Ultrathin organic device displays information directly on skin

The field of electronic skin (e-skin) has blossomed in recent years, with researchers demonstrating a range of practical healthcare applications for these wearable devices, ranging from detecting inflammation around wounds to reading the electrical activity of the brain. Various research groups are now working toward incorporating stretchable light-emitting diodes (LEDs) into their devices, which would allow users to read their vitals straight from their skin.

In a step toward this goal, materials scientists at The University of Tokyo have created an ultrathin and ultraflexible optoelectronic skin with organic photodetectors and highly efficient polymer light-emitting diodes (PLEDs). In a practical test of the technology, published recently in *Science Advances* (doi:10.1126/sciadv.1501856), the team demonstrated a skin-laminated device that could serve as an accurate pulse oximeter to measure oxygen concentration of blood, and that is stable enough to survive in ambient air.

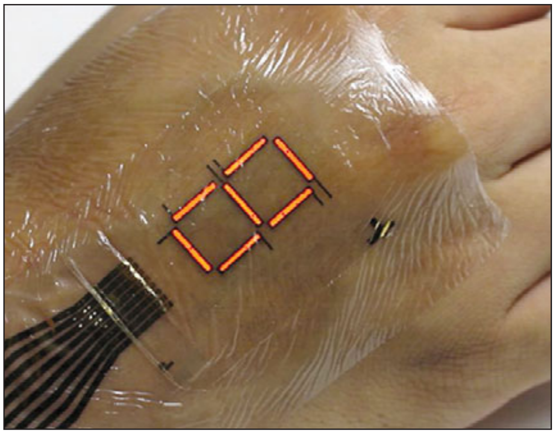
"Future technology is growing from wearable (e.g., smartbands, smart watches) to skin-attachable sensors and devices," says Hyunhyub Ko, a chemical engineer at Ulsan National Institute of Science and Technology in South Korea who was not involved in the study. "The advantage of skin-attachable e-skins is the improvement of biosignal detection accuracy and unobtrusive monitoring of daily healthcare signals. In this regard, I think that this work [has] opened a pathway to skin-attachable organic optoelectronic skins."

A key aspect of e-skin is its ability to flex and stretch with the skin without becoming damaged. Previously, researchers successfully created stretchable devices using very thin inorganic LEDs, but the fabrication process was unsuitable for creating inexpensive, large-area devices. Organic LEDs are an attractive alternative. In addition to the natural flexibility of organic materials, "we can fabricate the device using [a] low-temperature process and printing," says the study's first author and University of Tokyo engineer Tomoyuki Yokota, adding that this reduces fabrication costs and allows for the use of very thin substrates.

In a previous study, the research group of Takao Someya, lead author of the current paper, developed ultrathin (2-μm-thick), highly flexible, stretchable PLEDs, but these devices were driven in a nitrogen atmosphere and were not stable in air. For the new work, Yokota, Someya, and their colleagues sought to solve this issue by optimizing the fabrication of their PLEDs—while still employing materials and processes currently used in the production of organic LEDs—and developing a protective, transparent film, or "passivation layer."

To create their PLEDs, the team began by creating a parylene substrate, a process that involved depositing a 1-μm-thick parylene layer onto a glass plate surface using chemical vapor deposition (CVD). They then spin-coated a 500-nm-thick polyimide planarization layer, which created a very flat, smooth surface to attach a transparent indium tin oxide (ITO) electrode. They further created an active layer with blue, green, or red light-emitting polymer materials, and completed their PLEDs by depositing a NaF/Al cathode.

After fabricating their PLEDs, the researchers added a flexible and thin passivation layer comprised of five alternating inorganic (SiON) and organic (parylene) layers deposited with plasma-enhanced CVD and CVD, respectively. Altogether, the PLED was just 3-μm thick, but still exhibited high flexibility and brightness, even when crumpled up or bent over the tip of a 100-μm-thick razor. Furthermore, the new PLED was over six times more efficient than previously reported ultrathin PLEDs, and did not degrade after 1000 cycles with 60% stretching. The passivation layer also extended the in-air operational half-life of the PLEDs from 2 hours to 29 hours.



Researchers have developed an ultrathin, ultraflexible electronic skin that contains a digital display with either red, blue, or green polymer light-emitting diodes. Credit: The University of Tokyo, Someya Group Organic Transistor Lab.

These PLEDs could also be laminated onto skin as an analog or a seven-segment numerical digital display (similar to clock displays).

"Air-stable organic light-emitting devices are harder to realize because some of the materials are usually air-sensitive," says Zhenan Bao of Stanford University, who was not involved in the research. "The ultrathin device here is impressive to have such a good air stability. We can imagine bright flashing tattoos."

Yokota and his colleagues similarly fabricated organic photodetectors on 1-μm-thick parylene substrates with an active layer of poly(3-hexylthiophene):(6,6)-phenyl-C61-butyric acid methyl ester and a 1-μm-thick parylene passivation layer. Like the PLEDs, these photodetectors were also hardy, showing little current and voltage differences after being compressed by 40%. They also did not degrade after 300 stretching cycles.

In a real-world test, the team laminated three layers—one green PLED, one red PLED, and one organic photodetector—on skin, creating an ultraflexible reflective pulse oximeter that was used to measure a person's blood oxygen concentration and pulse rate. When wrapped around a finger, the PLEDs emitted light into the body, and the reflected light was picked up by the photodetector. Pulses in the signal correspond to heart rate, and the amount of light absorbed by the body differs depending on how much oxygen is in the blood, allowing the device to read blood-oxygen concentrations.

The device performed as well as previous organic pulse oximeters that use glass and thick plastic substrates, but had the added benefit of being ultrathin and being strongly adhesive. "Our device is very thin, so we can achieve [a] very stable signal," Yokota says.

"The flexibility of organic semiconductors makes them a natural technology for wearable sensors," says Ifor Samuel of the Organic Semiconductor Optoelectronics Group at the University of St. Andrews, who was also not involved in the study. "This work shows a promising advance in flexible encapsulation of such devices."

Dae-Hyeong Kim, a researcher in flexible electronics at Seoul National University, adds that the work is "extremely important" for the field of deformable optoelectronics used in long-term health monitoring due to the softness of the device. As mobile electronics become more prevalent, "this technology would become one of [the] good technological solutions toward fully integrated wearable electronics and bioelectronics," Kim says.

The researchers are now looking to fabricate a full-color e-skin display and further improve the stability of the device in air. "If we succeed [in improving] stability, we want to use our device in water," Yokota says.

Joseph Bennington-Castro

Adaptive design loop ushers exploration of new materials

Advanced materials discovery is essential to economic security, technological advance, and social welfare. However, it often takes a few decades from initial discovery to market due to challenges in finding the optimal materials properties for certain applications. The enormous degree of complexity in manipulating materials properties comes with countless "knobs" to turn when designing the experimental approach. Strategies are needed for a design-feedback loop to locally exploit and globally explore optimized solution from hundreds of thousands of possible candidates. The adaptive design loop incorporates statistical inference, machine learning, design, and experimental feedback to optimally guide future experiments. The loop is finding relevance in materials design ranging from metal alloys to polymers, ceramics or nanomaterials, where researchers are searching for target materials properties.

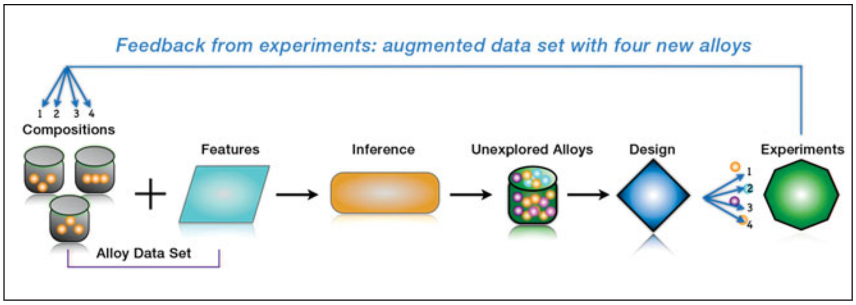


Illustration of adaptive design loop experiment of NiTi-based alloy system. Credit: Turab Lookman.

In a recent issue of *Nature Communications* (doi:10.1038/ncomms11241), a research team, led by Turab Lookman in the Theoretical Division at Los Alamos National Laboratory, developed a data-driven framework to effectively navigate new materials discovery with targeted property optimization. "The key question we addressed is the following: how do we optimally guide the next experiment(s) or calculation(s), in search of materials with a desired response, especially when the search space is vast?" Lookman says. The answer is to incorporate uncertainties in designing the experiments. The predicted data points with larger uncertainties will facilitate greater exploration in global search, even when the mean predicted values are far from desired. In contrast, the predicted data points with smaller uncertainties favor exploitation in local search.

The strategy was used to find very low thermal hysteresis (ΔT) in NiTi-based shape memory alloys with nearly 800,000 potential alloys in the $Ni_{50-x-y-z}Ti_{50}Cu_xFe_yPd_z$ family to choose from. The demonstrated adaptive design loop starts with collecting prior knowledge of 22 initial alloys with measured materials properties. Statistical models are used to predict materials properties and suggest new experiments. On the sixth of nine iterations of the adaptive design loop, an alloy with the lowest ΔT was discovered at an optimized materials composition. In each iteration, four alloy compositions were predicted and synthesized. Fourteen out of 36 alloy compositions were found to have ΔT smaller than the best data points in the original data set. "A key component of our strategy is 'design,' recommending the next experiments to be performed in order to improve subsequent model predictions, hence adaptive design," Lookman says.

"This type of approach brings the best statistical ideas to materials discovery and design. While the methods have seen substantial use in genomics and machine learning, their use in materials research opens the door to better targeted experiments; improved materials models; and a deeper, more effective science of materials," says James A. Warren, the director of the Materials Genome Program at the National Institute of Standards and Technology. "These methods, and their successors, will accelerate new materials-into-application, finding use across the breadth of materials research."

YuHao Liu