Ultrasensitive and stable all graphene field-effect transistor-based Hg\(^{2+}\) sensor constructed by using different covalently bonded RGO films assembled by different conjugate linking molecules

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Abstract
As "molecular bridge," coupling agents can not only realize the covalent connection of composites, but also affect their properties, thus affecting the properties of devices based on them. Herein, leveraging differences in charge conduction properties of the (3-aminopropyl)trimethoxysilane and 5,10,15,20-tetrakis(4-aminophenyl)-21H,23H-21H,23H-porphine caused by conjugacy structural differences, two kinds of layer-by-layer assembled smart carbon materials with different electrical properties are obtained at the same reduction temperature. The two graphene ultrathin films are then “planted” on Si/SiO\(_2\) substrates, respectively, as semiconductor layer and source/drain electrodes to fabricate an ultra-stable all-graphene field effect transistor (AG-FET). Enabled by the covalent functionalized configuration and the functionally diverse of coupling agents, the AG-FET obtained by this simple method won the high electrical characteristics, the hole, electron mobility, and the shelflife could reach 3.79 cm\(^2\)/(V·s), 3.78 cm\(^2\)/(V·s), and 18 months, respectively. In addition, good material stability and excellent device structure endow the device exceptional stability, electrical stability, and solvent resistance, improving its application prospect in solution phase sensing/detection. Such characteristics could be used to sense, transduce, and respond to external stimuli, especially in solution...
phase to monitor the important analytes, such as Hg$^{2+}$ in a flowing sewage environment. We believe that such easy-to-manufacture AG-FETs with ultrahigh performance and ultrahigh stability could also show great application prospects in other significant fields.

**KEYWORDS**
coupling agent, covalent bond, field effect transistors, Hg$^{2+}$ detection, ultra-stable

1 | INTRODUCTION

Field effect transistor (FET) has emerged as a novel multi-parameter, high-sensitivity, portable biochemical detection platform for its typical three-terminal structure, working principles, and processing methods.$^{1-7}$ Despite FETs constructed by photolithography are maturely implemented in various electronic devices, most FET devices are connected by weak interactions between the electrodes and semiconductor layers by physical deposition techniques,$^{8-10}$ their structure are vulnerable to damage during multistep post-production treatment. This vulnerability exacerbates the instability of electrical signal output, especially in applications such as biochemical sensor that require humid environments. To overcome these concerns, a series of optimization methods have been innovated, such as the design and synthesis of solution-resistant polymer semiconductor materials,$^{11-14}$ the introduction of protective structures on FETs,$^{15,16}$ and the construction of epitaxial grids.$^{17,18}$ While these methods prevent the interference of external forces on electrical signals to some extent, they typically suffer from complicated preparation steps, expensive processing techniques, and the degeneration of device performance.$^{19}$

In addition, the materials used as electrodes for the traditional FETs are very limited, and most of them are precious metals.$^{20,21}$ Due to limitations on the structures and properties of materials, a perfect match between the work functions (WFs) of the precious metals and the energy levels of the lowest unoccupied molecular orbitals (LUMOs)/conduction bands (CBs) and the highest occupied molecular orbitals (HOMOs)/valence bands (VBs) of the semiconductors is generally impossible, which leads to the large barrier between the electrode and the active material and lowers the efficiency of charge injection and transmission.$^{22}$ Although, through molecular engineering, crystal engineering, and interface engineering, researchers have made great efforts to achieve good injection and transmission of carriers in such devices,$^{23}$ the high price caused by precious metals remains a problem. Therefore, crafting FETs to maintain high stability while also being low-cost, and ease of manufacture characteristics remains a formidable challenge.

Reduced graphene oxide (RGO) is a kind of graphene derivative materials possessing multiple oxygen-containing functional groups, which exhibit distinguished advantages for electronic applications.$^{24-26}$ For one thing, it can greatly reduce the cost of the preparation of electronic components because of its facile preparation and mass production.$^{27}$ For another thing, by taking the distinguished merits of easy to chemically modified, RGO can react chemically with organic substances to form stable covalent bonds and finally construct super-stable films. Moreover, due to the electrical properties of RGO films are different under different reduction levels or different assembly modes, all graphene FET could be fabricated by using high conductivity RGO as electrodes and low conductivity RGO as a semiconductor.$^{28,29}$

Therefore, in this paper, leveraging differences in charge conduction properties of $(3$-aminopropyl)trimethoxysilane (APTMS) and 5,10,15,20-tetrakis (4-aminophenyl)-21H,23H-porphine (TAPP) caused by structural differences, two kinds of smart carbon materials, as shown in Scheme 1. Then an ultra-stable all-graphene (AG-FET) were fabricated by means of easy solution-processing techniques together with a one-step mild annealing in situ reduction. The AG-FET obtained by such simple method has remarkable electrical characteristics, with hole mobility, electron mobility, and shelflife potentially reaching 3.79 cm$^2$/V·s, 3.78 cm$^2$/V·s, and 18 months, respectively, which can be the ideal sensor platform for the detection of mercury ions in sewage, even in pipeline monitoring. The limit of Hg$^{2+}$ detection can reach as low as 16 pM, with a detection error of less than 17% in simulated sewage.

2 | EXPERIMENTAL SECTION

2.1 | Assembly of GO films

A coupling agent solution was created by dissolving 2 vol% APTMS in a 5:95 (v:v) water/ethanol solvent.
A hydroxylated SiO$_2$/Si wafer was alternatingly and repeatedly immersed in the coupling agent solution for 30 min and a GO solution (0.1 mg/ml) for 45 min. Multilayered GO LBL films called (APTMS/GO)$_N$ were obtained, where $N$ was the number of assembly layers. The APTMS/GO-modified substrate was then immersed in a DMSO solution of TAPP for 60 min at 70°C and then in a GO solution for 60 min at room temperature. This entire sequence of operations was conducted alternatingly and repeatedly to obtain (APTMS/GO)/(TAPP/GO)$_N$, where $N$ was the number of assembly layers.

### 2.2 Construction of AG-FET, all-covalent-rooted FET (ACR-FET), and Au/RGO/Au FET

The multilayered (APTMS/GO)/(TAPP/GO)$_N$ films were formed via above method. Then, using a copper gate as a mask, an aluminum film of 40 nm was deposited on the multilayer GO film, and the unmasked area of the GO film was removed using an oxygen plasma cleaner. The multilayered GO films were patterned on the Si/SiO$_2$ substrate by removing the aluminum film via a wet etching method. After that, the (APTMS/GO)$_N$ films were again grafted onto the pattern-multilayered GO arrays via covalent LBL assembly of GO. Subsequently, the whole device was reduced via a one-step reduction method, that is, annealing at 160°C for 6 h, so as to construct a AG-FET in which (APTMS/RGO)/(TAPP/RGO)$_N$ films with relatively good electrical properties acted as the electrodes, and (APTMS/RGO)$_N$ films with relatively poor electrical properties acted as the semiconductor layer. AG-FET electrical performance was measured using a semiconductor test platform (Cascade EPS150TRIA + Tektronix 4200SCS) at room temperature.

The ACR-FET electrodes were fabricated by assembling (APTMS/GO)$_N$ films, reducing at 240°C for 6 h, and patterning the (APTMS/RGO)$_N$ films via the above method. Sequentially, the (APTMS/GO)$_N$ films were again assembled on the ACR-FET electrodes and reduced at 160°C for 6 h to construct ACR-FETs.

As for the Au/RGO/Au FET, its semiconductor layer of the Au/RGO/Au FET was same as that of the ACR-FETs. The electrodes were Au constructed by vacuum-deposition after masking.

### 2.3 Modification of AuNPs and immobilization of ssDNA

AG-FETs were immersed in a 10 mmol/L HAuCl$_4$ solution for 30 min to fabricate AuNPs-decorated AG-FETs. AuNPs-modified AG-FETs were immersed in a 10 μmol/L ssDNA (5'-SH-TCA TGT TTG TTT GTT GGC CCC CCT TCT TTC TTA-3') solution and incubated for 90 min,
followed by incubation in a saline solution (10 mmol/L Tris-HCl, 1 mmol/L EDTA, 1 mol/L NaCl, pH = 7.5) for 15 min to obtain ssDNA-AuNPs-decorated AG-FETs.\textsuperscript{31}

2.4 Detection of Hg\textsuperscript{2+} based on ssDNA-AuNPs-decorated AG-FETs

For sensing experiments, our ssDNA-AuNPs-decorated AG-FETs were exposed to solutions with various cations (Hg\textsuperscript{2+}, K\textsuperscript{+}, Co\textsuperscript{2+}, Ca\textsuperscript{2+}, Zn\textsuperscript{2+}, and Hg\textsuperscript{2+} in simulated sewage) and incubated for 15 min. A blank control test was performed with Milli-Q water. The $V_G$-$I_{SD}$ characteristics of the ssDNA-AuNPs-decorated AG-FETs were measured by sweeping $V_G$ from +15 to $V_{SD}$ of 0.1 V, with the $\Delta V_{CNP}$ of the ssDNA-AuNPs-decorated AG-FETs induced by the analytes used as the sensing parameter.

For details on the materials and methods, please refer to the Supporting Information.

3 RESULTS AND DISCUSSION

3.1 Characterization of coupling agents cross-linked graphene oxide and RGO ultrathin films and AG-FETs

In FET devices, the morphology and characteristics of the electrodes and semiconductor are closely related to the performance of the device, so it is necessary to investigate the properties of the (APTMS/GO)$_N$ and (APTMS/GO)/(TAPP/GO)$_N$ films. According to the Fourier-transform infrared (FT-IR) spectra displayed in Figure 1A, the (APTMS/GO)$_N$ films (Figure 1A) show many characteristic bands such as Si–O–C stretching at 1036 cm$^{-1}$, Si–O–Si asymmetric stretching at 1109 cm$^{-1}$, aliphatic C–N stretching at 1225 cm$^{-1}$, and C–NH deformation and C–N stretching at 1575 cm$^{-1}$. Other characteristic GO bands at 1730, 2851, 2925, and 3400 cm$^{-1}$ are also apparent and are respectively attributed to the characteristic stretching vibration of C=O, the symmetric and antisymmetric vibrations of CH$_2$, and the...
O–H vibration of C–OH. These observations demonstrate the successful assembly of (APTMS/GO)_N films through a covalent reaction between APTMS and GO. As for the (APTMS/GO)/(TAPP/GO)_N films (Figure 1Aiii), the abovementioned broad bands, such as the asymmetric stretching of Si–O–Si and the symmetric and antisymmetric vibrations of CH_2, are still observed. This finding is due to the use of APTMS as a coupling agent in the film’s first layer. In addition, several new vibrations are apparent at 1525, 1620, 3220, and 3350 cm\(^{-1}\), assigned respectively to the bending and scissoring vibrations of N–H and to symmetric and asymmetric N–H stretching.\(^{29,32,33}\) The GO-related groups are also clearly observable. These experimental data indicate that TAPP and GO were successfully integrated in the films via the covalent LBL method.\(^{34}\) The Raman spectra of the two formulated GO films are displayed in Figure 1Bii,iii. Here, the two formulated GO films exhibit the Raman features of GO nanosheets at 1352 and 1589 cm\(^{-1}\), respectively forming corresponding (APTMS/RGO)/N films. As for composite materials, the coupling agent acts as a molecular bridge that not only plays a coupling role, but also influences its performance in various aspects.\(^{36–40}\) Therefore, the electrical properties of both films were investigated. As can be observed in Figure 1D, the conductivity of (APTMS/RGO)/(TAPP/RGO)_N was more prominent than that of (APTMS/RGO)_N at the same reduction temperature, which is owing to the unique planar, rigid molecular geometry and aromatic electron configuration of TAPP facilitated efficient charge carrier transport between RGO layers while the APTMS with electrically non-active, as shown in Scheme 1. It also demonstrates the feasibility of constructing our FET devices by a one-step reduction method. On the basis of the successful construction of the above two different electrical performance thin films using the construction method illustrated in Figure 1E, we obtained well-defined (APTMS/GO)/(TAPP/GO)_4 and large-area (APTMS/GO)_5 on the same substrate. As displayed in Figures S5 and 1F, the thickness difference between these two films was maintained at ca. 10.3 nm, in accordance with the measured thickness of (APTMS/GO)/N films have been removed. Moreover, the CH_2 vibrations, the Si–O–C stretching, the Si–O–Si asymmetric stretching, the aliphatic C–N stretching, and the C–NH deformation and C–N stretching of (APTMS/RGO)/(TAPP/RGO)_N films survived. The FT–IR spectra of (APTMS/RGO)_N films show changes similar to those of (APTMS/RGO)/(TAPP/RGO)_N films. These observations demonstrate the successful reduction of two GO films and the survival of covalent coupling agents with thermal annealing. According to the UV–vis spectra of the two as-fabricated RGO films shown in Figure 1C, the absorption at 231 nm shifted to 267 nm in both cases, confirming the achievement of annealing-induced reduction. In addition, the Soret band absorption of TAPP at 436 nm still appears for the (APTMS/RGO)/(TAPP/RGO)_N, suggesting that TAPP survives well after thermal annealing. These results are further verified by the two RGO films’ declines in the G-band/D-band intensity ratio (I_G/I_D) in their Raman spectra (Figure 1B) and their declines in the oxygen/carbon atom ratio (O/C) in their XPS spectra (Figure S3 and S4). All the above characterization indicates that the GO and RGO films can be and indeed were assembled in a controllable manner in morphology and thickness using two coupling agents.

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$$\mu = \frac{L}{W C_{Vi}} \Delta I_{SD}/\Delta V_G,$$

where $L$ is the channel length (20 $\mu$m), $W$ is the channel width (200 $\mu$m), $C_i$ is the gate capacitance and $I_{SD}/V_G$ is the transconductance in the linear region.\(^4\) Through measurement and calculation for 50 devices (Figure S6), the highest recorded hole mobility ($\mu_h$), electron mobility ($\mu_e$), and on–off ratio ($I_{on}/I_{off}$) reached 3.79 cm\(^2\)/(V·s), 3.78 cm\(^2\)/(V·s), and 9900. As communicated statistically in Figure 2B,C, the average values for $\mu_h$, $\mu_e$, and $I_{on}/I_{off}$ were 1.52 cm\(^2\)/(V·s), 1.51 cm\(^2\)/(V·s), and 3100, respectively. Comparing with the reported works about various graphene‐based FETs summarized in Table S1, the mobility of our AG‐FET is comparable with theirs, while our FET has significant bipolarity and high $I_{on}/I_{off}$.

To investigate why our AG‐FET performed so well, two systems were constructed as controls: an ACR‐FET, for which the electrodes and the semiconductor layer were both (APTMS/RGO)/(TAPP/RGO)\(_N\) films, respectively reduced at 240°C and 160°C for 6 h; and an Au/RGO/Au FET, for which the electrodes and semiconductor layer were Au and (APTMS/RGO)\(_N\) films, respectively. For details on the construction process, please refer to the supplementary information. Based on the data summarized in Figure 2D,E and Table S2, ACR‐FET and Au/RGO/Au FET both exhibited excellent ambipolar characteristics, but their mobilities and $I_{on}/I_{off}$ values were far from those of AG‐FET. This disparity follows chiefly from the following facts: (i) The reduction temperature of the (APTMS/RGO)/(TAPP/RGO)\(_N\) electrode we used was lower than that of the (APTMS/RGO)\(_N\) of ACR‐FET. However, as displayed in Figure 1E, the resulting electric conduction performance of (APTMS/RGO)/(TAPP/RGO)\(_N\) was outstanding owing to different layer charge transport functions of two coupling agents. (ii) The contact resistances between the electrodes and semiconductor layers of AG‐FET and Au/RGO/Au FET were measured via the transfer length method (TLM).\(^1\) As shown in Figure 2F, the contact resistance between electrodes and semiconductor in AG‐FET is 4.5 × 10\(^3\) $\Omega$·cm much smaller than the contact resistance of 9.5 × 10\(^4\) $\Omega$·cm between electrodes and semiconductor in Au/RGO/Au FET. This is due to although the coupling agents used for electrodes and semiconductor in our AG‐FET were different and manifested different conductivities, they were both RGO of almost identical material, thus facilitating the transfer of charges and making for relatively low contact resistance.

Such good carrier injection and transmission were the main reasons why the performance of AG‐FET was much better than that of Au/RGO/Au FET and ACR‐FET.
These data demonstrate that by using cheap GO and a simple one-step reduction method, a high-performance FET can be fabricated. This design concept can be extended to other materials.

### 3.3 Stability of AG-FETs

Excellent electrical performance is crucial, but it is also very important for the FET device to maintain stable performance during subsequent processing and application. Thanks to the layer-to-structure covalent bonding of our device, the instability found in most FETs in the market has been overcome here. We compared microscopy images (Figure 3A–C) and UV–vis spectra (Figure 3D) of our devices in their original state, after 500 min of continuous ultrasonic treatment in water, and at room temperature after 18 months without any protection. We found only negligible changes to the morphology of the AG-FETs and the properties of the electrodes and semiconductor, whether subjected to the strict physical action of ultrasonication or the long storage process. These results suggest that the (APTMS/RGO)/(TAPP/RGO)$_N$ electrodes and (APTMS/RGO)$_N$ semiconductor layer do not fall off, and rather that the whole device maintains a remarkable covalent connection. FT-IR spectra (Figure S7), Raman spectra (Figure S8), and AFM images (Figure S9) further confirm these conclusions. More importantly, good device structure maintenance lays a solid foundation for good device performance maintenance. Figure 3E,F show the transfer characteristics of our AG-FETs after 500 min of sonication and 18 months of storage, respectively. Only trivial performance degradation is observed, again indicating devices with excellent stability and exceptional shelflife.

### 3.4 Hg$^{2+}$ detection based on AG-FETs

With the excellent electrical stability and ultrahigh solvent resistance of the AG-FET in mind, we implemented one potential application, namely as a sensitive and selective sensor. Mercury ion (Hg$^{2+}$), as an extremely poisonous and widespread environmental contaminant, can cause a series of severe health problems such as brain damage, kidney failure, and various cognitive and motor impairment.$^{43-46}$ Thus, we focused on sensitive and selective Hg$^{2+}$ detection based on AG-FETs as a proof of concept. We designed selective binding sites for mercury ion into the AG-FETs by decorating the AG-FETs with ssDNA-functionalized gold...
nanoparticles (AuNPs) using a previously reported technique.\textsuperscript{43–46} Scheme 3 elucidates the sensing principle of the AG-FETs for Hg\textsuperscript{2+} detection. The AG-FET was immersed in an HAuCl\textsubscript{4} solution for 30 min to obtain AuNPs of uniform size and shape as controlled using the advantageous and unique properties of RGO. Subsequently, the AuNPs modified AG-FET was functionalized with a 33-base thiolated ssDNA probe, previously reported to be selective for Hg\textsuperscript{2+}.\textsuperscript{47} Given that no other reducing agent was used to obtain AuNPs and given the good biocompatibility of RGO, the DNA could maintain its activity very well. Various concentrations of Hg\textsuperscript{2+} were introduced for Hg\textsuperscript{2+} detection, leveraging Hg\textsuperscript{2+}'s distinctive property of binding specifically to two DNA thymine bases (T) to form a thymine–Hg–thymine (T–Hg\textsuperscript{2+}–T) base pair.\textsuperscript{48} The two ends of the ssDNA would be brought close to each other by the T–Hg\textsuperscript{2+}–T base pair, bringing the negative charge of the ssDNA closer to the FET. N-type doping then occurs on the device, leading to a leftward shift of the Dirac point. Thus Hg\textsuperscript{2+} detection can be achieved via monitoring the Hg\textsuperscript{2+} sensors' shift in $V_{\text{CNP}}$, that is, the $V_{\text{G}}$ corresponding to the minimum conductance.

To investigate the decoration of AuNPs and the immobilization of ssDNA, we first measured the electrical characteristics of the AG-FETs. As displayed in Figure 4A,
the $V_{\text{CNP}}$ of the olive curve obtained after AuNPs decoration was shifted to the right compared with the original $V_{\text{CNP}}$, due to p-doping of the device by AuNPs. This result demonstrates successful modification by AuNPs. The scanning electron microscope (SEM) images (Figure S10) confirm the electrical characteristics. Then, owing to n-doping of the ssDNA, the $V_{\text{CNP}}$ of the black curve obtained after the immobilization of ssDNA was shifted to the left. This experimental result implies the achievement of ssDNA immobilization, as further verified by the fluorescence images shown in Figure S11. We then proceeded to use the ssDNA-AuNPs-decorated AG-FETs as Hg$^{2+}$ sensors to investigate the Hg$^{2+}$ sensing capability. As shown in Figure 4A, the $V_{\text{CNP}}$ of the red curve obtained upon the addition of Hg$^{2+}$ was shifted to the left. These observations demonstrate the successful occurrence of sensing events. Then various concentrations of Hg$^{2+}$ were introduced to explore the sensitivity of the ssDNA-AuNPs-modified AG-FETs, and a blank was introduced as a reference. Table S3 summarizes the detailed $\Delta V_{\text{CNP}}$ behavior for the above process, namely the shift in $V_{\text{CNP}}$ after incubating with Hg$^{2+}$ relative to the $V_{\text{CNP}}$ of ssDNA, with other ions, or with the blank solution. As shown in Figure 4B, $\Delta V_{\text{CNP}}$ as the sensor response provides us with a calibration curve, in which the blue dashed line represents a signal/noise ratio (S/N) of 3. Here $\Delta V_{\text{CNP}}$ increases as the concentration of Hg$^{2+}$ increases from $1 \times 10^{-10}$ to $1 \times 10^{-6}$ mol/L, yielding a regression equation of $\Delta V_{\text{CNP}} = -0.62116 - 0.05474 \log C_{\text{Hg}^{2+}}$ with $R^2 = 0.9961$. A detection limit of the Hg$^{2+}$ sensor as low as $1.61 \times 10^{-11}$ mol/L (at S/N = 3) is obtained. The AG-FET as a sensing platform is preferable to the most standard/existing methods summarized in Tables 1 and S1.

We further evaluated the specificity of Hg$^{2+}$ recognition for our sensor. Table S4 summarizes the $\Delta V_{\text{CNP}}$ behavior in the detection of various metal ions based on ssDNA-AuNPs-modified AG-FETs. As indicated in Figure 4C, the $\Delta V_{\text{CNP}}$ values for $1 \times 10^{-6}$ mol/L Hg$^{2+}$ (red column), K$^+$ (blue column), Co$^{2+}$ (cyan column), Ca$^{2+}$ (magenta column), Zn$^{2+}$ (yellow column), and blank (green column) were $-293.87 \pm 9.24$, $-47.77 \pm 13.01$, $-40.00 \pm 16.12$, $-41.8 \pm 11.92$, $43.03 \pm 15.89$, and $33.87 \pm 10.12$ mV, respectively. Here the $|\Delta V_{\text{CNP}}|$ of Hg$^{2+}$ is significantly larger than those of K$^+$, Co$^{2+}$, Ca$^{2+}$, and Zn$^{2+}$. Indeed, the $\Delta V_{\text{CNP}}$ values for K$^+$, Co$^{2+}$, Ca$^{2+}$, and Zn$^{2+}$ differ only negligibly from that of the blank solution. Along with the above data, these observations imply that the recognition between Hg$^{2+}$ and ssDNA is specific and that our Hg$^{2+}$ sensor distinguishes Hg$^{2+}$ from K$^+$, Co$^{2+}$, Ca$^{2+}$, and Zn$^{2+}$ with good selectivity.
### TABLE 1  Comparison of different methods for Hg$^{2+}$ detection

| Method                  | Limit of detection (LOD)   | Linear range     |
|-------------------------|---------------------------|------------------|
| Colorimetry             | H$_2$TCP/CoS/ZnS/CoS$^{49}$ | 17.74 nmol/L     | 40 – 1000 nmol/L |
|                         | MoS$_2$                   | 0.5 μmol/L       | 2 – 200 μmol/L   |
|                         | DNA–AuNPs$^{51}$          | 50 nmol/L        | \               |
| Electrochemical         | SnO$_2$/rGO NCs$^{52}$    | 279 pmol/L       | 0.4 – 1.2 μmol/L |
|                         | ssDNA-Au$^{53}$           | 0.6 zmol/L       | 5 zmol/L–55 pmol/L |
| FET                     | rGO/TGA–AuNP$^{54}$       | 25 nmol/L        | \               |
|                         | MoS$_2$/AuNP$^{55}$       | 100 pmol/L       | 100 pmol/L–10 nmol/L |
|                         | MoS$_2$/CD-DNA$^{56}$     | 0.65 amol/L      | 1 amol/L–10 pmol/L |
| All graphene/DNA (this study) | 16 pmol/L             | 100 pmol/L–1 μmol/L |

Abbreviation: FET, field effect transistor.

### TABLE 2  Determination of Hg$^{2+}$ in simulated sewage

| Samples | Added  | Found   | RSD (%) | Recovery (%) |
|---------|--------|---------|---------|--------------|
| Sewage 1 | 1.00 μmol/L | 0.95 μmol/L | 6.6     | 95.0          |
| Sewage 2 | 5.00 nmol/L  | 5.82 nmol/L  | 4.8     | 116.4         |
| Sewage 3 | 500 pmol/L    | 459 pmol/L    | 6.4     | 91.8          |

Abbreviation: RSD, relative standard deviation.

### FIGURE 5  Simulating Hg$^{2+}$ detection into using Hg$^{2+}$ sensor. (A) Schematic drawing and (B) optical image of experimental setup. (C) Transfer characteristics of Hg$^{2+}$ sensor for detecting H$_2$O (black curve), K$^+$ (blue curve), and Hg$^{2+}$ (red curve) into the pipeline.
Considering the electrical and solvent stability of AG-FETs, Hg\textsuperscript{2+} detection in sewage based on our as-fabricated Hg\textsuperscript{2+} sensor was investigated. Since we lacked access to sewage with real-life concentrations of Hg\textsuperscript{2+}, we simulated sewage by spiking deionized water with 1 μmol/L of each of K\textsuperscript{+}, Co\textsuperscript{2+}, Ca\textsuperscript{2+}, and Zn\textsuperscript{2+}. Hg\textsuperscript{2+} was added to the simulated sewage to prepare detection samples with three different concentrations of Hg\textsuperscript{2+}: 1 × 10\textsuperscript{-6}, 5 × 10\textsuperscript{-9}, and 5 × 10\textsuperscript{-10} mol/L. As displayed in Figure 4D, as the Hg\textsuperscript{2+} concentration in sewage increased, |ΔV\textsubscript{CNP}| increased significantly. Table 2 summarizes the recovery and relative standard deviation (RSD) of Hg\textsuperscript{2+} detection in simulated sewage. Here the recoveries range from 91.8% to 116.4%, and RSDs are under 6.6%, indicating an Hg\textsuperscript{2+} sensor with a strong electrical and solvent resistance to simulated sewage across a range of Hg\textsuperscript{2+} concentrations.

In addition to good stability, sensitivity, and selectivity, our AG-FET devices as sensors also possess the advantages of miniaturization, easy integration, and ease of low-temperature, large-scale manufacturing via solution processing. This key advantage allows these sensors to be placed into several kinds of pipelines, such as sewage pipelines, to monitor Hg\textsuperscript{2+} levels. To simulate Hg\textsuperscript{2+} detection in the pipeline, an AG-FET fabricated on a 6 × 15 mm silicon substrate was packaged and placed inside a silicone pipeline with an interior diameter of 8 mm (Figure 5A,B). The liquid flowing within contained either Hg\textsuperscript{2+} or K\textsuperscript{+}. As shown in Figure 5C, the ΔV\textsubscript{CNP} for the pipeline filled with Hg\textsuperscript{2+} containing liquid was more robust than the ΔV\textsubscript{CNP} for the pipeline filled with K\textsuperscript{+}-containing liquid at an identical concentration. This result, highly consistent with shifts in an atmospheric environment, is due to the electrical stability and solvent insusceptibility of the AG-FETs, further implying that our Hg\textsuperscript{2+} sensors possess the potential for practical applications in the field of pipeline inspection.

4 | CONCLUSION

In summary, AG-FETs were constructed via the covalent LBL assembly method to more easily obtain a high-performance, economical, practical FET device. In this method, one coupling agent was used in the construction of the electrode, and another was used in the construction of the semiconductor. On the one hand, this design choice realizes outstanding stability for the device through coupling of layer-to-structure covalent bonds, given the different properties pertaining to the different structures of the two coupling agents. On the other hand, this design choice realizes the one-step reduction construction of the all-graphene FET device by leveraging the different electrical transmission characteristics of the two coupling agents. Meanwhile, the use of RGO, for both electrodes and semiconductors of our AG-FETs allows the structure to be built without precious metals as electrodes, not only greatly reducing the cost of device construction but also reducing contact resistance between electrodes and semiconductors, thus greatly improving device performance. As a result, the hole mobility, electron mobility, and shelflife of our device could respectively reach 3.79 cm\textsuperscript{2}/(V·s), 3.78 cm\textsuperscript{2}/(V·s), and 18 months. At the same time, by leveraging their outstanding electrical stability and ultrahigh solvent insusceptibility, our AG-FETs can function as sensors to monitor Hg\textsuperscript{2+} with a LOD of 16 pmol/L. Detection error in simulated sewage using our sensor was less than 17%. Hg\textsuperscript{2+} detection even works in a simulated pipeline environment. The features of our AG-FETs lay the foundation for further functionalization, integration, industrialization, and commercialization, showing broad prospects in areas such as environmental monitoring.

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CONFLICT OF INTEREST

The authors declare no conflict of interest. [Correction added on 29 June 2021, after first online publication: Conflict of Interest section has been added.]

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