論 文 の 内 容 の 要 旨

論文題目 High-frequency organic thin-film transistors for conformable large-area electronics

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Background

Organic thin-film transistors (OTFTs) are one of the most important building elements to realize large-area flexible electronics. Recent material advancements allow organic transistors to be environmentally and thermally stable and provide good electrical performance. This progress enabled the fabrication of electronic circuits for practical applications, such as displays and processing units. High frequency operation of single devices and oscillators could also be demonstrated on thick plastic foils. In addition to their improved electrical properties, the mechanical flexibility and conformability of such devices can be improved by reducing the thickness of the base films. Organic elements manufactured on ultrathin substrates can provide smaller bending radii and better conformability to complex objects including human skins.

Despite the impressive development of organic electronics, no demonstration for large area electronics that provides both excellent conformability, large bandwidth (cutoff frequency > 100 kHz) and good uniformity (~10% device variation) has been made yet. Organic transistors on thin films suffer from narrow bandwidth due to limited scaling methods and inherently low carrier mobility. So far, only low frequency OTFTs and circuits were demonstrated on ultrathin and conformable substrates. Thermally stable organic devices could be achieved only by the use of specific transistor structure on rather thick plastic foils. Uniformity of organic transistors over large area is another main issue towards reliable implementation of practical applications.

Objective

In this study we develop a novel method for fabrication of high frequency organic transistors and circuits on large area and ultrathin foils, which shows high degree of uniformity, mechanical and environmental stability. The technology is based on the fabrication of high performance, short channel OTFTs in the bottom contact architecture to enhance devices bandwidth in low operation voltages. We aim to demonstrate large area compatibility on ultrathin substrates to enable the realization of future conformable sensor sheets for wearable or implantable devices.

Method

Our method for devices improvement towards reliable and conformable OTFTs is based on three main principles, demonstrated simultaneously for the first time in this study:

Substrate thickness

Reduction of the base film down to ~1 μm thick substrates, allows devices to conform to any three-dimensional object. Small device thickness also facilitates the stress applied on the transistors and enhances their mechanical flexibility.

Bottom contact structure

The bottom contact structure allows reliable high resolution patterning using photolithography to increase transistor's cutoff frequency.

Vapor deposited polymeric gate dielectric

The choice of flexible vapor deposited polymeric gate dielectric allows uniform device performance and ultrathin, pinhole free interface with the organic semiconductor for reduction of operation voltage.

We utilized those principles in our device fabrication and discuss their effect on device performance.

First, we adapt photolithography to be used on ultrathin parylene diX-SR foils and fabricate OTFTs for the first time using this technology on 1 μm substrates. By utilizing the bottom contact architecture we pattern channel lengths down to 2 μm to realize air stable, dinaphtho[2,3-b:2´3´-f]thieno[3,2-b]thiophene (DNTT) based OTFTs. We improve device performance by applying contact modification techniques using pentafluorobenzenethiol (PFBT) self-assembled-monolayer (SAM) and optimize mobility with different deposition conditions. We compare this method to other available contact modification techniques and discuss the challenges when applying to ultrathin films.

Second, we perform thorough electrical device characterization and analyze important OTFT parameters. Thanks to the effective contact modification we could realize saturation mobility of ~0.2 $cm^2/V \cdot s$ for short channel transistors (~2 μm channel length) in bottom contact architecture with transfer length of only 1 μm. The obtained contact resistance in our devices was found to be 5.5 kΩ·cm which is comparable to top contact devices with the same materials. The fine patterning method allowed us to reduce the overlap capacitance of less than 0.5 pF per channel were achieved. High ratio between ON and OFF currents of 10^7 was demonstrated which is important for practical sensing applications. We could reduce operation voltage down to 8V by reduction of dielectric thickness to less than 100 nm and still keeping leakage current lower than 100 pA during transistor operation. Theoretical and measured transistor cutoff frequency exceeded 100 kHz on ultrathin foils. We discuss the successful results in light of the top gate architecture implementation and fabrication technique.

Further stress characterization were conducted on the ultrathin devices. Transistors were tested under severe mechanical bending and crumpling and showed good durability against the mechanical stress. While only 2 μm thick, devices could be rolled down to 600 μm bending radius and be crumpled several times with showing practically no degradation in performance (< 10% in mobility). We discuss the theoretical considerations in applying stress on the bottom contact structure in the ultrathin form.

Additionally, we evaluated devices environmental stability by performing thermal annealing tests. We showed that ultrathin OTFTs in the bottom contact structure could endure heating up to 170°C in N_2 and 150°C when annealed in air. We provide evidence to cyclic stability of those devices after repeated annealing steps. We calculated that thanks to the short channel implementation, cutoff frequency of devices could be kept above 100 kHz even after annealing at 170°C. We analyze the changes in channel and contact resistances and by electrical and topological means.

Finally, we demonstrate the reliability of our OTFTs by the realization of electrical circuits and large area active matrix. We designed and successfully fabricated pseudo-CMOS inverters with gain of as high as 30 dB. We also showed that inverter gain can be improved by at least 150%

after simple post annealing step at 150°C. Organic amplifier with improved frequency response of up to 25 kHz (f_{3dB}) and 45 kHz unity gain could be achieved in the AC coupled architecture. For the closed loop design we added parylene capacitors in total active area of 45 mm^2, which achieved constant frequency response of from 3Hz to 3kHz. By integrating our devices with aluminum-oxide capacitors (~500 nF/cm^2) we reduce effective amplifier area to only 15 mm^2 which is suitable to implementation for high resolution sensing devices. Integrated amplifiers showed a closed loop gain of ~6 dB and stable operation between 5Hz-700Hz. The large area uniformity of our devices was successfully demonstrated by the fabrication of 6x6 cm^2 transistor array which showed excellent yield (98.6% out of 144 transistors) and only 0.019 cm^2/V·s standard deviation from the average value of 0.12 cm^2/V·s. The transistors were used as a large area pressure sensor to demonstrate their applicability.

Summary

We have successfully demonstrated the highest frequency OTFTs reported on ultrathin films (~1 µm) in the bottom contact architecture. Channel patterns down to 2 µm length with cutoff frequencies greater than 1 MHz were realized. Using ultrathin parylene diX-SR gate dielectric, we could maintain low voltage operation for our devices. OTFTs showed excellent durability to mechanical and thermal stress thanks to our design and material selection. Transistors reliability was demonstrated by the fabrication of ultrathin inverters and amplifier circuits, showing record high bandwidth (25 kHz) for a single organic amplifier. The smallest organic closed-loop amplifier (15 mm^2 of active area) was realized by integrating the transistors with AlOx capacitors, presenting uniform gain up to 700Hz. Excellent uniformity (~15% of mobility standard variation) and device yield (98.6%) was realized on 36 cm^2 active matrix with 144 transistors, utilized as an ultrathin pressure sensor.