Robust High-Capacitance Polymer Gate Dielectrics for Stable Low-Voltage Organic Field-Effect Transistor Sensors

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Organic field-effect transistors (OFETs) have shown great promise for use as chemical sensors for applications that range from the monitoring of food spoilage to the determination of air quality and the diagnosis of disease. However, for these devices to be truly useful, they must deliver reliable and stable low-voltage operation over extended timescales. An important element to address this challenge is the development of a high-capacitance gate dielectric that delivers excellent insulation with robust chemical resistance against the solution processing of organic semiconductors (OSC). The development of a bilayer gate dielectric containing a high-k fluoropolymer relaxor ferroelectric layer modified at the OSC/dielectric interface with a photo-crosslinked chemically resistant low-k methacrylate-based copolymer buffer layer is reported. Bottom-gate OFET chemical sensors using this bilayer dielectric operate at low-voltage with exceptional operational stability. They deliver reliable sensing performance over multiple cycles of ammonia exposure (2 to 50 ppm) with an estimated limit-of-detection below 1 ppm.

Portable and wearable low-power consumption chemical sensors that generate rapid and reliable quantitative information, particularly in resource-limited settings, are in tremendous demand for application in areas such as medical diagnostics, food and environmental monitoring.[1] Sensors based on organic field-effect transistors (OFETs) are attractive candidates due to the intrinsic advantages of low-power consumption, light weight, substrate flexibility (e.g., paper or plastic) and direct electrical transduction of the analyte response.[2] Furthermore, these devices can be processed from solution under ambient conditions and the sensitivity/selectivity of the response can be optimized by changing the chemical composition of the organic semiconductor (OSC).[3] The OSC plays a vital role during sensing for recognition of the analyte via physical and chemical interactions that perturb the output of the OFET to produce a measurable electrical response. Another essential component of the OFET is the gate dielectric; this material provides electrical insulation from the gate electrode and modulates the number of charge carriers in the conduction channel of the OSC layer per applied voltage. Two important considerations for fabrication of a low-voltage OFET chemical sensor are i) the OSC layer has to be placed in a position that permits exposure to the analyte, that is, in a bottom-gate device geometry, and ii) the gate dielectric material requires a large areal capacitance ($C_{ai}$) and good electrical insulation, but it needs to be chemically robust to withstand the solvents used to solution process the OSC layer in this geometry.[4] A gate dielectric that meets these requirements would enable the development of low-powered sensing platforms such as electronic noses for multi-analyte detection where an array of OSC sensing elements is required.[5]

OFET devices are generally fabricated using conventional metal oxide (e.g., SiO$_2$) gate insulators, as these have been used as a platform to study the effect of OSC molecular design and film structure on the sensitivity, selectivity, and stability of the sensing performance of OFETs.[6–8] However, these materials are brittle in nature and typically require high processing temperatures that are unsuitable for fabrication on flexible plastic substrates. Room temperature anodization of ultra-thin (<10 nm) oxide gate dielectrics,[9] or vapor phase deposition of self-assembled nanodielectrics[10] overcome these drawbacks, but such methods require complex fabrication steps that may be challenging for scale-up.[11] In contrast, polymer-based dielectric insulators can be processed from solution over large areas on top of OSC layers.[12] Conventional polymer insulators give films with low $k$ values (2–5) and $C_{ai}$ unsuited for low voltage operation at conventional film thicknesses (>100 nm). A
simple way to increase the $C_i$ is to downscale the film thickness to ultra-thin layers of polymers such as poly(4-vinylphenol).[13] Due to the thicknesses required (<30 nm), crosslinking of these thin films is needed to impart better electrical insulation and chemical robustness for subsequent OSC processing, and this approach has been used to fabricate low-voltage OFET-based chemical sensors.[14,15] However, in these devices, unreacted hydroxyl groups act as charge trapping sites that are undesirable for stable device operation[16] and large-area processing of robust and uniform ultra-thin films poses a significant challenge for scale-up of these devices.[16]

An alternative method is to use high-$k$ polymer gate dielectrics,[17] such as polyelectrolytes or derivatives of poly(vinylidene fluoride-trifluoroethylene) (PVDF). The hygroscopic nature of polyelectrolytes present a severe disadvantage toward operational stability in ambient conditions.[18] In contrast, PVDF polymers are hydrophobic in nature, solution processable at low temperatures and have extremely high permittivity values ($k > 50$).[19] The use of these materials as the gate dielectric in an OFET enables low operating voltages ($<5$ V) due to the high $C_i > 100$ nF cm$^{-2}$ achievable for films of thicknesses >100 nm.[20] One concern is that the large dipole moment of the C–F bonds induces ferroelectric behavior in these thin films under an applied electric field, and this causes hysteresis in the device characteristics that makes the operation of these devices more suitable for use in non-volatile memory.[21,22]

To reduce the hysteresis, a bilayer-type dielectric configuration (see Figure 1a) has been employed where the surface of the high-$k$ fluoropolymer is modified by a low-$k$ polymer that acts as a buffer layer at the interface with the OSC to suppress the influence of the ferroelectric effect of the fluoropolymer. This buffer layer needs to be thin enough to maintain a high $C_i$ so that the OFET operates at low voltages.[23–27] Several buffer layers have been previously reported including cross-linked PVP films with a high-$k$ hybrid PVDF inorganic nano composite layer.[28] However, inhomogeneous film morphologies caused by the inorganic nanoparticles and unreacted hydroxyl groups lead to charge trapping at the OSC/dielectric interface causing performance instabilities in these devices. Photo cross-linked poly(vinyl cinnamate) layers have also been used,[29] but the thermal reversibility of the $[2 + 2]$ photodimerization reaction of the cinnamoyl group causes unstable film morphologies reducing reproducibility and preventing prolonged transistor operation.[30] Poly(methyl methacrylate) PMMA (see Figure 1b) has also been employed as a suitable buffer layer. In top-gated devices, vertical phase segregation of a blend with the fluoropolymer forms a thin low-$k$ PMMA layer at the OSC/dielectric interface,[26] while in bottom-gated devices, the OSC was generally vacuum deposited due to the PMMA layer easily dissolving in the solvents typically used to process OSCs (e.g., chlorobenzene, toluene, chloroform).[27] To avoid this problem and enable devices to be solution processed, the PMMA can be crosslinked using chloro-silanes.[31] Unfortunately, the moisture sensitivity of the resulting film and the formation of HCl as a by-product has adverse effects on device performance.[13] Synthetic approaches have been proposed to solve these problems by attaching

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Figure 1. a) Schematic of bottom-gate/top-contact OFET with a bilayer (low-$k$/high-$k$) gate dielectric configuration; molecular structures of the gate dielectric components used in this study: b) PMMA, c) P(BMA-co-MMA); d) Michler’s ketone (MK); e) P(VDF-TrFE-CFE); f) DSC thermograms of the 1st heating scan of (from top down) MK, PMMA:MK and P(BMA-co-MMA):MK with 12.5 and 25.0 wt% of MK, respectively. * indicates $T_g$ while red and blue box highlights the region of the MK transition blended with the polymers; AFM images of thin films of g) bare P(VDF-TrFE-CFE); and UV-illuminated bilayer films with buffer layers PMMA:MK and P(BMA-co-MMA):MK containing 12.5 wt% of MK h,i) before solvent washing and j,k) after solvent washing.
pended crosslinkable functional groups as side-chains, or by using diblock-copolymers composed of a fluoropolymer and a PMMA segment, however, the additional synthetic steps introduces complexity to the overall fabrication process of the device and a simpler approach is required for manufacturing at scale.

To overcome these various limitations, we have developed a facile photo-crosslinking strategy to form robust and chemically resistant, buffer layers on top of high-k fluoropolymers. The process relies on the intermixing of a commercially available low-k methacrylate copolymer derivative, poly(butyl methacrylate-co-methyl methacrylate) (P(BMA-co-MMA)) (see Figure 1c), with a photoinitiator, Michler’s ketone (MK) (see Figure 1d). Using this approach, a bilayer dielectric with excellent chemical resistance and a large capacitance (169 nF cm$^{-2}$ at 1 kHz) was obtained (see Figure 1e). In this system, utilization at 365 nm leads to covalent bond formation between neighboring macroradical polymer chains that are produced by hydrogen abstraction with a photo-excited MK (see Figure S1, Supporting Information, for the absorption spectra of MK and plausible crosslinking mechanism). The polymer chain thermal property (i.e., whether it is the glassy or rubbery state) has a substantial effect on the intermixing with the photoinitiator and the addition of butyl side chains increases the number of reactive sites for hydrogen abstraction. This may affect the crosslinking efficiency primarily due to the need for sufficient interfacial area between photoinitiator and polymer to generate macroradicals to facilitate the covalent bond formation between polymer chains.

To test this hypothesis a glassy polymer, PMMA, and a rubbery polymer P(BMA-co-MMA) were mixed with various loadings of MK (in weight percentage at 0, 12.5, and 25 wt%) and the thermal behavior investigated by differential scanning calorimetry (DSC, Figure 1f). MK has a distinct endothermic melting transition at $T_m = 176$ °C while PMMA has a glass transition peak at $T_g = 122$ °C and P(BMA-co-MMA) at 63 °C. Blending 12.5 and 25 wt% of MK with PMMA showed a clear broadening of the endothermic melting transition related to MK and a shift of approximately 12 °C to lower temperatures, the transition associated with the $T_g$ of PMMA was absent. Upon adding 12.5 and 25 wt% of MK to P(BMA-co-MMA) the phase transitions were significantly suppressed revealing a single homogenous amorphous phase. It is evident that there is a difference in phase behavior between the two polymer:MK blends that may be attributed to dissimilar glass transition properties. The glassy nature of PMMA leads to MK segregating from the polymer matrix (as reflected from the presence of a melting transition of MK), while higher polymer chain mobility in P(BMA-co-MMA) (indicated from a lower $T_g$) permits a larger degree of inter-mixing with the MK as reflected in the essentially amorphous DSC thermogram, increasing the likelihood of crosslinking upon UV exposure of the polymer:MK blend.

The chemical resistivity of the illuminated buffer layer was investigated for the MK blended with PMMA and P(BMA-co-MMA) (refer to Figure S2, Supporting Information, for a description of the solvent resistivity experiment). Formation of a cross-linked polymer network should render the entire film insoluble. Bilayer thin films were prepared by spin coating a 195 nm layer of P(VDF-TrFE-CFE) on a glass substrate, this film was annealed at 60 °C for 2 h under nitrogen atmosphere to give a distinctive crystalline grain-like morphology as shown from the atomic-force microscopic (AFM) image in Figure 1g (see Figure S3, Supporting Information, for a full AFM film morphology analysis). The surface of this film was then coated with the polymer:MK blends at various MK loadings (2.5, 12.5, and 25 wt%) using an orthogonal solvent mixture of toluene:anisole and the bilayer films were exposed to UV light to induce crosslinking (refer to Experimental Procedure for a detailed description). The polymer:MK (12.5 wt%) films on P(VDF-TrFE-CFE) had a total thickness of between 250–257 nm and showed smooth featureless surface morphologies in AFM as shown in Figure 1h,i (see Figure S3, Supporting Information, for other MK wt%). The surface of exposed films were then fully covered with chlorobenzene (typical solvent used to solution process an OSC) for 5 min before removal of the solvent by spin-coating and the films annealed at 100 °C for 10 min to evaporate any remaining solvent residues. After solvent removal, substrates coated with PMMA:MK (12.5 wt%) revealed the underlying P(VDF-TrFE-CFE) layer (see Figure 1j) as the buffer layer had been removed by the solvent wash. For substrates coated with P(BMA-co-MMA):MK (12.5 wt%), a uniform film with no significant morphological changes from the original coated substrate was seen as shown in Figure 1k. Control experiments on unexposed films showed that these were all removed after solvent washing, confirming that MK is essential to promote a chemically resistant polymer film (see Figure S3, Supporting Information). In addition, exposed PMMA:MK films were removed by solvent washing irrespective of MK loadings (2.5 and 25 wt%) while films of P(BMA-co-MMA):MK were solvent resistant at higher loadings of MK (12.5 and 25 wt%) but were removed at lower loadings (2.5 wt%). In these films, it is possible that insufficient macroradical generation results in a lower degree of crosslinking. Washing with chloroform or toluene showed similar solvent resistivity properties as chlorobenzene (see Figure S4, Supporting Information). These results correspond well with the DSC analysis and it appears that only strongly intermixed P(BMA-co-MMA):MK (12.5 and 25 wt%) blends give stable thin films.

The dielectric properties of the bilayer films were investigated by fabrication of metal–insulator–metal (MIM) capacitors using the same solution processing steps as described in the solvent resistance trials (see Experimental Section, Supporting Information). Thin films of P(VDF-TrFE-CFE) exhibited a large leakage current density of 1.72 × 10$^{-5}$ A cm$^{-2}$ at −5 V (see red line in Figure 2a). When coated with a layer of P(BMA-co-MMA) the leakage current density was significantly reduced to 5.53 × 10$^{-9}$ A cm$^{-2}$ (see blue line in Figure 2a). UV exposure of these buffer layers decreased the leakage current density further to 1.78 × 10$^{-9}$ and 1.63 × 10$^{-9}$ A cm$^{-2}$ with 12.5 and 25 wt% of MK, respectively (see brown and green lines in Figure 2a). This is presumably due to an increase in film density on crosslinking the capping layer. Capacitance-frequency ($C$–$F$) measurements (see Figure 2b) for bare P(VDF-TrFE-CFE) films showed a $C_F$ of 202 ± 8 nF cm$^{-2}$ at 100 Hz. These films show a rapid decrease in measured capacitance with increasing frequency due to the inability of the remnant polarization of the $C$–$F$ dipoles to respond to the change in electrical field at higher frequencies. Coating the surface of the P(VDF-TrFE-CFE) film with the P(BMA-co-MMA):MK layer leads to a dramatic decrease in the $C_F$ measured at 100 Hz to 82 ± 6 and
91 ± 7 nF cm$^{-2}$ for MK loadings of 12.5 and 25 wt%, respectively. The reduced dependence of capacitance on the change in frequency is likely a result of a more static dipole of the low-k polymer in the buffer layer.$^{[23,25,26]}$

Bottom-gated OFETs on flexible polyethylene-naphthalate (PEN) plastic substrates with a p-type diketopyrrolopyrrole (DPP) based polymer semiconductor (DPPT-TT, see Figure S5, Supporting Information, for the molecular structure) were fabricated to evaluate the merits of the bilayer gate dielectric (see Figure 1a for schematic of the OFET structure and Experimental Procedure for device fabrication steps). All devices were tested in the dark under ambient conditions and were able to operate at driving voltages of −4 V ($V_{SD} = V_{GS}$) as reflected from the device transfer characteristics shown in Figure 2d (see Figure S6, Supporting Information, for output characteristics and Table S1, Supporting Information, for full device performance parameters). Pristine devices with bare P(VDF-TrFE-CFE) as the gate dielectric showed a large hysteresis loop in the transfer curves (refer to solid line in Figure 2c for 1st cycle) and an average hole mobility $\mu_{h,avg} = 0.34 ± 0.01$ cm$^2$ V$^{-1}$ s$^{-1}$, on/off ratio of 10$^4$ and threshold voltage $V_{th} = −1.9$ V were extracted from the transfer characteristics. On cycling these devices from $V_{GS} = 1$ to −4 V under a constant $V_{SD} = −4$ V for 100 transients (see dotted line in Figure 2c), the max $I_{SD}$ increased by 67% while the $V_{th}$ shifted by 38% in the positive direction to −1.2 V (see Table S1, Supporting Information, for absolute values and Figure S7, Supporting Information, for % changes in $I_{SD}$ and $V_{th}$ against number of cycles). In comparison, pristine devices fabricated with a buffer layer of P(BMA-co-MMA) at 12.5 wt% MK loading showed negligible hysteresis (refer to solid line in Figure 2d for the 1st cycle) and improved performance with $\mu_{h,avg} = 0.16 ± 0.02$ cm$^2$ V$^{-1}$ s$^{-1}$ and an on/off ratio of 10$^4$. These devices showed an improved stability after 100 cycles of bias sweep (see dotted line in Figure 2e) with minimal changes of 7% in max $I_{SD}$ and 2% negative $V_{th}$ shift (see Figure S7, Supporting Information). Pristine OFETs with buffer layers containing 25 wt% of MK showed a lower performance with $\mu_{h,avg} = 0.16 ± 0.02$ cm$^2$ V$^{-1}$ s$^{-1}$ with a more negative $V_{th}$ of −1.6 V and an on/off ratio of 10$^4$ (Figure 2e). These devices also exhibited lower device stability with an increase of 9% in max $I_{SD}$ and a negative $V_{th}$ shift by 14% (see Figure S7, Supporting Information). The operational stability of the OFET devices under ambient conditions at a constant applied bias (at $V_{SD} = V_{GS} = −4$ V) was tested by continuously measuring $I_{SD}$ for 10 h (see Figure 2f). OFETs with P(VDF-TrFE-CFE) as the gate dielectric showed a significant increase in normalized $I_{SD}$ of almost 1200% (red line) from the pristine state. Devices capped with the buffer layer at a 12.5 wt% loading of MK (green line) remained stable over this period with only a minor decrease in $I_{SD}$ by 10%. In contrast, devices with buffer layers containing 25 wt% of MK showed an increase in $I_{SD}$ by 85% (brown line).

From these results, it is evident that hysteresis and bias stability of the transistor characteristics can be dramatically improved by inclusion of a buffer layer on the P(VDF-TrFE-CFE) gate dielectric. The instability of devices with a bare P(VDF-TrFE-CFE) film as dielectric has previously been explained by the highly polar surface induced at the interface with the OSC caused by the remnant polarization of the polar C–F units in P(VDF-TrFE-CFE) under gate bias.$^{[21,23]}$ This induces additional mobile charges into the conducting channel at the
OSC/dielectric interface. As a result, it leads to higher channel currents (max \( I_{SD} \)) and a positive shift in \( V_{th} \) under prolonged bias (see Figure 2c). Incorporation of the buffer layer with the lower polarity of the ketone groups in P(BMA-co-MMA) and the higher film density (due to a cross-linked polymer matrix) suppresses disorder of the transport energy states at the OSC/dielectric interface and passivates the influence of the ferroelectric P(VDF-TrFE-CFE) film leading to higher charge carrier densities and stable device performance (see Figure 2c; Figure S7, Supporting Information).[26,27] It was observed that incorporation of very high loadings of MK (25 wt%) in the buffer layer lead to devices with lower \( \mu_{h,avg} \) and poorer bias stability when compared to buffer layers with MK loadings of 12.5 wt%. A possible explanation could be that excess MK remains in the buffer layer (as shown from the optical absorption spectra of the respective bilayer gate dielectric films in Figure S8, Supporting Information) may act as charge trapping sites for charge carriers at the dielectric/OSC interface.

To demonstrate the wide utility of the bilayer dielectric layer several other p-type polymers such as the amorphous polymer poly(indacenodithiophene-benzothiadiazole) (IDTBT)[21,18] and ambipolar DPP-based benzothiadiazole copolymer (DPPTBT) were employed as the OSC layer in the OFET devices (see Figure S9, Supporting Information, for molecular structures, device transfer and output characteristics). Incorporation of the buffer layer with the IDTBT and DPPTBT achieving \( \mu_{h,avg} \) of 0.26 ± 0.04 and 0.22 ± 0.02 cm² V⁻¹ s⁻¹, respectively.

To demonstrate the utility of these OFET devices as chemical sensors, the devices were exposed to various concentrations of ammonia gas, a common volatile organic compound which is toxic to patients with kidney, liver failure or diseases such as Alzheimer’s.[39] Ammonia was diluted at varying concentrations using compressed air at different levels of relative humidity (RH: 0(dry), 40, and 60%). The gas mixture was passed at a controlled rate into an enclosed chamber placed over an array of 8 OFETs (see Figure S10, Supporting Information, for sensor set-up). The devices were repeatedly exposed to ammonia (at concentrations ranging from 2 to 50 ppm) for 30 s and allowed to recover for 30 min before re-exposure while continuously measuring the dynamic \( \Delta I_{res}/I_0 \) response under an applied bias of \( V_{SD} = V_{GS} = -3 \) V. As shown in Figure 3a, the devices showed an increasing response (decreasing \( I_{SD} \)) with increasing exposure to ammonia gas with an estimated limit-of-detection (LOD) of \(~500\) ppb.[40] The sensing performance for ammonia gas is comparable to literature-reported values.[14,41–43] The observed decrease in \( I_{SD} \) is likely due to \( \text{NH}_3 \) molecules diffusing into the conductive channel at the OSC/dielectric interface and donating electrons via a charge transfer reaction, which decreases the number of charge carriers (holes).[44] Furthermore, the \( I_{SD} \) responses remained stable after five continuous cycles of dynamic ammonia exposure over a period of more than 10 h at various levels of %RH as shown in Figure 3b (Figure S11, Supporting Information, for change in current response vs time). The improved sensitivity at higher RH has been previously assigned to the mixture of ammonia and water forming ammonium hydroxide, which enhances the magnitude of \( \Delta I_{res}/I_0 \) response of the device.[45]

In conclusion, we report a high capacitance low-k/high-k bilayer polymer gate dielectric composed of a photocross-linked buffer layer P(BMA-co-MMA) combined with a high-k P(VDF-TrFE-CFE) polymer layer for use in robust, stable, and scalable OFET devices that operate at low voltage when fabricated on a flexible plastic substrate. The crosslinking strategy for the buffer layer relies on a methacrylate-based copolymer with higher chain mobility to enhance the intermixing with the photoinitiator. This generates sufficient macroradicals to deliver extensive crosslinking of the polymer film. Using this method, several OSCs can be processed from solution whilst maintaining the integrity of the dielectric layer. Excellent device performance was observed and a dramatic improvement in the operational stability of the devices was achieved. These devices were used as ammonia sensors that showed highly reproducible and stable sensing over prolonged measurement cycles in air and humidity. The straightforward fabrication of the bilayer gate dielectric with excellent chemical robustness, low-voltage operational stability and sensing capabilities enables the fabrication of OFET chemical sensors with a variety of OSC sensing elements for multi-analyte detection, and this is currently under active investigation.

**Experimental Section**

The experimental procedures and materials used are included in Supporting Information.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

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ammonia gas, chemical sensors, ferroelectric relaxor fluoropolymers, organic semiconductors, polymer crosslinking

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