DIRECT VAN DER WAALS INTEGRATION OF 2D MATERIALS FOR HIGH-PERFORMANCE CHEMICAL SENSORS

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ABSTRACT

We present a platform for direct van der Waals integration of two-dimensional (2D) materials into high-performance chemical sensors. In our approach, the fabrication of device layers and their integration with 2D materials are separated. Prefabricated device layers are engineered to have sufficient surface interactions to directly pick up the 2D layer upon contact. This enables a single-step 2D material-to-sensor integration that preserves pristine interfaces, while uniquely keeping the surfaces fully exposed to the environment, allowing for high-sensitivity detection. Using this platform, we fabricated and characterized MoS_2 devices for NO_2 gas sensing, demonstrating high sensitivity compared to conventionally fabricated devices. Our versatile one-step integration platform can be extended to other materials and analytes, and allow for rapid screening of emerging 2D materials for new sensing functionalities.

KEYWORDS

Chemical sensing, Gas sensing, Van der Waals integration, 2D materials, Bottom-up integration.

INTRODUCTION

Two-dimensional materials present an exciting platform for fabrication of novel chemical sensors [1]. Their atomically thin nature results in high surface area-to-volume ratios, allowing for high-sensitivity, low-power devices. Furthermore, the existing library of 2D materials includes diverse chemical and electronic properties, providing opportunities for selective sensing of different analytes in complex environments [1]. To take advantage of these properties in a sensing system, the 2D material needs to be integrated with patterned metal and dielectric features. Conventional approaches for this integration require transfer of the 2D material to a support substrate, and post-transfer processing on top. This can induce damage to the 2D material through exposure to polymers, solvents and high-energy processing, resulting in devices that are limited by processing artifacts rather than their intrinsic properties.

Van der Waals (vdW) integration, where layers are physically stacked and held together by vdW forces, presents an alternative integration approach that enables diverse heterostructures with clean interfaces [2]. Despite the clean interfaces, current vdW integration strategies typically result in encapsulated 2D materials where the top surface is not exposed for sensing applications. Chemical, thermal or physical removal of the surface encapsulating sacrificial layer is feasible, yet can introduce damage to the 2D film. Additionally, this is often followed by subsequent lithographic fabrication steps for device implementation, which can further exacerbate the induced material damage. Using our newly developed adhesive matrix transfer platform [3], here, we present an approach for single-step vdW integration of 2D materials into functional sensors. This allows for scalable, fast fabrication of 2D sensors with clean interfaces, in a design where the surface is fully exposed to maximize interaction with the analyte. The unique combination of these features provides an ideal platform for the development of high-performance chemical sensors, which we highlight by demonstrating sensitive MoS_2-based NO_2 gas sensors.

FABRICATION APPROACH

Our adhesive matrix transfer approach, summarized in Figure 1a, enables the direct integration of 2D materials into devices, with no post-transfer processing. It begins by fabricating the functional elements of a device, which here is in the form of a transistor with gate, source, and drain contacts, on a low-adhesion carrier wafer (i). These features are then delaminated from the carrier wafer (ii) and immediately contacted to a 2D material source (iii). By designing our device electrodes such that the low-adhesion gate dielectric is surrounded by a high-adhesion matrix (here, the metallic electrodes), we are able to use surface forces to directly transfer a monolayer of 2D material to the device electrodes (iv) through a single contact-and-release step, without the conventional need for sacrificial layers, high temperatures or solvents. The resulting device (Figure 1c) is fully pristine with atomically clean interfaces as evidenced in the transmission electron microscope (TEM) image in Figure 1d.

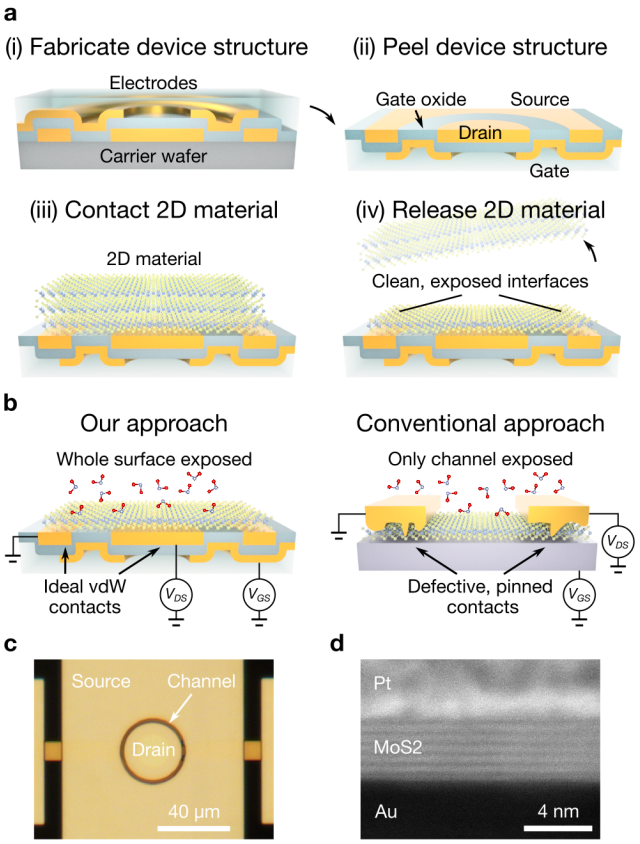

Figure 1: Direct integration of 2D materials-to-sensors with pristine interfaces and exposed surfaces. (a) Device structures are fabricated (i), delaminated (ii), and contacted with the 2D material source (iii), then released (iv) to form the sensor. (b) Schematic illustration of our approach compared to the conventionally fabricated designs. (c) Microscope image of a representative sensor fabricated from a monolayer MoS_2. The resulting interfaces are clean as evidenced by (d) TEM of a multilayer sample.

This single-step 2D material-to-device integration has several features which make it ideal for fabricating sensors based on 2D materials. This platform is compatible with diverse 2D materials and

978-1-940470-05-4/HH2024/$25©2024TRF
DOI 10.31438/TRF.HH2024.125

is scalable, while keeping surfaces and interfaces pristine. Additionally, it allows sensor designs where the 2D material sensing surface is fully exposed to the environment, with both the contact and channel regions available to interact with the analytes (Figure 1b). This is in contrast to conventional devices, where the contact is deposited on top of the 2D material (Figure 1b). Though this geometry is standard, it can be problematic for sensing applications. The high-energy contact deposition damages the 2D material and leads to Fermi level pinning, causing the contact to be largely insensitive to changes in carrier density. In contrast, vdW interfaces show suppressed Fermi level pinning [4], making them sensitive to changes in carrier density induced by the analyte. Additionally, conventional designs often sandwich the 2D layer between the metal contacts and an underlying substrate, preventing direct exposure to the analyte and potentially reducing sensitivity. In contrast, our approach leaves the contacts clean and exposed, allowing for the analyte to directly and efficiently interact with the 2D semiconductor in this sensitive region.

DIRECT VDW INTEGRATION FOR GAS SENSING

To highlight the prospects of our fabrication approach for chemical sensing, we tested the sensitivity of the device shown in Figure 1c to gaseous NO_2. This benchmark gas is widely employed in the literature as an electron withdrawing analyte [1], and allows us to compare the sensitivity of sensors fabricated through our approach to devices which are fabricated through conventional post-transfer processing. The electrical characterization of our device in both dry air and 10 ppm NO_2 is shown in Figure 2. The transfer and output characteristics of our device (Figure 2a) show a pronounced decrease in device conductivity after exposure to NO_2. Examination of the transfer characteristics on a logarithmic scale (Figure 2b), shows that the threshold voltage shifts to higher values after NO_2 exposure and the off-state current decreases. The former is primarily due to a shift in the Fermi level in the channel, whereas the latter is due to reduction in carrier density at both the channel, and the exposed contacts in our device. This multimodal response can potentially be leveraged to discriminate between different gases which have different effects on the channel and contact regions in a multianalyte sensing scheme.

Figure 3 characterizes the sensitivity of our device by sequentially exposing it to increasing NO_2 concentrations. The normalized resistance of our device at a fixed gate/drain bias as a function of time is shown in Figure 3a. An increase in resistivity is observed with increasing gas concentrations. Figure 3b characterizes the response of our device as a function of concentration. We observe a linear device response within the tested

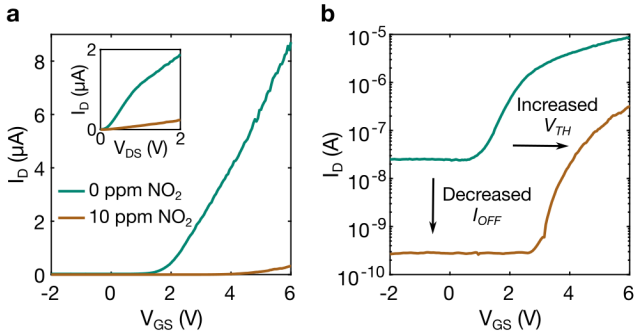

Figure 2: Electrical characterization. (a) Transfer characteristic (V_{DS} = 1 V) and output characteristic (inset, V_{GS} = 4V) in dry air and NO_2. (b) Transfer characteristic on logarithmic scale, showing device response consisting of both a shift in threshold voltage (V_{TH}) and decrease in off-state current (I_{OFF}).

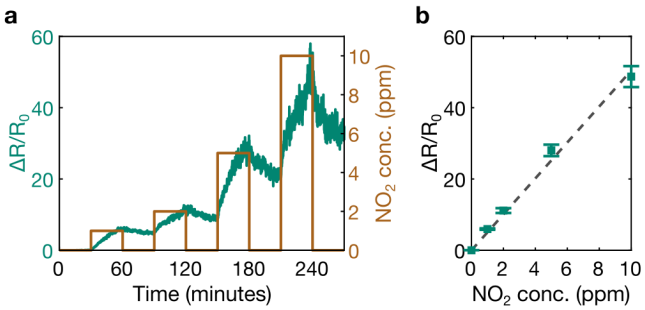

Figure 3: Sensing response of MoS_2 transistor. (a) Change in resistance as a function of time for MoS_2 device exposed to various concentrations of NO_2. (b) Device response vs. NO_2 concentration showing linear response with high sensitivity. Error bars represent standard deviation of measured resistance after stabilization.

concentration range, with a sensitivity $\Delta R/R_o$ = 504 %/ppm. In contrast, previously reported monolayer MoS_2 devices with deposited ohmic contacts show sensitivities of < 10 %/ppm for NO_2 exposure [5,6]. Our large enhancement in sensitivity compared to these conventionally fabricated devices shows the prospects of our approach as a platform for high-performance 2D material sensors.

CONCLUSION

We present adhesive matrix transfer as a platform for facile, single-step 2D materials-to-sensors vdW integration with pristine and exposed surfaces and interfaces. These features are ideal to maximize the electrical response of the 2D material device to a chemical analyte. Using NO_2 as a benchmark gas, we demonstrate that devices fabricated through our approach show high sensitivities compared to conventionally fabricated devices with a similar contact type. Future work will use the multi-modal response of our devices at both the contact and channel to improve selectivity when sensing in complex environments, and use this rapid integration approach to screen emerging 2D materials for chemical sensing applications.

ACKNOWLEDGEMENTS

This work was supported by the National Science Foundation (NSF) Award No. CMMI-2135846.

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