An adhering, pure conducting polymer hydrogel for medical applications

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Laser-fabricated conductive hydrogel micropatterns that can strongly adhere to various polymer substrates even after harsh mechanical stimuli. Credit: Daeyeon Won, PhD, SNU

The rapid advancement of electronics and artificial intelligence (AI)
tools have opened interesting opportunities for the development of technologies for a wide range of applications. These include implantable devices designed to support the treatment of medical conditions, monitor biological processes, or augment human abilities.

Researchers at Seoul National University, the Korea Advanced Institute of Science and Technology (KAIST), Konkuk University and Hanyang University recently created a new hydrogel based on a pure conducting polymer that could be used to create bio-compatible devices.

This hydrogel, introduced in a paper published in *Nature Electronics*, could be easier to produce and tailor for specific applications than other similar materials developed in the past.

"Electronics directly implanted into the body cannot yet escape conventional hard materials, so side effects such as immune response induced by mechanical mismatches with soft biological tissues are critical risks under prolonged implantation," Seung Hwan Ko, co-author of the paper, told Tech Xplore.

"To solve this problem, electronics are being developed using soft materials that have similar properties to our bodies (e.g., low Young’s modulus, high water content), but they face the limitations of poor device performance and weak mechanical stability in wet physiological environments."

Ko and his collaborators have been developing new soft materials for more than five years, using various processing techniques that could ensure their stability in wet environments, such as the inside of the human body.

Their recent works specifically focused on hydrogels, artificial materials that are most similar to the human body, as they are characterized by a
low so-called Young's modulus (i.e., the ability to withstand changes in length when a force is applied) and high-water content.

"To ensure the high electrical conductivity of conductive hydrogels, we used no insulating polymers, instead only treating the pure conducting polymer (PEDOT:PSS)," Ko explained.

"The primary goals of our research were the fabrication of extremely stable conductive hydrogel electronic devices, the assurance of electrical performance of the hydrogel device greatly exceeding current devices and the realization of these properties at the microscale with simple processes."

In a previous paper published in 2022, the researchers had introduced a process for the micropatterning of conductive hydrogels, which entails the laser-induced separation of PEDOT:PSS.

However, thick and dark PEDOT:PSS could easily absorb visible light in most wavelengths, thus they found that their proposed technique failed to deliver photothermal energy to the substrates and could not create strong bonds.

"Along with our paper in 2022, there were prevailing concerns that most existing conductive hydrogel electronics would not overcome the limitation of practical use due to easy delamination from the substrates inside the wet bodies," Ko said. "In our new paper, we found the inspiration to solve this problem at the 'interface.' The idea was to create direct bonds with PEDOT:PSS and the substrate by concentrating the photothermal energy of the laser at the interface."

As most soft polymer substrates can transmit the majority of visible light, Ko and his colleagues decided to flip over PEDOT:PSS-coated transparent substrates and irradiated them with a 532 nm laser beam.
This beam was transmitted through the transparent substrate, allowing the polymer PEDOT:PSS to absorb it and generate concentrated photothermal energy at the interface with the substrate.

"By subsequently immersing the laser-treated sample in water, only the region treated by laser remains highly stable in water due to the phase separation of PEDOT:PSS and strong bonding with the substrates," Ko said. "This unique PEDOT:PSS patterns become conductive hydrogels that can contain more than 80% water, and are composed only of pure conducting polymer, ensuring high conductivity of more than 100 S/cm."

The laser-assisted micropatterning strategy employed by the researchers merely entails laser irradiation, thus eliminating the need for complex pre-processing steps. Its only requirements are the careful drying of PEDOT:PSS on polymer substrates and well-defined laser scanning conditions.

"Essentially, we drop-cast PEDOT:PSS solution on various polymer substrates and dry it well," Ko said. "Then proper parameters of the laser beam are irradiated toward the transparent substrates, which induces phase separation of PEDOT:PSS and creates strong bonds with the substrate. Additionally, the phase separation of PEDOT:PSS can be further increased for electrical conductivity enhancements, and post-treatment with various organic solvents is possible."

As part of their recent study, the researchers specifically treated their hydrogel using ethylene glycol. Using their proposed strategy, they produced a hydrogel pattern with a resolution 5 μm, which is comparable to the resolution achieved using photolithography techniques.

"Various solution-based processes can also easily pattern conductive hydrogels," Ko said. "Usually, conductive hydrogels are synthesized in a solution state or made into mixtures, so they are patterned through
various solution processes such as 3D printing, inkjet printing, and screen printing. These processes have limitations in spatial resolution over 100 micrometers due to liquid spreading effects."

One of the most advanced methods for the high-resolution patterning of hydrogels is photolithography. While this technique can achieve good resolution, it also requires complex and expensive manufacturing processes, without guaranteeing the strong bonding of polymers to substrates.

"Normally, soft hydrogel electronics are considered very fragile, and there has been a tacit agreement in the research field that it is too early for practical applications," Ko said. "In addition, it was unclear whether conductive hydrogel microelectrodes could be used practically for prolonged implantation due to the critical risk of delamination due to their water-rich characteristics.

"Our work is of great significance, as we showed that microelectronics made only with pure conductive hydrogels adhere to various commercial polymer substrates with high bonding forces and can be used stably for long-term periods."

In initial tests, the hydrogel fabricated by Ko and his colleagues achieved remarkable results, exhibiting good adhesiveness and stability in wet conditions. In addition, they found that the hydrogel maintained its bonding strength even after strong ultrasonic cleaning, which could be advantageous for the development of implantable devices.

"The mechanism of this strong bonding was explored by various in-depth analyses at the interfaces," Ko said. "We believe our research will give good insights to various electronic applications operating in wet environments."
The recent study by this research team could soon pave the way for the development of new electronics that can operate inside the human body. Ko and his colleagues have already started using their micropatterning technique to fabricate bio-compatible soft hydrogel electronic components.

"In the next research, we are planning to identify specific clinical applications where our soft electronics can be used reliably," Ko added. "Additionally, one of the great advantages of our process is its rapid process speed. It enables rapid prototyping of devices in response to various organs with different shapes. Therefore, we plan to develop hydrogel microelectronics that can be applied to small organs that require shape optimization."


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