/substrate interface where the shear stress is zero. The depth of the crack can be predetermined by manipulating the thickness and residual stress of the Ni layer.

For formation of the controlled crack, the depositions of multiple thin films are completed before the peel off process so that the entire TFSCs are separated from the donor substrate simultaneously. For example, as shown in Figure 4b, InGaP/(In)GaAs tandem solar cells were first grown on a Ge substrate that served as an epitaxial template, followed by the deposition of the thick Ni stressor layer.[55] Afterward, mechanical peel off with the assistance of a flexible polyimide tape allowed to create a crack and peel off the entire InGaP/(In)GaAs tandem TFSCs from the Ge substrate. Both the top Ni stressor layer and the bottom residual Ge layer on the peeled surface were removed by chemical etching to complete the TFSCs. Representative light current density–voltage (*J*–*V*) characteristics of the transferred InGaP/(In)GaAs tandem TFSCs appear in Figure 4c with a conversion efficiency of around 28.1%.[55]

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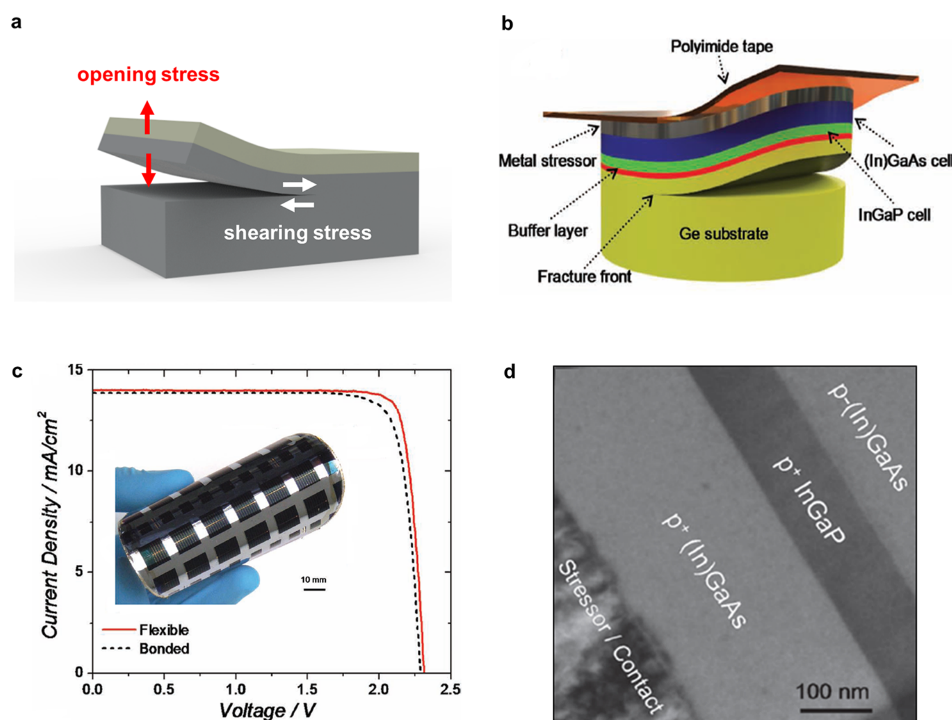

Figure 4. Working principles and solar cell applications the Controlled Spalling Method. (a) Schematic illustration of the spalling fracture mode of a substrate due to the presence of an opening stress and a shearing stress. Reprinted with permission from ref 53. Copyright 2012 IEEE Electron Devices Society. (b) Schematic illustration of the controlled spalling process to separate a fully fabricated InGaP/(In)GaAs tandem solar cell from a Ge donor substrate. Reprinted with permission from ref 55. Copyright 2013 Wiley-Blackwell. (c) A comparison of the representative light current density−voltage (*J*−*V*) curve of 10 mm × 10 mm InGaP/(In)GaAs tandem solar cells on a flexible substrate (red line) and on a Si wafer bonded using a silver-based conductive epoxy (black dotted line), measured under the simulated AM 1.5 solar spectrum with one sun intensity. Inset shows a photograph of the final transferred and flexible InGaP/(In)GaAs tandem solar cell arrays on plastic (diameter: 100 mm). Reprinted with permission from ref 55. Copyright 2013 Wiley-Blackwell. (d) A representative cross-sectional transmission electron microscopy image of the transferred thin film layers, which shows no visible defects after the controlled spalling process. Reprinted with permission from ref 55. Copyright 2013 Wiley-Blackwell.

The performances of the InGaP/(In)GaAs tandem TFSCs on the flexible substrate was comparable to that of a similar tandem TFSCs bonded on a Si wafer using a silver-based conductive epoxy. The transferred tandem TFSCs presented great mechanical flexibility (Figure 4c, inset), indicating that the bending had little influence on the *J*−*V* characteristics.

A notable benefit of this method arises from the ability to simultaneously transfer multiply stacked thin films with no visible crystalline defects (Figure 4d).[55] In addition, the donor substrate can be reused after a post-treatment, such as polishing, to reduce the surface roughness after the spalling process. Although this method is capable of transferring relatively large area (at least 10 mm × 10 mm) of TFSCs, a couple of challenges remain for scaling up. One is the high material and deposition cost associated with the thick metal stressor layer. When the materials of the TFSCs are different from the donor substrate, it would require the expensive and slow epitaxial growth process. In addition, a precise control of the residual stress in the metal stressor layer is challenging since it is sensitive not only to the deposition tools but also to the detailed temperature history of the deposition process, which would impose a great challenge for reliable and consistent manufacturing.

Transfer by Water-Assisted Thin Film Delamination. In 2012, our group demonstrated a new paradigm of transfer printing,[21] named as the peel-and-stick method or the water-assisted transfer printing, which is capable of peeling off fully fabricated TFSCs from a metal (*e.g.,* Cu or Ni) coated host substrate in water and then stick to arbitrary receiver substrates. This method involves three steps (Figure 5a).[21] The first step is to fully fabricate TFSCs on a Si host substrate by using their usual deposition methods and conditions. Here, the only additional step is to deposit a couple of hundred nanometers thick of metal film (*e.g.,* 300 nm thick Cu or Ni) on top of the Si host substrate by electron-beam or thermal evaporation before the fabrication of the TFSCs and the metal thin film serves as a separation layer in water. In the second step, a temporary holder (*e.g.,* thermal release tape) is attached to the top of the TFSCs. By soaking the whole system in a water bath at room temperature and gently pulling back the temporary holder, the entire TFSCs together with the underneath metal thin film are completely peeled off from the Si host substrate in water. The

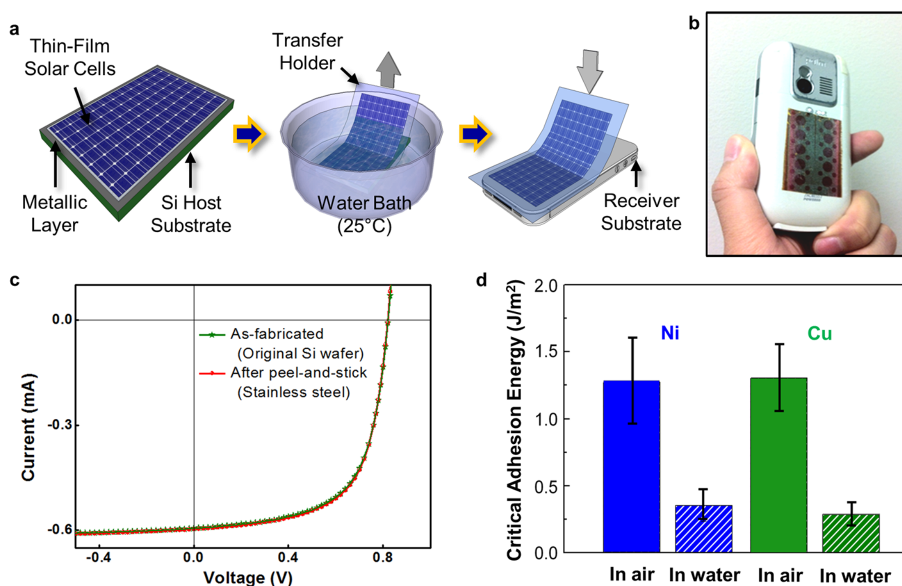

Figure 5. Working principles and solar cell applications of the Peel-and-Stick Method. (a) Schematic illustration of the peel-and-stick process. The TFSCs are fabricated on a Si wafer, then mechanically peeled off in a room temperature water bath, and finally attached onto a receiver substrate. Reprinted with permission from ref 21. Copyright 2012 National Institute of Science Communication and Information Resources. (b) A photograph of the transferred TFSCs on a cell phone case. Reprinted with permission from ref 21. Copyright 2012 National Institute of Science Communication and Information Resources. (c) Representative light current−voltage (*I*−*V*) curves of the TFSCs are identical before (green dotted line) and after (red dotted line) the peel-and-stick process, indicating little damage from the process. Reprinted with permission from ref 21. Copyright 2012 National Institute of Science Communication and Information Resources. (d) Measured critical adhesion energy between SiO_2 surface and Ni (blue) and Cu (green) in air with a 20% relative humidity and in water at 21 °C and it shows that water significantly lowers the critical adhesion energy. Reprinted with permission from ref 56. Copyright 2013 Nature Publishing Group.

metal thin film can be removed by chemical etching if necessary or kept as a metal contact layer. The peeled TFSCs are attached onto a receiver substrate by using commercial adhesive agents, such as polydimethylsiloxane (PDMS) or tapes. Finally, removal of the thermal release tape by heating at 90 °C completes the process with only TFSCs left on the receiver substrate, such as cell phone cases (Figure 5b).

This method was applied to the transfer printing of hydrogenated amorphous silicon (a-Si:H) TFSCs.[21] Figure 5c presents the identical (*I*−*V*) curves of the representative a-Si:H TFSCs before (green dotted line) and after (red dotted line) the transfer printing process. Average solar cell efficiencies of the a-Si:H TFSCs were $7.4 \pm 0.5\%$ and $7.5 \pm 0.5\%$ before and after the transfer printing process, respectively, implying that no damages were induced in the thin films during the mechanical peel off process in water. The high quality of transfer printing results from the phenomena of water-assisted subcritical thin film debonding, in which the presence of water reduces the critical adhesion energy of metal−SiO_2 interface by 70−80% (Figure 5d).[56] Specifically, the water-assisted subcritical debonding is actually caused by accelerated chemical reactions between the highly strained surface bonds at the crack-tip and the environmental species (*e.g.*, H_2O molecules).[57] Depositing Ni film on a Si wafer forms the Ni−O−Si bond, and during the peel-off process in water, the applied mechanical peel-off stress deforms the Ni−O−Si crack-tip bond that readily reacts with H_2O molecules to form Ni−O−H and Si−O−H on each side of the fractured surface. The delaminated surfaces are terminated with hydroxyl (−OH) groups and are hydrophilic, which facilitates the further adsorption of H_2O molecules and continuous reactions along the Ni−SiO_2 interface. As a result, the presence of water, together with moderate mechanical deformation, leads to clean debonding between the metal film together with the top TFSCs and the Si substrate.

This method provides several unique features. First, the entire process occurs at room temperature and requires no postfabrication processes, so therefore, the receiver substrates are never exposed to temperature higher than 90 °C (only 30 s) and harsh chemicals, which significantly broadens the choice of the receiver substrates, ranging from papers, rubbers, cell phone cases to existing building window. Second, the water-assisted subcritical interface debonding phenomenon is observed for a range of metal−SiO_2 interfaces and the critical adhesion energy can be further tuned by varying different environment conditions,[58−60] greatly increasing the flexibility of implementing this method. Third, this method can transfer materials and devices processed at relatively high temperatures for high-performance electronics, since both SiO_2 and metal thin film can sustain the relatively high temperatures. Our group recently succeeded in transferring the

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TABLE 1. Comparisons of Transfer Printing Methods Associated with (1) Transfer by Sacrificial Layers, (2) Transfer by Porous Si Layer, (3) Transfer by Controlled Crack, and (4) Transfer by Water-Assisted Thin Film Delamination

transfer method	applicable materials	strength	weakness	demonstrated applications
Sacrificial layer (chemical etching)	•GaAS •InP •Si	•Transfer of monocrystalline materials (high efficiency solar cells)	•Slow lateral etching of sacrificial layer •Toxic chemical use	•Solar cells[34,38] •Field-effect transistors[34] •Light-emitting diodes[41]
Porous Si layer (mechanical transfer)	•Si	•Transfer of monocrystalline Si (high efficiency solar cells) •Well-established Si and SOI technology and industry	•High cost for the epitaxial Si growth •Potential mechanical breakage for large area transfer •Post-treatment of the remaining porous Si for reuse	•Solar cells[22,23] •Silicon-on-insulator wafer[50]
Controlled crack (mechanical transfer)	•InGaP •(In)GaAs	•Transfer of monocrystalline materials (high efficiency solar cells) •Transfer of fully fabricated TFSC	•Additional cost of using thick metal stress layer •Precise control of the residual stress in the metal stress layer •Potential mechanical breakage for large area transfer	•Solar cells[53,55] •Integrated circuits[54]
Water-assisted thin film delamination (chemical delamination)	•Si	•Transfer of fully fabricated TFSCs •Use of water (no toxic chemicals)	•Not applicable for transferring monocrystalline materials •Additional cost of using thin metal film	•Solar cells[21] •Nanoelectronics (graphene,[61] nanowires,[33] carbon nanotubes[63])

polycrystalline Si thin film processed at 620 °C onto a polyimide sheet.[56] Nevertheless, the main limitation arises from the fact that the metal deposition is the first step executed on top of the silicon supporting substrate, which limits the growth of monocrystalline materials. Thereby this method would be only applicable to transfer printing of amorphous and/or polycrystalline thin film devices. Lastly, the water-assisted transfer printing method has also been used for the fabrication of flexible devices based on graphene,[61] nanowires,[33,62] and carbon nanotubes,[63] for applications in biomedical and transient electronics.

CONCLUSION

Remarkable progress has been made in the development of various transfer printing methods for transfer printing either the absorber materials or the entire TFSCs onto flexible substrates. These transfer printing methods, in comparison to the direct deposition of TFSCs on flexible substrates, overcome the incompatibility issues between the thermal, mechanical and chemical properties of these substrates and the fabrication conditions. In this review, we discussed the history, working principles, and potential and challenges associated with applications in TFSCs of four major transfer printing methods associated with (1) transfer by sacrificial layers, (2) transfer by porous Si layer, (3) transfer by controlled crack, and (4) transfer by water-assisted thin film delamination. Key remarks of each method are summarized in Table 1 for comparisons. The first two methods use the sacrificial layers (*e.g.*, AlGaAs and porous Si) as the seed layers of the epitaxial growth of monocrystalline absorber materials of the solar cells, and the sacrificial layers are separated from their donor substrates by chemical etching or mechanical cleavage. Both methods have produced high efficiency flexible TFSCs (*i.e.*, GaAs TFSCs with a conversion efficiency of ≥20%[34] and Si TFSCs with a conversion efficiency of ≥17%[24]). Nevertheless, both methods face the great challenge of high fabrication cost for applications in flexible TFSCs. On the other hand, the latter two methods utilize mechanical peel-off either to generate cracks in bulk materials or to debond interfaces, leading to the separation of prefabricated TFSCs from their host substrates. These methods successfully transfer fully fabricated TFSCs with unchanged efficiency (*i.e.*, InGaP/(In)GaAs solar cells of ≥28%[55] and a-Si:H TFSCs of ≥7%[21]). Main benefit of these two methods is the capability to transfer partially/completely fabricated TFSCs, so the postfabrication processes are minimized and the choices of receiver substrates are significantly broadened. The method using controlled crack is capable of transfer monocrystalline based solar cells but the method also has a challenge regarding to the high fabrication cost. The method using water-assisted thin film delamination phenomena can only deliver amorphous and polycrystalline TFSCs, but its fabrication process is much simpler.

Further developments are needed for all these transfer printing methods to achieve economic viability. The method of transfer by sacrificial layers needs to significantly increase both the etching speed of the sacrificial layers and the transferred film area, which can be helped by developing special transfer printing tools. The method of transfer by porous silicon needs to justify the high manufacture cost by approaches, such as using the bottom porous Si structures on the monocrystalline Si film to increase the efficiency of solar cells, and reusing the growth Si wafer with minimal amount of treatments (*i.e.*, polishing the remaining porous Si structures on the top surface of the growth wafer). The methods of transfer by controlled crack and transfer by water-assisted thin film delamination need to reduce the material and deposition cost of the metal stressor layer and the metal thin film, and one potential method is to directly use the metal layer as electrodes of TFSCs. In addition to cost reduction, all the methods reviewed here need to further investigate the yield of transfer printing larger area TFSCs when repeatedly using the donor substrates in a statistical manner. Moreover, the mechanical stability, reliability and stability of the transferred TFSCs also need to be systemically characterized for practical applications. Finally, it should be noted that all of these transfer printing methods have application potentials beyond TFSCs, such as in the emerging field of bioelectronics that requires flexible and biocompatible substrates for conformal integration with the soft, curvilinear surfaces of biological tissues,[64−67] which can open up exciting opportunities in many biomedical applications, ranging from brain-machine interfaces,[65] advanced surgical devices,[66] to epidermal electronic monitoring systems.[67]

Conflict of Interest: The authors declare no competing financial interest.

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