PAPER

Chemical and topographical patterns combined with solution shear for selective-area deposition of highly-aligned semiconducting carbon nanotubes†

Jonathan H. Dwyer,a Anjali Suresh,b Katherine R. Jinkins,b Xiaoqi Zheng,b Michael S. Arnold,*b Arganthaël Berson*c and Padma Gopalan*ab

Selective deposition of semiconducting carbon nanotubes (s-CNTs) into densely packed, aligned arrays of individualized s-CNTs is necessary to realize their potential in semiconductor electronics. We report the combination of chemical contrast patterns, topography, and pre-alignment of s-CNTs via shear to achieve selective-area deposition of aligned arrays of s-CNTs. Alternate stripes of surfaces favorable and unfavorable to s-CNT adsorption were patterned with widths varying from 2000 nm down to 100 nm. Addition of topography to the chemical contrast patterns combined with shear enabled the selective-area deposition of arrays of quasi-aligned s-CNTs (~14°) even in patterns that are wider than the length of individual nanotubes (>500 nm). When the width of the chemical and topographical contrast patterns is less than the length of individual nanotubes (<500 nm), confinement effects become dominant enabling the selective-area deposition of much more tightly aligned s-CNTs (~7°). At a trench width of 100 nm, we demonstrate the lowest standard deviation in alignment degree of 7.6 ± 0.3° at a deposition shear rate of 4600 s⁻¹, while maintaining an individualized s-CNT density greater than 30 CNTs μm⁻¹. Chemical contrast alone enables selective-area deposition, but chemical contrast in addition to topography enables more effective selective-area deposition and stronger confinement effects, with the advantage of removal of nanotubes deposited in spurious areas via selective lift-off of the topographical features. These findings provide a methodology that is inherently scalable, and a means to deposit spatially selective, aligned s-CNT arrays for next-generation semiconducting devices.

1. Introduction

Semiconducting single-walled carbon nanotubes (s-CNTs) are excellent candidates for next-generation field-effect transistors (FETs) due to their outstanding properties such as ballistic transport, excellent charge mobility, and high thermal conductivity.¹⁻⁷ However, to date, the vast majority of s-CNT-based FETs have underperformed compared with conventional Si- and GaAs-based FETs due to two main factors. One of these factors is the need to achieve greater than 99.99% semiconducting CNTs from an electronically heterogenous CNT mixture. This material processing challenge has been largely overcome through a number of sorting agents in both aqueous and organic solvents.⁸⁻¹⁰ The second issue, which has yet to be reliably solved, relates to the difficulty of scaling single s-CNT devices to s-CNT array devices. An ideal s-CNT array requires deposition to be spatially controlled with a pitch of 5–10 nm (corresponding to 100–200 CNTs μm⁻¹ density), while tightly aligning s-CNTs, preventing their overlap and achieving parallel alignment to each other.⁵⁻⁷,¹⁸

Two primary pathways are typically utilized in the literature to obtain aligned s-CNT arrays: (1) direct growth of s-CNT array through chemical vapor deposition (CVD), and (2) s-CNT deposition from solution. CVD growth uses CNT growth precursors on catalytic substrates to fabricate aligned s-CNT arrays.¹¹⁻¹⁵ Advantages to CVD method include high degrees of s-CNT alignment in arrays as well as the relative ease of patterning catalytic materials for localized s-CNT growth. Densities as high as 130 CNTs μm⁻¹ have been achieved,¹⁴ which is in the predicted optimal density range for s-CNT-based electronics. However, the major disadvantage of the CVD growth method is the concurrent growth of both s-CNTs and metallic CNTs (m-CNTs), hence lowering current on/off ratios.¹⁶ Although progress has been made in selectively synthesizing s-CNTs using CVD¹⁷,¹₈ and removing m-CNTs post-synthetically,¹⁵ the purity levels do not approach those required for high-performing s-CNT-based devices.⁷ In addition, most CVD s-
CNT growth mechanisms require specific substrates such as sapphire and quartz. Thus, an additional CNT array transfer step is needed to deposit CVD grown s-CNTs on traditional MOSFET substrates like Si wafers.

In contrast to CVD growth mechanisms, s-CNT purities greater than 99.99% are attainable by dispersing s-CNTs in solution to create “inks”. To overcome inter-CNT π–π interactions to individualize and deaggregate s-CNTs, typically dispersing agents are necessary. Dispersants such as aromatic conjugated polymers which interact non-covalently with CNTs are also able to sort CNTs into high-purity, electronics-grade s-CNT inks. From these inks, alignment of s-CNTs on substrates has been achieved through various methods including Langmuir–Blodgett/Schaefer, vacuum filtration, electric fields, shear, evaporation, 3D printing, and at liquid/liquid interfaces. While these studies have made progress in fabricating continuously aligned s-CNTs on wafer-scale areas, selective-area deposition and controlling their pitch in a scalable manner are still unresolved.

Current selective-area CNT deposition methods include covalent bonding with the substrate, tailored electrostatic interactions between the polymer wrapper and the substrate, and use of DNA-based nanotrench guides. Park et al. used a blade shearing method to deposit polypyrrole-wrapped s-CNT arrays selectively on a substrate with alternating patterns of 500 nm wide solvent wetting and dewetting regions. These arrays are among the densest, selectively deposited arrays in the literature with a density of 150–200 CNTs μm\(^{-1}\). However, considering the high s-CNT density, the devices made from these s-CNT arrays have a notably low current. Kumar et al. used diazonium chemistry to pattern HfO\(_2\) trenches with diazonium head groups to covalently react with s-CNTs. While the authors were able to selectively deposit s-CNTs down to a pitch of 50 nm, the density of s-CNTs in each trench is low (~1 CNT per 50 nm trench), thereby reducing the current drive in the FETs. In addition, covalently reacting with s-CNTs to obtain spatial control leads to sp\(^3\) defects and deterioration of electronic properties. Sun and coworkers used DNA nanotrenches to selectively capture the preassembled DNA templated s-CNT arrays, thereby reducing the current drive in the FETs. A corresponding density of just under 100 CNTs per micron-scale target deposition area using sub-50 nm wide solvent-mediated a and imaged on a TEM grid. In a subsequent study, the authors patterned a widely spaced array of 200 nm wide PMMA cavities to selectively capture the preassembled DNA templated s-CNT arrays over a 0.35 cm\(^2\) substrate. The authors demonstrated single s-CNT per micron-scale target deposition area using sub-50 nm wide DNA templates. This approach is advantageous as the DNA trench periodicity can be varied by supramolecular assembly as the s-CNTs are preassembled in solution. However, the area of aligned s-CNTs arrays is dependent on the size of the DNA template, and the alignment of the s-CNTs is dependent on the aspect ratio of the cavities. Hence, simultaneously achieving high density of perfectly aligned s-CNTs in selective regions of the substrate on a wafer-scale without compromising on electronic properties is still an outstanding challenge.

Recently through a systematic study, we reported that varying the substrate chemistry by using self-assembled monolayers (SAMs) impacts the number density of deposited s-CNTs from solution. While substrate water contact angle alone was insufficient to explain the observed s-CNT adsorption trends on these chemically modified substrates, molecular dynamics (MD) simulations provided useful mechanistic insights. In particular, the solvent-mediated affinity of polymer-wrapped s-CNTs played a key role in s-CNT deposition onto a surface. Specifically, the global minimum of the s-CNT adsorption free energy occurred in a configuration where the minimum of the solvent density around the s-CNT coincides with the minimum of the solvent density near the SAM-grafted substrate. Hence, the oscillatory solvent structure near a SAM-grafted surface determines the adsorption free-energy landscape driving s-CNT deposition trends.

Building on these studies in this work, we develop chemical and topographical patterns to guide selective shearing deposition of aligned array of s-CNTs from organic solvents. High shear rate deposition on chemically and topographically contrasted patterns lead to the selective-area deposition of arrays of quasi-aligned s-CNTs (~14°) in patterns that are wider than the length of the individual nanotubes (>500 nm). However, as the width of the patterns is reduced below the length of the individual nanotubes, confinement effects dominate in the deposition process, leading to selective-area deposition of more tightly aligned s-CNTs (7°). We characterize these arrays for s-CNT density via scanning electron microscopy (SEM) image analysis and s-CNT alignment degree via a two-dimensional fast Fourier transform (2D FFT) methodology. We also demonstrate that these surface patterns can be removed after s-CNT deposition resulting in aligned, spatially selective s-CNT arrays for devices.

2. Experimental

2.1 Poly[[9,9-dioctylfluorenyl-2,7-diyl]–alt-co-(6,6’-{2,2’-bipyridine})] wrapped s-CNT solution preparation (s-CNT ink)

Chloroform s-CNT inks are prepared from isolating s-CNTs from CNT soot using a previously established procedure. Briefly, a 1 : 1 ratio by weight of arc-discharge CNT soot (Sigma-Aldrich) and poly[[9,9-dioctylfluorenyl-2,7-diyl]–alt-co-(6,6’-{2,2’-bipyridine})] (PFO-BPy) (American Dye Source, Inc., Quebec, Canada; #ADS153-UV) are each dispersed at a concentration of 2 mg mL\(^{-1}\) in ACS grade toluene. This solution is sonicated with a horn tip sonicator (Fisher Scientific, Waltham, MA; Sonic Dismembrator 500) and then centrifuged in a swing bucket rotor to remove undispersed material. After centrifugation, the supernatant containing polymer-wrapped s-CNTs is collected and centrifuged for an additional 18–24 h to sediment and pellet the s-CNTs. The collected s-CNT pellet is dispersed in toluene with horn tip sonication and again centrifuged. The centrifugation and sonication process is repeated a total of three times. The final solution is prepared by horn tip sonication of the s-CNT pellet in chloroform (stabilized with ethanol), s-CNTs prepared via this approach are characterized by a log-normal length distribution with an average length of 580 nm and diameters varying from 1.3 to 1.8 nm. Concentration of s-CNT ink was determined using optical cross sections from the
2.2 Surface pattern fabrication on silicon oxide substrates

Surface patterns were fabricated using traditional electron-beam lithography techniques. Silicon [100] wafer substrates (Addison Engineering, Inc.) with 90 nm wet thermal silicon oxide were immersed in a 3 : 1 by volume H₂SO₄ : H₂O₂ piranha solution for 1 h at 85 °C. After piranha treatment, substrates were rinsed with deionized (DI) water and dried with N₂. For chemical patterns, ma-N 2401 resist (Micro Resist Technology) was spin coated onto the wafers and patterned using electron-beam lithography techniques. A RIE oxygen plasma for resist descum exposed the silicon oxide. The patterned substrates were submerged in octadecyltrichlorosilane (OTS) (Sigma-Aldrich, 104817) at a concentration of 5 mM in toluene for 12 h. Substrates were bath sonicated in toluene for 30 min, rinsed in toluene, and dried with N₂. Resist was stripped by submerging the substrates in anhydrous N-methyl-2-pyrrolidinone (NMP) (Sigma-Aldrich, 328634) for 24 h and drying with N₂. For topographical patterns, PMMA resist (MicroChem Corp.) was spin coated onto the piranha cleaned silicon substrates and an electron-beam lithography system (Elionix ELS-G100) exposed the PMMA resist with the desired pattern. After PMMA development and oxygen plasma descum, metals were evaporated onto the exposed silicon oxide. PMMA lift-off in acetone resulted in metal features on the silicon substrates. Additional Au chemical functionalization was performed using thiol-based chemistry developed from a previous procedure. Substrates with Au features were submerged in 1-octadecanethiol (OTh) (Sigma-Aldrich, O1858) at a concentration of 1 mM in ethanol for 24 h, rinsed with additional ethanol, and dried with N₂. For both OTS and OTh samples, a control water contact angle measure determined the effectiveness of SAM grafting to substrates. A 7 µL DI water droplet was dispensed using a Dataphysics OCA 15 optical contact angle measuring system on the SAM surface. Once the water droplet was fully formed, the static water contact angle (WCA) of the droplet was immediately measured. Samples were considered fully functionalized if the WCA was greater than 110°. Functionalized silicon oxide substrates were stored under N₂ until s-CNT deposition.

2.3 s-CNT array fabrication and characterization

Details of the shear system, and the shear rate control used to deposit s-CNTs, are described elsewhere. Chloroform s-CNT ink was sheared across surface patterned silicon substrates at a set shear rate. Additional chloroform solvent was immediately sheared across the same substrate to remove residual s-CNTs. Substrates were boiled in toluene at 110 °C for 1 h and dried with N₂ to remove excess polymer wrapper. Substrates were stored under N₂ until characterization. Scanning electron microscope (SEM) images were taken with a Zeiss LEO 1550VP SEM. These SEM images were processed using a two-dimensional fast Fourier transform (2D FFT) algorithm to...
determine s-CNT alignment. The 2D FFT methodology is described in detail in ESI Fig. S2–S5.

2.4 Topographical pattern removal

Topographical pattern trenches were removed by etching away the metal between the s-CNT arrays. A thin layer of PMMA was spin coated onto the s-CNTs prior to metal removal to protect the s-CNTs from the metal etchant. Any s-CNTs crossing over the gold mesas were first removed using an oxygen plasma reactive-ion etching procedure (see ESI Fig. S16†). Metal trenches (Au/Cu) were removed by submerging substrates in a standard gold etchant (Sigma-Aldrich, 651818) for 5 min and soaking the samples in DI water for 10 min. The iodine-based gold etchant converted Cu to a copper iodine complex that is insoluble in aqueous solution. This process was repeated once to completely convert all Cu. PMMA protective layer and copper iodine complex was removed by boiling acetone at 110 °C for 15 minutes.

Raman spectroscopy measurements of s-CNTs were taken on a Thermo Scientific DXRxi Raman imaging microscope using the mapping function. Raman maps of the s-CNT characteristic bands over 34 × 34 μm² areas were taken consisting of 1156 pixels where a pixel represents a 1 μm² area.

3. Results and discussion

3.1 Chemical patterns for s-CNT arrays

We first explored the effectiveness of chemical pattern contrast for selective deposition of s-CNT arrays by shear from an organic ink. The polymer-wrapped s-CNTs prefer to adsorb on SiO₂ compared to octadecylsilane (OTS)-grafted silicon oxide due to solvent structure effects. To take advantage of s-CNT adsorption preferences to drive selective-area deposition, alternating stripes of SiO₂ and OTS were patterned using an electron-beam lithography (EBL) process illustrated in Fig. 1a. Initial studies using PMMA resist for patterning resulted in the penetration of the OTS through the resist to the substrate during the liquid deposition, hence a negative tone resist was selected to mitigate this problem. The crosslinking in the negative tone resist prevented penetration of OTS into the resist during selective functionalization (ESI Fig. S1†). After OTS functionalization, the negative tone resist was removed resulting in a chemically patterned silicon substrate consisting of alternating stripes of SiO₂ and OTS depicted in Fig. 1a. Individual stripes widths were varied between 250 and 2000 nm where the width of the SiO₂ stripes in the chemical pattern is defined as w (illustrated in Fig. 1a). The OTS stripe width in a chemical pattern is also equal to w at a given SiO₂ stripe width.

On these chemically patterned substrates, 375 μL of s-CNT ink at a concentration of 240 μg mL⁻¹ was deposited using a previously established shear deposition method²⁷ at a high shear rate of 46 000 s⁻¹. These experimental deposition conditions were based on our prior work on optimizing CNT alignment by shear deposition, which resulted in the best alignment.²⁷ Fig. 2a shows the SEM images of s-CNTs deposited on alternating SiO₂ and OTS stripes fabricated using EBL. From these SEM images, as expected the s-CNT density was significantly higher on SiO₂, the favorable s-CNT adsorption surface, compared to OTS, the unfavorable adsorption surface.

Fig. 2 shows s-CNT deposition on the chemically patterned substrates. Alignment of s-CNTs in these arrays was characterized by two-dimensional fast Fourier transform (2D FFT)

![Fig. 2](image-url)
3.2 Topographical patterns for s-CNT arrays

We hypothesized that addition of a physical barrier to the chemical patterns on the silicon substrate would both significantly improve s-CNT alignment on SiO₂ stripes as well as limit pinning across OTS stripes. The design and fabrication of the topographical surface patterns with integrated chemical patterns are illustrated in Fig. 1b and c. The trench floors were bare SiO₂ acting as the favorable s-CNT deposition surface while the mesas acted as the unfavorable deposition surface. The mesas needed to be fabricated from a material that could be selectively functionalized without modifying SiO₂ on the trench floor. For fabrication simplicity, we picked gold for the mesas as it could be selectively functionalized with octadecanethiol (OTh), a thiol terminated OTS. To improve adhesion of gold to the mesas acting as the unfavorable deposition surface. The studies presented here uncover important guidelines to achieving exceptionally aligned s-CNT arrays while selectively depositing them in desired regions of the substrate. These studies confirm that, as on planar substrates, increasing the shear rate during s-CNT deposition does not indefinitely increase their alignment in the trenches. When the patterns are wider than the length of the s-CNT (>500 nm), increase in shear rate leads to quasi aligned CNTs, with a σ of ~14°. For example, while increasing shear rate from 46 s⁻¹ to 4600 s⁻¹ dramatically improved the alignment from 28.5 ± 6.3° to 16.2 ± 1.3° in 1000 nm wide trenches, a further increase to 46 000 s⁻¹ resulted in a marginal improvement to a σ of 13.3 ± 1.0°. When the pattern width is reduced to less than the length of the individual s-CNTs (<500 nm), confinement effects dominate over shear rate, leading to a dramatic enhancement in the alignment degree. For example, using a low shear rate of 46 s⁻¹ with a 100 nm wide trench achieved an alignment degree of 7.6 ± 1.3°. This alignment degree is remarkable in comparison to the alignment degree of 19.3 ± 3.5° at the high shear rate (46 000 s⁻¹) on planar SiO₂ (ESI Fig. S12†).
Chemical patterns alone consisting of alternating OTS and SiO₂ stripes resulted in a constant s-CNT alignment degree \( \sim 18^\circ \) regardless of the stripe width. Whereas the addition of a topographical pattern consisting of 25 nm tall metal stripes improved s-CNT alignment from 19.3 ± 3.5° at a deposition shear rate of 4600 s\(^{-1}\) on bulk SiO₂ to 8.5 ± 2.8° in 100 nm wide trenches. Hence, the alignment can be improved by decreasing the trench width provided both the trench width is sufficiently narrow (<500 nm) and the trench height is sufficiently high to prevent s-CNTs from depositing on multiple SiO₂ stripes. Based on experimental results, trench heights over 25 nm did not further improve s-CNT alignment degree (ESI Fig. S13†). s-CNT alignment on these patterned substrates is uniform across the 2 × 3 cm\(^2\) SiO₂/Si substrates (ESI Fig. S14†) demonstrating the inherent scalability of this process. Larger area deposition can be achievable in future by scaling up of the shear deposition system as shown in our prior work on unpatterned substrates.\(^{27}\)

Another important criterion for this pattern design to be compatible with device fabrication is to completely remove any residual metals post-CNT deposition. For these experiments, Cr, a traditional adhesion layer for Au, was substituted with Cu in the fabrication scheme shown in Fig. 1c because standard Cr etchants attack a PMMA protective layer on the CNTs unlike Cu etchants.\(^ {26}\) SEM images of s-CNT arrays before (Fig. 4a) and after (Fig. 4b) trench removal confirm that the alignment of the s-CNT was preserved (ESI Fig. S15 and S16†), making this removal process compatible with FET device fabrication. Another consequence of the trench removal process is that any crossing nanotubes that might bridge between the SiO₂ stripes, are also removed.

To ensure the electronic properties of the s-CNTs are preserved, we examined the Raman spectrum of s-CNTs before and after exposure to Au/Cu trench removal treatments. Analysis of the ratio of D to G band intensities (\( I_D/I_G \)) is commonly used in the literature to examine electronic defects in CNTs.\(^ {31}\) We use spin coated s-CNTs on 90 nm SiO₂/Si substrates for these tests to increase the signal of G, D, 2D, and Si Raman peaks. These samples were subjected to the same trench removal process as used in Fig. 4. Raman spectra of s-CNTs were taken over a 34 μm\(^2\) area and average into a single spectra (ESI Fig. S17†). Fig. 4d shows the averaged Raman spectra of the s-CNTs before and after trench removal. Before processing, the \( I_D/I_G \) of the s-CNTs is 0.20 ± 0.02. After the trench removal process, s-CNTs have an \( I_D/I_G \) of 0.15 ± 0.02. These data show the trench removal process does not adversely affect the electronic properties of the CNTs. The slight improvement in the \( I_D/I_G \) is likely due to increased removal of residual polymer-wrappers due to the gold etchant. Adsorbates on CNTs will also suppress G band intensity,\(^ {35}\) hence lowering \( I_D/I_G \). These results confirm that the electronic quality of the starting s-CNTs is preserved.
throughout the processing steps, making this removal process compatible with FET device fabrication.

4. Conclusion

In conclusion, we have exploited chemical and topographical patterns in concert with shear forces to selectively deposit a tightly aligned array of s-CNTs. Chemical contrast on a SiO$_2$/Si substrate was created by alternate stripes of SiO$_2$ and OTS-grafted SiO$_2$. During shear deposition, s-CNTs deposited preferentially on the SiO$_2$ stripes compared to OTS stripes. However, the alignment degree of s-CNT arrays was similar to unpatterned substrates and could not prevent pinning of CNTs between adjacent SiO$_2$ stripes. This problem was addressed by introduction of topography with the chemical pattern to deposit s-CNTs by shear. The role of shear and confinement by the topographical features was delineated through a series of studies. Pre-alignment of s-CNTs via shear plays a major role when the trench width is greater than 500 nm or larger than the length of the s-CNTs. However, when the trench width decreases below 500 nm, the confinement effect dominates over shear. At a trench width of 100 nm, exceptional degree of alignment (with a $\sigma$ of 7.6 $\pm$ 0.3° at a shear rate of 4600 s$^{-1}$), was achieved while maintaining a density greater than 30 CNTs $\mu$m$^{-1}$. The rotational diffusion coefficient is expected to quickly decrease, as the s-CNT length increases, thereby aiding in shear alignment. Hence, the pre-alignment by shear forces and confinement effect in trenches can both be potentially enhanced by increasing the average s-CNT length. Additionally, these topographical patterns can be removed post-CNT deposition using standard solution processing steps to obtain aligned s-CNT arrays on SiO$_2$ for device fabrication without altering the alignment. While chemical contrast alone enables selective-area deposition, chemical contrast in addition to topography enables more effective selective-area deposition, stronger confinement effects, and the removal of nanotubes deposited in spurious areas via selective lift-off of the topographical features. Since this methodology relies on traditional fabrication and substrate modification methods, it is inherently scalable and provides a means to fabricate spatially selective, aligned s-CNT arrays for next-generation semiconducting devices.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

J. H. D., A. S., K. R. J., M. S. A., A. B., P. G. acknowledge funding from NSF SNM-IS award no 1727523. J. H. D. acknowledges...
partial support from the PPG Industries sponsored fellowship. Raman spectroscopy and mapping conducted by X. Z. and M. S. A. supported by U.S. Department of Energy, Office of Science, Basic Energy Sciences, under award no. DE-SC0016007. We acknowledge S. Foradori for insightful discussions on removal of the topographical patterns. The authors gratefully acknowledge use of facilities and instrumentation at the University of Wisconsin–Madison Wisconsin Centers for Nanoscale Technology partially supported by the NSF through the University of Wisconsin Materials Research Science and Engineering Center (DMR-1720415). Portions of this work were done using an electron-beam lithography tool at the University of Wisconsin Nanoscale Fabrication Center. We acknowledge support from the NSF (DMR-1625348) for the acquisition of this tool.

References

1. P. Avouris, Z. Chen and V. Perebeinos, Nat. Nanotechnol., 2007, 2, 605–615.
2. T. Dürkop, S. A. Getty, E. Cobas and M. S. Fuhrer, Nano Lett., 2004, 4, 35–39.
3. Y. I. Jhon, C. Kim, M. Seo, W. J. Cho, S. Lee and Y. M. Jhon, Sci. Rep., 2016, 6, 20324.
4. S.-Y. Yue, T. Ouyang and M. Hu, Sci. Rep., 2015, 5, 15440.
5. G. S. Tulevski, A. D. Franklin, D. Frank, J. M. Lobez, Q. Cao, H. Park, A. Afzali, S.-J. Han, J. B. Hannon and W. Haensch, ACS Nano, 2014, 8, 8730–8745.
6. J. Zhang, A. Lin, N. Patil, H. Wei, L. Wei, H. P. Wong and S. Mitra, IEEE Trans. Comput. Aided Des. Integrated Circ. Syst., 2012, 31, 453–471.
7. S. J. Kang, C. Kocabas, T. Ozcel, M. Shim, N. Pimparkar, M. A. Alam, S. V. Rotkin and J. A. Rogers, Nat. Nanotechnol., 2007, 2, 230–236.
8. D. Janas, Mater. Chem. Front., 2018, 2, 36–63.
9. A. Nish, J.-Y. Hwang, J. Doig and R. J. Nicholas, Nat. Nanotechnol., 2007, 2, 640–646.
10. F. Léonard, Nanotechnology, 2006, 17, 2381–2385.
11. L. Kang, S. Zhang, Q. Li and J. Zhang, J. Am. Chem. Soc., 2016, 138, 6727–6730.
12. J. Si, D. Zhong, H. Xu, M. Xiao, C. Yu, Z. Zhang and L.-M. Peng, ACS Nano, 2018, 12, 627–634.
13. W. Liu, S. Zhang, L. Qian, D. Lin and J. Zhang, Carbon, 2020, 157, 164–168.
14. Y. Hu, L. Kang, Q. Zhao, H. Zhong, S. Zhang, L. Yang, Z. Wang, J. Lin, Q. Li, Z. Zhang, L. Peng, Z. Liu and J. Zhang, Nat. Commun., 2015, 6, 6099.
15. S. H. Jin, S. N. Dunham, J. Song, X. Xie, J.-h. Kim, C. Lu, A. Islam, F. Du, J. Kim, J. Felts, Y. Li, F. Xiong, M. A. Wahab, M. Menon, E. Cho, K. L. Grosse, D. J. Lee, H. U. Chung, E. Pop, M. A. Alam, W. P. King, Y. Huang and J. A. Rogers, Nat. Nanotechnol., 2013, 8, 347–355.
16. G. S. Tulevski, A. D. Franklin and A. Afzali, ACS Nano, 2013, 7, 2971–2976.
17. S. Zhang, L. Kang, X. Wang, L. Tong, L. Yang, Z. Wang, K. Qi, S. Deng, Q. Li, X. Bai, F. Ding and J. Zhang, Nature, 2017, 543, 234–238.
18. M. He, X. Wang, S. Zhang, H. Jiang, F. Cavalca, H. Cui, J. B. Wagner, T. W. Hansen, E. Kauppinen, J. Zhang and F. Ding, Sci. Adv., 2019, 5, eaav9668.
19. T. Fujigaya and N. Nakashima, Sci. Technol. Adv. Mater., 2015, 16, 024802.
20. Q. Cao, S.-j. Han, G. S. Tulevski, Y. Zhu, D. D. Lu and W. Haensch, Nat. Nanotechnol., 2013, 8, 180.
21. X. Li, L. Zhang, X. Wang, I. Shimoyama, X. Sun, W.-S. Seo and H. Dai, J. Am. Chem. Soc., 2007, 129, 4890–4891.
22. V. Sgobba, G. Giancane, D. Canoletta, A. Operamolla, O. Hassan Omar, G. M. Farinola, D. M. Guldí and L. Valli, ACS Appl. Mater. Interfaces, 2014, 6, 153–158.
23. X. He, W. Gao, L. Xie, B. Li, Q. Zhang, S. Lei, J. M. Robinson, E. H. Hrzoz, S. K. Doorn, W. Wang, R. Vajtaj, P. M. Ajayan, W. W. Adams, R. H. Hauge and J. Kono, Nat. Nanotechnol., 2016, 11, 633.
24. S. Kaida, J. Matsu, T. Sagae, Y. Hoshikawa, T. Kyotani and T. Miyashita, Carbon, 2013, 59, 503–511.
25. C. Zamora-Ledezma, C. Blanc, M. Maugey, C. Zakri, P. Poulin and E. Anglaret, Nano Lett., 2008, 8, 4103–4107.
26. E. M. Remillard, Z. Branson, J. Rahill, Q. Zhang, T.Dasgupta and C. D. Vecitis, Nanoscale, 2017, 9, 6854–6865.
27. K. R. Jinkins, J. Chan, R. M. Jacobberger, A. Berson and M. S. Arnold, Adv. Electron. Mater., 2019, 5, 1800593.
28. S. Park, G. Pittner, G. Giri, J. H. Koo, J. Park, K. Kim, H. Wang, R. Sinclair, H. S. P. Wong and Z. Bao, Adv. Mater., 2015, 27, 2656–2662.
29. M. Engel, J. P. Small, M. Steiner, M. Freitag, A. A. Green, M. C. Hersam and P. Avouris, ACS Nano, 2008, 2, 2445–2452.
30. G. L. Goh, N. Saengchirat, S. Agarwala, W. Y. Yeong and T. Tran, Nanoscale, 2019, 11, 10603–10614.
31. G. L. Goh, S. Agarwala and W. Y. Yeong, ACS Appl. Mater. Interfaces, 2019, 11, 43719–43730.
32. C. Lan, M. Guo, C. Li, Y. Qiu, Y. Ma and J. Sun, ACS Appl. Mater. Interfaces, 2020, 12, 7477–7485.
33. G. L. Goh, S. Agarwala and W. Y. Yeong, Adv. Mater. Interfaces, 2019, 6, 1801318.
34. B. Herren, T. Gu, Q. Tang, M. Saha and Y. Liu, ASME International Mechanical Engineering Congress and Exposition, 2019, V012T10A019.
35. J. M. D’Arcy, H. D. Tran, A. Z. Stieg, J. K. Gimzewski and R. B. Kaner, Nanoscale, 2012, 4, 3075–3082.
36. Y. Joo, G. J. Brady, M. S. Arnold and P. Gopalan, Langmuir, 2014, 30, 3460–3466.
37. L. Liu, J. Han, L. Xu, J. Zhou, C. Zhao, S. Ding, H. Shi, M. Xiao, L. Ding, Z. Ma, C. Jin, Z. Zhang and L.-M. Peng, Science, 2020, 368, 850.
38. C. Klinke, J. B. Hannon, A. Afzali and P. Avouris, Nano Lett., 2006, 6, 906–910.
39. B. Kumar, A. L. Falk, A. Afzali, G. S. Tulevski, S. Oida, S.-J. Han and J. B. Hannon, ACS Nano, 2017, 11, 7697–7701.
40. W. Sun, J. Shen, Z. Zhao, N. Arellano, C. Rettner, J. Tang, T. Cao, Z. Zhou, T. Ta, J. K. Streit, J. A. Fagan, T. Schauss, M. Zheng, S.-J. Han, W. M. Shih, H. T. Maune and P. Yin, Science, 2020, 368, 874.
41 M. Zhao, Y. Chen, K. Wang, Z. Zhang, J. K. Streit, J. A. Fagan, J. Tang, M. Zheng, C. Yang, Z. Zhu and W. Sun, *Science*, 2020, **368**, 878.
42 J. H. Dwyer, Z. Shen, K. R. Jinkins, W. Wei, M. S. Arnold, R. C. Van Lehn and P. Gopalan, *Langmuir*, 2019, **35**, 12492–12500.
43 G. J. Brady, A. J. Way, N. S. Safron, H. T. Evensen, P. Gopalan and M. S. Arnold, *Sci. Adv.*, 2016, **2**, e1601240.
44 G. J. Brady, Y. Joo, M.-Y. Wu, M. J. Shea, P. Gopalan and M. S. Arnold, *ACS Nano*, 2014, **8**, 11614–11621.
45 H. Yeon, C. Wang, R. C. Van Lehn and N. L. Abbott, *Langmuir*, 2017, **33**, 4628–4637.
46 C. E. Ayres, B. S. Jha, H. Meredith, J. R. Bowman, G. L. Bowlin, S. C. Henderson and D. G. Simpson, *J. Biomater. Sci., Polym. Ed.*, 2008, **19**, 603–621.
47 E. E. Morrill, A. N. Tulepbergenov, C. J. Stender, R. Lamichhane, R. J. Brown and T. J. Lujan, *Biomech. Model. Mechanobiol.*, 2016, **15**, 1467–1478.
48 E. Brandley, E. S. Greenhalgh, M. S. P. Shaffer and Q. Li, *Carbon*, 2018, **137**, 78–87.
49 J. Zhang, J. Cui, F. Wei, W. Wang, X. He and X. Mei, *Appl. Surf. Sci.*, 2020, **504**, 144397.
50 K. R. Williams, K. Gupta and M. Wasilik, *J. Microelectromech. Syst.*, 2003, **12**, 761–778.
51 M. J. Shea, J. Wang, J. T. Flach, M. T. Zanni and M. S. Arnold, *APL Mater.*, 2018, **6**, 056104.
52 C. Kanimozi, G. J. Brady, M. J. Shea, P. Huang, Y. Joo, M. S. Arnold and P. Gopalan, *ACS Appl. Mater. Interfaces*, 2017, **9**, 40734–40742.
53 M. R. Kagan and R. L. McCreery, *Langmuir*, 1995, **11**, 4041–4047.
54 M. M. Tirado, C. L. Martinez and J. G. de la Torre, *J. Chem. Phys.*, 1984, **81**, 2047–2052.