Supporting Information

Memristive Behavior of V_2C MXene/PANI:PSS Composite and Its Applications in Ammonia Detection

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In the Materials synthesis and testing section of the paper, we previously included a simple flowchart of the composite material preparation process. We have now added the specific experimental procedures and testing setup:

1. **Preparation of V_2CT_x MXene:** First, a mixed solution of (20 mL (40%) HF + 20 mL 12M HCl + 10 mL deionized (DI)) was added to a 250 mL polytetrafluoroethylene (PTFE)-lined reactor. Then, 2 g of V_2AlC was weighed and added slowly in small portions to the reactor over approximately 15 minutes. The reactor speed was adjusted to 500 rpm, and the reaction was carried out at 40 °C for 48 h. After the reaction, the resulting slurry was poured into two 50 mL centrifuge tubes. Centrifugation was performed 7-8 times at 8500 rpm for 5 minutes each time, until a viscous precipitate was obtained. The supernatant liquid was then decanted. Next, 50 mL of a 5 wt% TMAOH solution was prepared and added to the viscous precipitate. After manual mixing, the mixture was poured into a 100 mL beaker and stirred at room temperature for 6 h (sealed with plastic wrap). After stirring, the solution was poured into a 50 mL centrifuge tube and centrifuged twice at 8500 rpm for 5 minutes. The solution was then filtered using a 0.22 μm aqueous filter membrane. After drying, the sample was placed in a vacuum drying oven at 40 °C for 2 h, then removed and ground into a powder to obtain V_2CT_x MXene.
2. **Preparation of V_2CTx/PANI:PSS nanocomposite:** First, 20 mg of multilayered V_2CT_x powder was dispersed in 30 mL of deionized (DI) water, followed by the addition of 40 mg of poly (sodium 4-styrenesulfonate) (PSS) acting as both a dopant and a dispersion stabilizer, along with 40 mg of aniline monomer. The mixture was ultrasonicated at room temperature for 10 min to ensure adequate monomer intercalation and the formation of a homogeneous suspension. Subsequently, the reaction system was transferred to an ice-water bath (0–4 °C) to regulate the polymerization rate. Under vigorous magnetic stirring, an aqueous solution containing 100 mg of ammonium persulfate (APS) was slowly added dropwise as

the initiator (maintaining an aniline-to-APS molar ratio of approximately 1:1). The reaction proceeded under ice-bath conditions for 4 h, during which the solution color gradually transitioned to dark green due to the formation of the polyaniline emeraldine salt. Upon completion of the reaction, the product was washed alternately with DI water and anhydrous ethanol at least three times to thoroughly remove oligomers and residual oxidants, finally yielding the V_2CT_x /PANI:PSS nanocomposite.

3. **Preparation of gas sensor:** First, the Au interdigitated electrodes were ultrasonically cleaned in acetone, anhydrous ethanol, and deionized water for 5 minutes each to remove surface organic residues and dust, and then dried with a nitrogen gun. Next, 5 μL of the prepared V_2CT_x/PANI:PSS nanocomposite dispersion was drop-cast onto the sensing area of the cleaned interdigitated electrodes using a micropipette. Finally, the drop-cast device was allowed to dry naturally at room temperature for 1 h, and then transferred to a vacuum oven and heat-treated at 60 °C for 4 h to remove residual solvent and enhance the interfacial contact between the film and the electrodes, forming a sensor with a uniform and dense sensing film.
4. **Testing setup:** In the testing setup, a Keithley 2614B System Source Meter was primarily employed to conduct comprehensive electrical characterization and testing of the device. Specifically, the Source Meter was utilized to: (1) acquire static current-voltage (I-V) characteristic curves to evaluate the bipolar resistive switching behaviors; (2) serve as a pulse generator to apply voltage stimuli for simulating synaptic plasticity; and (3) monitor the real-time dynamic resistance/current responses of the sensor upon exposure to various gas environments.

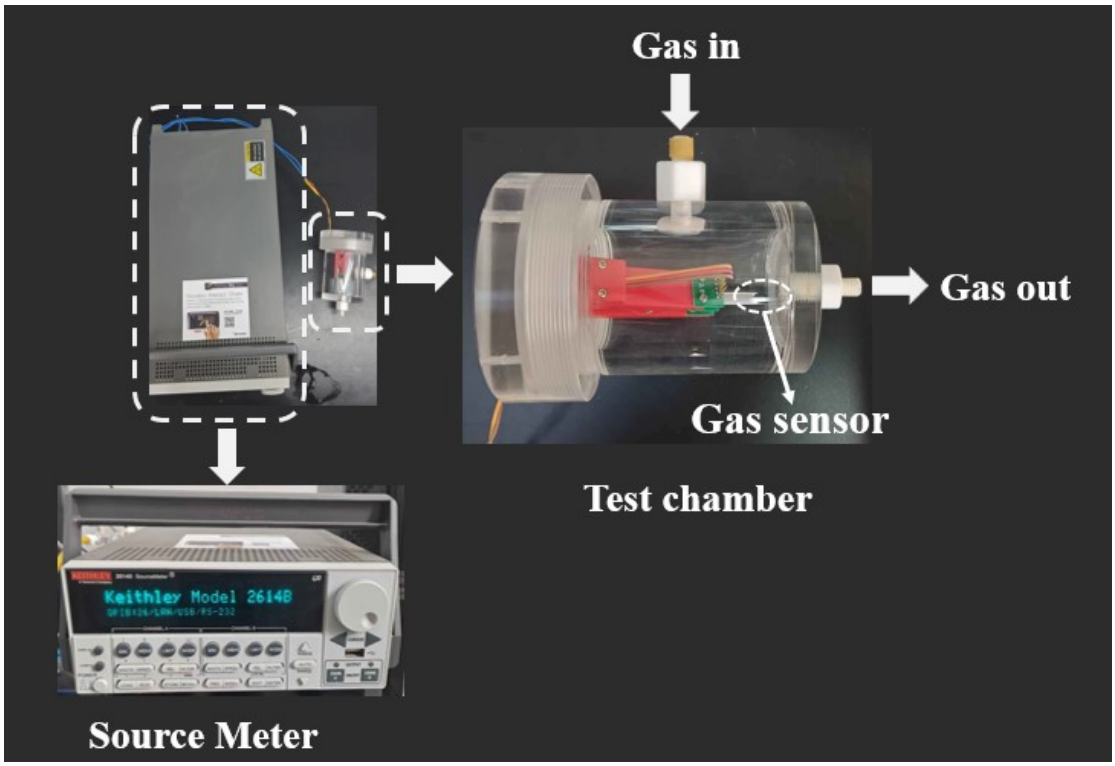

Figure S1. A photograph of the gas-sensitive memristor device and a schematic diagram of its assembly are shown. The inset illustrates the details of the device's connection to the external circuit.

As shown in the updated Figure S2, we increased the number of switching cycles to 10^4. The test results demonstrate excellent stability and a distinct switching window over 10^4 consecutive cycles. Both the High Resistance State (HRS) and Low Resistance State (LRS) currents remained stable at approximately 10^{-6} A and 10^{-4} A, respectively, maintaining a robust On/Off ratio of $>10^2$. Throughout the test, the data exhibited a flat distribution profile with negligible window drift or device fatigue.

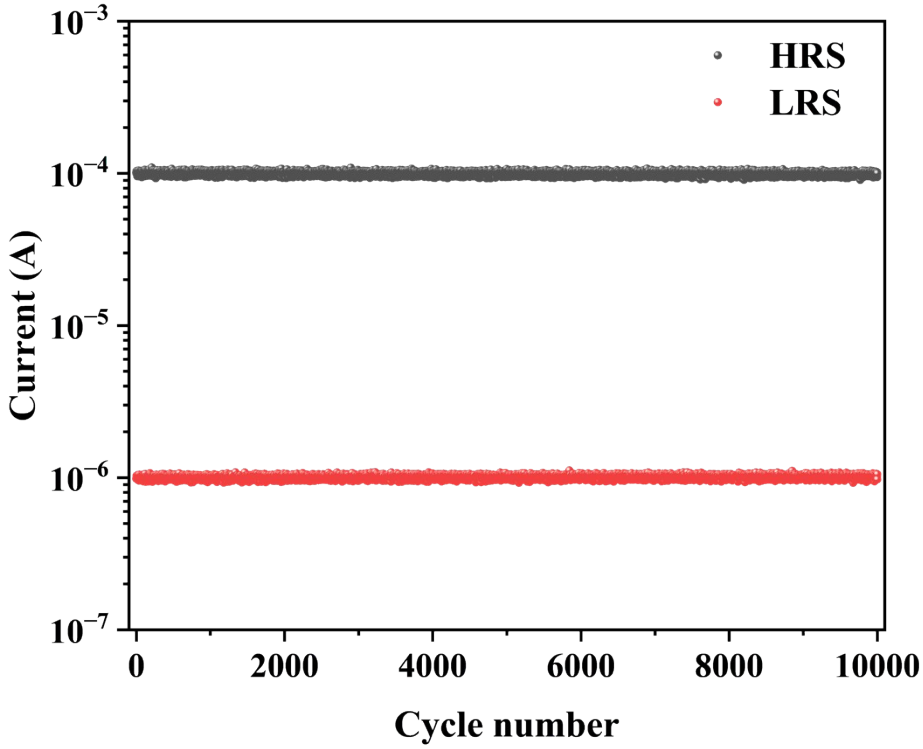

Figure S2. Endurance of the two resistance states with tight HRS/LRS distributions (ON/OFF > 10^2) over 10^4 switching cycles.

The XRD patterns show that the pure conductive PANI:PSS exhibits a broad amorphous diffraction peak in the 20 – 30° range, which is a typical XRD characteristic peak of conductive PANI, different from undoped intrinsic polyaniline. The XRD patterns of the V_2CT_x/PANI:PSS composite material show characteristic peaks of both V_2CT_x and PANI, indicating that no significant chemical changes or further oxidation occurred in the composite material throughout the gas sensing and memristive cycling processes.

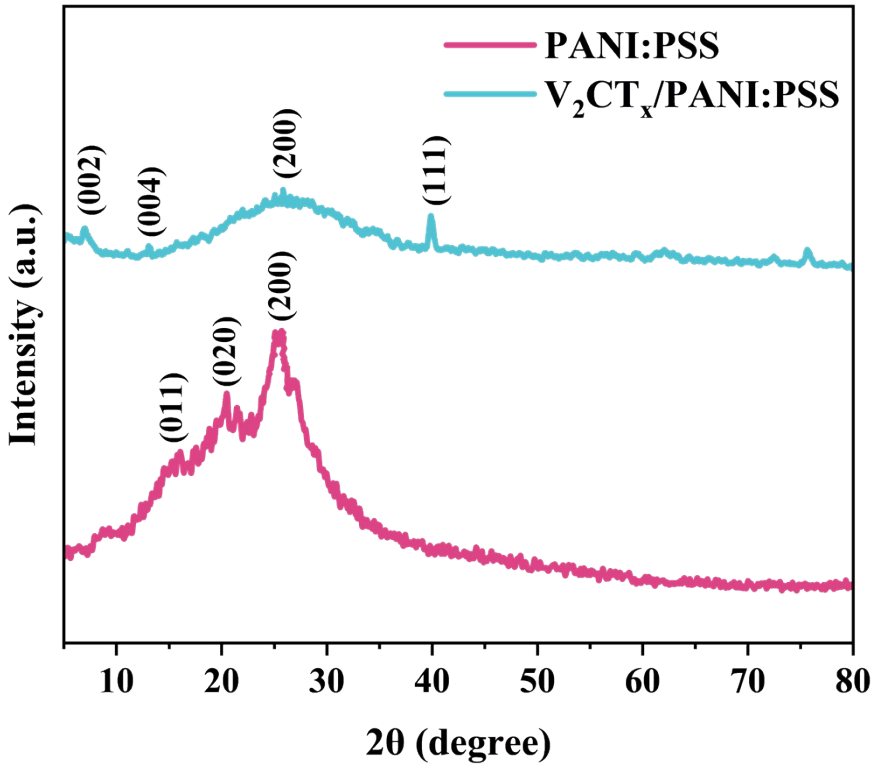

Figure S3. XRD patterns of PANI:PSS and V_2CT_x /PANI:PSS.

As illustrated in Figure S4, we evaluated the sensor's response to 5 ppm NH_3 under relative humidity (RH) conditions ranging from 30% to 80%. The experimental data reveal a slight downward trend in the response value as RH increases from 30% to 80% (decreasing from 32.5% at 30% RH to 30.2% at 80% RH). This minor decline aligns with surface adsorption theory. In high-humidity environments, water molecules in the atmosphere tend to adsorb onto the material surface, occupying a fraction of the active sites that would otherwise be available for ammonia molecules. This competitive adsorption mechanism partially hinders the effective interaction between the target gas molecules and the sensing material, thereby resulting in the observed slight reduction in response.

Despite the existence of this physical competition mechanism, given that the impact of humidity on the sensor is minimal (with a fluctuation of < 2.5%), we conclude that the device possesses sufficient robustness to cope with variations in environmental humidity.

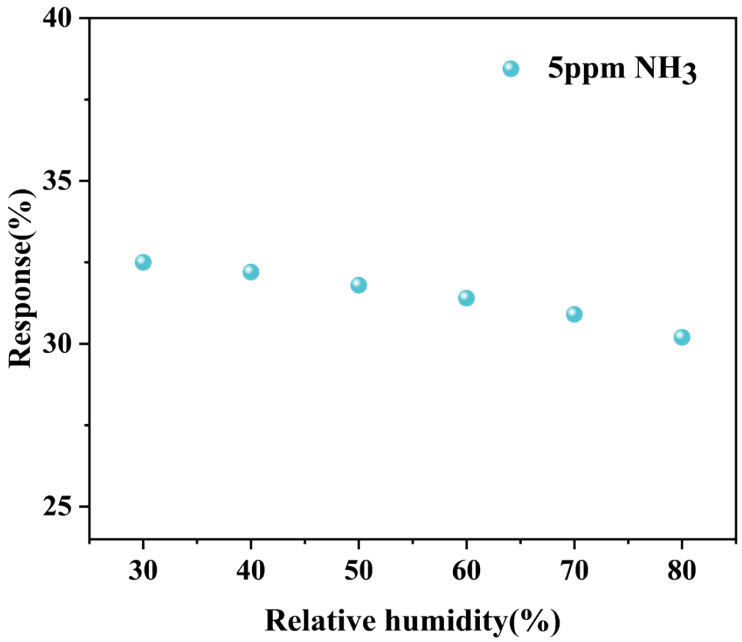

Figure S4. The relationship between response values and RH, measured at 5 ppm NH_3, under a humidity range of 30 – 80% RH.

Figure S5 clearly illustrates the transition from STD to LTD: the device exhibits a distinct dependence on stimulation intensity. Fewer pulse cycles (2 cycles) induce behaviors characteristic of STD, whereas increasing the number of pulse cycles (to 8 cycles) effectively consolidates the weight change, thereby generating stable LTD. Upon applying the stimulation pulses, the Inhibitory Post-Synaptic Current (IPSC) decreases significantly. Although a slight spontaneous recovery occurs immediately after the pulses, the current stabilizes at a lower level as the number of pulses increases. This sustained suppressed state confirms that the synaptic weight has been effectively updated, successfully mimicking biological Long-Term Depression (LTD) behavior.

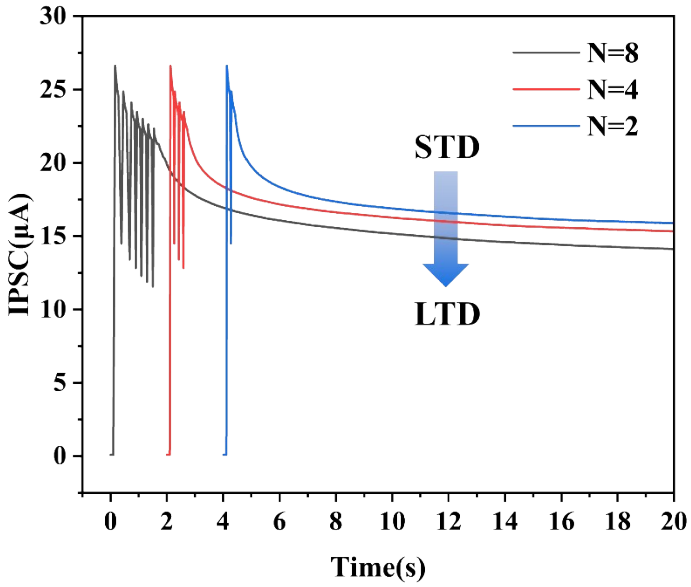

Figure S5. The transition from STD to LTD by increasing the number of pulses.