

Communication

Current Drops in CF₄ Plasma-Treated AlGaIn/GaN Heterojunction in Polar Gas Ambient

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Abstract: This letter reports the phenomenon of current drops in an AlGaIn/GaN heterojunction with CF₄ plasma treated in a polar gas ambient. Ungated AlGaIn/GaN HEMT with CF₄ plasma treatment was tested in ethanol, acetonitrile, and an acetic acid gas ambient, and the current dropped by 52%, 51% and 61%, respectively, which are much higher than the drops (6%, 7%, and 8%) in normal ungated HEMT. On the one hand, based on “ambient doping”, a possible model was proposed to explain this phenomenon. On the other hand, the lower initial current was very important to increase the sensitivity. This means that the initial current should be elaborately designed to achieve maximum sensitivity.

Keywords: AlGaIn/GaN heterojunction; CF₄ plasma; polar gas ambient



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1. Introduction

With the development of communications, computing, and other applications, integrated circuit systems continue to evolve toward being of a small size, having high power density, and having high operating frequency. AlGaIn/GaN heterojunction high-electron-mobility transistor (HEMT) devices, with high mobility, high two-dimensional electron gas concentrations, wide band gaps, chemical stability, and other excellent characteristics, show greater application potential in high frequency, high power [1], ultraviolet (UV) photodetector [2] and sensor areas [3–6].

AlGaIn/GaN HEMTs still have strong stability under extreme conditions such as high temperature, so more people use them as sensor devices [5,7–9]. Dropping polar liquid molecules on the surface of an AlGaIn/GaN heterojunction will lead to channel current drops of up to 60%, which can be used for detecting polar liquids, such as ethanol, acetone, methanol, etc. [4,10], and for detecting inorganic liquids, such as HCl [11]. Due to the excellent characteristics of the AlGaIn/GaN heterojunction's structure, it can also be used as a gas sensor device. For example, a Pt/AlGaIn/GaN HEMT structure was designed to detect H₂, CO, NO, NO₂, and NH₃ [3,5,12], and in addition, a Pt/n-GaN structure, GaN Schottky, and metal-oxide-semiconductor (MOS) diode were also used for gas detection [13]. The theory is that the molecules were catalyzed into atoms at the surface of the Pt gate, and formed a dipolar layer, causing a reversible lowering of the Pt work function. However,

the introduction of the expensive Pt metal makes the device's process more complex. Is it possible to directly apply the AlGaN/GaN heterojunction for gas detection without Pt metal?

We observed a new phenomenon that in a polar organic gas ambient, the current of the AlGaN/GaN heterojunction with CF₄ plasma treatment decreases, exhibiting higher sensitivity than the normal AlGaN/GaN heterojunction in both the dropping rate and the dropping amplitude. The current reduction is more than 50%. Based on the “ambient doping” [14] of the AlGaN/GaN heterojunction, we proposed a possible physical model, that is, the coupling of ambient dopants with surface polarization negative charges/fluorine negative charges to explain this phenomenon. This newly observed phenomenon probably provides a new approach for designing novel AlGaN/GaN heterojunction gas sensors that do not need Pt, which is expensive Pt. Incidentally, this phenomenon may also help people to understand the reason why the fluorine plasma-treated AlGaN/GaN heterojunction usually shows an unstable current in the air atmosphere.

2. Experiment

Two types of ungated HEMTs (A and B) were fabricated. Their structures are shown in Figure 1a,b. The epilayers were grown on a sapphire substrate via metalorganic chemical vapor deposition (MOCVD). From bottom to top, they consisted of a 6 µm thick unintentionally doped GaN buffer layer, 1 nm thick AlN interlayer, 21 nm thick Al_{0.26}Ga_{0.74}N barrier layer, and 2 nm thick GaN cap layer. The hall mobility was 1860 cm²/V s, and the 2DEG sheet density was 1.13×10^{13} /cm².

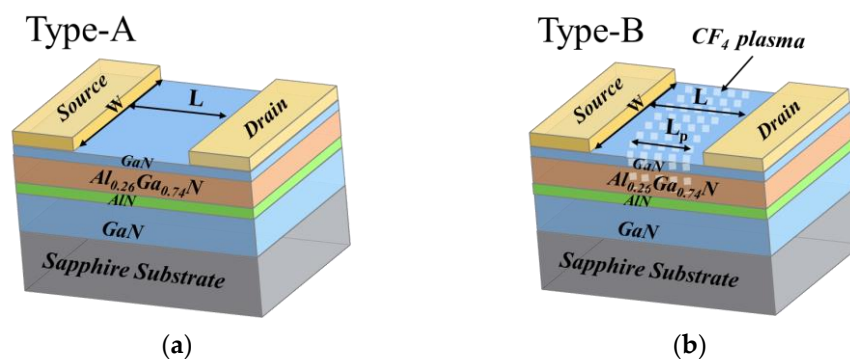


Figure 1. Schematic diagram of the device structures. (a) Type-A ungated HEMTs without plasma treatment. (b) Type-B ungated HEMTs with CF₄ plasma treatment.

The processes are as follows. Mesa isolation was performed using Cl₂/BCl₃ inductively coupled plasma (ICP). The Ohmic contacts were fabricated via e-beam evaporation of Ti/Al/Ni/Au (20/130/50/100 nm) stacks and then annealed at 870 °C for 30 s in a N₂ atmosphere. After that, the B-type devices were treated by CF₄ plasma to implant the F ions into the AlGaN barrier layer using APEX-ICP (inductively coupled plasma). The plasma treatment was performed with a RF power of 30 W, and ICP power of 600 W, and treating time was 2 min. The A-type devices were not treated by CF₄ plasma. After the fabrication and dicing of the devices, the 5 × 5 mm² chips were mounted on a printed circuit board (PCB). Wire bonding was carried out to connect the electrodes of the chips to the PCB electrodes.

There are two device geometries. L refers to the channel length. L_p refers to the length in the plasma treatment area. The L device is used for polar liquid testing, labeled as L, and the S device is used for polar gas testing, labeled as S. The L, L_p, and W of the L device are 1600 µm, 600 µm and 2200 µm, respectively. The L, L_p, and W of the S device are 100 µm, 60 µm and 400 µm, respectively. Hence, the ungated HEMTs are classified into four types, which are AL, AS, BL, and BS. The “L device” and “S device” have different dimensions, so we used the normalized current, I_{ds}/(W/L), in Figure 2a. The I–V curves of the four

samples are shown in Figure 2a. The linear region is located within the voltage range of $-7 \sim 7$ V.

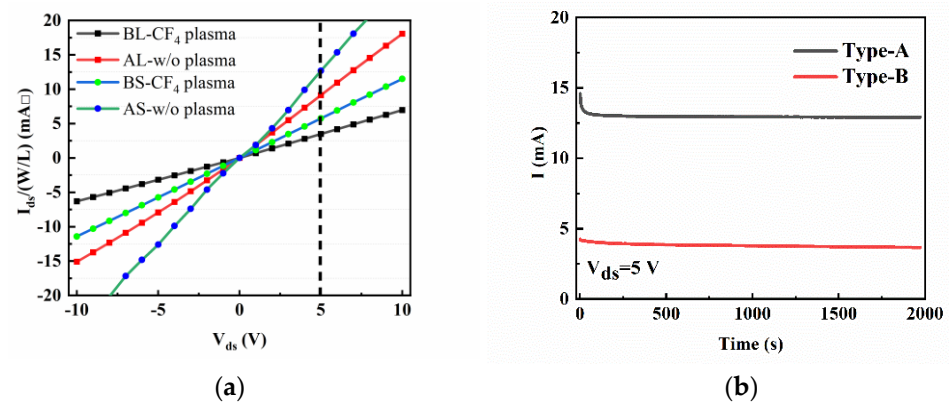


Figure 2. (a) The I–V curves of AL, AS, BL, and BS. (b) I_{ds} –t curves for type- A and -B device at $V_{ds} = 5$ V.

A voltage of 5 V was applied on the samples with a sampling rate of 1 time per second. The I_{ds} –t curves were recorded. In fact, there were trapping effects in our fabricated devices [15]. As shown in Figure 2b, at the beginning of the voltage bias, for type-A and type-B devices, the current is a bit high; over time, the current begins to drop, and after a while, the current stabilizes. For type-A devices without CF_4 plasma treatment, the time for the current to reach stability is ~ 1000 s. CF_4 plasma treatment increases the defect of the devices; therefore, trapping effects need to be excluded for a longer time, and the time for the current of type-B devices to reach stability is ~ 2000 s. In our experiments, the initial currents shown in the I_{ds} –t curves are the stable currents. Thus, the trapping effects were excluded in the experiments.

In the following tests, $V_{ds} = 5$ V was applied on the samples with a sampling rate of 1 time per second. After the current was stabilized, for the polar liquid testing, the polar liquids were dropped on the areas between the drain and source electrodes of AL and BL samples. For the polar gas test, the samples AS and BS were enclosed in a chamber, and the polar liquids were added along the side wall of the chamber, and they naturally volatilized to fill the chamber. The experimental results are shown in Figure 3. To observe the amount of current change, we normalized the current, as shown in Figure 3b,d.

As shown in Figure 3a,b, the currents of the AL sample without F plasma treatment were reduced by about 1.9 mA (16%), 2.2 mA (17%) and 2.7 mA (19%) by ethanol, acetic acid and acetonitrile liquids, respectively. The currents of the BL sample with F plasma treatment were reduced by 2.6 mA (54%), 4.1 mA (61%) and 4.17 mA (63%) by ethanol, acetic acid and acetonitrile liquids, respectively. Comparing the currents of the AL and BL samples in the polar liquids, three differences could be observed. First of all, the current dropping rate of the BL is a little faster than that of AL. Secondly, the absolute current reductions in the BL sample are a little higher than those in the AL sample. Finally, the current reductions in the BL sample are much higher (>3.2 times) than those in the AL sample. These differences indicate that AlGaIn/GaN heterojunctions with F plasma treatment are more sensitive to polar liquids than those without F plasma treatment are.

As shown in Figure 3c,d, the currents of the AS sample without F plasma treatment were reduced by about 3.25 mA (6%), 3.3 mA (7%), 3.5 mA (8%) by ethanol, acetic acid, acetonitrile gas ambient, respectively. The currents of the BL treatment with F plasma treatment were reduced by about 5.16 mA (52%), 4.71 mA (51%), 6.63 mA (61%) by ethanol, acetic acid, acetonitrile gas ambient, respectively. Compared with the AS and BS samples, the AL and BL samples show the same differences. However, the differences between the AS and BS samples are more significant. The current reduction rate of the BS sample (1000–2500 s) is more than 4 times that of the AS sample ($\sim 10,000$ s). The current reductions

in the BS sample (52%, 51%, and 61%) are more than 7 times those in the AS (6%, 7%, and 8%) sample. This means that AlGa_N/Ga_N heterojunctions with CF₄ plasma treatment are more sensitive to polar gases.

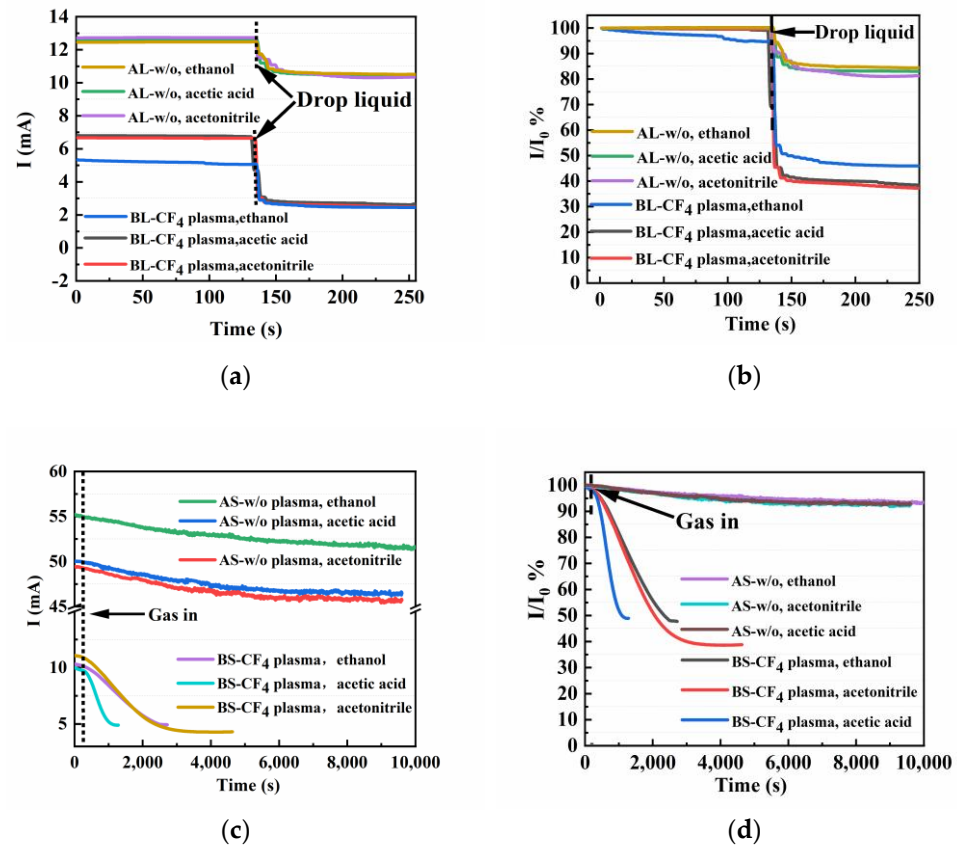


Figure 3. (a) I-t curves affected by polar liquids. (b) Normalized I-t curves affected by polar liquids. (c) I-t curves affected by polar gases. (d) Normalized I-t curves affected by polar gases.

3. Discussion

Secondary ion mass spectrometry (SIMS) was used to characterize the distribution of the F element in the samples, as shown in Figure 4a.

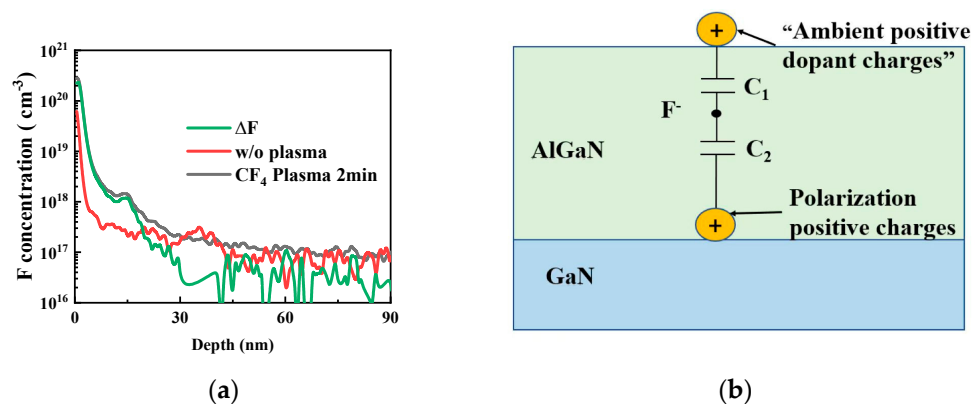


Figure 4. (a) Distribution profiles of F elements within the treated and untreated samples; (b) Schematic diagram of the equivalent circuit to specify the positive charges compensating for F⁻ ions.

Taking the F distribution of the sample without plasma treatment as the background, the F distribution of the sample with CF₄ plasma treatment was calibrated, as shown in

Figure 4. The red line is the F concentration of the device without plasma treatment, the black line is the F concentration of the device with CF₄ plasma treatment, and the green line is the difference between those two devices. The F elements are almost located in the AlGa_{0.3}N barrier layer with a peak concentration close to the AlGa_{0.3}N surface. The total sheet density was calculated to be about $4.3 \times 10^{13} \text{ cm}^{-2}$, which is much higher than the 2DEG sheet density ($\sim 1 \times 10^{13} \text{ cm}^{-2}$). As reported by previous literature [16], F has deep energy levels (2.8 eV and 3.25 eV below the conduction band) in AlGa_{0.3}N/GaN heterojunctions. In AlGa_{0.3}N/GaN heterojunctions, the energy levels of most F elements are below Fermi level, and as a result, F elements should be negatively charged, i.e., by F[−] ions.

In the thermal equilibrium state of the AlGa_{0.3}N/GaN heterojunction, electric neutrality is a basic requirement. Those F[−] ions should be compensated by the same amount of positive charges coming from the AlGa_{0.3}N/GaN interface and AlGa_{0.3}N surface. Based on the static electricity principle, as shown in Figure 4b, the number of positive charges in an AlGa_{0.3}N/GaN interface and AlGa_{0.3}N surface could be estimated by the following equations.

$$Q_P = -Q_F \times C_2 / (C_1 + C_2) \quad (1)$$

$$Q_A = -Q_F \times C_1 / (C_1 + C_2) \quad (2)$$

where C_1 and C_2 are the equivalent capacitors from the location of F[−] in an AlGa_{0.3}N/GaN interface and AlGa_{0.3}N surface, respectively. Q_F is the number of F[−] ions in the AlGa_{0.3}N barrier. Q_P is the number of polarized positive charges at the AlGa_{0.3}N/GaN interface. Q_A is the number of positive charges at the AlGa_{0.3}N surface. As shown in Figure 4a, the F[−] ions gather nearby the AlGa_{0.3}N surface, in which case, C_1 is larger than C_2 , resulting in $Q_A > Q_P$. Q_A and Q_P could be estimated to be $\sim 3.5 \times 10^{13} \text{ cm}^{-2}$ and $\sim 0.8 \times 10^{13} \text{ cm}^{-2}$. This explains why the 2DEG sheet density of $\sim 1 \times 10^{13} \text{ cm}^{-2}$ not be fully depleted by the higher F[−] sheet density of $\sim 4.3 \times 10^{13} \text{ cm}^{-2}$.

Although the origin of 2DEG in an AlGa_{0.3}N/GaN heterojunction is a controversial issue as yet, “ambient doping” sounds more reasonable, as states that “An important part of the origin of 2DEG in AlGa_{0.3}N/GaN heterojunction comes from the ambient doping [10,14,17]”. The high concentration of 2DEG in the AlGa_{0.3}N/GaN heterojunction obtained from environmental doping is associated with surface states and surface ions [14]. The occurrence of “ambient doping” implies that the Q_A in Equation (2) comes from the environmental positive ions (dopants) adsorbed on the AlGa_{0.3}N surface.

In an AlGa_{0.3}N/GaN heterojunction, as shown in Figure 5a, spontaneous and piezoelectric polarization induces positive and negative fixed charges at the interface and surface of the heterojunction [14,18]. Ambient doping causes positive ions to be adsorbed on the AlGa_{0.3}N surface, resulting in 2DEG generation in the channel. Two charge couplings have been established, i.e., polarization positive charges at the AlGa_{0.3}N/GaN interface with 2DEG in the channel, and the AlGa_{0.3}N surface’s polarized negative charges with the positive ions adsorbed on the AlGa_{0.3}N surface. For the fluorine plasma-treated AlGa_{0.3}N/GaN heterojunction, there were F[−] ions located in AlGa_{0.3}N barrier, as shown in Figure 5b. There were, additionally, two types of coupling dipoles. Some F[−] ions coupled with the positive polarization charges at the AlGa_{0.3}N/GaN interface, which reduced the 2DEG density. Other F[−] ions coupled with the positive surface ions. It should be noted that there were much more positive surface ions in the AlGa_{0.3}N/GaN heterojunction with fluorine plasma treatment than in the AlGa_{0.3}N/GaN heterojunction without fluorine plasma treatment.

When polar liquids were dropped on the untreated AlGa_{0.3}N surface, those polar liquids dissolved the surface ions, namely producing a cleaning effect, as shown in Figure 5c. The original charge couplings were broken, and new charge couplings are established. The surface’s polarized negative charges coupled with the polarized positive charges. As a result of electric neutrality, the corresponding electrons in the channel disappeared, leading to the current drop.

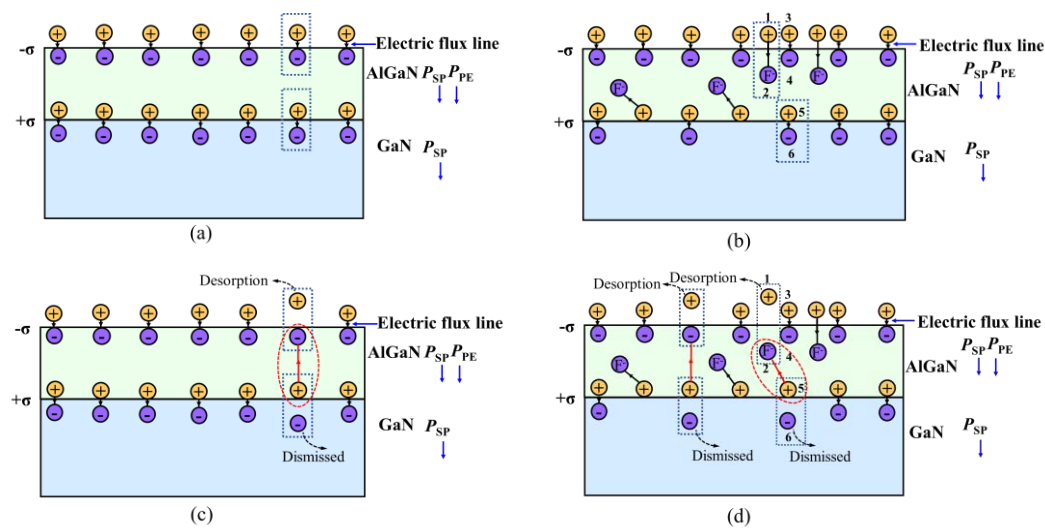


Figure 5. Physical model diagrams of surface of AlGaIn/GaN devices, (a) without plasma treatment, (b) with CF_4 plasma treatment (c) without plasma treatment being affected by polar molecules, (d) and with CF_4 plasma treatment being affected by polar molecules.

For the fluorine plasma-treated AlGaIn surface, as shown in Figure 5d, as long as the surface's positive ions were dissolved by polar liquids, the couplings of surface negative polarization charges with surface positive ions are broken, and the couplings of F^- charges with surface positive ions are broken too. As mentioned above, the dipole amount of the F^- charges with surface positive ions was much more than that of surface negative polarization charges with surface positive ions. Furthermore, the dipole moment (that is determined by the distance between the negative charge and positive charge) of the F^- charges with surface positive ions is larger than that of the surface's negative polarization charges with surface positive ions. The surface positive ions of the fluorine-treated AlGaIn/GaN heterojunction was more easily dissolved in a larger amount and at a faster rate than untreated one was. When surface positive ions were removed, the coupling of F^- charges and surface positive ions was broken, and then the F^- charges coupled with the polarized positive charges at the AlGaIn/GaN interface. Due to electric neutrality, the number of electrons in the channel were also reduced by equal amounts.

In the vapor ambient, the dissolving capabilities of polar gases were reduced significantly, depending on the collision rate between the polar gas molecules and surface positive ions. The 2DEG reduction of the vapor case exhibited a slower rate and a smaller amount, compared with the polar liquid case. Due to the higher amount of surface positive charges (the collision rate was higher) and the larger dipole moment (desorption with lower energy) between surface positive ions and F^- in the AlGaIn barrier, the current of the fluorine plasma-treated AlGaIn/GaN heterojunction shows a relatively faster dropping rate and a relatively larger reduction amount. For the fluorine plasma-treated AlGaIn/GaN heterojunction, the initial current was much lower than that in the heterojunction without treatment. Hence, the relative current reductions were enlarged, as shown in Figure 3d, implying a lower initial current in fluorine plasma-treated AlGaIn/GaN heterojunctions should exhibit higher sensitivity to polar gas vapors.

People have always found that the current of the fluorine plasma-treated AlGaIn/GaN heterojunction is not very stable even in an air atmosphere [19,20]. This is possibly attributed to the existence of polar gases in the air atmosphere [10], which remove the “ambient dopants” from the surface [14].

4. Conclusions

In summary, we found that the AlGaIn/GaN heterojunction with F plasma treatment was more sensitive to polar gas vapor than that without F plasma treatment. Based on

“ambient doping”, we proposed a model to explain this phenomenon. In the AlGaIn/GaN heterojunction with F plasma treatment, F[−] ions were incorporated in the AlGaIn barrier, attracting more surface “ambient dopants” with a larger dipole moment, which could be easily removed by polar gas molecules. The initial current is an important factor affecting the current’s sensitivity. This phenomenon could reasonably explain the current instability of the AlGaIn/GaN heterojunction with F plasma treatment in an air atmosphere. This phenomenon provides another possible approach to develop the AlGaIn/GaN gas sensors without Pt, which is expensive, with a simpler fabrication process.

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Data Availability Statement: The data that support the findings are available from the corresponding author upon reasonable request.

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Conflicts of Interest: The authors declare no conflict of interest.

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