Abstract

Detecting minute concentrations of gaseous pollutants in the atmosphere and human breath enables environmental monitoring and non-invasive detection of ailments. Consequently, due to graphene's single-molecule sensitivity, it is highly sought after for high sensitivity gas detection. However, its poor selectivity, huge p-doping in the atmosphere, and inability to identify adsorbed gases makes graphene less useful for practical applications in environmental and clinical gas sensors which are typically deployed in atmospheric conditions. Hence in this work, we develop a chemical vapour deposition (CVD) graphene-based sensor with extreme ammonia sensitivity and selectivity in atmospheric air. Furthermore, we demonstrate the van der Waals (vdW) bonding memory of adsorbed gases on graphene and consequently demonstrate a proof-of-concept, the Charge Neutrality Point Disparity (CNPD), that characterizes various gases in atmospheric air.

First, we demonstrate the gas sensing, electrical properties, and morphological characteristics of a 38 nm oxidized activated carbon functionalized graphene field-effect transistor (a-CF-GFET) sensor. We show that the activated carbon (a-C) on the graphene channel passivates the graphene channel against p-doping in the atmosphere while simultaneously enhancing ammonia selectivity. Consequently, 500 parts per trillion (ppt) of ammonia was detected in the atmosphere with a response time of 3 seconds making the a-CF-GFET sensor the most sensitive ammonia selective sensor so far reported. The extreme ammonia sensitivity makes the a-CF-GFET sensor suitable for environmental monitoring and non-invasive medical diagnosis of ailments such as ulcers and kidney problems using ammonia as a biomarker.

Furthermore, we demonstrate that the electric field induced graphene-molecule charge transfer is retained in the graphene-molecule vdW complex even after the electric field is turned off, with the retained charge still unique to the applied electric field magnitude and direction.

Consequently, the vdW bonding memory of adsorbed molecules on graphene was observed. This bonding memory is important for the molecular identification of gases based on their electrically tunable charge transfer.

Following the observation of the vdW bonding memory, we demonstrate a proof of concept for a charge transfer based molecular identification technique, the CNPD method which measures the charge-transfer induced by consecutive applications of $\mp$tuning voltages ($\mp V_t$). The difference between the $-V_t$ and $+V_t$ induced charge transfer obtained from the shift of graphene's charge neutrality point was characteristic of various gas environments detecting parts per billion concentrations of ammonia and acetone.

In conclusion, we demonstrate a facile fabrication route for the simultaneous activation of the graphene channel towards ammonia selectivity while passivating it against atmospheric p-doping. Thereafter we show that adsorbed gases on graphene possess a tuning voltage induced van der Waals bonding memory lasting over 2h. Finally, following the demonstration of this vdW bonding memory, a proof-of-concept for a charge-based molecular identification technique, CNPD, which characterizes adsorbed gases based on their electrically tunable charge transfer is demonstrated.

Keywords: CVD graphene, ammonia selectivity, activated carbon, scattering, molecular identification