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Memristive model of electro-osmosis in skin

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We show that some of the nonlinear conductance properties of electro-osmosis in sweat-duct capillaries may be modeled by a memristive circuit. This includes both the observed phase shift and amplitude modulation of the electrical current response to a simple harmonic driving potential. Memristive sytems may therefore be expected to play a role in modeling the electrical properties of skin, and perhaps also in other systems where nonlinearities are observed in their bioimpedance.

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I. INTRODUCTION

It is well known that noninvasive measurements of the electrical conductance of skin, obtained by placing flat electrodes on top of the epidermis, depend on the current flowing through the skin. This may, at least in part, be due to electro-osmotic transport of water through the sweat-duct capillaries [1]. Our purpose here is to reexamine this explanation by showing that electro-osmosis can be described using a *memristive* model, whose defining property is that it has memory of the current having passed through it, in which the impedance is not constant but is some nonlinear function of the current history.

The *memristor* is treated as an elementary passive circuit element, complementing resistors, coils, and capacitors (RLC), whose *memristance* $M(q) = d\varphi/dq = v/i$ is defined by how the magnetic flux $\varphi = \int v(t)dt$ responds to the charge $dq = i\,dt$ transported by the electrical current $i(t)$. Chua [2] first recognized the potential usefulness of employing a "memristive" circuit element almost four decades ago, but this passed largely unnoticed until a team from Hewlett-Packard realized that their nano-device could be modeled as an electronic memristor [3]. We want to show that the memristor concept may be useful in describing biophysical systems, and in particular that the electro-osmotic behavior observed in human skin can be thought of as a memristive system. Such memristive properties are now believed to be rather common in very small systems (cf. Ref. [3]). This includes the extremely tiny electronic components built from semiconductors, but also includes the scale where the collective degrees of freedom used in biophysics and biology start to emerge from the underlying molecular chemistry. It is believed [4,5] that the memristor is also capable of mimicking neural networks.

Memristive systems are generalizations of the memristor concept where the memristance is controlled by a number of state variables [6]. This study extends the preliminary results reported in an earlier study [7] to include two microscopic state variables, whose physical origin is explained and which explains further properties of electro-osmosis.

In dry skin, most pores and capillaries are empty or only partly filled, but conductive films are believed to be present also in the otherwise empty parts of these ducts [8,9]. With dry electrodes applied on the skin surface on the ventral forearm, an increase in conductance may occur if water is dragged by the applied electric field from deeper layers toward the surface where the electrode is located. The amount of filling in the capillary will be influenced by the electro-osmotic transport of water, as illustrated in Fig. 1.

When the mobile part of the conductive double layer is subjected to an externally applied electric field, the ions are set in motion and consequently drag along the fluid column in the filled part of the duct because of viscous forces. Consequently the volume V of water in the capillary changes at a rate (flux) dV/dt, since fluid is supplied or extracted at the bottom of the duct from or to the surrounding tissue. This flux is proportional to the cross-section area $A = \pi r^2$ of the capillary, the applied electric field E, the electro-kinetic potential ζ at the interface between the duct wall and the fluid, and the relative permittivity ϵ, and inversely proportional to the viscosity η of the fluid [10]. Lumping the systemic parameters into one with suitable dimensions for our problem, $\alpha = \zeta\epsilon/(4\pi\eta)$, we obtain Glasstone's fundamental equation

$$dV = \alpha A E\, dt. \quad (1)$$

Since $E(t) = v(t)/L$, with L the total length of the capillary, $v(t)$ the applied potential, and $\alpha < 0$ because $\zeta < 0$ in a sweat duct [11], we see that the volume of the water column increases when the electrode potential is negative. A rising column reduces the total resistance so we expect the current to increase in value.

A typical electrical current response measured on human skin using dry disk electrodes is shown in Fig. 2. As expected, the value of the current increases as the applied field changes from positive to negative polarity at the skin surface electrode because of the electro-osmotic transport of the filled part of the ducts. A negative potential attracts the water column, whereas a positive potential depresses it.

Furthermore, the current amplitude was found to increase from one cycle in the applied periodic voltage signal to the next, described as effects of "wetting" in Ref. [1]. This effect is thought of as a buildup of fluid in the duct over a series of current cycles. As the fluid column oscillates, it deposits more water on top of the thin film covering the wall (the duct wall is "wetted"), leading to reduced duct resistivity, and hence increased current. The height $w(t)$ of the water table will be a function of the amount of charge having passed through the duct; therefore we recognize this as a memristive system, i.e., a system with some "memory" of its electrical history.

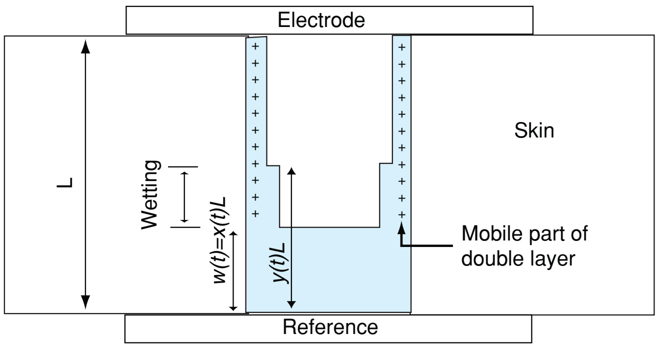

FIG. 1. (Color online) Schematic illustration of a sweat-duct capillary with wetting layers.

II. MEMRISTIVE MODELING OF ELECTRO-OSMOSIS

A partial explanation of these data was proposed in Grimnes [1] by relating the observed changes in skin conductance to fluid motion through electro-osmosis. An improved understanding, and a better quantitative analysis, may be obtained by using the concept of memristance. The memristance M defined below in Eq. (2) is physically distinct from ordinary bioelectric impedance since it has memory, and is therefore an example of the memristance of a memristive system defined by Chua [2,6]. More precisely, by deriving expressions for the charge function $q(t)$ and the corresponding current $i(t)$, we shall see that an electro-osmotic capillary behaves like a memristive system.

From Fig. 1 it is clear that an electrical current flowing through a capillary of total length L meets a total *instantaneous* (i.e., at any instant in time) resistance $M(t)$ composed of three resistances coupled in series,

$$M(t) = R[x(t),1] + R[y(t)-x(t),\tau_F] + R[1-y(t),\tau_f], \tag{2}$$

where $x(t)$ and $y(t)$ are state variables as presented in Fig. 1, satisfying $x \in [0,1]$ and $y \in [x(t),1]$ since $y(t) \geqslant x(t)$. Observe that not only are these "resistors" time dependent, their time evolutions are inextricably intertwined with each other.

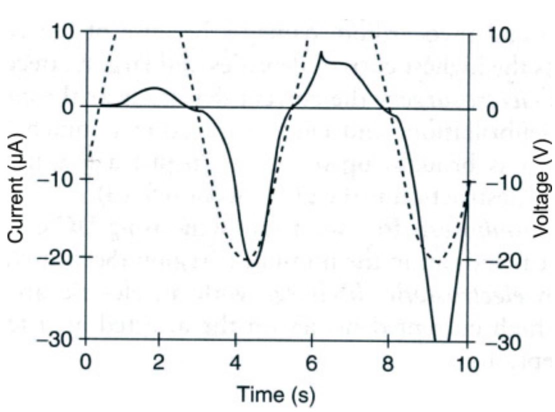

FIG. 2. Current response (solid curve) and applied voltage (dashed curve) on ventral forearm using dry disk electrodes (data from Ref. [1]).

The electrical resistance of each section of the capillary depends on the length l and thickness d of its wet volume. We normalize so that the (unobservable and microscopic) state variable is the longitudinal filling fraction $z = l/L$, while the transverse filling fraction $\tau(d)$ for a film of thickness d is the fraction of the total cross-sectional area of the capillary that is filled by the film,

$$\tau(d) = \frac{A(d)}{A(r)} = \frac{d}{r}\left(2 - \frac{d}{r}\right) \xrightarrow{d \ll r} \frac{2d}{r}. \tag{3}$$

Thus $\tau(d=r) = 1$ in the full part, while $\tau \ll 1$ for both the thin film (f) of thickness $d_f \ll r$ and the fat film (F) of thickness $d_F \ll r$. We then have

$$R(z,\tau) = \frac{z(t)}{\tau(d)} R_\bullet, \tag{4}$$

where $R_\bullet = \rho L/A$ is the minimum possible total resistance, which is obtained when the capillary is completely filled ($z = \tau = 1$) with the electrolyte (sweat), which has electrical resistivity ρ. The maximum possible total resistance $R_o = R_\bullet/\tau_f \gg R_\bullet$ is reached when the capillary is completely empty except for the thin film coating the capillary wall. The longitudinal fractional filling of water is called $x(t) = w(t)/L$, and $y(t)$ is the fractional height reached by the thick film (cf. Fig. 1).

The wetting problem is most naturally parametrized by

$$\delta = \frac{\tau_f}{\tau_F} \approx \frac{d_f}{d_F} \leqslant 1, \tag{5}$$

with $\delta = 1$ when there is no wetting, i.e., $d_F = d_f$. We can use δ to express the total resistance (actually, memristance) in terms of a single dimensionless function (measured in the natural unit R_o) as

$$M[R_o] = \frac{M}{R_o} = 1 - \delta x(t) + (\delta - 1)y(t). \tag{6}$$

Note that this memristance depends on time through the two microscopic and unobservable state variables $x(t)$ and $y(t)$, and in order to progress we must now deal with these. We first trade x for a measurable quantity (charge or current) by exploiting Glasstone's law.

During a small time interval dt, the volume of water in the capillary changes by $dV = Adw = ALdx = V_\bullet dx$, where $V_\bullet = AL$ is the total volume of the capillary.

Another expression for dV follows from Glasstone's law, given by Eq. (1), if we assume that all of the current $i(t) = dq/dt$ passes through the water column. The potential drop over this part of the capillary is $Ew = \rho wi/A$, which is used to eliminate the electric field strength from Glasstone's law, giving $dV = \alpha\rho dq$. Equating the two expressions for dV, we find the simple differential equation $dx = -c_1 dq$, where $c_1 = -\alpha\rho/V_\bullet > 0$ is a systemic constant.

This immediately integrates to $x(t) = x_0 - c_1 q(t)$, where x_0 is the initial position of the water table at $t = 0$. The sign of q is chosen so that a negative current gives the water table a positive velocity $dx/dt > 0$, as required by the experimental findings in Ref. [1]. Inserting this expression for $x(t)$ into Eq. (6) gives the memristance

$$M[R_o] = c_0(\delta) + \delta c_1 q(t), \tag{7}$$

where the new function

$$c_0(\delta) = 1 - \delta x_0 + (\delta - 1)y > 0 \tag{8}$$

at first sight appears to depend on a time-dependent function $y(t)$. However, this is a general expression for the memristance in the duct, valid with or without wetting, and it will turn out that $y = y_0$ is a constant in the former case.

In the latter case, $\delta = 1$ and y drops out. This describes the first half cycle after a negative driving potential $v(t) = -v_0 \sin(\omega t)$ $(v_0 > 0)$ is applied. This ensures that the water table that initially only is filling the lowermost parts of the sweat duct is attracted to the electrode. Electro-osmosis will, as long as the driving potential remains negative, drag the water table in the duct toward the electrode located at the skin surface [1]. When the driving potential after a time $T/2$ changes from negative to positive, the water table is at its maximum height $x(T/2) = y_0$, and the memristance is at its lowest value.

With a positive potential, the electro-osmotic force flips sign, pushing the water table back into the sweat duct and leaving behind a thickened film. This additional film is the wetting effect, and now the total memristance will consist of a series of three volume conductors: two films and the decreasing water table. The total memristance is now lower than it would have been without wetting. The current flow is therefore larger, pushing the water table deeper down in the duct, beyond its starting point x_0.

In other words, after a full period $(t = T)$, in which the first half is without wetting and the second half is with wetting, we find that $x(T) < x_0$.

If we now assume that further wetting does not take place, i.e., that the film can only have two thicknesses, then the subsequent periods are quite different since the half cycles are now symmetric. The water table will therefore return to the same maximum height y_0 at times $t_n = nT/2$ for each odd integer n, and to its minimum value $x(T)$ at times t_n for each even positive integer n. This heuristic description will be verified analytically below.

In this model we have ignored the so-called "clipping" effect, which is expected to happen when the driving potential is positive and the water table is repelled by the surface electrode. Then the film eventually loses contact with the electrode on top of the duct, yielding no current for a short period until the film diffuses back and remains in contact with the electrode, which again repels the film, etc. This repeats as long as the potential is positive and leads to a reduced current during this interval, which can be seen in the data reproduced in Fig. 2. There is therefore less current to drive the water table downward by electro-osmosis and the table does not reach back to its lowest position where it was one period earlier. With $x(t_n) > x_0$ (n odd), the water table will reach higher up in the duct at its next maximum, giving a higher value of the current due to an increase in the wetting effect for each period of the driving potential. This is also as reported in the original paper by Grimnes [1].

In summary, in our mathematical model, which includes wetting but not clipping, we need a negative initial potential in order to account for the observed increase in current amplitude with time. When clipping is present, the current amplitude will increase for each cycle as discussed above, and a positive initial potential could have been applied.

We now look in detail at the two basic intervals of interest, where we have either one-film or two-film memristance.

A. Memristance without wetting (thin film with constant thickness)

This is the first half-period $0 < t < T/2$, after the driving potential is switched on. The memristance during this time is simply

$$M(q) = R_o(c_0 + c_1 q), \tag{9}$$

with $c_0 = c_0(1) = 1 - x_0 > 0$.

By integrating Ohm's law, $v(t)dt = M(q)dq$, we find the charge as a function of magnetic flux $\varphi = \int v(s)ds$:

$$c_1 q(t) = -c_0 + \sqrt{c_0^2 + 2c_1\varphi(t)/R_o}. \tag{10}$$

As explained in Sec. I, we choose a driving potential $v(t) = -v_0 \sin(2\pi t/T)$ that starts out negative $(v_0 > 0)$, which gives the flux $\varphi(t) = -(v_0 T/\pi)\sin^2(\pi t/T)$. The flux-charge characteristic is shown in the inset in Fig. 3.

The current is given by $i = Gv$, where the *memductance* $G = 1/M$ is obtained from Eq. (9) by inserting our result from Eq. (10) for $q(t)$:

$$M = R_o\sqrt{c_0^2 + 2c_1\varphi(t)/R_o}. \tag{11}$$

B. Memristance with wetting (film with varying thickness)

As soon as $t > t_1 = T/2$, wetting starts to modify the total memristance of the system,

$$M(q)[R_o] = c_0(\delta) + \delta c_1 q, \tag{12}$$

where $\delta = d_f/d_F$ is the ratio of the thickness d_f of the thin film and the thickness d_F of the thick film left behind by wetting the thin film. We have no restrictions on δ in our model, apart from $\delta \leqslant 1$.

This M governs the current for all $t > t_1$, and the charge and current functions are obtained by the same argument used

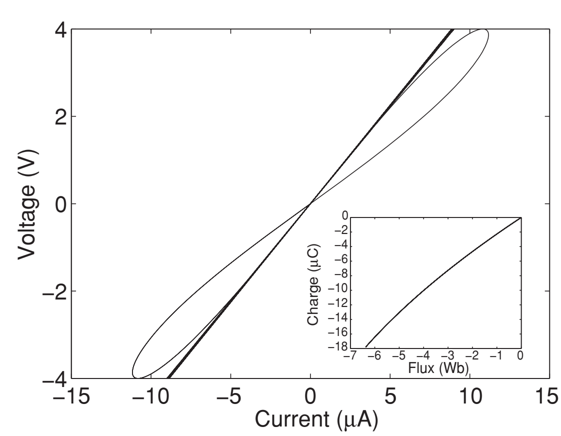

FIG. 3. Current-voltage characteristic of the memristive model deduced for skin sweat ducts, shown for $\omega = 2\pi/T = 0.2$ Hz and $\omega = 5$ Hz. Inset: Charge-flux characteristic of the memristor.

in the nonwetting case discussed above. We find the charge valid for $t > t_1 = T/2$,

$$\delta c_1\, q(t) = -c_0(\delta) + \sqrt{c_0(\delta)^2 + 2\delta c_1 \varphi(t)/R_o}, \qquad (13)$$

where the $c_0(\delta)$ term has been updated by the fractional height of the thick film, which is $y = x_{\max} = x(t_1) = x_0 - c_1 q(t_1)$, in order to include also the effects of wetting.

The current is given by $i = Gv$, where the memductance $G = 1/M$ is obtained from Eq. (12) by inserting our result from Eq. (13) for $q(t)$:

$$i(t) = v(t)/R_o \sqrt{c_0(\delta)^2 + 2\delta c_1 \varphi(t)/R_o}. \qquad (14)$$

It is now easy to verify that $q(t_3 = 3T/2) = q(t_1)$, from which it follows that $x(t)$ returns to the same value for all later times $t_k = (2k+1)T/2$, with $k = 0,1,2,\ldots$, as claimed above. The expression for $i(t)$ deduced from $M(q)$ is therefore valid for all $t > T/2$.

Figure 3 shows this current plotted against the potential $v = -v_0 \sin(\omega t)$, with $\omega = 2\pi/T$ and $\delta = 0.5$. For a suitable choice of parameters, hysteresis in the iv characteristic, which is the fingerprint of memristance, is in evidence at low frequencies, with curves running through the origin in double loops. At higher frequencies, the iv characteristic degenerates to a straight line, i.e., the memristive system developed in our model behaves like an ordinary resistor.

The inset in Fig. 3 shows the charge-flux characteristic with the harmonic driving potential, for which the flux is $\varphi(t) = -(2v_0/\omega)\sin^2(\omega t/2)$. Evidently the $q\varphi$ characteristic is single valued, as it must be for charge-controlled memristive systems [2].

A smaller δ will result in a reduced memristance M and an increased current $i(t)$. If $\delta = 1$, then there is only one single film and no wetting at all, meaning no increase in the current amplitude. Figure 4 illustrates how the current in the sweat duct given by our memristive model behaves for a suitable choice of system parameters, for some possible thicknesses of the wetting layer. The current response is similar to the behavior seen in the data displayed in Fig. 2. The current amplitude increases with wetting when the applied potential is negative, and the phase shift is positive so that the current lags behind the driving potential. The positive current amplitude is larger

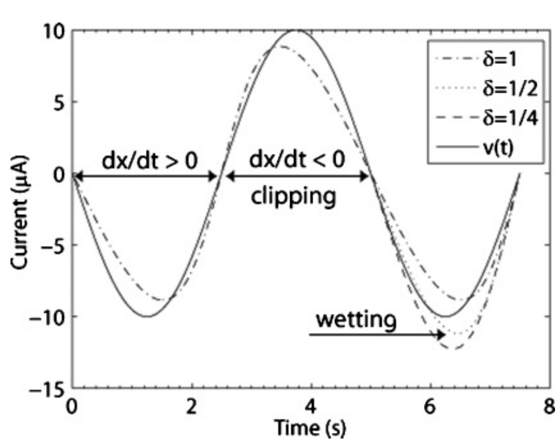

FIG. 4. Scaled applied voltage (solid curve) and current responses for different thicknesses of the wetting layer.

in our model than seen in Fig. 2 since the depressing clipping effect is not accounted for.

III. DISCUSSION

We have demonstrated that an improved understanding of skin conductance can be obtained by using the memristive paradigm defined by Chua [2], which goes beyond the traditional "resistive (RLC) framework." This shows that the memristor circuit component, as well as the more general memristive systems, may be suitable for modeling biophysical systems whose electrical parameters depend on their (electrical) history, in particular when hysteresis and phase shifts are present. The expression for the memristance of sweat-duct capillaries obtained in Eq. (7) clearly exhibits the explicit charge dependence of this "device," showing that it is memristive rather than resistive.

A closer examination of the memristance M is instructive. Resurrecting the dependence on systemic parameters, we can write Eq. (7) as

$$M[R_o] = c_0(\delta) - \frac{\alpha \delta R_\bullet}{L^2}\, q(t), \qquad (15)$$

which exhibits how M depends on the scale L of the device. It is clear that M will only deviate significantly from a resistance when L is small, as the charge dependence is suppressed by L^2. Since there are many biophysical systems, including skin capillaries, that are very small, we suggest that memristance may be useful for explaining a variety of nonlinearities seen in biophysics and bioelectricity—effects that in many cases are not yet fully understood.

The frequency band where memristive properties are seen will in general depend on the "speed" of the system, that is, how rapid the water table responds to changes in the driving potential. High frequencies will depress the memristive character of the system, since the liquid column has inertia and therefore needs time to respond to the applied field.

Figure 3 shows the typical feature of memristors when studied in the $q\phi$ plane, giving a single-valued and nonlinear characteristic. This agrees with Ref. [3], which also reported a nonlinear $q\phi$ characteristic in the semiconductor nanomemristor that they described.

The effect of "clipping" has been left out of our model due to its complexity, but it should probably be taken into account in order to explain the continued increase in current amplitude over several cycles as observed in Grimnes [1]. The net effect of clipping is a reduced average current, which is actually a highly jagged current smoothed out at the small frequency ($\omega = 0.2$ Hz) used in the experiment shown in Fig. 2. At higher frequencies, the resolution improves, and at $\omega = 500$ Hz, the current curve appears to be ragged around the peaks, but not elsewhere [1]. While his observation supports the clipping explanation, the inclusion of clipping in our model would increase its nonlinearity substantially, beyond our ability to analyze the model properly.

Figure 4 shows how the value of the wetting parameter $0 < \delta = d_f/d_F \leqslant 1$ determines the amount of current passing through the capillary duct. Since the thickness of the wetting film is unknown, it is hard to reach a definite conclusion about the importance of the wetting mechanism in real systems. A reasonable estimate is perhaps that wetting doubles the film thickness ($\delta \approx 0.5$), in which case it should not be neglected.

Another uncertain aspect of our model is that we assume that the wetting film remains more or less constant in height y during a cycle, i.e., that diffusion processes do not have sufficient time to smear out the film.

Our model does not so far include electrical activity in the epidermal stratum corneum itself, which usually acts in parallel to the sweat ducts [11]. Some of the activity in the stratum corneum is conventionally modeled using a theoretical device called "the constant phase element (CPE)." This is not a real device which can be built from a finite number of conventional (RLC) passive circuit elements [11,12]. It would be of great interest to study how the new memristive circuit element or memristive systems such as the memcapacitor, defined in [13], can be used in this type of more sophisticated modeling.

IV. CONCLUSION

We have shown that the memristor circuit element, first described theoretically by Chua [2], can be used in modeling electro-osmotic properties of capillaries in skin. The memristive paradigm should be useful in describing other nonlinear phenomena that appear at small scales. Such phenomena have frequently been observed in electronics, physics, and biological systems, but have often remained unexplained. Memristors clarify and enable circuit modeling of nonlinear systems, and therefore promise to be useful in circuit modeling of biophysical mechanisms, particularly bioelectrics.

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