Self-organized organic electronic and photonic materials by micro-dewetting

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Techniques based on rupture of a thin liquid film to form droplets and line structures create submicrometer-size patterns that are used in making organic LEDs and field-effect transistors.

Many electronic and photonic devices require patterning of some of the materials involved. There are three main challenges in this field: decreasing pattern size, increasing throughput, and lowering production cost and energy. The first task is tackled by top-down light- and electron-beam lithographies, as well as bottom-up biomimetic methods using, for example, DNA and protein templates. As a result, pattern sizes are now down to a few nanometers. However, the direct-writing techniques necessary for the highest resolution are very time-consuming, with typical writing speeds of a few tens of micrometers per second. This drawback can be partially resolved by lithographically produced nanoimprint molds that allow roll-to-roll patterning and thus massively parallel production. Still, this approach suffers from technological hurdles, such as requiring complex steppers and lasers.

Increasing throughput and reducing cost can also be achieved through solution processes. But the surface tension of liquids does not allow formation of arbitrarily small droplets. Only recently was it reported that a liquid printing process could break the micrometer barrier. Moreover, ink-jet printing is a serial process. Even though arrays of printing heads can be used, it still takes time to prepare large-area patterns.

But surface tension does not pose only problems. The very fact that a liquid film or jet contracts and forms droplets can, in principle, be used to make a pattern. If a homogeneous liquid is placed on an unstructured substrate and the liquid breaks up into droplets, a pattern may form where no pattern existed before. Ilya Prigogine was a pioneer in the field of nonequilibrium dynamics and dissipative structures. In applying his theory to an evaporating solution, we observed clearly that dynamic structures emerged during solvent evaporation. Fluctuations in surface tension are stabilized by a positive feedback mechanism,

Continued on next page
which produces ordered structures with certain sizes. By adding a nonvolatile solute, we were able to fix these structures on the substrate. Figure 1 shows a microscope image of the edge of an evaporating polystyrene-solution droplet. The dissipative structures form a regular fingering instability that leads to the deposition of micrometer-sized aggregates of the polymer. The key advantages are the narrow size distribution of the aggregates, their equidistant spacing, and the speed of formation. It only takes a few milliseconds for each aggregate to form. The receding speed of the liquid can be controlled and is typically in the range of several centimeters per minute, so that an A4 (European letter paper)-size substrate can be coated within a few minutes.

Many different amorphous materials can thus be coated onto a variety of substrates, including mica, silicon wafer, glass, and indium tin oxide. Aggregate sizes are determined by the dewetting parameters, and aggregates with diameters as small as 200nm can be formed. We realized organic LEDs by dewetting a low-molar-mass amorphous hole (positive charge carrier) transport material, followed by ‘conventional’ vacuum evaporation of the electron conductor and top electrode. We have also fabricated microlens arrays with a feature size of \( \sim 1\mu m \).

We recently reported that crystalline materials can also form these nanoaggregates. Controlling crystallization at the receding liquid edge enables either single nanocrystals or high-aspect-ratio fibers. Pentacene, a hole-transport material for organic field-effect transistors, can form fibers with a diameter of 500nm, a height of 100nm, and a length of up to 1mm. We found that coating an electrode array with a pentacene solution resulted in fiber arrays between source and drain electrodes with charge carrier mobilities of up to \( 1.6 \text{cm}^2 \text{V}^{-1} \text{s}^{-1} \). In summary, we have shown the potential usefulness of liquid processing of organic materials in patterning the active ingredients of photonic and electronic devices. OLEDs, microlenses, and transistors may just be the beginning. It might be possible to create novel micrometer-scale patterned organic solar cells using this method. Biomedical sensor arrays can also be produced by dewetting. The technique does not involve stamping or lithography techniques. As a next step, we plan to use our microdewetting method for surfaces that are not readily accessible, such as in the channels of microfluidic devices.

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