Control of geometry in polymer dielectric enhances performance of organic field-effect transistors

Flexible electronic devices are yet to become dominant players in the electronics market, but a new development is bringing this highly desirable technology a step closer to everyday applications. Researchers managed to maximize the performance of organic field-effect transistors (OFETs) by tuning the chemistry of fluoropolymer dielectric materials used as gate insulators, as reported by Jwala M. Adhikari of The Pennsylvania State University, Marcos A. Reyes-Martinez of The Dow Chemical Company, and their colleagues, in Advanced Materials (doi: 10.1002/adma.201602873).

OFETs can be fabricated on plastic substrates at low temperatures and are thus expected to find use in low-cost flexible electronics. In an OFET, the gate electrode is insulated by a layer of a polymer with a high dielectric constant $k$. This polymer dielectric layer also separates the gate from the organic semiconductor (OSC) in which current flows. The current can be controlled by several parameters, including the dielectric constant of the gate insulator. Antonio Facchetti, professor of chemistry at Northwestern University, who was not involved in the study, says, “To enhance the OFET current it would be desirable to couple a high-$k$ dielectric with the organic semiconductor since this induces more charge carriers in the semiconductor layer.”

However, until now previous research has demonstrated an empirical inverse between the dielectric constant of the polymer gate insulator and the charge mobility in the semiconductor layer. Facchetti says this is because in such systems, “the trapping sites also increase dramatically, with the overall effect of reducing the OFET carrier mobility (thus the electrical current), which is problematic for practical applications.”

“The significance of our results is that it breaks this trend. It was very surprising to us that the chemistry of polymer dielectrics turned out to be so important to maximize the performance of organic transistors,” says Enrique D. Gomez, a professor at The Pennsylvania State University and leader of the team that conducted the research. The team has shown that in rubrene single-crystal FETs, high charge mobility (12 cm$^2$ V$^{-1}$ s$^{-1}$) can be achieved with a high-$k$ fluoropolymer dielectric material by controlling the chemistry of the latter.

Rubrene is an OSC frequently used in OFETs. The fluoropolymer used in the study was P(VDF-BTFE), a vinylidene fluoro (VDF) copolymerized with bromotrifluoroethylene (BTFE), a high-$k$ material with great stability. In OFETs based on conjugated polymers, polar groups, like fluorine in this case, can induce chain conformations near the OSC/dielectric interface, which hinder the charge mobility in the device. But BTFE allows cross-linking of the polymer chains through thermal- or photocuring, resulting in low density of charge traps.

Cross-linking of P(VDF-BTFE) has little effect on its dielectric properties and is realized through adding cross-linkers (dicumyl peroxide and triallyl isocyanurate) to P(VDF-BTFE) and exposing the film to ultraviolet light for several hours. The cross-linking is controlled through shadow masks and the uncross-linked polymer is removed through solvent exposure.

The role of cross-linking on P(VDF-BTFE) was investigated by quantifying the populations of P(VDF-BTFE) chain conformations by means of Fourier transform infrared spectroscopy. The results showed that TTTG (T: trans, G: gauche) conformations mostly dominate. The researchers believe that enhancement of trans chain conformations and the lower heterogeneity in the dielectric layer demonstrates a reduction of chain twisting induced by misoriented dipoles, and thus less energetic disorder and minimal trap density.

According to Facchetti the extension of this work to other organic semiconductors will open unprecedented opportunities for unconventional electronics. Oana Jurchescu, a professor at Wake Forest University, also considers the work of Gomez’s team as a significant achievement that paves the way for the high-performance operation of OFETs at low voltages. “These results are extremely promising as they show that organic semiconductors can yield efficient devices even at low voltages upon minimizing the trap densities in the device channel,” Jurchescu says.

Gomez believes that the control of the semiconductor–dielectric interface has the potential to transform OFETs either “through changes to chemical structure of the polymer dielectric or through changes specific to the interface.” He says the group is planning to continue their work “to achieve performance that will make these devices relevant for flexible displays or bioelectronic applications.”

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Hole mobilities of rubrene single-crystal devices versus $k$ of the dielectric layer. The dotted line is a guide to the eye. The highest mobilities and highest dielectric constants are achieved with cross-linked P(VDF-BTFE) used in this study. Credit: Enrique D. Gomez.