Sugar transfer of nanomaterials and flexible electrodes

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\textbf{ABSTRACT}

Nanomaterials with various dimensionalities (e.g., nanowires, nanofilms, two-dimensional materials, and three-dimensional nanostructures) have shown great potential in the recent development of flexible electronics. Conventionally, organic solvents are inevitable while integrating nanomaterials onto flexible substrates, where polymer mediator-assisted transfer techniques are involved. This often damages the flexible substrate and thus hamper the large-scale application of nanomaterials. Here we report a method using water-soluble sugar as a mediator to facilely transfer nanomaterials onto rigid or flexible substrates. This method requires no organic solvent during transfer. More importantly, the morphology and properties of transferred nanomaterials, such as shape, microstructure, resistivity, and transmittance are well preserved on the target substrate. We believe that this universal and rapid transfer method can greatly advance the applications of nanomaterials in the field of flexible devices and beyond.

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Introduction

Nanomaterials (e.g., nanowires, nanofilms, 2D materials and 3D nanostructures) have been widely used in the field of flexible electrodes, optoelectronics, and nanoelectronics, due to their exceptional optical and electrical properties [1–6]. Transfer of nanomaterials from its original substrate is one of the key components for the fabrication of the heterogeneously integrated functional systems [7]. Most transfer technologies require mediator-assisted materials including macromolecular polymers and small organic molecules [8–10], which inevitably lead to process-specific drawbacks [11–15], limiting the performance of the final flexible devices. For example, polymethylmethacrylate (PMMA) and thermal-release tapes are widely used mediators, for which the polymer/tape residuals have to be removed using organic solvents and perhaps subsequent high-temperature annealing [16]. The use of organic solvents and high-temperature annealing is not only time consuming and pollutive but also destructive to the flexibility of the final device. This is because all current flexible substrates, such as poly(dimethylsiloxane) (PDMS), poly(ethylene terephthalate) (PET), and propylene carbonate (PC), can easily swell in organic solvent, and most of them deform or melt during the high-temperature annealing. Moreover, polymer-assisted transfer are not as effective on three-dimensional (3D) nanomaterials as on 2D nanofilms. It is challenging but necessary to find a transfer method which is not only suitable for flexible substrate but also effective for nanomaterials with all kinds of dimensionality, including nanowires, nanofilms, and 3D nanomaterials.

Here we present a simple but effective method using water-soluble sugar as the mediator for the transfer of nanomaterials with different geometries and dimensions. No organic solvent and high-temperature annealing are required. The morphology as well as the mechanical, optical and electrical properties of the nanostructured materials are preserved excellently. Furthermore, this method works for 2D materials, including graphene and other 2D transition-metal sulfide (TMDC) materials. We demonstrate that even monolayer graphene retains its good electrical property after sugar transfer.

Results

Transfer of nanomaterials by using sugar transfer

Figure 1(a) shows the schematic diagram of the sugar transfer method. We first drop melted sugar onto the surface of the nanomaterials with their original substrate. The sugar quickly cools to a solid state. We then peel off the sugar film together with the nanomaterials and transfer them onto any target substrates. Because of its good water-solubility, the sugar film can easily dissolve in de-ionized water, leaving the transferred nanomaterials on the receiver substrate. The whole transfer process is simple and efficient, involving no organic solvent or high-temperature annealing.

Among various one-dimensional (1D) and two-dimensional (2D) nanomaterials, metal nanowires and metal nanofilms are widely used in flexible electronics because of their high conductivity and excellent mechanical flexibility [17–19]. Highly efficient transfer method is very useful for the fabrication of flexible devices. Printing transfer has been an effective method for transferring such nanomaterials. This method, however, highly depends on the adhesion difference between the original, the donor, and the receiver substrates, as well as
the size of the target materials [7,20]. Especially, silver nanowire electrodes typically consist of loosely stacked and intertwined nanowires, which could not be fully transferred by existing methods. Here, our sugar transfer is capable of transfer materials of various dimensionality with all kinds of complex topography. As shown in Figure 1(b,c), elastically squeezed wavy Ag nanowires are completely transferred from a PDMS substrate to a receiver PDMS substrate with high fidelity: the integrated structure and relative position of the wavy Ag nanowires before and after sugar transfer have not been changed at all. It is worth noting that such elastic serpentine often easily get straightened and lose the high stretchability by using other transfer methods. In addition, the transfer result of Au nanodisc array and patterned Au nanofilm is shown in Figure 1(d–g). The Au nanodisc array is transferred from a Si wafer substrate to a PDMS substrate without any visible feature loss (Figure 1(d,e)), and the patterned Au nanofilm transferred from a PET substrate to a polyethylene (PE) substrate also exhibits good fidelity (Figure 1(f,g)). By comparing the optical microscopy (OM) images of the Au nanodisc array on the original and target

Figure 1. Schematic diagram of sugar transfer and transferring of nanomaterials with different dimensions and geometries. (a) Schematic of the transfer procedure. (b, c) OM images of wavy Ag nanowires transferred from a PDMS substrate (b) to another target PDMS substrate (c). Scale bars are 50 μm. (d, e) OM images of Au nanodiscs array transferred from a Si wafer (d) to a receiver PDMS substrate (e), and scale bars are 50 μm. (f, g) Photographs of a patterned Au nanofilm transferred from a PET substrate (f) to a PE substrate (g). Scale bars are 5 mm. (h, i) SEM images of PS nanoparticles transferred from a PDMS substrate (h) to a PET substrate (i). Scale bars are 5 μm.
substrates, it is evident that the interval distances of Au discs remain the same before and after transfer (Figure 1(d,e)).

Despite the wide range of applications of 3D nanomaterials in various fields such as energy, sensing, and electronics [21], the transfer of 3D nanomaterials remains difficult as they exhibit a complex concave-convex structure on the substrates. We successfully transferred self-assembled polystyrene (PS) spheres from PDMS substrate to PET substrate, using our sugar transfer, as shown in Figure 1(h,i). The transfer has shown good integrity and high quality (Figure 3(h,i)). The integrated structure of transferred nanomaterials demonstrates the versatility of the sugar transfer method for transferring nanomaterials with various dimensionalities.

**The properties of transferred nanomaterials**

Our previous work has reported potential applications of Au nanomeshes in flexible transparent electrodes due to their high optical transparency and electrical conductivity, and excellent mechanical stretchability [22–24]. However, the network microstructure of the Au nanomeshes is easily destroyed or forms surface wrinkles or ridges during the transfer process, which greatly impede the application of Au nanomeshes. Using the sugar transfer method, Au nanomeshes can be transferred with high quality and integrity (Figure 2(a,b)). The type of target substrate has little to no influence on the cleanliness and quality of the transferred Au nanomeshes (Supplementary Fig. S1). No sugar residual is

![Image](https://example.com/image2.png)

**Figure 2.** The properties of transferred nanomaterials. SEM images of Au nanomesh transferred from a PDMS substrate (a) to Si wafer (b), the scale bar is 1 μm. The resistance of the Au nanomesh only has little change after transfer (c). (d) OM image of the donor PDMS substrate after transfer, no residual Ag nanowires were observed. Scale bar is 10 μm. (e) OM image of transferred Ag nanowires film. Scale bar is 100 μm. (f) Transmittance spectra of the Ag nanowire film. In the wavelength range of 400–800 nm, the optical transmittance of Ag nanowire film slightly changed at 550 nm after transfer. The insets show the large-area and high transparency of the Ag nanowire film before and after transfer. The scale bar is 5 mm.
detected on the surface of transferred Au nanomeshes. The resistance of Au nanomesh is maintained well after transfer (Figure 2(c)).

Importantly, sugar transfer can be applied on the transfer of Ag nanowire films, which usually requires delicate transfer techniques as the Ag nanowires are randomly and loosely stacked. Figure 2 shows the result of transferring Ag nanowire films. Optical microscopy (OM) images of PDMS substrate after transfer show that the Ag nanowire film has been completely peeled from the PDMS substrate (Figure 2(d)). The film remains integrated and compact after transfer (Figure 2(e)). The transmittance and sheet resistance of the Ag nanowire film are key properties for its application as flexible transparent electrodes. Figure 2(f) shows the transmittance spectra of the Ag nanowire film before and after transfer. The optical transmittance of Ag nanowires film remains 88% at 550 nm after transfer. The sheet resistance of the sugar transferred Ag nanowire film remains the same order with a slight increase from 10 Ω/sq to 18 Ω/sq, which still fulfills the requirement for flexible transparent electrode application [25,26]. The insets in Figure 2(f) are the OM pictures of the large-area and high transparent Ag nanowire film before and after transfer. Seeing through the transferred Ag nanowires film, green, red, and blue are as easily distinguished as through the films before transfer.

In addition, sugar transfer is capable of transfer patterned Ag nanowire films, such as a Ag nanowire mesh, which can be obtained by using a patterned substrate [27]. However, the transfer of such an electrode is challenging due to the concave-convex

Figure 3. Raman spectroscopy, photoluminescence and transfer characteristics of the sugar processed 2D materials. (a) Raman spectrum of mechanically exfoliated monolayer graphene after sugar transfer. Typical 2D peak and low D peak are shown for the monolayer. (b) Photoluminescence (PL) spectra and (c) Raman spectra of the CVD grown monolayer MoS_2 before (upper panel) and after (lower panel) the sugar transfer. About 10 nm PL blue shift is observed after the transfer. (d) Gate dependence of the resistivity of as transferred monolayer graphene device measured at T = 2 K. (e) Transfer curves of the CVD MoS_2 field-effect transistor after sugar process obtained at V_{sd} = 0.1V (black), 0.2V (red), 0.5V (blue), 1.0V (cyan), respectively. Device images of (d) and (e) are shown in the inset. Scale bar is 10 μm. (f) Output curves of the switching FET at different gate voltages. Red arrow marks the increasing gate from 0 V to 20 V with a step of 2 V.
structure of the patterned substrate. Using the sugar transfer method, Ag nanowire meshes can be transferred easily from the patterned PDMS substrate with high efficiency (Supplementary Fig. S2b) and its structure is preserved well on the target substrate (Supplementary Fig. S2c).

**Transfer of 2D van der Waals materials using sugar transfer method**

2D materials have been the recent focus as emerging materials for future electronics. Transferring 2D nanosheets between different substrates are inevitable for their device applications. We perform sugar transfer of graphene and other 2D transition metal dichalcogenide (TMDC) materials. Their optical and electrical properties are discussed below. Supplementary Fig. S3 shows the sugar transfer of mechanically exfoliated 2D graphene nanosheets of graphene and WS$_2$ thin layers. The atomic force microscopy (AFM) images show the surface of transferred graphene (Supplementary Fig. S3). Raman spectroscopy is used to confirm the quality of graphene. Figure 3(a) shows the Raman spectra of the transferred graphene piece, where the positions of G band and 2D band are centered at 1852.3 cm$^{-1}$ and 2674.9 cm$^{-1}$, respectively. The low $I_D/I_G$ value indicates low defects in the transferred graphene. The intensity ratio of the 2D to G is larger than 2, further indicating the high quality of the transferred graphene. The resistance and mobility of transferred graphene on 300 nm SiO$_2$/Si substrate have been measured at $T = 2$K to demonstrate the electrical property of the transferred graphene. The device, as shown in the inset of Figure 3(b), was fabricated by standard e-beam lithography and metal deposition. As shown in Figure 3(b), the transverse resistance plot has shown the Dirac behavior as back gate tunes the Fermi level of graphene, yielding a field-effect mobility of 20,000 cm$^2$ V$^{-1}$s$^{-1}$ at 2 K. The peak resistance is on the order of $\sim$1.5 KΩ at $V_{\text{dirac}}=11.3$ V. The origin of this initial electron doping could be from the polymer residue in the e-beam lithography or the sugar residue during the sugar transfer process. The characteristic curves of a monolayer CVD MoS$_2$ field-effect transistor (FET) after similar sugar exposure are shown in Figure 3(e,f), with a considerable on-off behavior ($>10^6$). It is worth to note that both devices have shown an increased roughness after dissolving sugar (Fig. S3), and the motility also drops about one order after in contact with sugar (Fig. S4). Nevertheless, the residue does not hinder much the Dirac nature of graphene and the FET behavior of the semiconductor MoS$_2$, inferring the applicability of this transferring method.

In flexible device application, patterning of large-scale 2D materials synthesized by chemical vapor deposition (CVD) is required [28–30]. We then corroborate that such sugar transfer application on patterned 2D materials, in which little quality decay is observed. Since the bonding between CVD graphene and its original metal substrate is usually strong, we first use the conventional wet chemical etch method to transfer CVD graphene onto silicon wafer substrate, and then apply sugar transfers for the subsequent tests. Figure 4(a) shows the sugar transfer result of unpatterned CVD graphene onto PDMS substrate, where the red dashed line marks the faint boundary of the sugar transferred CVD graphene because of the excellent transparency. Raman spectra of the PDMS background and as-transferred graphene (Figure 4(b)) indicate successful transfer, as graphene can be identified by the signature Raman peaks of G (1580 cm$^{-1}$) and 2D (2690 cm$^{-1}$). As shown in Figure 4(c), the CVD graphene is patterned by e-beam...
lithography and subsequent dry etch on a silicon substrate, and then sugar transferred onto a PET substrate. The sheet resistance of the patterned graphene on PET after two times sugar transfer is as low as 575 $\Omega$/sq. This resistance is similar to the original value tested on silicon substrate, and is also comparable to CVD graphene transferred by the conventional wet etch method. By the same process, we also demonstrate sugar transfer of a patterned CVD MoS$_2$ monolayer onto flexible PET substrate. With the results from the multiple sugar transfer for patterned 2D materials, we are confident that the sugar transfer method can meet the requirement of multi-step transfer of 2D materials for complex assembly in flexibly electronics application.

**Conclusion**

In conclusion, we have demonstrated that sugar transfer can be a general method for transferring nanomaterials with various dimensionality and shapes onto both rigid and flexible substrates. The transfer process is quick, facile, and low-cost. The electrical and optical properties of nanomaterials are almost retained after the process. In addition, the high carrier mobility and low sheet resistivity of the as-transferred graphene reveal a low level of external impurity introduced by the sugar transfer process. This method could
greatly advance the applications of nanomaterials, from 1D to 3D, in the field of flexible devices.

**Methods**

**Transfer of nanomaterials**

The transfer process includes the following steps: Sugar (Sucrose, analytic) was melted at 200°C in a beaker until becoming saffron yellow. Then, the temperature cooled down to 130°C in case scorching the sugar. Melted sugar was dropped on the surface of nanomaterials supported on an original substrate. The thickness of sugar is tens of micrometer to centimeter, depending on the flexibility of sugar film which the experiments needed. After solidification, the sugar piece which adheres nanomaterials were peeled off. Then, the sugar piece was transferred to a target substrate, followed by washing away the sugar using de-ionized water (18.25 MΩ) to leave the nanomaterials on the target substrate. The sugar can be washed away in 2–3 min.

Au nanomeshes were made by grain boundary lithography. Au nanofilms and Au nanodiscs were fabricated by electron-beam lithography. Before transfer, the Au nanomeshes, Au nanofilm, and Au nanodiscs were adherent on PDMS, PET, and Si substrate, respectively. The Ag nanowire films were spray-coated on PDMS substrates before transfer. PS spheres were self-assembled and floated on water, and then they were picked up using a PDMS substrate. The hollow hemispheres were fabricated by e-beam evaporating (TF 500) a thin layer of Au on a patterned PDMS substrate. The graphene sheets were prepared using both mechanical exfoliation (small size) and chemical vapor deposition (large area), depending on the size needed.

**Characterization**

Optical microscopy, scanning electron microscopy (TESCAN MIRA 3 LMH) and atomic force microscopy (Oxford/Cypher ES) were used to characterize the morphology of the nanomaterials before and after transfer. Optical spectra were measured using a UV-Visible spectrophotometer (Lambda 650S) in the wavelength range from 400 to 800 nm. Raman spectrum was recorded with a HORIBA LabRAM HR Evolution confocal Raman microscopy system.

**Author contributions**

T. H., Y. Z. and C. G. conceived the idea. T. H. carried out nanomaterials transfer, property test and wrote the manuscript. Y. Z. and C. G. directed experiments and contributed to the manuscript writing. J. Y. assisted in material fabrication, performed optical simulation, and helped write the manuscript. T. S. and H. P. conducted AFM analysis of 2D materials, as well as the test of electrical properties. J. Z., Y. H., W. Y., Z. Q. and G. L. assisted in materials fabrication and property tests. S. C. contributed to data analysis and manuscript preparation.

**Disclosure statement**

No potential conflict of interest was reported by the authors.
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