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Application of graphene vertical field effect to regulation of organic light-emitting transistors*

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The luminescence intensity regulation of organic light-emitting transistor (OLED) device can be achieved effectively by the combination of graphene vertical field effect transistor (GVFET) and OLED. In this paper, we fabricate and characterize the graphene vertical field-effect transistor with gate dielectric of ion–gel film, confirming that its current switching ratio reaches up to 10^2. Because of the property of high light transmittance in ion–gel film, the OLED device prepared with graphene/PEDOT:PSS as composite anode exhibits good optical properties. We also prepare the graphene vertical organic light-emitting field effect transistor (GVOLEFET) by the combination of GVFET and graphene OLED, analyzing its electrical and optical properties, and confirming that the luminescence intensity can be significantly changed by regulating the gate voltage.

Keywords: graphene vertical field effect transistor, organic light-emitting transistor, ion–gel film, gate voltage regulation

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1. Introduction

Recently, organic light-emitting transistor (OLED), as a kind of light-emitting diode, has great development potential and broad application prospects because of its wide viewing angle, high color saturation and contrast, short response time, large light-emitting area, and low energy consumption.[1–3] Owing to the advantages of electrical conductivity, light transmission, stability, and mechanical properties,[4–7] graphene has been successfully used in OLED device preparation.[8–10] Vertical field effect transistor (VFET) is a kind of field effect transistor, whose channel direction is perpendicular to the surface of substrate, forming heterojunction structure between material layers.[11] Vertical electric field channel length can be shortened down to nanometer level that facilitates carriers' recombination.[12–14] Carrier accumulation in graphene and semiconductor layers can be regulated by the gate electrode due to the adjustability of graphene work function, so that Fermi levels can be rearranged to change the interface resistance between graphene and semiconductor, which significantly influences the switching ratio of VFET. In addition, by combining VFET structure with OLED device, a new method of regulating the luminescence intensity can be achieved by changing gate voltage.[15]

In this paper we use ion–gel to prepare an ITO/ion gel film/graphene OLED composite structure. The ion–gel is a mixture of organic polymer and electrolyzable salt electrolyte material with good thermal stability and high dielectric value[16,17] and widely used in flexible devices.[16,18] Under the action of an external electric field, anion and cation in polymer will migrate and diffuse to form electric-double-layer distribution with charge layer on the surface of insulating layer, which is similar to a capacitive effect.[19] In previous study,[20] we used an ion–gel film as a dielectric layer which significantly improved the performance of graphene field effect transistor (GFET). In the present experiment we achieve the effectively regulation of luminescence intensity of OLED devices due to GVFET.

2. Experimental materials and methods

2.1. Materials

Single-layered graphene was synthesized on copper foil with CVD process; 6-wt% PMMA was dissolved in anisole; ion–gel solution was prepared by dissolving 10-wt% PVDF and 2.5-wt% [EMIM]TF2N in N,N-Dimethylformamide (DMF) at 60 °C by water bath for 7 h until a clear solution was obtained; poly(3,4-ethylenedioxythiophene)-poly (styrenesulfonate) (PEDOT:PSS) aqueous solution was purchased from J & K. Corp; organic luminescent material:1,1-Bis[(di-4-tolylamino)phenyl] cyclohexane (TAPC), 1,3-bis-(carbazol-9-yl)benzene (mCP), Bis(4,6-difluorophenylpyridine-N,C2') pyridinecarboxamide (FIrpic) and 1,3,5-tri[(3-pyridyl)-phen-3-yl]benzene (TmPyPB) were purchased from BanHe Tech. Corp.

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2.2. Graphene vertical field effect transistor

Ion–gel film was used as gate dielectric in VFET. The prepared ion–gel solution was spin-coated (3000 rpm, time 15 s) on the surface of ITO glass, ion–gel was transparent after being dried at temperature 160 °C for 30 min to form a 2-μm-thick film tested by step meter, graphene was characterized after being trasferred to the surface of ion–gel film by Raman spectrometer and light transmittance tester. The 5-wt% dimethylformamide was added into PEDOT:PSS solution to make PEDOT:PSS film smoother on the surface of graphene, then the solution was spin-coated (1500 rpm, time 30 s) and dried at temperature 120 °C for 30 min. A 100-nm-thick aluminum layer was deposited on PEDOT:PSS film, serving as a top electrode and the device structure was prepared to be ITO/ion–gel/graphene/PEDOT:PSS/Al (100 nm).

2.3. Graphene vertical organic light-emitting field effect transistor

The OLED structure prepared in this study was graphene/PEDOT:PSS/TAPC (50 nm)/mCP (10% FIrpic) (30 nm)/TmPyPB (50 nm)/LiF (0.5 nm)/Al (100 nm) (Fig. 4(e)): Graphene/PEDOT: PSS was used as a composite anode, the TAPC was a hole transport layer, and mCP was the host material of light-emitting layer. The FIrpic was doped with phosphorescent dye. The TmPyPB was electron transport layer. The electron injection layer and cathode material were LiF and Al, respectively. All functional layers except composite anode were deposited using vacuum deposition in vacuum of less than 4×10^{-4} Pa.

2.4. Detection

After preparation, the voltage–luminance–current density characteristic curve of GOLED was detected at room temperature by using an electroluminescence test system consisting of Keithley Model 2400 current source and PR 655 spectrometer. The electrical characteristics of GVFET were measured by using Keithley 2612a dual channel source meter.

3. Results and discussion

Transmittance of dielectric layer material has an effect on the luminescence intensity of graphene OLED, showing that the transmittance of visible light (wavelength 390 nm–760 nm) in grapheme/ion–gel/ITO/glass composite structure reaches up to 90% (Fig. 1(b)), glass and ITO are transparent, indicating that the ion–gel film is highly transparent and has little effect on light extraction efficiency of device. The Raman characterization diagram (Fig. 1(a)) shows that the Raman spectrum of ion–gel film (black) has an obvious peak only at 2980 cm^{-1}, and the characteristic peaks of red curve peak at 1580 cm^{-1} (G peak) and 2680 cm^{-1} (2D peak), which are graphene characteristic peaks caused by graphene atom lattice vibration,[21] which, combined with SEM images (Figs. 1(c) and 1(d)), indicates that graphene is successfully transferred to the ion–gel membrane surface.

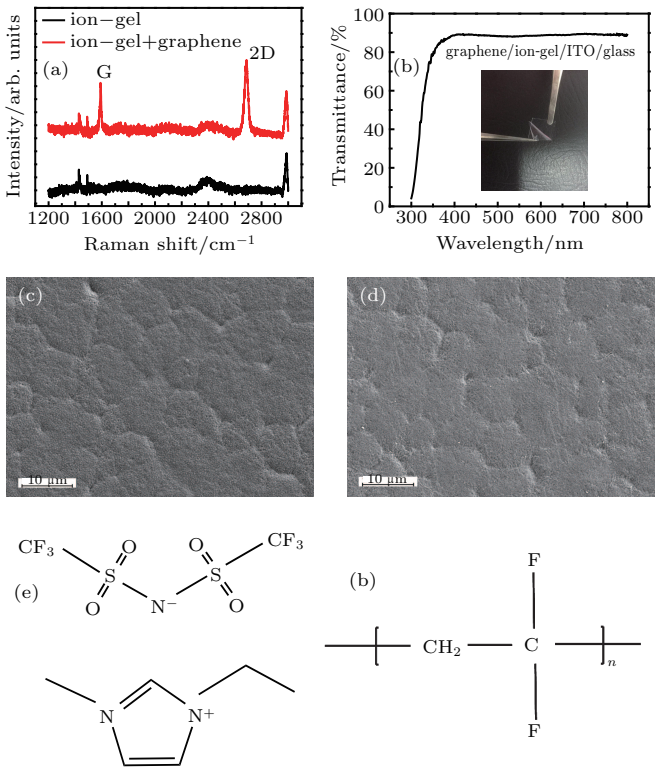


Fig. 1. (a) Comparision of Roman spectra before and after transferring graphene on ion–gel; (b) transmittance *versus* wavelength of grapheme/ion–gel/ITO/glass composite structure, with inset showing SEM image of ion–gel film on glass substrate; (c) ion–gel film without graphene; (d) ion–gel film with transferred graphene; molecular formula of (e) [EMIM]TF2N, and (f) PVDF.

In addition, we also measure the specific capacitance of 2-μm-thick ion–gel film sandwiched between gold electrode and ITO as a function of voltage frequency ($c = -1/2\pi fZ$, where Z denotes the impedance and f the frequency), and the results are shown in Fig. 2(a). The measured capacitance reaches up to 9 μF/cm^2 at low frequencies. Under the action of external electric field, the charge distribution in the ion–gel is asymmetric, and the electric charge layer on the surface of external insulating layer forms an electric-double-layer distribution, resulting in high capacitance value in per unit area with a micron-thickness, whichis different from the capacitance values of other materials with low dielectric constants.[15–18,20–23] The capacitance value decreases as frequency increases due to the interaction between charge in solid electrode and counter ion in electrolyte.[23]

PEDOT:PSS is often used as OLED hole injection material because of its high work function and high transmittance.[24] In this experiment, the PEDOT:PSS is spin-coated on the surface of graphene to form a heterojunction structure.[8] In addition, the ion–gel gate structure is introduced to form G-VFET, and the energy band of graphene

can be opened by gate regulation, so that the Fermi levels of graphene and barrier height between graphene and PEDOT:PSS are changed, and thus affecting the interface resistance and current output. In the transfer characteristic curve, the source–drain voltage (V_{sd}) is constant (at 5 V), and the gate voltage varies from 0 V to 10 V. According to the circuit superposition principle,[25] equivalent value of gate voltage (V_{eg}) is the difference value between gate bias and source–drain voltage, *i.e.*, (V_g–V_{sd}), the actual gate voltage variation ranges from −5 V to 5 V. In the output characteristic curve, source–drain voltage varies from 0 V to 2 V, gate voltage variation is in a range from 1 V to 9 V (step 2 V). Switching ratio (J_{on}/J_{off}) can be obtained from transfer characteristic curve to be about 10^2 (Fig. 3(a)). Output characteristic curve of G-VEFT shows that source–drain current density (J) changes with V_g variation and increases exponentially, which is in accordance with the volt-ampere characteristics of heterojunction curve. In the equilibrium electric field state ($V_{eg} = 0$) (Fig. 2(b)), Fermi level (E_F) of graphene is intrinsic Fermi level (Fig. 3(d)), hole and electron carriers disperse in the dielectric layer. Under negative gate voltage ($V_g < V_{sd}$), hole accumulation in graphene forms a p-type channel (Fig. 2(c)), the Fermi level of graphene is below the intrinsic Fermi level (Fig. 3(e)) which is favorable for hole injection; as the gate voltage increases, the carrier concentration in graphene is changed and will be in a critical conduction state when electron concentration is equal to hole concentration; under positive gate voltage ($V_g > V_{sd}$), electron concentration in graphene increases (Fig. 2(d)), the Fermi level of graphene is above intrinsic Fermi level (Fig. 3(f)) and electrons accumulate in graphene, froming an n-type channel blocking hole injection.

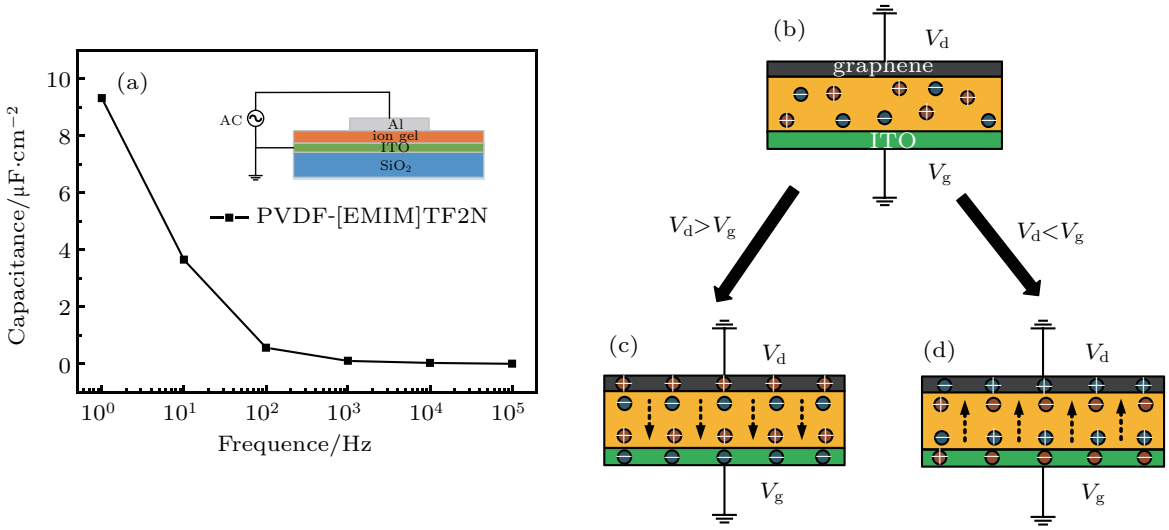


Fig. 2. (a) Frequency dependence of specific capacitance of ion–gel spin-coated on ITO glass; schematic diagrams of charge layer distribution: (b) anions are dispersed in dielectric layer; (c) anions are dispersed under negative gate voltage, with holes accumulating in graphene; (d) anions are dispersed under positive gate voltage, with electrons accumulating in grapheme.

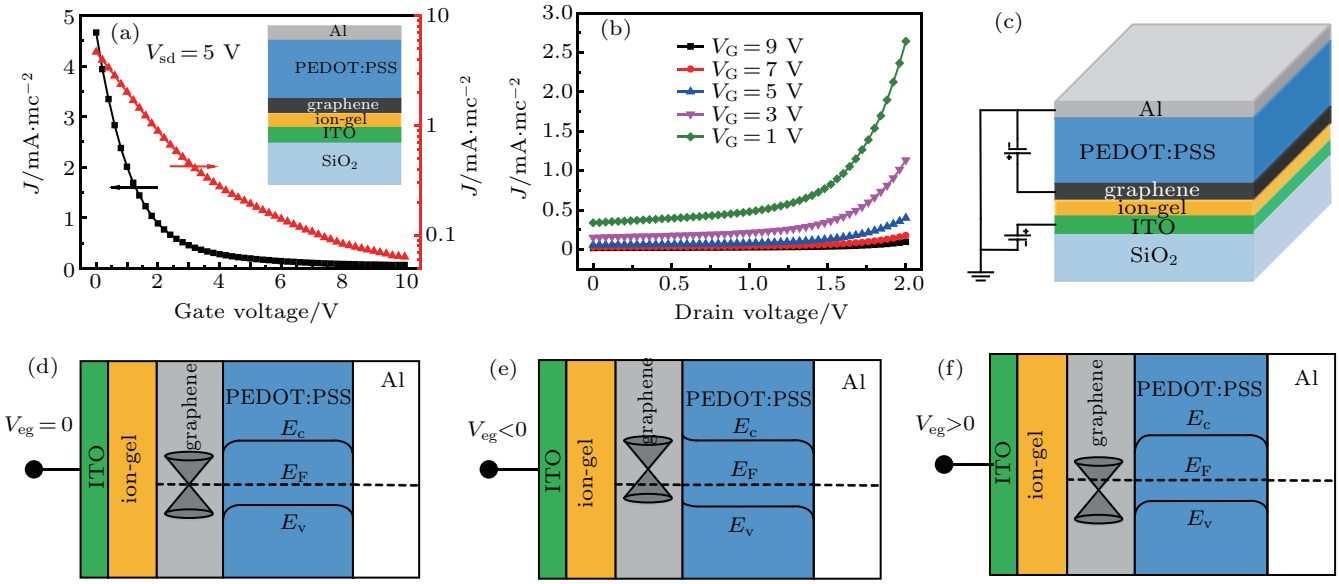


Fig. 3. (a) Transfer and (b) output characteristic curves of GVFET; (c) structure and schematic diagram of GVFET with ion–gel film gate; (d)–(f) Fermi level diagrams of GVFET under different gate voltages.

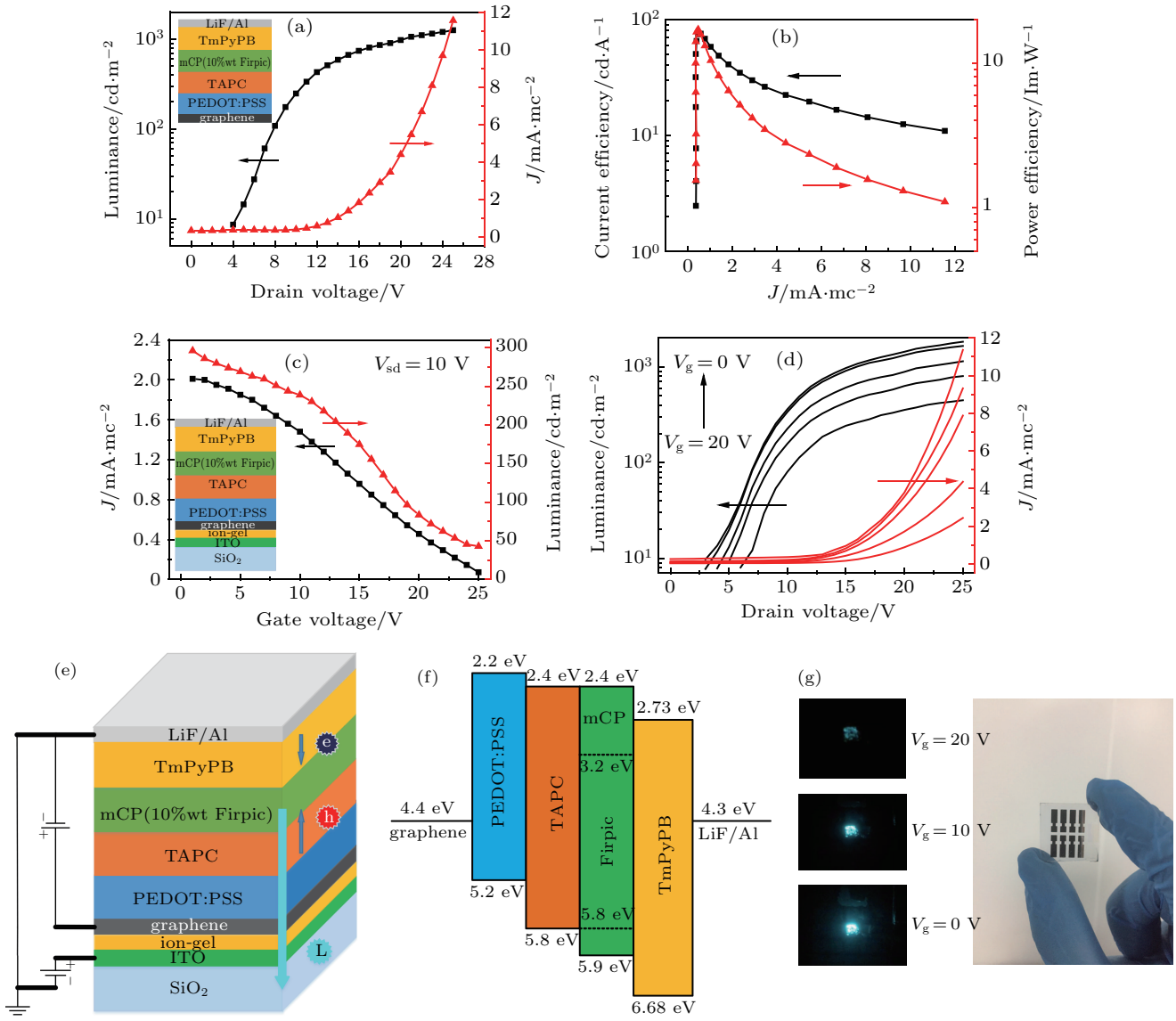


Fig. 4. (a) Characteristic curves of luminance–current density *versus* drain voltage of GOLED; (b) characteristic curves of current efficiency and power efficiency of GOLED; (c) transfer characteristic curves of current and luminescence intensity of graphene OLED; (d) output characteristic curves of current and luminescence intensity *versus* drain voltage of graphene OLED; (e) structure and schematic diagram of GVOLEFET; (f) device energy level diagram of graphene OLED; (g) comparison among illuminating images in the same area of graphene OLED.

Graphene needs to be hydrophilized by UV treatment for 30 min after being transferred to make PEDOT:PSS solution dispersed on the grapheme surface smoothly. In addition, UV treatment can improve graphene work function[26] and PEDOT:PSS film can reduce surface roughness of graphene, which are beneficial to the improvement of the luminescent properties of OLED device.[8,27,28] In order to confirm the luminescence properties of OLED device with graphene/PEDOT:PSS as the composite anode, we prepare an OLED structure as shown in Fig. 4(e) (device energy level diagram is shown in Fig. 4(f)). In the characteristic curves, the illuminating voltage (V_{on}) and maximum brightness (L_{max}) of OLED device are 4 V and 1259 cd/m^2, maximum current efficiency (luminous intensity per unit current) and power efficiency (luminous flux per unit power) are 75.3 cd/A and 17 lm/W, respectively. These results indicate that the OLED device with graphene/PEDOT:PSS as composite anode exhibits the good luminescence properties.

According to the GVFET structure, we add OLED structure between graphene and aluminum layer, and design the structure of GVOLEFET.[15,29] When the source–drain voltage is 10 V, the OLED luminous intensity decreases as the gate voltage increases (from 0 V to 25 V). The inset in Fig. 4(g) shows the comparison among the illuminating images in the same area at 0-V, 10-V, 20-V gate voltages respectively. The maximum luminous intensity of device is 296 cd/m^2 under the negative equivalent gate voltage ($V_{eg} < 0$), holes accumulating in graphene facilitates its injection into organic layer. As gate voltage increases, the carrier density in graphene layer changes from hole accumulation to electron accumulation. Under the action of the positive equivalent gate voltage ($V_{eg} > 0$), electron accumulation in graphene blocked hole injection can re-

sult in luminous intensity of device decreasing to 43 cd/m^2. Figure 4(d) shows that the comparison among the changes in light intensity and current output curves at different gate voltages (from top to bottom: $V_g = 0$ V, 5 V, 10 V, 15 V, 20 V). As the gate voltage increases gradually, the turn-on voltage increases, and the luminous intensity at the same source–drain voltage decreases, which is mainly due to the holes accumulating in the graphene at an equivalent negative gate voltage, and the hole conductivity turns better, which is beneficial to implementing the more hole injection at low source–drain voltage, thus recombining with electrons to emit light, which has a positive correlation with the change of current output curve.[30] In general, the results show that the output current of graphene is significantly affected by gate voltage, which further affects the current density and luminescence intensity of OLED device.

In addition, we also test the repeatability of device. We can see that the short-term (within 10 min) repeatability is better (Fig. 5(b)), the maximum error of light intensity value under the same source–drain voltage is within 3%. But the long-term repeatability shows a significant decrease in performance after 3 days (Fig. 5(c)). We also perform a long-term detection of specific capacitance of the ion–gel dielectric layer (Fig. 5(a)), find that it has not changed significantly, verifying the stability of ion–gel film and also explaining that the decline in device performance is mainly due to the preparation and packaging. In addition, the OLED device is a multilayered structure, although it is beneficial to reducing the energy level barriers crossed by the transition of electrons and holes, the process is complicated and repeatability will also be affected. Therefore the device structure design should be further improved in the future researches.

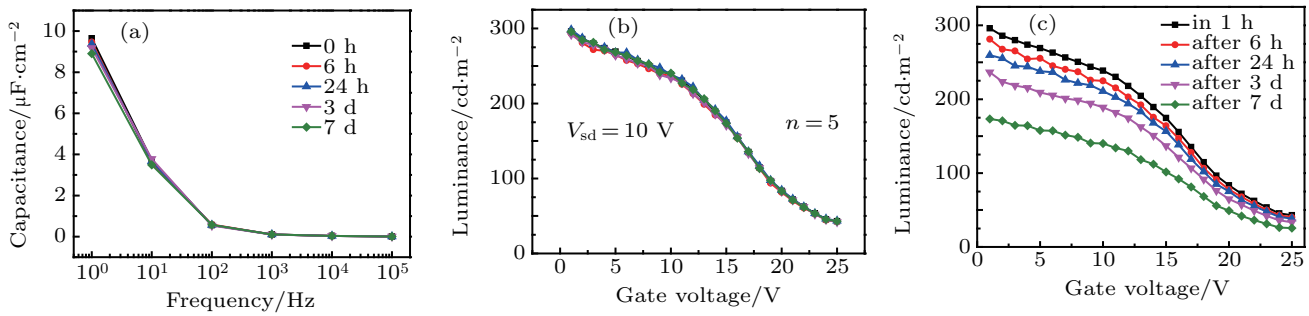


Fig. 5. (a) Long-term detection of specific capacitance of ion–gel dielectric layer; (b) repeated detection of luminous intensity curve under gate voltage regulation in short-term; (c) repeated detection of luminous intensity curve under gate voltage regulation in long-term.

4. Conclusions

In this research, we fabricated and characterized the graphene/PEDOT:PSS vertical field effect transistor with ion–gel film gate dielectric, which shows current switching ratio up to 10^2. Heterojunction structure formed with graphene and PEDOT:PSS is affected by the gate voltage to regulate its interface resistance and current output. The graphene OLED prepared by using the graphene/PEDOT:PSS serving as a composite anode exhibits good luminescence performance. The GVOLVEFT is prepared by combining GVFET and graphene OLED, and the luminescence intensity is significantly changed by regulating the gate voltage. In recent years, graphene have been widely used as transparent electrodes in OLED research and development. Combined with the high light transmittance of gate dielectric materials, GVFET will be used as an important device platform for developing various photoelectric sensors.

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