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Use of Field Concentration for Electroluminescent Devices

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Field concentration is often regarded as a problematic issue in soft electronics applications, especially when using curved electrodes, and in particular, those with sharp edges. However, field concentration can be turned into an advantage with appropriate device design. Herein, the applications of field concentration in hydrogel-elastomer devices are explored. Three different types of electroluminescent hydrogel-elastomer devices are fabricated using different types of electrodes and different light patterns. In addition, the effect of the field concentration can be extended into the bulk of the elastomer by preparing porous silicone elastomers and filling them with silicone oil. These devices are shown to be flexible and possess both good luminance and a long lifetime.

1. Introduction

As wearable technologies and hand-held devices gain more and more importance in our daily lives, the demand for cheap and reliable technologies enabling soft, flexible screen displays is increasing. This has led to increased research in stretchable electronics, with a focus on innovative soft, stretchable luminescent devices that can be used for the next generation of wearable electronics.[1,2] and soft screens.[3,4] The recent progress in super-stretchable elastomers is also believed to further new applications for unprecedented flexibility devices.[5–7]

Light sources, such as light-emitting diodes (LEDs) and organic light-emitting diodes (OLEDs), are commonly integrated into wearables and softscreen devices. However, the preparation processes for both LEDs and OLEDs are complex, requiring vapor deposition of metals and/or a clean room environment. As an alternative, electroluminescent (EL) technologies can be integrated in a facile manner into alternating current (AC)-powered soft electronic devices.[2] Compared to LEDs and OLEDs, alternating current electroluminescent (ACEL) device technology is still considered particularly promising for flat and flexible large-area light sources.[8]

In EL devices, light emission results from phosphorescent materials in an alternating electric field. Zinc sulfide (ZnS) is commonly used as an electroluminescent phosphor that emits light under an alternating electric field. When a high electric field is applied, electrons and holes are generated and recombined in the phosphor to produce visible light when the electric field is reversed.[9–11] The emitting color of a given EL device depends on type and concentration of the co-dopant. Copper (Cu) doping in ZnS results in a blue–green light, manganese (Mn) doping in a red light, and aluminum (Al) doping in a blue light.[2,9,10,12–15] Copper-doped zinc sulfide (ZnS:Cu) has been studied extensively and its working principle is thoroughly understood, making it an excellent candidate for use in ACEL devices.[15] However, the use of dopants produces weak spots in the luminescent powder, as some of the doping atoms and the ZnS itself can be oxidized or start hydrolyzing over time, causing the ACEL device to degrade. Despite these weak spots, ACELs are still less susceptible to degradation than LEDs in the presence of oxygen and water.[14] Moreover, to overcome this issue, phosphor powders are imbedded in the elastomer layer in most ACEL devices via mixing with elastomer, thereby reducing the water contact.[10] Also, field concentration damages may occur because of the deformation of these flexible materials.[16,17]

The ACEL devices' high operating voltage and high frequency are the main obstacles to their use as wearable EL devices. It has previously been reported that, with an applied electric field higher than 5 V μm^{-1} and a frequency higher than 1 kHz, brightness with luminance above 100 cd m^{-2} can be achieved.[10] The luminance (L) of an ACEL device can be empirically correlated to the applied voltage (V) using equation 1:[9,10,12,15]

$$L = L_0 \exp\left(\frac{-b}{\sqrt{V}}\right) \tag{1}$$

where the parameters L_0 and b depend on the particle size of the phosphor, the concentration of the phosphor in the dielectric,

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the dielectric constant of the embedding medium and the device thickness. From this equation, it is obvious that luminance increases with higher voltage and materials characteristics.

Overall, ACEL devices possess good brightness, high resolution, uniform light emission and low power consumption compared to other EL devices.[8] In addition, the soft polymer substrate used in most ACEL devices makes it possible to design them to be extremely flexible, allowing them to be twisted, bent, and rolled.[14,15]

EL device preparation is quite simple. Initially, EL devices were prepared as flat plate capacitors by sandwiching the dielectric layer with ZnS particles between two electrodes.[9] While ACEL devices result from a variety of fabrication methods, the overall concept still has not changed drastically. Dielectric elastomer actuators (DEAs) have been reported to be ideal for combination with EL devices to provide flexibility and stretchability.[4,15] EL devices can also be made by placing the luminescent material between two dielectric layers and then further sandwiching the structure between electrodes.[1,12,14] Another approach is to use a multistep dip-coating method that results in EL fibers.[18] Finally, printed EL devices have also been reported.[2]

Regardless of the preparation process used, material choice is crucially important when making EL devices. Since ACEL devices are prepared with an EL layer in the middle, the electrode placed on top of that layer needs to be transparent. Unsurprisingly, the development of the transparent electrode material tin oxide (SnO_2) led to a growth in ACEL device research.[9] However, because the brittle nature of these electrodes hindered stretchability, the introduction of stretchable transparent electrodes, such as graphene electrodes.[8] and ionic conductors (hydrogels)[19] was necessary to further advance EL device technology. The resulting soft electroluminescent devices have been shown to possess great versatility:, e.g., as displays, sensing or lighting devices, soft electronic devices, and soft screen devices.[2,8–11,14,15,20]

In this work, ACEL devices are constructed using a silicone elastomer as well as hydrogel and copper electrodes. The silicone elastomer, Sylgard 184, is transparent across the entire visible spectrum (360-780 nm), with a transparency up to 94%, and remains transparent when stretched up to 50% strain.[21] Sylgard 184 can be mechanically extended to a strain of 140%. The hydrogel, which consists of water and polymer chains, is stretchable up to 2000% strain[3,12,22] and it is 99% transparent.[20,23,24] These properties allow any desired device shape and/or size to be achieved while also eliminating any transparency issues.

While it was reported previously.[17] that the development of field concentration is an inherent problem when preparing soft devices, the soft electroluminescent devices fabricated here demonstrate that field concentration can in fact be used constructively. We previously tested DEAs made of a hydrogel and an elastomer.[17] and found that such hydrogel-elastomer devices possess short lifetimes due to the large electric field concentration around the electrode edges. In this work, field concentration is used to prepare three new electroluminescent hydrogel elastomer devices using different types of electrodes (hydrogel electrodes, and a combination of hydrogel and copper electrodes) to create different electrode patterns. Lifetime tests were performed on each device while applying defined voltage at a frequency of 1 kHz for 90 min. The influence of voltage and frequency on the devices' luminance and color was tested, and the luminance of each device was reported. Finally, we showed how field concentration can be extended to light up the bulk material.

2. Experimental Section

Soft EL devices were made using commercial silicone rubber (Sylgard 184, Dow Corning) as the dielectric. To prepare the polyacrylamide (PAAm) hydrogel electrodes, acrylamide (AAm, Sigma–Aldrich), cross-linking agent N,N' methylenebisacrylamide (MBAA, Sigma–Aldrich), photoinitiator α ketoglutamate acid (α-keto, Sigma–Aldrich), and lithium chloride (LiCl, Sigma–Aldrich (no. 746 460 worked well here)) were used. 500 µm thick hydrogel sheets were prepared as previously described[17] In the wire devices, hydrogel electrodes were coupled with copper wires (ø500 µm, McMaster-Carr); in the silicone oil lamps,

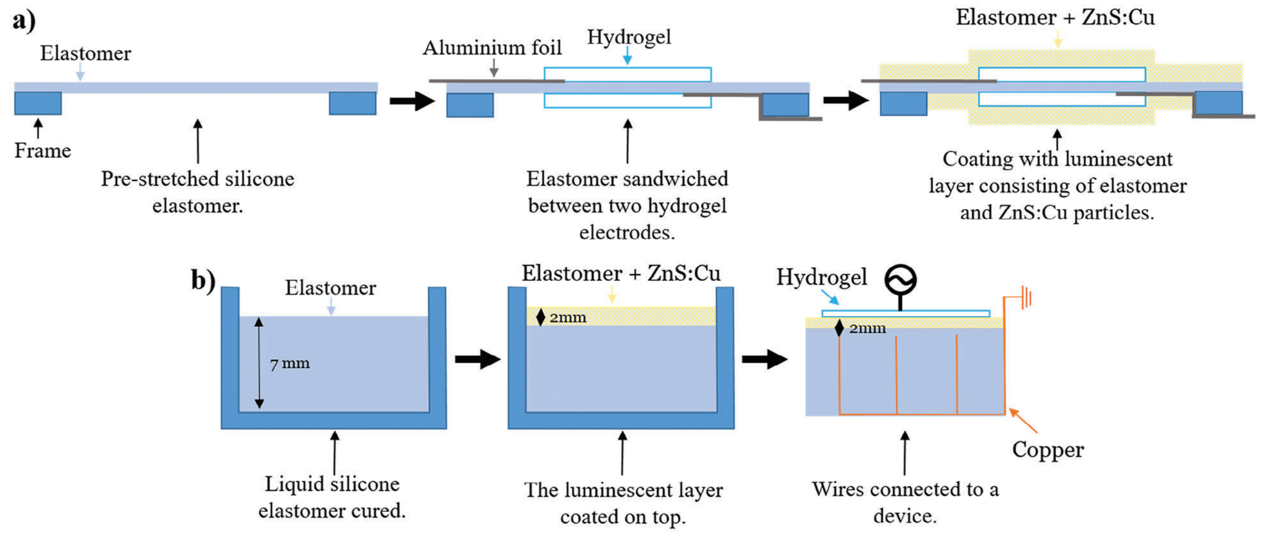

Figure 1. Schematic of ACEL device preparation processes: a) preparation process for hydrogel device; b) preparation process for wire device, in which electrodes are inserted into the previously cured silicone layer.

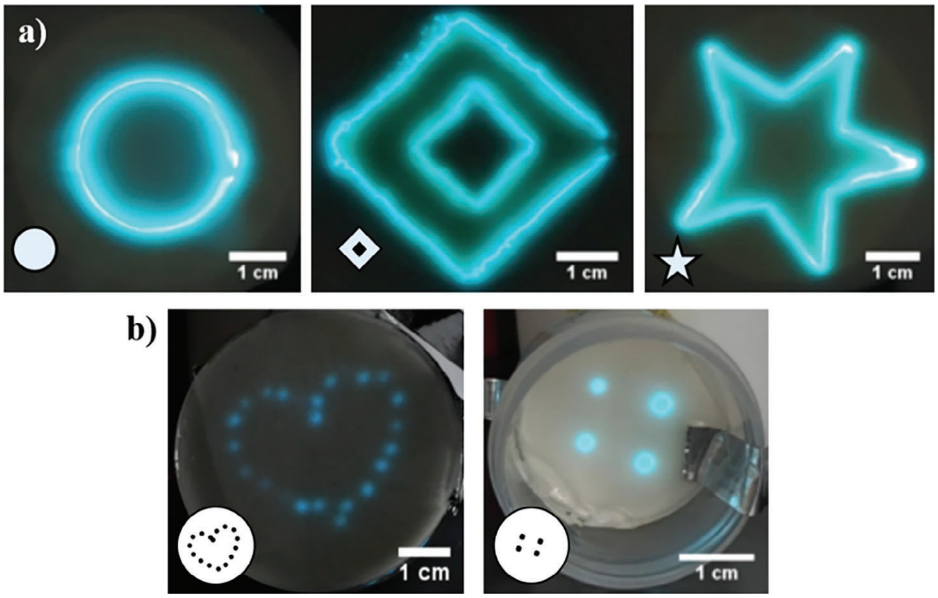

Figure 2. ACEL device lighting patterns. The electrode shape is presented in the left bottom corner of the picture. a) Patterns for hydrogel electrodes under a voltage of amplitude 3 kV and frequency 1 kHz. From left: ring, diamond, and star-shaped electrodes. b) Patterns for wire electrodes under a voltage of amplitude 7.5 kV and frequency 1 kHz. From left: heart (wires punched into silicone), dots (wires nailed into cup).

glycerol (Emmelev A/S) and silicone oil (POWERSIL Fluid TR 50, Wacker Chemie) were used. Zinc sulfide particles doped with copper atoms (ZnS:Cu, Shanghai Keyan Phosphor Technology Co., LTD.) were used in the electroluminescent layer to create blue–green color luminescence. To begin, two different devices were made: one with hydrogel electrodes and another with a combination of copper wire and hydrogel electrodes. The hydrogel device consists of a pre-stretched dielectric elastomer layer sandwiched between hydrogel electrodes and coated with a luminescent layer from both sides. **Figure** 1a presents a schematic of the device preparation process (a more detailed description of this process is presented in the Electronic Supplementary Information (ESI)).

In these ACEL devices, the high field concentration along electrode edges causes light emission. This fact was used to design the devices to create different lighting patterns. Variously shaped hydrogel electrodes were cut out of a hydrogel sheet using a Yueming laser cutter (GD Han's Yueming Laser Tech CO., LTD., China); ring, star, and diamond shapes were tested (**Figure** 2a).

The wire device consists of copper wires inserted into one side and a transparent hydrogel electrode on the other side and was prepared in two different ways: it could be made by curing the elastomer layer and then inserting the wires into it, or by first nailing wires into the cup and then curing silicone elastomer layers inside it. Figure 1b presents a schematic of the first preparation process, in which the wires were inserted into a previously prepared elastomer layer (a more detailed description of this process can be found in ESI, along with that of the second preparation process, in which the wires are nailed into the cup, Figure S1, Supporting Information).

A list of all the devices prepared is presented in Table S1 (Supporting Information). All devices were tested by applying a voltage with a high amplifier (Trek, 30/20A) for 90 min to assess device lifetime. A sinusoidal voltage of amplitude 3 kV and frequency of 1 kHz was applied to the hydrogel devices, while a voltage of amplitude 7.5 kV and frequency of 1 kHz was applied to the wire devices. The difference in applied voltages was due to the different distances between the electrodes, and thus the applied electric field was different ($E = V/d$).[10] Trek amplifies the input voltage generated by a waveform generator (KEYSIGHT, 33500B) by a factor of 3000. By changing frequency and voltage, device performance could be observed under different conditions. The luminance distribution of the whole sample area was measured with an imaging luminance measurement device (ILMD) (LMK mobile, Technoteam: DXM6058B11) with a narrow angle lens adjusted at 50 mm (LMK mobile, 17–50 mm, ser.no 15 254 504). Luminance images were generated from three high dynamic range images of each sample. Luminance and chromaticity coordinates of the emitted light were measured with a luminance and color meter (CS-200, Konica Minolta, ser.no. 1 003 936) at the positions of highest luminance identified from the luminance images. Chromaticity coordinates (x,y) according to CIE (Inter-

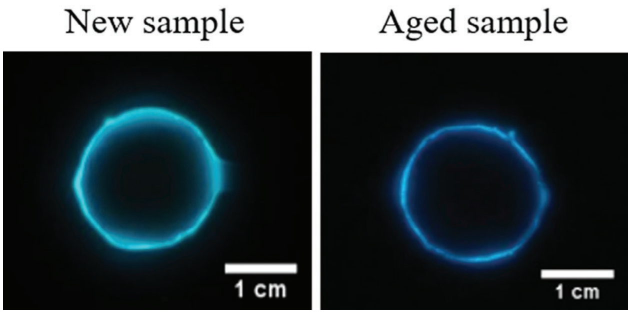

Figure 3. Example of ageing of hydrogel devices. (Left) New device with circle electrodes, and (right) the same device aged 14 months on the right. Working conditions are 3 kV voltage and 1 kHz frequency for both devices.

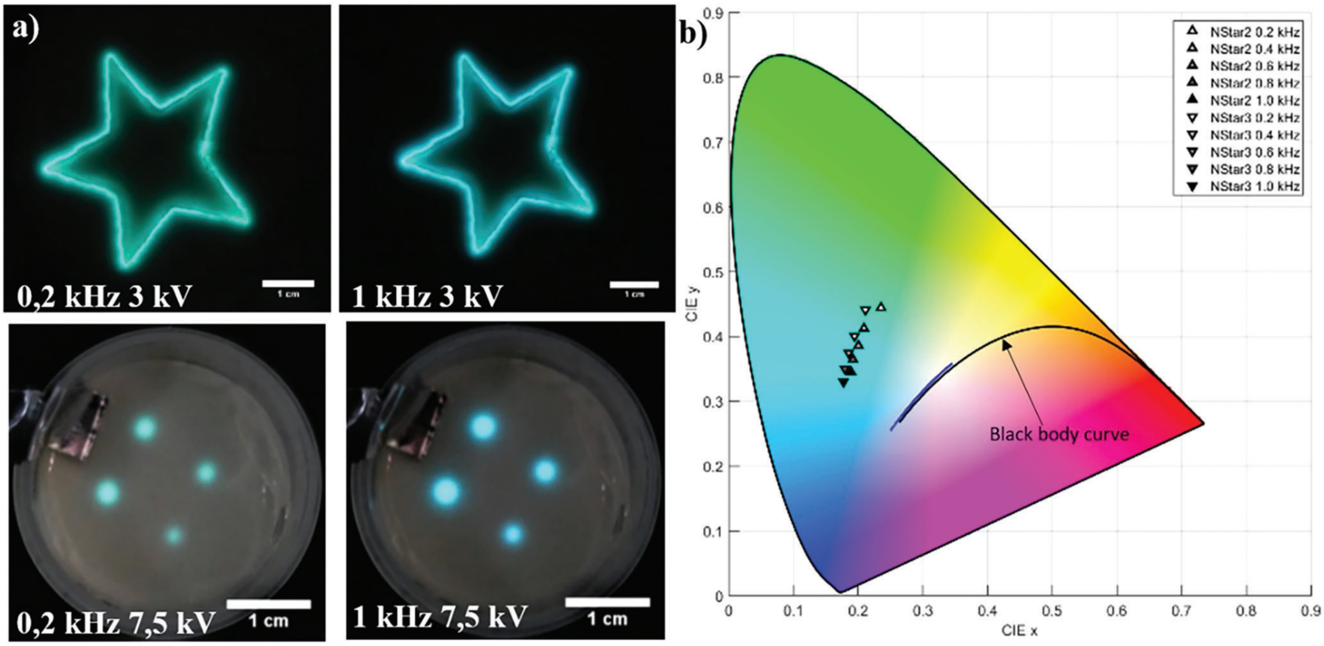

Figure 4. Devices at different frequencies: a) photos of hydrogel and wire devices in a frequency range between 0.2 and 1 kHz; b) color space CIE 1931 (x,y) chromaticity diagram showing CIE measured (International Commission on Illumination) chromaticity coordinates of device light emission at different frequencies.

national Commission on Illumination) 1931 2° color matching functions were reported. In addition to newly prepared hydrogel and wire devices, hydrogel devices aged for 14 months in ambient conditions in the lab were also tested. Aged (A) hydrogel devices are denoted as AStar when containing a star-shaped hydrogel electrodes and ACircle when containing a circular hydrogel electrodes.

3. Results and Discussion

ACEL devices emit light in response to applied voltage, making the pattern created by the electrodes visible. The lighting patterns of the prepared ACEL devices are displayed in Figure 2.

When a voltage of 3 kV amplitude and 1 kHz frequency is applied to the hydrogel devices, only ZnS particles located around the hydrogels' edges gain adequate electric field to emit light. The electric field generated from the middle parts of the hydrogel electrodes is not strong enough to trigger EL particles for light emission. Different electrode shapes have different lighting patterns, as the examples presented in Figure 2a demonstrate.

When a voltage of amplitude 7.5 kV and frequency 1 kHz is applied to the wire devices, light is only emitted from the part of the device where the tip of the wire is close to the luminescent layer (Figure 2b). The gap between wire tips and the hydrogel electrode is ≈2 mm, so these devices require higher voltage than hydrogel devices to gain the electric field necessary to trigger light emission by the luminescent layer. It was reported[9,10] that, for a 1 mm thick electroluminescent layer sandwiched between two electrodes, a voltage of amplitude 20 kV (20 MV m^{-1}) is required to pass the required threshold of 10 MV m^{-1}. The field concentration on the wire tip allows us to reduce the working voltage by almost one order of magnitude, as our applied electric field is 3.75 MV m^{-1} for wire devices.

The pictures of the prepared wire devices (Figure 2b) show that not all dots in a given pattern light equally (or at all) and that the intensity is decreased compared to the hydrogel analogues. This phenomenon is a result of the device preparation process since it is not possible to place all the wires at precisely the same depth and same angle in our specific process. The local distances between wires and the luminescent layer therefore change throughout the material and therefore cause a non-uniform field. The distance between the wire tip and electroluminescent layer is very important in such devices, as larger gaps between electrodes necessitate a higher operating voltage[10] so even small differences in this distance affect device efficiency as well as brightness.

3.1. Lifetime Tests

Over the course of 90 min, the luminescent devices completed 5.4 million cycles. Digital pictures were taken after 1 cycle, 3.6 million cycles, and 5.4 million cycles in order to capture any visible changes to the device. Results for devices with different electrodes are presented in Figure S2 (Supporting Information). No changes within experimental uncertainty were observed in the luminance, suggesting that the tested luminescent devices are stable.

Hydrogel devices with star and circle electrodes in duplicates were aged in petri dishes for 14 months while stored in a cabinet in ambient conditions. The aged hydrogel devices showed no degradation resulting from storage (**Figure 3**), providing good performance and thereby further confirming EL device stability.

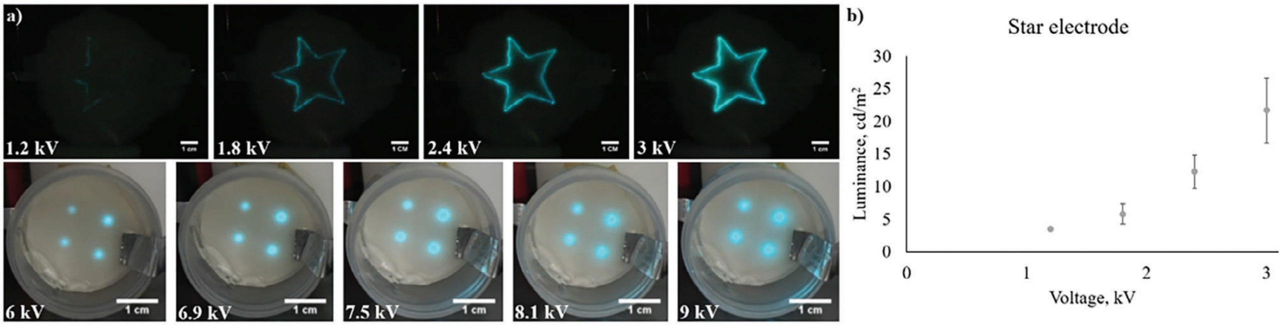

Figure 5. Device reactions to different applied voltages: a) photos of hydrogel and wire devices under different voltages; b) measured brightness of hydrogel devices with star electrodes.

These aging properties are comparable or even better than the current state of art within aging-optimized perovskite LEDs.[25,26]

3.2. Dependence on Voltage and Frequency

It was previously shown[4,9,12,27] that by changing the applied frequency, emitted light color and the luminance of the EL device will change. When Cu-doped ZnS luminescent powder is used, device color depends on frequency, as expected, changing between green and blue. **Figure** 4a presents examples of devices working at different frequencies. When the frequency changes from 0.2 to 1 kHz, the color changes from green to blue. This observed color change is further confirmed via chromaticity coordinate measurements (Figure 4b).

In addition, luminance measurements at different frequencies for the hydrogel device with a star electrode are presented in Figure S3 (Supporting Information). When a frequency of 0.2 kHz is applied, the measured luminance is 8.6 cd m^{-2}; this changes to 25.5 cd m^{-2} on average when a frequency of 1 kHz is applied. This can be explained by the increased likelihood of excitation of Cu^{2+} ions with increased frequency.[27]

Changing the voltage, on the other hand, only alters a device's luminance. This can be explained by Equation 1. Yang et al.[12] previously showed that, with an applied voltage of amplitude 3 kV, the luminance is highest at a frequency of 1 kHz when using hydrogel electrodes. The same results were found for the devices tested in this work (**Figure** 5). When a lower voltage of 1.2 kV is applied to the hydrogel device with a star electrode, its brightness is only 3.5 cd m^{-2}; however, when the voltage is increased to 3 kV, brightness increases to 22 cd m^{-2}. The wire device was observed to display lower brightness at 6 kV compared to 9 kV (Figure 5a). When varying the applied voltage at constant frequency, device luminance changes but color remains the same. Chromaticity coordinates were measured for the hydrogel devices with a star electrodes and are shown in Figure S4 (Supporting Information); all the devices remain the same color when different voltages are applied at a given frequency.

3.3. New Wire Devices

Due to the sensitivity of the lighting pattern to the manner of device preparation, a new wire device was prepared to enhance the lighting emission from the small wire tips to the bulk of the device. This novel device was termed a "silicone oil lamp". The preparation process for silicone oil lamps is identical to that of wire devices, except for the substitution of a translucent silicone oil filled silicone sponge for the transparent bulk silicone. However, this small change in the fabrication process affects the working principle of the device as a whole. When transparent dielectric silicone is used, it allows light to travel through it, so only the pattern created by the wires is visible. However, a translucent dielectric layer scatters light as it passes through, causing the whole device to emit light.[28] A schematic preparation process for silicone oil lamps is presented in Figure S5 (Supporting Information), and a detailed description of this process is presented in the ESI. A silicone oil lamp under AC voltage is shown in **Figure** 6.

3.4. Luminance Measurements

The studied devices' brightness were measured and the results are presented in **Figure** 7 alongside those from previous studies of both hydrogel electrodes[18,29] and silver nanowire electrodes.[15,27,30]

As Figure 7 makes clear, the novel silicone oil lamp is the brightest of the reported devices. Where the tip of a wire is close

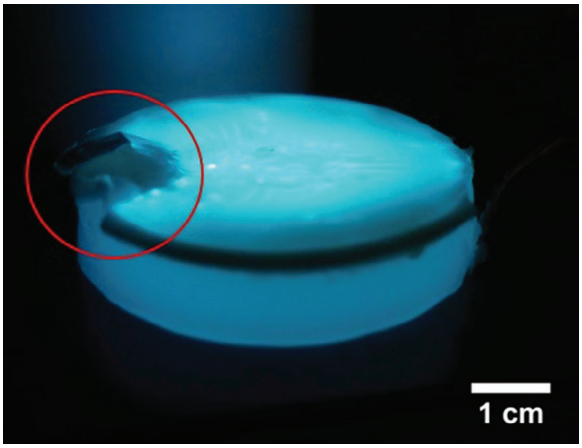

Figure 6. Silicone oil lamp under 6 kV applied voltage at 1 kHz frequency. The red circle marks the part of the device that does not light up due to the aluminium electrode's connection to the hydrogel electrode.

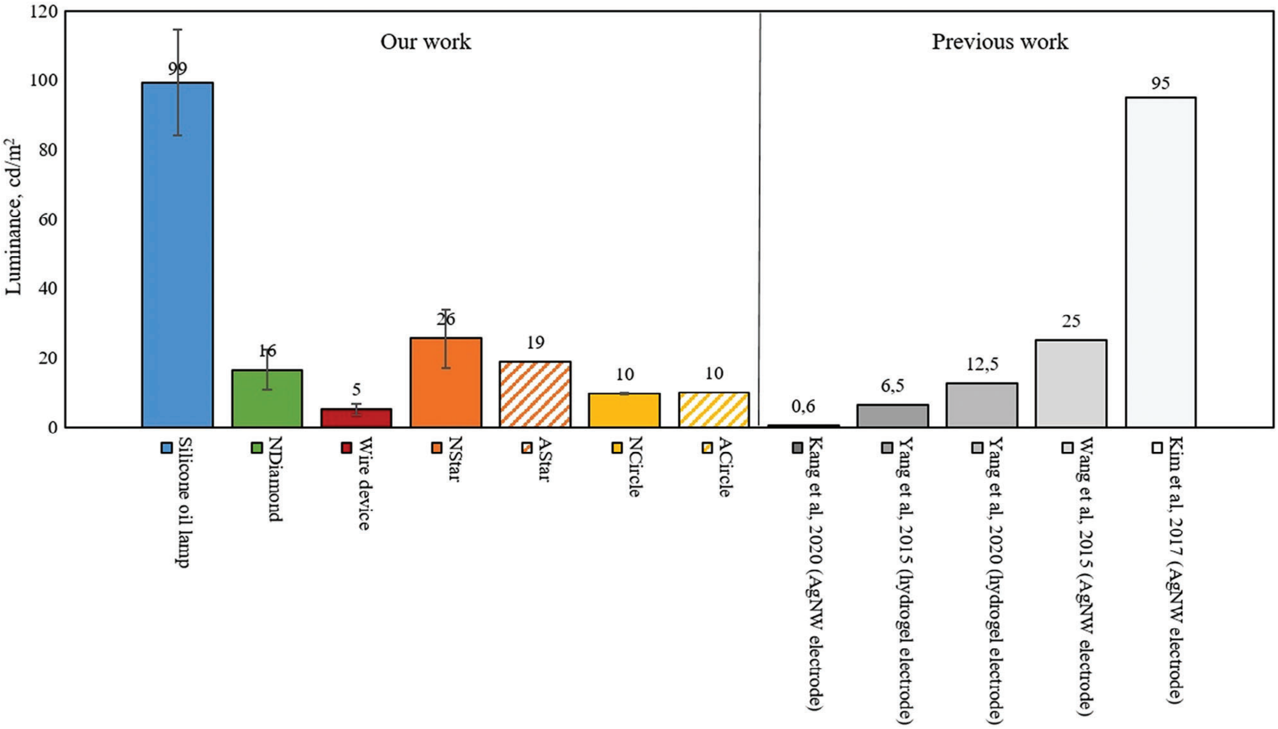

Figure 7. Luminance measurements for our devices (left) compared those from other studies (right). N– new device; A – aged device. Our devices were tested in duplicates.

to the EL layer, the lamp displays a luminance of ≈99 cd m^{-2}, while its background luminance is 15 cd m^{-2}. The hydrogel device with star electrodes has a luminance of 26 cd m^{-2}, compared to 16 cd m^{-2} for device with diamond electrodes and 10 cd m^{-2} for the circle electrode device; the wire device has the lowest luminance of 4 cd m^{-2}. The aged hydrogel devices with star electrodes lost ≈27% of their luminance after 14 months, whereas the luminance of aged devices with circle electrodes remained the same.

The luminance of the devices with hydrogel electrodes studied here is similar to what has been reported by others.[18,29] The silicone oil lamps have high luminance, comparable to that of a previously reported device with silver nanowire electrodes. However, ACEL device luminance varies drastically depending on the preparation process when silver nanowire electrodes are used: from 0.6 to 25 cd m^{-2} and 95 cd m^{-2}, respectively.[15,27,30] When ACEL devices with silver nanowires are prepared as DEAs—i.e., by layering a luminescent layer between two electrodes—they display similar luminance to that of the devices studied here.[15] Even though our devices do not defeat the results from previous works, our technology works with novel design and demonstrated similar luminance.

Measured luminance image is presented in Figure S6 (Supporting Information), where the hydrogel device in working condition is shown. The brightest light being emitted from the edge of the electrode—i.e., where the electric field is concentrated. The luminance of the AStar device is 19 cd m^{-2}. The strongest light emanates from the edge of electrode, while there is no luminance signal emanating from the interior of the electrode. Pictures of other devices under various working conditions are presented in Figures S7–S43 (Supporting Information).

3.5. Stretching and Bending of Devices

All our fabricated EL devices are flexible, though to varying extents, and the hydrogel devices must be constructed without a rigid frame in order to be stretchable or bendable. After coating, the hydrogel devices are ≈3 mm thick, making them difficult to stretch. They can still be bent, however. **Figure 8** shows that the hydrogel device with a circle electrode works normally while bent. During bending, due to the dissimilar nature of the materials

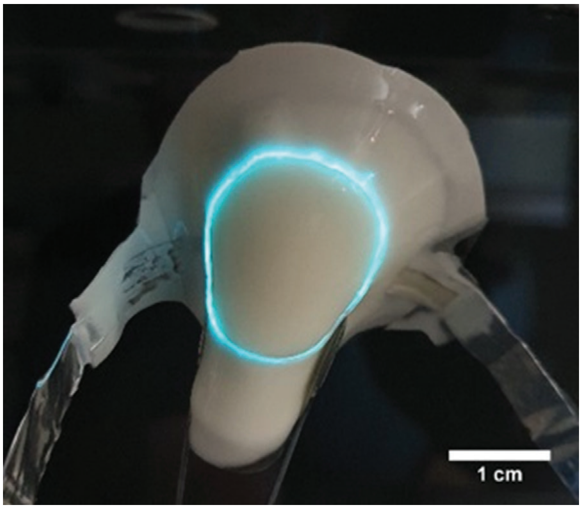

Figure 8. Bended hydrogel device at 3 kV and 1 kHz.

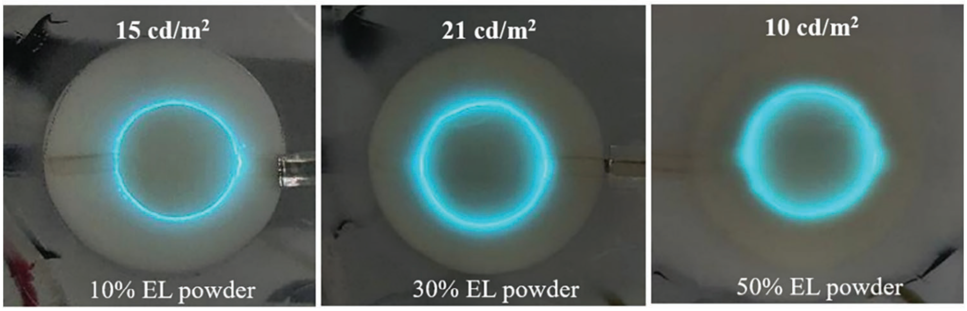

Figure 9. Different luminescent powder concentrations in hydrogel devices at 3 kV and 1 kHz. The rings have diameters of 2 cm.

used (hydrophilic hydrogel and hydrophobic elastomer) in their construction, the hydrogel devices were observed to undergo a delamination effect, in which the EL layer separates from the hydrogel layer. The same effect is observed when trying to stretch such devices. No chemical bonds were created between hydrogels and elastomers in the devices presented here, but the introduction of chemical bonding may be necessary in order to maintain device structure. It was previously reported that silane condensation between hydrogels and elastomers can be used to adhere these two materials via covalent bonds.[18,31] This bonding would potentially strengthen EL device performance, and hydrogel devices could hypothetically be bent and stretched without delamination.

3.6. Luminescent Powder Concentration

An EL powder concentration of 50% by weight was used to make the ACEL devices presented here. It was previously reported that higher concentrations of EL powder result in increased luminance.[15] Tests carried out as part of our study confirmed this same trend in our devices.

Hydrogel devices with different EL powder concentrations are presented in **Figure 9**. When lower EL powder concentrations are used, the resulting hydrogel device presents a very fine, thin lighting pattern, with a luminance of 15 cd m^{-2}. At increasing powder concentrations, this line expands and blurs, while its luminance increases to 21 cd m^{-2}. At the highest EL powder concentration studied, however, device luminance is only 10 cd m^{-2}, while the lighting pattern is even less clear. For such devices, then, an EL powder concentration of 30% by weight is preferable to 50%.

4. Conclusion

In soft electronics applications, field concentration is often regarded as problematic, especially when using electrodes with greater curvature and sharp edges. However, it is also possible to use field concentration advantageously in such applications. In this work, we used field concentration along electrode edges to fabricate three easily prepared soft hydrogel-elastomer EL devices: a hydrogel device, a wire device, and a silicone oil lamp. Different types of electrodes were used to create different lighting patterns. All fabricated devices were stable, with no changes observed over the course of 5.4 million cycles. In addition, these devices possess long storage lifetimes, and their color can be changed by altering the operating frequency. When the operating voltage changes, so do the color and luminance of emitted light from the device. To transfer the light emitted by the device into the bulk material, a translucent oil-filled silicone sponge was successfully prepared and shown to possess exceptionally high luminance (99 cd m^{-2}). Hydrogel device luminance varies depending on the shape of the electrode used, with wire devices displaying the lowest luminance of all those tested here.

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.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

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