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Soft and ion-conducting hydrogel artificial tongue for astringency perception

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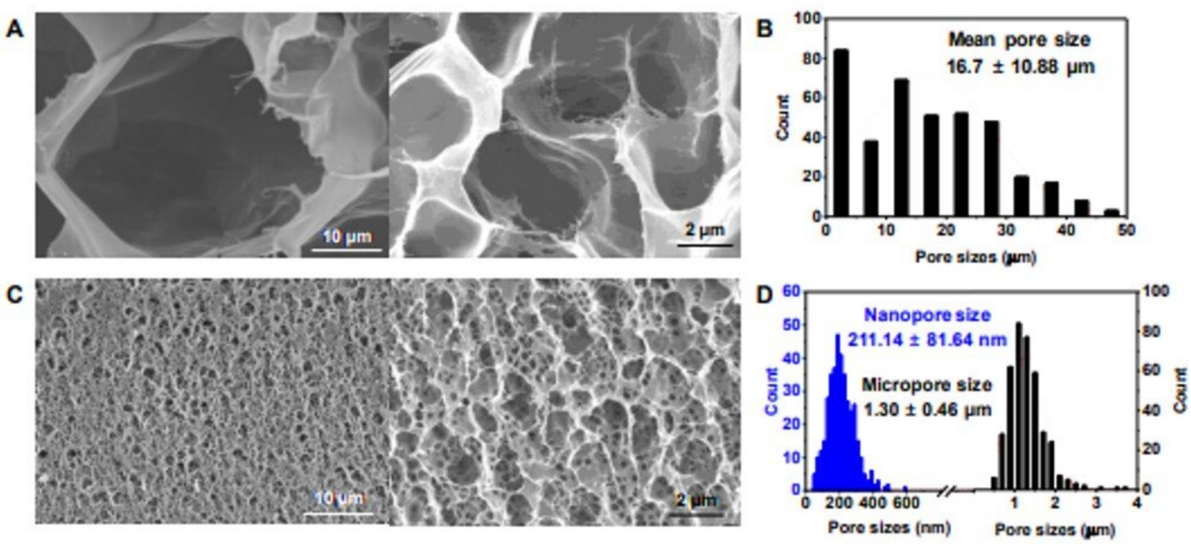
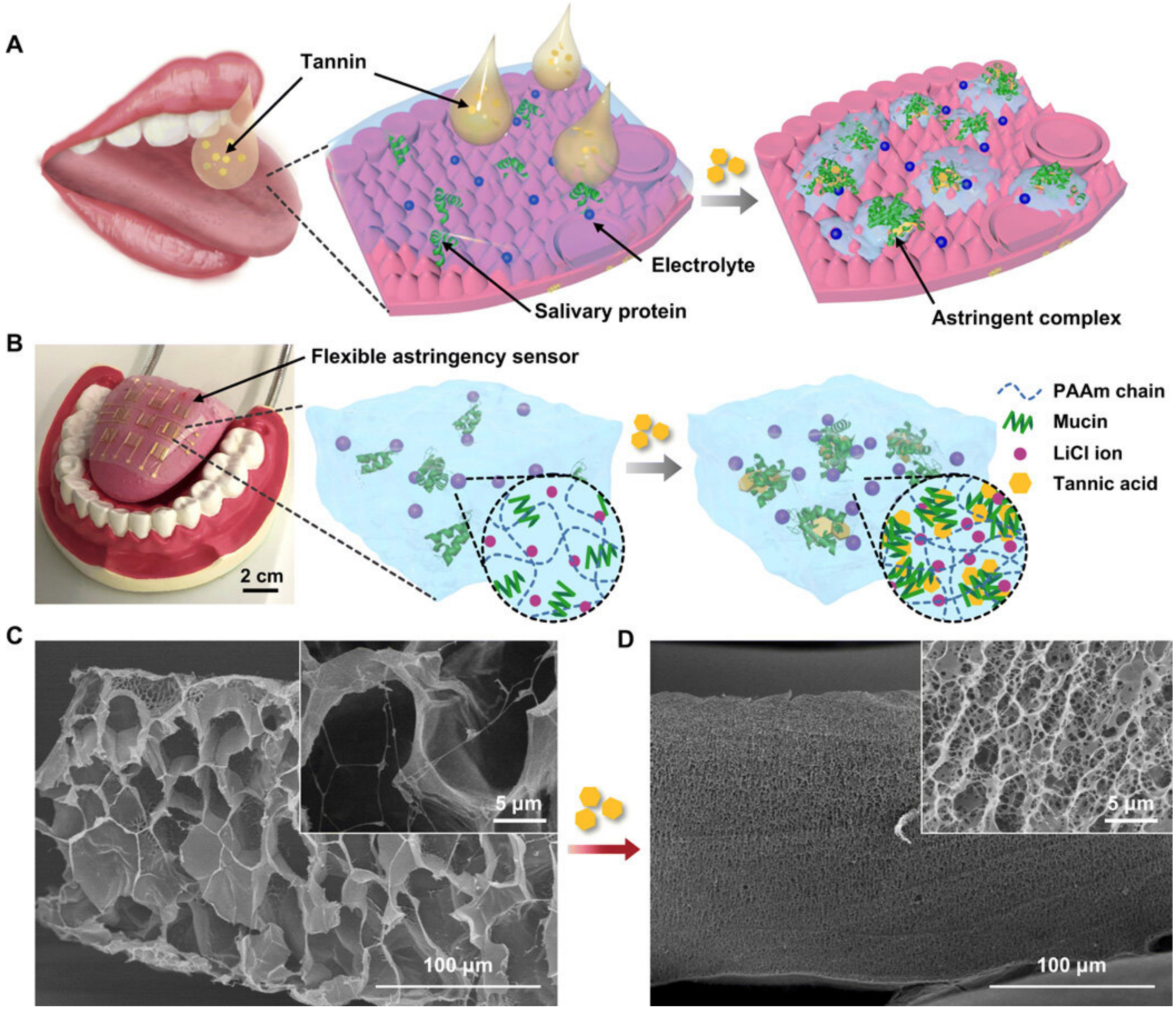


Changes in pore size of human tongue mimicked hydrogel under TA treatment. (A) SEM images of human tongue mimicked hydrogel before TA treatment. (B) Histogram of the mean micro pore sizes of the artificial tongue before TA treatment. (C) SEM images of the human tongue mimicked hydrogel after TA treatment. (D) Histogram of the mean micro/nanopore sizes of the artificial tongue after TA treatment. Histograms mean and error bars (B and D) are measured from 390 pores. Credit: Science Advances, doi: 10.1126/sciadv.aba5785

Artificial tongues have received increased attention due to their ability to detect the five basic tastes, but until now scientists have been unable to fully enable human tongue-like biomimicry for astringency in the lab. To mimic the mechanisms of human tongue-like perception of astringency, Jeonghee Yeom and a team of scientists in energy engineering and chemical engineering at the Ulsan National Institute of Science and Technology in the Republic of Korea, used a saliva-like, chemiresistive ionic hydrogel anchored to a flexible substrate to create a soft artificial tongue. They exposed the construct to astringent compounds and allowed hydrophobic aggregates to form in the microporous network, transforming it into a micro/nanoporous structure with improved ionic conductivity. Using the unique human tongue-like structure, they detected tannic acid (TA) across a wide spectrum (0.0005 to 1 weight percentage) with high sensitivity and a fast response time. As a proof-of-concept, the sensor detected the degree of astringency in beverages and fruits based on a simple wipe-and-detect method. The platform will have powerful future applications in humanoid robots and as taste monitoring devices, the research work is now published on *Science Advances*.

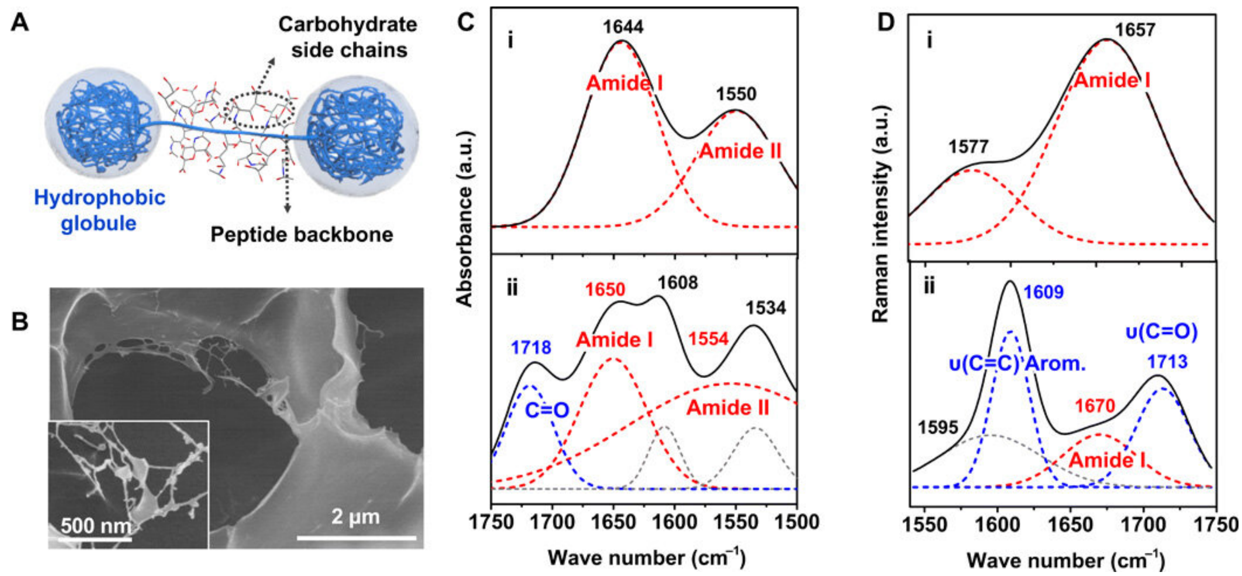
The tongue is a muscular organ that forms one of the softest, most flexible and sensitive body parts housing a range of mechanical receptors and ion channels. A thin salivary film of a few hundred microns thickness maintains the moisture of the tongue, and contains a mixture of 99 percent water, a mixture of electrolytes, immunoglobins and secretory proteins. Saliva plays a significant role during taste perception by dissolving tastants and allowing them to bind to receptor cells or efficiently flow through ion channels. Humans can distinguish five basic tastes, which include sweet, sour, bitter, salty and umami. The water-soluble tastes can be detected via taste receptor cells or ion channels, based on electrical signals that are generated due to depolarization of receptor cells after binding taste chemicals for sweet, bitter and umami sensations. For salty and sour tastes the process depends on the flow of sodium or hydrogen ions through the ion channels.



Operating principle of the astringency detectable sensor. (A) Schematic illustration of astringency sensing principle of the human tongue. (B) Photograph of artificial tongue and schematic illustration of the astringency sensing principle of artificial tongue. Photo credit: J.Y., Ulsan National Institute of Science and Technology. (C) Scanning electron microscopy (SEM) image of the astringency detectable hydrogel before exposure to TA. (D) SEM images of the astringent detectable hydrogel after exposure to 1 wt % TA for 60 s; insets in (C) and (D) are magnified SEM images demonstrating micropores and micro/nanopores, respectively. Credit: Science Advances, doi: 10.1126/sciadv.aba5785

Humans can sense astringency through exposure to polyphenols mainly found in unripe fruits, wines and teas. They are a strong antioxidant and anti-inflammatory substance, but capable of provoking negative nutritional impacts or becoming lethal in high doses. Astringents can be detected due to the strong association of ingested astringent tastants and salvia proteins that cover the tongue. Inside the oral cavity, astringent tastants can bind with secreted proteins and form insoluble precipitates to shrink the epithelium causing a dry, puckered feeling. Thus far, bioengineers have not developed a fully flexible and soft artificial tongue selective for specific astringent tastants. In this work, Yeom et al. mimicked the mechanisms of human astringency perception by introducing a soft hydrogel-based artificial tongue. They were bioinspired by the thin salivary layer on the human tongue to create an equally soft and thin hydrogel film on a flexible polymer substrate via covalent bonding.

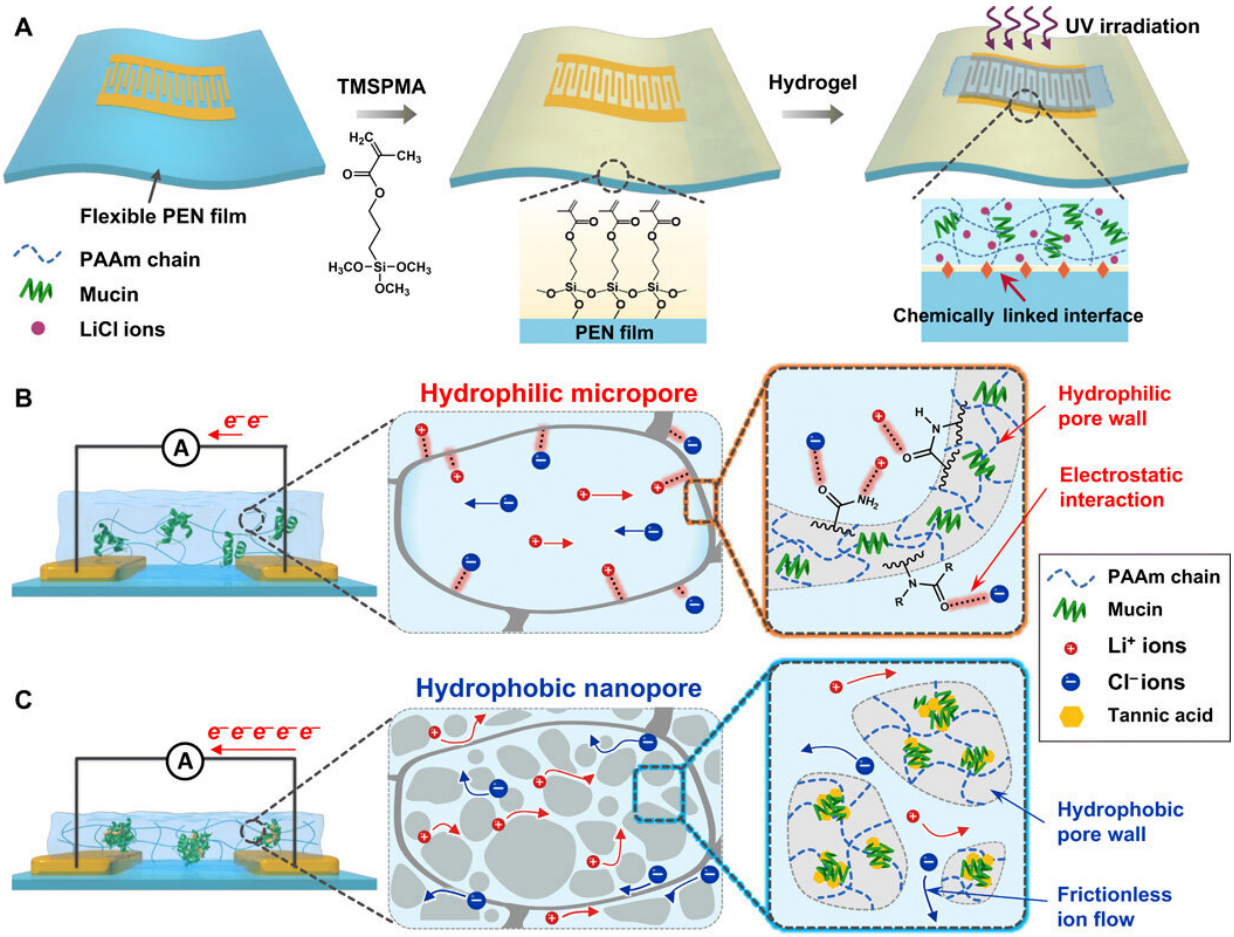
The artificial tongue contained mucin as a secreted protein, lithium chloride (LiCl), polyacrylamide (PAAm) and a three-dimensional (3-D) porous polymer network to allow the facile flow of electrolytes. The soft hydrogel thickness of 200 microns was comparable to an actual salivary layer on a human tongue and facilitated efficient adsorption and diffusion of astringents. As an example, Yeom et al. used tannic acid (TA) during the experiments. When TA diffused into the hydrogel matrix, incoming TA molecules bound and complexed with mucin to form hydrophobic aggregates. The process transformed the microporous gel into a hierarchical micro or nanoporous structure with enhanced ionic conductivity. The construct could successfully detect the degree of astringency in real beverages and also efficiently monitored the ripening of fruits.



Binding mechanism of mucin and TA. (A) Schematic illustration of mucin. (B) SEM image of the mucin polymer dispersed in the PAAm hydrogel network; inset image is magnified part of the pore edge. (C) FTIR spectra of (i) mucin and (ii) mixture of mucin and TA. (D) Raman spectra of (i) mucin and (ii) mixture of mucin and TA. a.u., arbitrary unit. Credit: Science Advances, doi: 10.1126/sciadv.aba5785

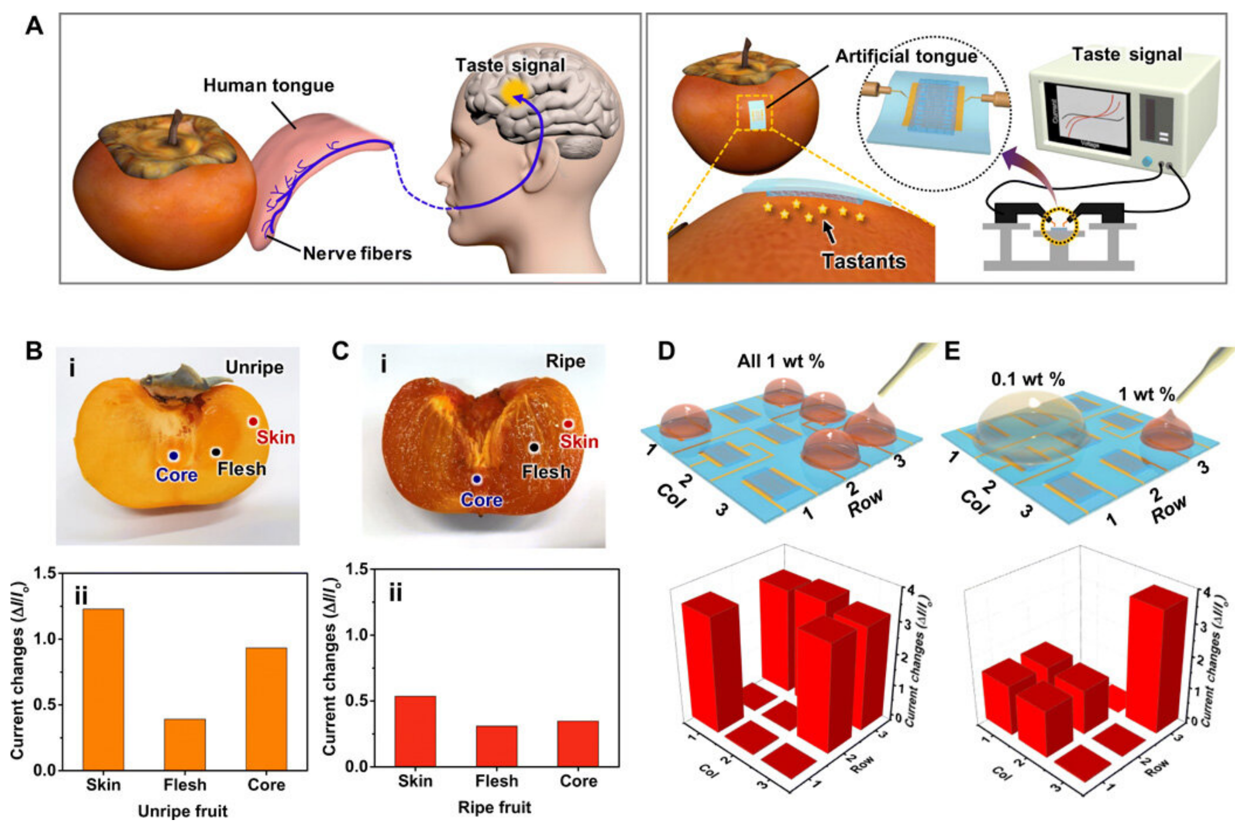
Yeom et al. examined the binding mechanisms of mucin and tannin and studied their chemical composition using Fourier-transform infrared (FTIR) and Raman spectroscopies. Vibrational peaks of mucin corresponded to the protein bands of amide I and amide II and the bound tannin caused a change in background conformation. To design a flexible chemiresistive sensor using a saliva-like hydrogel and flexible electrode substrate, the scientists used poly(ethylene naphthalate) (PEN), followed by oxygen plasma treatment to form a hydrophilic (water-loving) PEN surface for efficient surface attachment to the saliva-like PAAm hydrogel network. They then used a chemical anchoring agent under ultraviolet (UV) polymerization for covalent bonding between the substrates.

During its mechanism of action, mobile LiCl ions in the 3-D microporous network caused the artificial salivary film to exhibit moderate electrical conductivity, however, electrolytes adhered to the hydrophilic micropores for poor ion transport. When Yeom et al. introduced TA to the artificial tongue, mucin and TA complexed to form hydrophobic aggregates that enhanced ion transport throughout the hierarchical pore structure. This transition facilitated astringency perception via increased ionic conductivity. The team quantified the sensory performance by monitoring the relative changes of current under various concentrations of TA. The sensor had a wide sensing range and high sensitivity with many potential advantages in practice. To test the astringency of real beverages, the scientists used three different types of wine, including red, rosé and white, as well as black tea with different brewing times. As with TA before, they monitored the specific current changes to assess standard astringency, where red wine had the highest degree of astringency due to its concentration of tannins.



Design of the flexible artificial tongue. (A) The fabrication process of the flexible astringency sensor. (B) Schematic illustration of the working principle of the astringency sensor before TA treatment (left); a hydrophilic micropore in the hydrogel (center); magnified pore walls visualizing the electrostatic interaction–limited ion flow (right). (C) Schematic illustration of the working principle of astringency sensor after TA treatment (left); a hierarchical micro/nanopore in the hydrogel (center); magnified pore walls of hydrophobic nanopore visualizing enhanced ion flow (right). Credit: Science Advances, doi: 10.1126/sciadv.aba5785

The scientists then considered the stability of sensors for real-world applications. To prevent dehydration of saliva-like hydrogels that are primarily composed of water, they adopted LiCl on the artificial tongue as a conductive and hydrating agent. The artificial tongue showed stable sensing performances across a broad sensing temperature range due to its constituent mucin. While a human tongue can detect traces of a compound by licking it, artificial tongues have limited capacity to detect trace analytes. In contrast, the new astringency sensor developed here directly analyzed liquid analytes via a wipe-and-detect scheme in a flexible wiping process built in the sensor device. The team then tested unripe persimmon using the setup, a fruit that naturally contained a large amount of tannin to evoke astringency. When they attached the artificial tongue to the core of the persimmon, they detected relatively high astringency. Upon ripening the fruit, it displayed relatively low astringency. The new device detected varying degrees of astringency and can therefore be used as a portable taste mapping device based on electric changes within specific regions.



Applications of the artificial tongue. (A) Schematic illustration of wipe-and-detection of the human tongue and artificial tongue. (B) Astringency detection of an unripe persimmon: (i) photograph of unripe persimmon and (ii) current changes at different parts of the unripe persimmon. Photo credit: J.Y., Ulsan National Institute of Science and Technology. (C) Astringency detection of a ripe persimmon: (i) photograph of ripe persimmon and (ii) current changes at different parts of the ripe persimmon. Photo credit: J.Y., Ulsan National Institute of Science and Technology. (D) Scheme of the arrayed artificial tongue with five drops of 1 wt % TA and resulting taste mapping of the arrayed artificial tongue. (E) Scheme of the arrayed artificial tongue with 0.1 and 1 wt % of TA and corresponding taste mapping data; the size of sensing elements for taste mapping (D and E) is 6 × 10 mm for each pixel. Credit: Science Advances, doi: 10.1126/sciadv.aba5785

In this way, Jeonghee Yeom and colleagues developed an artificial tongue fully inspired by the human sensing mechanism. They prepared the experimental construct using UV polymerization on a flexible substrate to observe extraordinary sensing capabilities. The human tongue-like device had a wide sensing range and a low limit of detectable concentrations, as well as high selectivity from other specific tastes. The team exposed the device to astringent compounds and recorded its mechanism of action. They intend to further optimize the proteins constituting the artificial construct to improve its universal sensing capability. The outstanding results obtained for the artificial tongue sensor make it attractive for taste quantification or evaluation, to study taste disorders, and for integration within humanoid robots.

More information: Jeonghee Yeom et al. Soft and ion-conducting hydrogel artificial tongue for astringency perception, *Science Advances* (2020). DOI: 10.1126/sciadv.aba5785

Sae Ryun Ahn et al. Duplex Bioelectronic Tongue for Sensing Umami and Sweet Tastes Based on Human Taste Receptor Nanovesicles, *ACS Nano* (2016). DOI: 10.1021/acsnano.6b02547

Hyunwoo Yuk et al. Tough bonding of hydrogels to diverse non-porous surfaces, *Nature Materials* (2015). DOI: 10.1038/nmat4463