An artificial organic skin that can secrete liquid and wet its surface is demonstrated. The secretion is triggered by an alternating electric field at radio frequency. The polymer skin is constructed of a porous liquid crystal polymer network with the embedded dielectric liquid. The electric field accumulates the liquid in between the electrodes. By network contraction, the liquid is ejected at the surface of the polymer skin.

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HIGHLIGHTS
Liquid ejection by RF electric field
Release of functional liquid droplets
Spontaneous reabsorption of the secreted liquid at field-off state

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Demonstrate
Proof-of-concept of performance with intended application/response
SUMMARY
Living organisms enhance their survival rate by excreting fluids at their surface, but man-made materials can also benefit from liquid secretion from a solid surface. Known approaches to secrete a liquid from solids are limited to passive release driven by diffusion, surface tension, or pressure. Remotely triggered release would give active control over surface properties but is still exceptional. Here, we report on an artificial skin that secretes functional fluids by means of radiofrequency electrical signals driven by dielectric liquid transport in a (sub-)microporous smectic liquid crystal network. The smectic order of the polymer network and its director determine the flow direction and enhance fluid transport toward the surface at pre-set positions. The released fluid can be reabsorbed by the skin using capillary filling. The fluid-active skins open avenues for robotic handling of chemicals and medicines, controlling tribology and fluid-supported surface cleaning.

INTRODUCTION
A considerable part of recent “smart materials” research is dedicated to coupled sensing and actuation in response to changes in environment. However, to make materials really “smart,” we presently face the challenge of making materials exchange chemical information to humans, to devices, or between materials mutually. Besides, providing mechanisms to extend the lifetime of material and device, for instance by adjusting tribological properties, will also provide communication and feedback mechanisms in the context of robotics, virtual reality, and adjusted drug delivery. For example, taking mammal skin as inspiration, coatings that chemically alter their surface properties by secreting a chemical substance interacts with their environment by affecting adhesion, wettability, temperature, or touch experience. However, most approaches are limited to passive release whereas, inversely, administering fluids by triggered release in combination with refilling by absorption is still exceptional. Here, we introduce an artificial skin consisting of a (sub)microporous thin coating that can be filled with liquid that secretes upon actuation. In the case of the liquid being a chemical substance, interfacial chemical reactions can be initiated that are visualized by providing color to the reaction product.

The active excretion is commanded by a radio frequency (RF) alternating electric field. RF with the frequencies ranging from 20 kHz to 300 GHz, including radio waves and microwaves, is considered a low-energy and non-ionizing radiation. RF has branched out into the medical context of various modern electrotherapies and has recently been exploited in the aesthetic field, for instance, skin tightening and cellulite reduction. Despite its benign character and, therefore, extensive...

Progress and Potential
We developed an artificial skin that administers fluids by a triggered release. The principle is based on a radio frequency actuated liquid crystal polymer network that squeezes out the initially locked liquid from its nano-/microsized pores. We demonstrate that chemical reagents or medicine can also be released upon secretion of the fluids. The polymer skin can be refilled by absorption in a controlled manner. We anticipate this trigger-induced liquid secretion to be integrated into soft robotics and microfluidics, for instance to control chemical reactions, drug delivery, and to tune surface wettability and adhesion.
medical applications, RF’s potential in stimulating responsive organic and soft materials is still poorly understood and underestimated by materials scientists.

In a very recent study, we discovered that the RF electric field affects the material properties of highly ordered dielectric liquid crystal polymer networks (LCNs). Under the RF alternating-current (AC) field, the aligned polarizable molecules are subjected to a continuous oscillatory force. Although these affected moieties are embedded in the polymer network, and therefore very limited in their rotational response, the high-frequency deviation of the molecules from their initial positions creates dynamic molecular voids (free volume). This formation of molecular voids is in competition with free volume relaxation, which explains why it is only observed at the high frequencies where the voids form faster than refilling them by rearrangement of the polymer chains. Under the appropriate conditions it translates into macroscopic effects such as mechanical morphing of the coating surfaces.

RESULTS AND DISCUSSION

The polymer skin itself is solid and the liquid to be secreted is initially stored in micrometer-sized pores. The secretion concept based on field-stimulated lateral transport of the liquid component building up local pressure inside the porous polymer, which ultimately leads to excretion of the liquid at the polymer surface. To reallocate the liquid by an RF electric field, we employ the mechanism of dielectric diffusion, which describes the interaction between the electric field and a medium with inhomogeneous dielectric constant. Similar to the switching of liquid dielectric lenses, the dielectric force exerting on the liquid media can be expressed as 

$$ F = \varepsilon_0 \varepsilon \nabla (E \cdot E) $$

where $E$ is the electric field, $\varepsilon_0$ is the permittivity of free space, and $\varepsilon$ are the permittivity of the polymer network and the mobile liquid stored in the pores, respectively.

Based on this newly acquired knowledge, in this work we exploit the use of RF AC fields to alter the chemical properties at a coating surface through excreting and absorbing functional liquids. These functional artificial skins may administer medicine (e.g., for wound treatment), adjust tribology by controlled release of lubricants, or carefully dose chemicals for interfacial chemical reactions.

To initiate in-plane liquid displacements we use a lateral (in-plane) electric field generated by interdigitated electrodes (IDES) buried underneath the dielectric material, as shown in Figures 1A and 1B. We select the cyan group containing molecule 1 (Figure 1C) as the dielectric liquid, which couples strongly to the electric field because of large dielectric anisotropy. Naturally, without any additional anchoring force, this molecule tends to pack in a smectic homeotropic configuration. For reference, a liquid film of 4 μm is spin-coated on the glass substrate provided with IDE. The homeotropic alignment, with the molecular long axes on average perpendicular to the substrate, is confirmed by polarized optical microscopy (POM) showing that the film remains black between crossed polarizers irrespective of the sample orientation (Figure 1D and Video S1). We also characterize the surface of the liquid coating by digital holographic microscopy (DHM) (Figures 1E–1G and Video S2). Initially, the film exhibits a flat surface. Upon applying an RF AC field at 2 MHz, the flat liquid film morphs firstly into a wavy structure with elevations near the electrode edges as caused by dielectric diffusion, in which molecule 1 flows to the region that has the highest electric field intensity. In this case, the force on the liquid is now simplified as

$$ F = \varepsilon_0 \varepsilon_2 \nabla (E \cdot E) $$

This profile further develops into a symmetrical corrugation of 0.2 μm, which is centered between the electrodes, thereby

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minimizing its surface free energy. This process of fluid flow from flat to corrugated takes place within 80 ms. Under POM, we observe that the molecule 1 realigns to be parallel to the electric field line as caused by its positive dielectric anisotropy (Figure 1D). Between the crossed polarizers, the areas between electrodes appear as birefringent bright-blue lines as caused by the uniaxial planar orientation. On top of the electrodes, the lines remain dark, corresponding to a homeotropic alignment of the molecules. At the edges, the film lightens up because of the optical retardation change due to the alignment transition from planar to homeotropic, which occurs over a finite distance as determined by the elastic constants of molecule 1 as well as by the electric field lines. We conclude from these experiments that we can transport the anisotropic liquid 1 by RF AC field into the direction along the electric field lines and collect it in the area between the electrodes.

Next we perform the same experiment, but confine dielectric liquid 1 in the voids of the (sub-)microporous LCN that is applied as coating on the IDE substrate. LCNs are polymers that form by polymerizing aligned liquid crystal monomers, thereby

**Figure 1. Transport of Dielectric Liquid under an RF AC Field**

(A) Schematic representation of the interdigitated electrodes (IDEs).
(B) Representation of the in-plane electric field provided by the array of indium tin oxide IDEs at a glass substrate. Each electrode is 10 μm wide and separated by a gap of 10 μm.
(C) Materials used in the entire work. Dielectric liquid 1 was used as porogen and secreting liquid, diacrylate 2 and monoacrylate 3 were applied to form LCNs, photoinitiator 4.
(D) Optical microscopic images taken between crossed polarizers show the initial homeotropically aligned dielectric liquid 1 (left) and the realignment of the liquid when the RF AC field is switched on (right).
(E) Migration speed of molecule 1 measured at positions 1 and 2 given in (D).
(F and G) (F) 3D images of surface topographies measured by DHM show dielectric liquid transports to the gap area and (G) its corresponding 2D profile. The applied RF field is 62.0 Vrms at the frequency of 2 MHz.
See also Videos S1 and S2.
maintaining their molecular orientation. Recently, they have been explored for use as actuators and soft robotic applications because they change shape when triggered by light, temperature, and electricity. Here, we form a sponge-like LCN coating by photopolymerizing a smectic mixture of monomers 2 and 3 in the presence of the non-reactive LC 1 and photoinitiator 4. The composition (Experimental Procedures) is optimized on phase behavior in the monomeric state (Figure S1) and the mechanical and secretion properties (Figure S2). By means of an alignment layer coated on the substrate, the molecules are aligned perpendicular to the substrate surface, which means that the smectic layers are parallel to the surface. During the formation of the polymer network of monomers 2 and 3, the non-reactive component LC 1 phase separates to form (sub-)micrometer voids. The micropores are filled with pure molecule 1, as supported by the differential scanning calorimetry (DSC) measurement showing the phase transitions being identical to that of molecule 1 (Figure S3). We characterize the structural morphology of the porous LCN by grazing-incidence small-angle X-ray scattering (GISAXS) (Figures 2A and 2B). The diffraction pattern confirms the smectic order and the homeotropic alignment. The two-dimensional (2D) and one-dimensional (1D) profiles show the presence of three diffraction signals confirming the phase separation of LC 1 from the network. The peak at 2.1 nm⁻¹ is attributed to the bilayer smectic layer periodicity known for molecule 1, while the primary scattering peak (q*) is attributed to the polymer network. The secondary peak (2q) belongs to both network and molecule 1. To reveal the morphology of the polymer porous structures, we perform scanning electron microscopy (SEM) imaging on the cross-section of the coating (Figure 2D).
The sample was prepared by extracting 1 by immersing the sample in cyclohexane for 48 h. This reduces the thickness by 50% while the LC-1-filled pores of the LCN largely collapse. SEM images show the remaining nanometer-sized pores that are relatively homogenously distributed over the sample cross-section. Taking the volume reduction into account, we conclude that the diameter of the original pores is between 100 and 200 nm.

Figure 2C shows the photo of the device we made to study the RF-induced liquid secretion. A 4-μm-thick film is formed on top of the glass substrate provided with IDEs. The coating has an average surface modulus of 2 MPa as characterized by atomic force microscopy (AFM), as shown in Figure S4. DHM reveals that the initial coating surface has a flat profile with only a small surface relief of around 40 nm, corresponding to the underlying electrode pattern (Figure 3A). When switching on the RF field (62.0 Vrms, 2 MHz), with the fluid-filled coating on the electrodes, we made the following observations. Directly at the onset of the RF field, still prior to liquid secretion occurring, the thickness of the coating decreases at the position on top of the electrodes while it increases in between the gap. This can also be seen from the left side of the curves in Figure 3C showing a relatively small increase and decrease in the areas between and on top of the electrodes. We attribute this elastic deformation of the coating surface to accumulation of 1 in the gap area between the electrodes in correspondence with the dielectric diffusion observed for the unconfined liquid coating (supra vide). Then, after 7 s, 1 is released forming droplets in the gap area (Figure 3B and Video S3) and the height of the solid coating...
consequently decreases (Figure 3C). During this process the coating thickness on top of the electrodes also decreases further though without fluid release at that location, indicating a continuation of the dielectric diffusion of liquid 1 to the gap areas. Prior to the RF actuation LC 1 aligns homeotropically, parallel to the LCN matrix molecules. As soon as the liquid is released, however, it realigns along the electrical field lines resulting in birefringent dots shown in Figure 3D (Video S4). Simultaneously, a parallel experiment determining the contact angle of the water (Figure S5) reveals a slightly enhanced surface wettability.

The GISAXS measurements in Figure 2B demonstrate that under the RF field the diffraction peak that corresponds to the 1 dimer is largely reduced, while the peak corresponding to 1 immersed in the LCN network to some extent remains. Simultaneously, Fourier transform infrared spectroscopy (FTIR) measurements (Figure S6) reveal that under RF exposure there is still a considerable amount of 1 dissolved in the LCN. This leads to the model in which during the formation of the porous LCN coating two specific morphological region types are formed in the coating. The first one consists of the liquid crystal network containing a low concentration of molecule 1 dissolved on a molecular-length scale and corresponding to the 2q peak of Figure 2B. The second region is the voids exclusively filled with 1. Upon RF actuation, 1 excretes predominantly from the latter regions. After this partial removal of 1 the LCN coating, including the remaining 1, remains black between the crossed polarizers with only a minor color change to dark blue indicating that the remaining 1 in the network, although not covalently bonded in the network, does not realign under the influence of the field. This also implies that the order of LCNs has barely changed. This LCN order preservation is also recognized by in situ GISAXS measurement, as shown in Figures 2A and 2B.

Based on the observation that the location of secreting droplets is solely between the electrodes and the observation that molecule 1 in the absence of the LCN also diffuses to the center in between the electrodes, we postulate the mechanism whereby dielectric diffusion plays a dominant role. LC 1 in the network diffuses from the sides and from the area above the electrodes to the center between the electrodes, builds up pressure, and escapes. The pressure built up is confirmed by the initial surface deformation before the actual fluid release takes place. In this process the direction of the molecular orientation, and thereby the direction of the layers, also appears to be important. In a series of reference experiments, we examined the secretion efficiency and the dielectric diffusion of the coatings with various molecular alignment (Figure S7). The secretion efficiency is evaluated by the amount of the secreted liquid (Figure S8). In an isotropic coating prepared by polymerizing the mixture (Figure 1C) at elevated temperature in the isotropic state, no measurable amount of molecule 1 is excreted under the influence of RF electric field. However, dielectric diffusion still occurs, as measured by the increase in surface height in the gap area and the height decrease on top of the electrodes (Figure S7). This indicates that the molecular order of the LCN decisively contributes to the secretion but, additionally, the director orientation also appears to be important, as is illustrated in Figure S7. When comparing homeotropic smectic with planar aligned smectic films, only the homeotropic film shows secretion. Apparently, escape through the LCN surface is only made easy for the homeotropic films whereby the pathway of secreted liquid is parallel to the molecular orientation. This is also confirmed by a nematic equivalent that also shows surface secretion, though to a lesser extent than the smectic coating. We anticipate that the dipole alignment of the homeotropic order maximizes in-coupling of the RF energy in combination with easy escape pathways throughout the network. In situ POM and GISAXS experiments reveal that
RF-induced deflections from the common molecular axes are very small, as the order parameter remains constant within the accuracy of the experiments. We postulate that upon applying the RF electric field, molecules are subjected to an ultrafast oscillatory dielectric torque from the RF electric field, especially when molecules are perpendicular to the electric field that couples maximum torque. The GISAXS measurement, the minor shift of the LCN peaks ($q^*$ and $2q$), might point to this vibrational excitation.

By modulating the electric field strength and the frequency, we are able to adjust the amount of the excretion. Figure 4A shows that there is a threshold voltage $V_{th}$ of approximately $2.6 \text{ V} \mu\text{m}^{-1}$ above which excretion of molecule 1 takes place. Above $V_{th}$, the excretion increases with increasing voltage. This can be understood as increasing dielectric diffusion with larger electric field strength. The coating is also subjected to a frequency sweep (Figure 4B). We observe an increase in the quantity of the excreted liquid with increasing frequency. It is also known that an RF electric field generates heat. We monitored the temperature rise by employing an infrared (IR) camera (Figure 4C). To estimate the influence of the temperature increase, we actuate the coating by temperature alone in the absence of the electric field (Figure S9). The liquid excretion due to the thermal effect contributes approximately 40% of the excreted liquid (Figure 4D). The data points in Figure 4D relate to the frequency-dependent temperature as derived from Figure 4C.

An important feature is that, after excretion of the liquid, the coating can reabsorb the liquid again, as demonstrated in Figures 2A, 2B, and 3A–3D. When the secreted droplets are left on the surface directly after removing the RF field, the droplets reabsorb within several seconds. We have repeated this for several cycles, as shown in Figure 4.
Figure S10. This implies that, under the combined action of polymer elasticity and capillary forces, the coating can be “reloaded” with similar or different liquids. Thinking in terms of applications, this will lead to surfaces that on command can be “wet” or “dry,” for example, by varying the level of lubrication or surface adhesion.37–39

To demonstrate the administration of functional liquids, we have chosen a demonstrative acid-base interfacial reaction providing color upon release of an excreted substance. The process is shown in Figures 5A and 5B, Videos S5 and S6, and Figure S11. A pH-sensitive dye (4-(dimethylamino)azobenzene) (Figure 5C) is dissolved (1.5 wt %) in 1, and the combination is added to the coating. To avoid early release of the dye from the coating in the absence of electric field, we create the LCNs with a denser network at the surface. For this purpose, we include a UV absorber (Tinuvin 328, Figure S12) in the mixture to induce polymerization-induced diffusion of the network forming diacylate 2 to the surface driven by the light-intensity gradient along the coating thickness.40 Upon applying the RF electric field, the pH-sensitive dye is excreted from the coating entering into an acidic environment conducting the acid-base reaction. Following the Beer-Lambert law, we correlate the absorbance to the mass of the released dye over time by UV-visible spectroscopy (Figure 5D).41

Our coating is also capable of releasing medicines. As shown in Figures 5E and 5F, ibuprofen, a well-known painkiller,42 is loaded by carrier liquid molecule 1 in the coating and excreted from the coating when applying an RF AC field. We monitored
the releasing process by tracking the absorption of the protonated ibuprofen in an aqueous solution. We choose the solution with a pH condition (pH 7.4) similar to that of human blood. Another example is shown in Figure S13, demonstrating the release of cinnarizine, an antihistamine, by following of the UV-visible absorbance of an acidic contact liquid over time.

Conclusions
In conclusion, we used an RF alternating electric field to command liquid excretion on a porous polymer coating. In the field-off state, the liquid could be reabsorbed reversibly by network elasticity in conjunction with capillary forces. The porosity of the coating is controlled by the addition of a smectic porogen in the reactive mixture prior to its polymerization. We performed preliminary studies on releasing functional liquids such as medicine. Furthermore, we demonstrated that the coating interacts with the environment by performing chemical reactions at its surface. We anticipate that those reactions can also be carried out in the bulk of the coating, eventually, after (re-)absorption of a second reagent. Due to the robust nature of the polymer, its function is not limited as a stand-alone film but can be integrated into other devices as functional coating. This opens new avenues toward not only robotic handling of reactions, medicine release, and transfer of chemicals but also alteration of tribological properties of surfaces or removal of unwanted species (e.g., sand, ice, high-viscosity liquids).

EXPERIMENTAL PROCEDURES
Resource Availability
Lead Contact
Further information and requests for resources and reagents should be directed to and will be fulfilled by the Lead Contact, Danqing Liu (D.Liu1@tue.nl).

Materials Availability
This study did not generate new unique reagents.

Data and Code Availability
This study did not generate/analyze any datasets/code.

Materials
Molecules 1 and 3 were purchased at Synthon Chemicals. Molecule 2 was kindly provided by Philips Research Organics. Molecule 1 shows the following phase behavior: C, 21.5°C SmA 33.5°C N 40.5°C I. Molecule 4 was purchased from Ciba. Typically, thin polymer films were fabricated from a mixture containing 69 wt % molecule 1, 15 wt % monomer 2, 15 wt % monomer 3, and 1 wt % photoinitiator 4. Tinuvin 328 was purchased from Ciba. 4-(Dimethylamino)azobenzene (methyl yellow), ibuprofen, and cinnarizine were purchased from Sigma-Aldrich. Sulfuric acid (95 wt %, Sigma-Aldrich) was diluted with deionized water to an aqueous solution of 0.1 M. Glass substrates with interdigitated indium tin oxide electrodes were purchased from Suzhou Walthy.

Coating Formation
Coatings with smectic alignment were prepared by using the cell construction. To establish homeotropic alignment, we used the glass plate with pattern IDEs provided with a monolayer of 3-(trimethoxysilyl)propyl methacrylate to enhance the adhesion. For planar (parallel) alignment, glass plate with electrodes were coated with polyimide (Optimer AL1051, JSR), which was unidirectionally rubbed by a polyester fabric. In both cases, the cell is formed by attaching a second plain glass plate.
modified with release agent 1H,1H,2H,2H-perfluorodecyl-triethoxysilane. This construction results in the coating being attached to the side with patterned electrodes upon opening the cell. The coating thickness is controlled by the defined spacers. In this work, we choose spacers with 4 \( \mu \text{m} \) diameter. The liquid crystal monomer mixture was filled into the cell by capillary force at a temperature where the mixture is in the isotropic phase. Subsequently, it is slowly cooled to the smectic state at 28°C of the mixture (Figure S1), at which it is photopolymerized using a mercury lamp (Omnicure EXFO S2000).

A nematic coating with homeotropic alignment was prepared by spin-coating process. The substrate with IDEs was coated with polyimide, which provides homeotropic alignment (AL7511, Sunever; Nissan Chemical). The liquid crystal monomer mixture was spin-coated at 1,000 rpm (acceleration rate 300 rpm \( \text{s}^{-1} \)) from tetrahydrofuran (THF) (25 wt %) solution onto IDE substrate and subsequently polymerized by UV illumination at 45°C for 30 min in \( \text{N}_2 \) environment by using a mercury lamp (Omnicure EXFO S2000). The final coating has the thickness of 4 \( \mu \text{m} \).

A coating that releases an active component or drug was prepared from a mixture containing 2 wt % 4-(dimethylamino)azobenzene or ibuprofen, 67 wt % molecule 1, 14.5 wt % molecule 2, 14.5 wt % molecule 3, 1.5 wt % Tinuvin 328, and 1.0 wt % photoinitiator 4. The monomer mixture was spin-coated from THF solution (25 wt %) on the IDEs at the speed of 800 rpm (500 rpm/s acceleration). IDE substrate was coated with polyimide (SE-5661, Nissan Chemical), which provides homeotropic alignment. The final polymer coating is approximately 7 \( \mu \text{m} \) in height.

**Characterization**

Phase-transition temperature of anisotropic liquid 1, liquid crystal mixtures, and glass transition temperature (\( T_g \)) of liquid crystal network after removal of molecule 1 were measured by DSC (Q2000, TA Instruments) at a rate of 5°C/min and 20°C/min, respectively. For SEM, molecule 1 was removed by immersing the coating in cyclohexane for 48 h. The removal of molecule 1 is confirmed by FTIR with an attenuated total reflectance attachment (Varian 670-IR FTIR spectrometer). The alternating electric field with sinusoidal wave function was provided by a function generator (Tektronix AFG3252C). The electric signal from the function generator was amplified by a high-voltage linear amplifier (Falco Systems WMA-300). The output voltage was measured by an oscilloscope (InfiniiVision DSO-X 3032T, Keysight). Coatings thickness was measured by an interferometer (Fogale Nanotech Zoomsurf). Surface topography of the coating was measured by DHM (Lyncée Tec). Secretion behavior of the coating was investigated under a polarized optical microscope (Nikon Ci Eclipse) equipped with a hot stage (LTS420 system, Linkam). Temperature increase during actuation was monitored by infrared thermometers (Fluke Ti32 thermal imager, and Gobi-646-GigE, Xenics). GISAXS was performed on an instrument from Ganesha Lab. The flight tube and sample holder were all under vacuum in a single housing, with a Genix-Cu ultra-low divergence X-ray generator. The source produces X-rays with a wavelength (\( \lambda \)) of 0.154 nm and a flux of \( 1 \times 10^8 \text{ph s}^{-1} \). Scattered X-rays were captured on a two-dimensional Pilatus 300K detector with 487 \( \times \) 619-pixel resolution. The sample was placed at an angle (\( \theta \)) of 0.5°. The nanoscale elastic modulus was measured by Multimode AFM in the PeakForce Quantitative Nanomechanical Mapping (PF-QNM) mode (Bruker) using HQ: NSC19/No Al (MikroMasch) cantilevers. The DMT (Derjaguin, Muller, and Toporov) model of contact mechanics and the “relative modulus determination method” were employed to quantify the elastic modulus, referring to the polydimethylsiloxane calibration sample of 2 MPa (Bruker).
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AUTHOR CONTRIBUTIONS
D.J.B. and D.L. conceived the research and supervised the project. Y.Z. conducted experiments, analyzed the data, and drafted the manuscript. B.A.G.L. and M.M.R.M.H. performed the GISAXS experiments and analyzed the GISAXS data. F.L.L.V. performed data analysis for calculation for amount of secreted liquid. Y.Z., B.A.G.L., F.L.L.V., D.L., and D.J.B. contributed to the writing of manuscript. G.Z., D.L., and D.J.B. financially supported the project.

DECLARATION OF INTERESTS
The authors declare no competing interests.

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