Electronic Supplementary Material (ESI) for Soft Matter.

1. Detail derivation of the governing equations.

We use s^{-} to denote the nominal stress when the DE membrane is in its equilibrium state without the voltage and s^{+} as the nominal stress after the voltage applied. According to the first two assumptions,

$$s^{+} = s^{-} + s^{M}, \qquad \text{* MERGEFORMAT (1)}$$

where s^{M} is the Maxwell stress.

Following the ideal DE field theory[1], The electric displacement is defined as

$$D = \varepsilon E, \qquad \text{* MERGEFORMAT (2)}$$

where ε is the permittivity of DE, and is assumed to be constant during the deformation, E is the electric field given as,

$$E = \frac{\phi}{h}. \qquad \text{* MERGEFORMAT (3)}$$

Denoting H as the thickness in the reference state and considering the incompressibility of the material, h is obtained as

$$h = H / (\lambda_1 \lambda_2), \qquad \text{* MERGEFORMAT (4)}$$

where λ_1 and λ_2 are the stretch ratios along the longitudinal and latitudinal directions. The electric filed can be obtained by Eqs. * MERGEFORMAT (3) and * MERGEFORMAT (4) as

$$E = \lambda_1 \lambda_2 \frac{\phi}{H}. \qquad \text{* MERGEFORMAT (5)}$$

According to the former study[2], the Maxwell stress components caused by the electric filed, and defined as

$$s_1^{M} = \varepsilon E^2 / \lambda_1, \qquad \text{* MERGEFORMAT (8)}$$

$$s_2^M = \varepsilon E^2 / \lambda_2 .$$ * MERGEFORMAT (9)

Combining Eqs. * MERGEFORMAT (5), * MERGEFORMAT (8), and * MERGEFORMAT (9), we obtain

$$s_1{}^M = \frac{\varepsilon \lambda_1 \lambda_2{}^2 \phi^2}{H^2},$$ * MERGEFORMAT (10)

$$s_2{}^M = \frac{\varepsilon \lambda_1{}^2 \lambda_2 \phi^2}{H^2}.$$ * MERGEFORMAT (11)

To account for the stiffening effect of the DE membrane[2], the material is modeled using Gent model with the following free energy density,

$$W = -\frac{\mu}{2} J_{\lim} \ln(1 - \frac{\lambda_1{}^2 + \lambda_2{}^2 + \lambda_3{}^2 - 3}{J_{\lim}}),$$ * MERGEFORMAT (12)

where μ is the shear modulus, and $J_{\lim}$ is a constant that represents the stretch limit of the elastomer. When the DE membrane is in its equilibrium state without the voltage, the nominal stress components s_1^- and s_2^- can be obtained by

$$s_1^- = \frac{\partial W(\lambda_1, \lambda_2)}{\partial \lambda_1} = \frac{\mu(\lambda_1^2 - \lambda_3{}^2)}{\lambda_1 (1 - (\lambda_1{}^2 + \lambda_2{}^2 + \lambda_3{}^2 - 3) / \mathrm{J}_{\lim})},$$ * MERGEFORMAT (15)

$$s_2^- = \frac{\partial W(\lambda_1, \lambda_2)}{\partial \lambda_2} = \frac{\mu(\lambda_2^2 - \lambda_3{}^2)}{\lambda_2 (1 - (\lambda_1{}^2 + \lambda_2{}^2 + \lambda_3{}^2 - 3) / \mathrm{J}_{\lim})}.$$ * MERGEFORMAT (16)

Deformation theory for thin membrane structures under mechanical loading are well developed in the literature. Here we follow the work by Adkins and Rivlin[3] to derive the governing equations. We assume that the DE membrane is axisymmetric along the *z* direction as shown in the schematic in Fig. 5. The deformation of the DE membrane is represented by two functions $z(R)$ and $r(R)$, where z and r are the coordinates of a material point in the deformed state, and R is the coordinate of the same material point in the reference state. The stretch along the latitudinal direction is defined as $\lambda_2 = r / R$.

As demonstrated in Fig. 5, a material particle with radius form R to R+dR in the reference state, but with new radius form $r(R)$ to $r(R+\mathrm{d}R)$ and height form $z(R)$ to $z(R+\mathrm{d}R)$. We define the slope angle of the particle relative to the latitudinal direction as $\theta(R)$. The geometric relation yields the following two equations,

$$\frac{\mathrm{d}r}{\mathrm{d}R} = \lambda_1 \cos\theta \,, \qquad \text{* MERGEFORMAT (17)}$$

$$\frac{\mathrm{d}z}{\mathrm{d}R} = -\lambda_1 \sin\theta \,. \qquad \text{* MERGEFORMAT (18)}$$

Force balance along z direction (Fig. 5(c)) requires that

$$\frac{d}{dR}\left(HRs_1^{-} \sin\theta\right) = \lambda_1 \lambda_2 RP \cos\theta \,. \text{* MERGEFORMAT (19)}$$

Also, force balance along the direction normal to z direction (Fig. 5(d)) is enforced by the following equation

$$\frac{d}{dR}\left(HRs_1^{-} \cos\theta\right) + \lambda_1 \lambda_2 RP \sin\theta = s_2^{-} H \,. \text{* MERGEFORMAT (20)}$$

Eqs. * MERGEFORMAT (19) and * MERGEFORMAT (20) give the following two equations,

$$\frac{d\theta}{dR} = -\frac{s_2^{-}}{s_1^{-}} \frac{\sin\theta}{R} + \frac{\lambda_1 \lambda_2}{s_1^{-}} \frac{P}{H} \,, \qquad \text{* MERGEFORMAT (21)}$$

$$\frac{ds_1^{-}}{dR} = \frac{1}{R}(s_2^{-} \cos\theta - s_1^{-}) \,. \qquad \text{* MERGEFORMAT (22)}$$

Eqs. * MERGEFORMAT (17), * MERGEFORMAT (18), * MERGEFORMAT (21) and * MERGEFORMAT (22) are the governing equations that are solved for the four variables, $r(R)$, $z(R)$, $\theta(R)$ and $\lambda_1(R)$, which require suitable boundary conditions.

We set the origin of the coordinates at the apex of the inflated DE membrane,

$$z(0) = 0,\ r(0) = 0\ . \qquad \text{* MERGEFORMAT (23)}$$

To maintain axisymmetric deformation of the inflated membrane, the slope angle θ at the apex is fixed as

$$\theta(0) = 0\ . \qquad \text{* MERGEFORMAT (24)}$$

Finally, the radial stretch ratio at the circular edge of the DE membrane is assumed to be constant and equal to the prestrech ratio throughout the deformation $r(R_0) = R_C$.

The boundary-value problem is solved as an initial-value problem using the standard shooting technique. The initial values at the apex of the DE balloon for the four governing equations are $r(0) = 0$, $z(0) = 0$, $\theta(0) = 0$, $\lambda_1(0) = \lambda_{apx}$ and they are used for the numerical integration of the governing equations. The value λ_{apx} is the prestretch at the apex of the DE balloon and is obtained by satisfying the boundary condition $r(R_0) = R_C$.

Once the four governing equations are solved, the state of the DE membrane is determined.

1. Z. Suo, *Acta Mechanica Solida Sinica*, 2010, **23**, 549-578.
2. T. Li, C. Keplinger, R. Baumgartner, S. Bauer, W. Yang and Z. Suo, *Journal of the Mechanics and Physics of Solids*, 2013, **61**, 611-628.
3. J. E. Adkins and R. S. Rivlin, *Philosophical Transactions of the Royal Society of London. Series A, Mathematical and Physical Sciences*, 1955, **248**, 201-223.