Oscillating surfaces fueled by a continuous AC electric field

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Mechanically responsive materials have attracted recent attention, varying from shape-memory materials delivering a single-time deformation in response to a trigger (often temperature) to stimuli responsive materials that respond reversibly to a trigger (often temperature, light, or pH).[1–9] However, until present the fabrication of oscillating materials that dynamically alter their shape upon a continuous flow of energy or “chemical fuels” is still in infancy. These materials could lead to functions such as peristaltic gas pumping through membranes or controlled movement of molecules or particles at surfaces. Only a very limited number of such materials is known, which shows their great potential on one hand, but also shows the challenges that have to be overcome to bring them to the prospective level of sophistication.[10,11]

So far, the most successful example for autonomous movement of materials is based on the Belousov–Zhabotinsky (BZ) reaction generating chemical oscillations used in gels.[10,12,13] The first biomimetic gel with an autonomous self-oscillating function, resembling heart muscles, was developed by Yoshida et al. in 1996. The self-oscillating polymer is composed of a poly(N-isopropylacrylamide) network in which the catalyst for the BZ reaction is covalently immobilized.[14,15] The other example known in literature is an oscillating beam of a laser-powered azobenzene-modified liquid crystal network (LCN).[16] The mechanical coupling of momentum and directional addressing leads to a fast oscillation. Clamped at one side, the beam oscillates with its eigenfrequency between 5 and 100 Hz depending on its dimensions, normally a few millimeters. If progress is to be made, new approaches are needed in this challenging field of materials science.

Here, we propose an approach to generate an oscillating wave in a soft polymer network under a continuous AC electric field. The surface motilities demonstrated open options to new applications, such as self-cleaning, adjustable tribology, deceiving, haptics, and mobile transport of species. To further support our understanding of the actuation mechanisms, a finite element model is employed to analyze the autonomous surface topographic changes of the elastic dielectric coating, which shows good qualitative agreement with our experiments.

Surface deformation is produced by a periodic electrostatic attraction based on Maxwell stresses. Maxwell stresses are well-known and extensively studied and applied for actuation of electroactive polymers.[17–24] Generally, a two-plate capacitor configuration is established where a thin elastomeric film is coated at both sides with flexible electrodes. By applying an electric field, the electrostatic attraction between the electrodes causes the polymer film to contract in thickness while expanding in the
lateral direction.[23,24] We expanded on this concept and trigger the Maxwell stresses locally on a thin crosslinked film firmly adhering to a rigid substrate. The device configuration is shown in Figure 1a. When the coating is compressed, the global in-plane displacement is prohibited by the rigid substrate. However, locally we create lateral escape routes of the compressed excess of volume by adjacent expanding areas as schematically illustrated in Figure 1c. This is achieved by employing patterned bottom electrodes (E1) made by conventional lithographic procedures. On top of E1, we created a coating consisting of an ultra-soft siloxane polymer network (Note S1, Supporting Information) provided with an 11 nm silicon oxide top film (Note S2, Supporting Information). This silicon oxide top is obtained by an UV/ozone treatment of the siloxane network.[25,26] On top of that a compliant second electrode (E0) is directly coated. Details of the device dimensions and the process are provided in the Experimental Section. The combination of the gold top electrode and the silicon oxide provides the robustness needed but is still flexible enough to deform together with the polymer coating. To initiate surface waves, E0 and E1 are supplied with electric field with opposite polarity (Figure 1c), typically the voltage is 150 V. We carried out a finite element simulation (Note S3, Supporting Information) to analyze the electric field distribution over the polymer coating. The result is given in Figure 1b suggesting the electric potential is built across the coating thickness in between E0 and E1. We demonstrated the proposed actuation principle by monitoring the time-resolved electro-mechanic response of the coating by digital holographic microscopy (DHM) at two adjacent locations. The results are given in Figure 1d. Location 1 is activated by the Maxwell stress which leads to a negative surface deformation. Location 2 is inactive and directly connects to location 1. Although location 2 was not subjected to any measurable electric field, we measured a surface expansion that is of the same order as observed for the contraction caused by the mechanical stress–strain from the contraction at the connecting areas (location 1). Keeping E1 and E0 at the same polarity of +150 V (Figure 1e), there is no potential established suggesting a zero electrostatic potential.
compression. The kinetic measurements as carried out under isothermal conditions reveal that there is no electrostatic repulsion to expand the coating (Note S4 and Figure S3, Supporting Information). Subsequently, by reversing the polarity of E0, we create a coating whose surface transfers from flat to the corrugated wave, Figure 1f.

Building further on this idea, surface waves are brought into oscillation under a continuous AC electric field. The principle of generating oscillating standing waves is based on spatially periodic and continuous polarity inversion in the elastic dielectric coating. Based on the device configuration given in Figure 1a, we create two patterned indium tin oxide (ITO) interdigitated bottom electrodes (E1, E2) as seen in Figure 2a. During activation, the tri-electrode structures are simultaneously provided with electric signal. The polymer film functions as two individual capacitors provided by a continuous electrode and two discrete electrodes, the corresponding equivalent circuit is given in Figure 2b. The patterned electrodes (E1, E2) are supplied with a direct electric field (DC) with opposite polarity. An alternating electric field (AC) is applied on the continuous top electrode (E0) and thus E0 repeatedly inverts its polarity. The simulated electric field distribution over the polymer coating is given in Figure 2c. It shows a snapshot of the field distribution at the moment when electrode E1 and E0 have opposite charges while E2 and E0 have equal charge. A potential is thus built between E1 and E0, which is indicated as $\Delta V_1$ in Figure 2b, while between E2 and E0 the potential is zero ($\Delta V_2$). At a consequence, the Maxwell's attraction compresses polymer film between E1 and E0 via $\Delta V_1$, simultaneously, because of the conservation of the volume the adjacent area (between E2 and E0) expands. As a result a sinusoidal wave appears at the coating surface. This is accompanied by a strain in the silicon oxide-gold top layer as the surface area continuously expands and relaxes. By flipping the polarity of E0, the wave reverses its peaks and valleys (Figure 2e). The oscillation frequency is thus governed by the electric signal applied to E0.

**Figure 2.** Design principle of an oscillating surface wave in an elastomer with time, fueled by an electric field. a) Schematic representation of the device configuration. b) Equivalent electric circuit of the tri-electrode driving scheme. The activated polymer works as a double capacitor when placed in the external electric field. c) Snapshots of the simulated distribution of electric field across the polymer film when all three electrodes are connected to the power source and an internal potential is acting between E0–E1 and E0–E2 alternately. d) Illustration of initial surface and e) the actuation principle based on the inversion of polarity in the dielectric elastomeric coating.
The oscillating standing wave is characterized by DHM. The profile of the wave, as well as its corresponding oscillation kinetics, is extracted from the measurement. Prior to actuation, the coating surface is flat and smooth and does not change with time (Figure 3a,c (black curve)). Upon applying the electric field, the surface corrugates into a sinusoidal wave and oscillates temporally following the alternating electric field (Figure 3b,c). Typically, both DC and AC power sources supply 150 V to generate the electric field. The peak-to-peak voltage value of the AC field is measured and is further denoted as $V_{pp}$.

Under this applied potential, the wave exhibits a peak-to-valley height difference of 150 nm on top of an 8 µm coating. The dynamics of the coating are shown in Movie S1 in the Supporting Information.

Taking a closer look at the oscillating wave, we discovered a deformation asymmetry when the polarity of the $E_0$ switches from positive to negative. To be more specific, the height difference between point 1 and 2 is 30% larger when $E_0$ is positive than when $E_0$ is negative. To solve the origin of this height difference, we investigate the influence of the in-plane electric field on the deformation profile. By applying a DC electric field across electrodes $E_1$ and $E_2$, we observe a small surface relief of 50 nm in which the material above the positive electrode 1) forms a peak while at the location above negative electrode 2) the material forms the valley (Figure 3d,e, Note S5, Supporting Information). We attribute this phenomenon to the ionic impurity possibly from the platinum catalyst in the curing agent of the polysiloxane, in which the negatively charged material accumulates at the positive electrode. This is further verified by a measured in-plane current of 315 nA (Note S5 and Figure S4c, Supporting Information).

To gain more insight into the deformation mechanics, we modeled the field-induced deformation. For this study, we used the Marc Mentat nonlinear finite element analysis with a coupled electrostatic-structural analysis (Note S6, Supporting Information).

Figure 4a,b illustrates the results of the modeling of an actuation following the driving scheme of Figure 2b for a frequency of 1 Hz (Movie S2, Supporting Information). The amplitude values of the surface wave is measured and modeled as a function of the applied electric field strength, shown in Figure 2c. This response can also be intuitively understood in terms of the larger Maxwell force exerted on the coating with increasing field strength. In the first approximation, this observation can be qualitatively analyzed by Equation (1)

$$\delta = \frac{\varepsilon_0 \varepsilon_r E}{E} \left( \frac{V}{d} \right)^2$$

where $\delta$ is the indentation depth, $\varepsilon_0$ is the vacuum permittivity, $\varepsilon_r$ is the relative permittivity, $E$ is the elastic modulus, $d$ is the thickness of the coating, and $V$ is the applied potential. The indentation ($\delta$) forced by Maxwell stresses leads to
an expansion in the adjacent area due to volume conservation forming a protrusion. The deformation amplitude is then calculated as the height difference between the valley and the peak and scales with the quadratic of the applied voltage which is in agreement with the measurement and the modeled data of Figure 4c. The mechanical properties of the siloxane network...
also influence the deformation, as presented in Figure 4d. The deformation scales inversely with the modulus. At a modulus above 1 MPa, barely any deformation occurs. This observation is explained as higher modulus prohibits lateral displacement (Note S7, Supporting Information). Details of the modulus as a function of composition and presence of silicon oxide top layer measured by atomic force microscopy (AFM) is shown in Note S1 and Figure S1 in the Supporting Information.

Next, we investigate the oscillating behavior of the standing wave in terms of the viscoelastic behavior of the soft siloxane network. We randomly selected two adjacent areas above the electrodes as indicated in Figure 2e and traced their relative height changes under the continuous electric field input in time. The results are shown in Figure 4e. The polarity of E0 reverses every 2 min, as indicated by vertical red lines. In response to this change, the surface of the coating inverts its original topography. The inversion initiates with a rapid elastic-like deformation, followed by a gradual change as controlled by viscoelastic properties. A similarly shaped response curve is measured at a frequency of the oscillation to 1 Hz, as demonstrated in Figure 4f. In order to simulate the viscous response, we performed viscoelastic measurements using a cone-plate rheometer. Details are provided in Note S8 in the Supporting Information. The values of the frequency-dependent storage ($G'$) and loss ($G''$) moduli are shown in Figure 4h, which show the presence of a strong viscous component (Note S9, Supporting Information). With the input of the viscoelastic data into the finite element model, we obtain a reasonably good agreement between experiment and simulation as is shown in Figure 4f. Despite the viscous component, the maximum protrusion height is obtained within 100 ms by switching the polarity of E0 corresponding to a vertical displacement speed of 5 µm s$^{-1}$. In comparison, the surface deformation of coatings in other systems generally require 10 s or more.[27–29]

We then monitor the deformation amplitude during a frequency sweep. The results are shown in Figure 4g. Both simulation and experimental results indicate that at the frequency above 5 Hz, the deformation amplitude decreases rapidly. The oscillatory mechanical frequency sweep in Figure 4h reveals that $\tan(\delta) > 1$ at a frequency of 5 Hz. Above this frequency, the value of the loss modulus exceeds the storage modulus indicating that the energy dissipation by the viscous component of the material starts to dominate. As a result, the strain can no longer keep up with the applied stresses.

The energy consumption of the surface dynamics is determined by measuring the in-plane and out-of-plane current values (Figure S4c, Supporting Information). With a potential difference of 150 V, the in-plane current is 315 nA while the value of the out-of-plane current is only 12 nA. Considering also the surface area of our sample (1 cm$^2$), a remarkably low energy consumption of 168 mW m$^{-2}$ is required to bring the surface to a mechanical oscillation. In comparison, electrically responsive LCN surface topographies require about 90 W m$^{-2}$ and UV responsive LCN coatings 2–7 kW m$^{-2}$, more than five orders of magnitude higher.[27,29,30]

In conclusion, we describe a method to generate oscillation in complex motion figures such as waves in a thin solid coating triggered by a continuous AC field. In this new approach, surface deformation is achieved by localized periodic electrostatic attraction based on Maxwell stresses. Our innovation is that a tri-electrode configuration is designed to initiate the oscillation. A finite element model is developed to predict the profile and kinetics of the wave as a function of the viscoelastic properties of the soft siloxane network, the applied field strength, and frequency. We demonstrate that the waves can be brought into motion at a frequency up to 5 Hz. This value is significantly larger than that of, for instance, LCN coatings which is generally in the range of 0.1 Hz.[27–29] We envision that this newly proposed fast oscillating wave can be used for haptic applications, such as human–machine interface, refreshable braille display as well as used to interact with the environment, for instance, transporting materials and particles, mechanically de-icing through surface vibrations, and regulating light. For many of the practical applications, we need to optimize the coating further, e.g., isolating the top electrode by a flexible top elastic coating, balancing the viscoelastic component in the siloxane for a preferred deformation amplitude and kinetics. Moreover, it is anticipated the motion figure can be further tuned by introducing complex electrodes patterns.

**Experimental Section**

**Materials**: Interdigitated ITO electrodes were provided by Merck. The poly(dimethyl siloxane) (PDMS) silicone elastomer and curing agent (5Silgard 184) were obtained from Dow Corning.

**Sample Preparation**: The PDMS curing agent was added to the silicone elastomer in a concentration of 2 wt% to obtain a soft and flexible network. After thorough mixing of the two components, the trapped air in the sample was removed under reduced pressure. The substrates with the interdigitated ITO electrodes ($10 \times 10 \mu m$) were cleaned by ultrasonication for 20 min in acetone and isopropanol, respectively, and dried with air flow. Subsequently, the clean substrates were subjected to an UV-ozone treatment for 20 min. The PDMS mixture was applied on the substrates by spin coating at 7000 rpm (acceleration rate 1000 rpm s$^{-2}$) for 2 min, which resulted in a thickness of 8 µm. The resulting layer of PDMS was cured overnight at 70 °C. After subjecting the PDMS surface to a 20 min UV-ozone treatment, a thin layer of gold (10 nm) was sputter coated on top of the PDMS layer at a current of 65 mA for 11 s.

**Device Configuration**: The sample made by the above described methods is shown in Figure 5. The electric activated area had the dimension of 1 cm $\times$ 1 cm.

**Characterization**: The alternating electric field with a square pulse function was provided by a function generator (Tektronix AFG3252).

![Figure 5. The photo of a sample. E0 is the continuous gold electrode. E1 is interdigitated ITO electrodes on a glass substrate. PDMS actuating coating is in between the bottom and top electrodes. Two wires are attached to the contact pad of the interdigitated electrode and gold electrode, respectively, to connect to the power source.](image-url)
The electric signal from the function generator was then amplified with an amplifier (Falco Systems WMA-300). The in-plane potential difference was generated by a DC Power Supply (3B Scientific U33000). The output voltage and current were measured with an oscilloscope (Keysight InfiniiVision DSO-X 3032T). The surface topographies were measured with a Digital Holography Microscope (Lynée Tec.). During the experimental period, no obvious fatigue was observed in the sample. The thickness of samples was measured by an interferometer (Fogale Nanotech Zoomsurf). The mechanical properties of PDMS were measured with an oscillatory frequency sweep by a rheometer (AR-G2, TA instruments) with a strain of 6%. AFM measurements were performed using a Multimode 8 AFM with a NanoScope V controller (Bruker) in the PeakForce Quantitative Nanomechanical Mapping mode. The electric field–induced surface dynamics of the coating were simulated using COMSOL Multiphysics (Bruker) in the PeakForce Quantitative Nanomechanical Mapping mode. The electric field distribution in a 2D cross-section of the coating was simulated using COMSOL Multiphysics 5.2. The electric field–induced surface dynamics of the coating were simulated in 3D using Marc Mentat 2014.0.0.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

AC electric fields, dielectric elastomers, dynamic surface topographies, oscillating waves, tri-electrode configurations

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