

RIGA TECHNICAL UNIVERSITY
Faculty of Materials Science and Applied Chemistry
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**DIELECTRIC BREAKDOWN OF HIGH
VOLTAGE LIQUID CRYSTAL DISPLAYS**

Summary of the Doctoral Thesis

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DECLARATION OF ACADEMIC INTEGRITY

I hereby declare that the Doctoral Thesis submitted for the review to Riga Technical University for the promotion to the scientific degree of Doctor of Engineering Sciences is my own. I confirm that this Doctoral Thesis had not been submitted to any other university for the promotion to a scientific degree.

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The Doctoral Thesis has been written in English. It consists of Introduction; 3 chapters; Conclusion; 90 figures; 11 tables; the total number of pages is 98. The Bibliography contains 122 titles.

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INTRODUCTION

Windows are the key to the design of any building and they are an essential part of new strategies to improve energy efficiency. Smart glass windows are about 70 % more energy efficient during the summer and 45 % in winter compared to standard dual-pane glass. This reduces overall energy spending for building of 25 %. According to the US Department of Energy buildings account for almost 40 % of all energy consumption in the United States. Total cost of energy used by buildings is more than USD 400 billion. Using smart glass can save USD 100 billion in US alone. Right now, there are two technologies available on the market: polymer dispersed liquid crystal and electrochromic. Polymer dispersed liquid crystal smart glass has high haze in open state and continuous energy supply is needed to maintain open state. Electrochromic smart glass also has a few disadvantages, like slow switching speed, coloration in open state and need for continuous energy supply to maintain closed state. It is expected for smart glass market to reach USD 8 billion by 2022.

Bistable smectic SmA liquid crystal has a potential to become the leading smart glass technology. Transparent state is characterized by very low haze; scattering state has opaque visual appearance. Both transparent and opaque states preserve their state after the removal of voltage, thus energy is required only for a short amount of time during switching from one state to the other. Bistability comes hand in hand with high viscosity, therefore high voltage (20–30 V/ μ) is needed to switch liquid crystal, and dielectric breakdown is likely to occur. Cholesteric liquid crystal fast switching shutters is another technology where high voltage is required. Fast switching shutters have a potential in various applications – as a projection screen for volumetric 3D display, 3D shutter glasses, switchable windows and also light modulating devices.

There is a very limited number of studies devoted to liquid crystal display dielectric breakdown suggesting the use of dielectric thin film and electrode patterning. Studies on other high voltage capacitive devices like electroluminescent displays show high dielectric breakdown field with use of high k dielectric thin and thick films. Studies of LEDs and capacitors show lifetime, and durability can be improved by reducing the roughness of electrode or using smoothing layer.

Aim of the Doctoral thesis

To develop high voltage liquid crystal display technology ensuring high optical transmittance and fast switching speed.

Tasks of the Doctoral thesis

1. To review literature on dielectric breakdown mechanism and methods to prevent it in liquid crystal displays and other similar high voltage devices.
2. To experimentally test various dielectric breakdown prevention methods and to evaluate their impact on electrical and optical properties.
3. To develop electrical and optical test methods to evaluate the possible cause and mechanism of dielectric breakdown.

4. Based on literature review and experimental results to propose the best liquid crystal display design.

Thesis statements

1. The dielectric thin film is a crucial part in the design of the device, which prevents dielectric breakdown and produces reliable high voltage liquid crystal display.
2. The main cause of dielectric breakdown in liquid crystal displays are pinholes formed during the deposition of dielectric thin film. Thin film works as a current limiting layer in liquid crystal display, therefore high quality defect free thin film is required.
3. Introduction of magnetron sputtered SiO₂ 200–300 nm thin film as a dielectric protective layer is the best way to produce reliable large area high voltage displays with high optical transmittance.

Scientific significance and innovation of the Doctoral thesis

1. Various dielectric breakdown preventive methods like electrode separation, smoothing electrode layer and dielectric thin film introduction are studied and evaluated. Dielectric thin film deposited by magnetron sputtering was found to be the most effective method yielding high dielectric strength and high optical transmittance.
2. For the first time origin and mechanism of dielectric breakdown in high voltage liquid crystal displays is described. It is demonstrated that the main cause for liquid crystal display breakdown is pinholes and defects in dielectric thin film introduced between the transparent conductive electrode and liquid crystal.

Practical significance of the Doctoral thesis

Inexpensive and scalable high voltage liquid crystal display technology for smart glass and 3D volumetric applications has been developed.

Thesis approbation

The main results of the thesis and scientific research is published in four full text papers and disseminated in two international scientific conferences.

LITERATURE REVIEW

A cross-section of liquid crystal display is shown in Fig. 1. Soda-lime glass with thickness 0,4–1,1 mm covered with transparent conductive oxide is typically used as a substrate. Indium tin oxide (ITO) is usually used as an electrode. To prevent Na^+ diffusion, SiO_2 barrier layer is placed between ITO and glass. The thin film is used as an alignment layer and electrode insulation. Typically, polyimide is used, but sometimes two thin films are used: SiO_2 and polyimide. Spacers maintain constant distance between electrodes. A gasket keeps substrates together and prevents liquid crystal from flowing out.

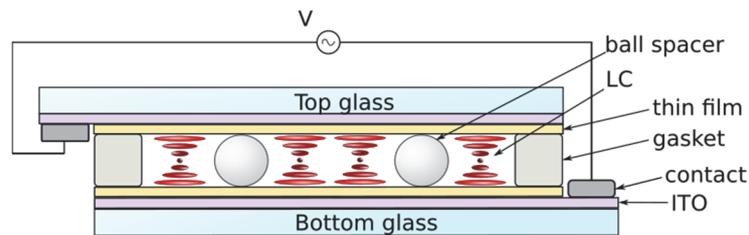


Fig. 1. Cross-section of liquid crystal display.

Working principle is shown in Fig. 2. Low frequency voltage (< 100 Hz) induces the motion of ionic additives through the layered smectic A liquid crystal and generates highly scattering focal conic structures, and the device becomes opaque. Typical feature size is in order of $1 \mu\text{m}$. Upon application of frequency beyond a critical value (~ 300 Hz–1 kHz), long axis of liquid crystal molecules rotates to dielectrically align with the field, and the device becomes clear with very low haze. After removal of the voltage both modes are preserved, owing to the high viscosity of the smectic A phase. Due to high viscosity, high voltage ($> 10 \text{ V}/\mu$) is needed to switch the liquid crystal, and dielectric breakdown is likely to occur [1].

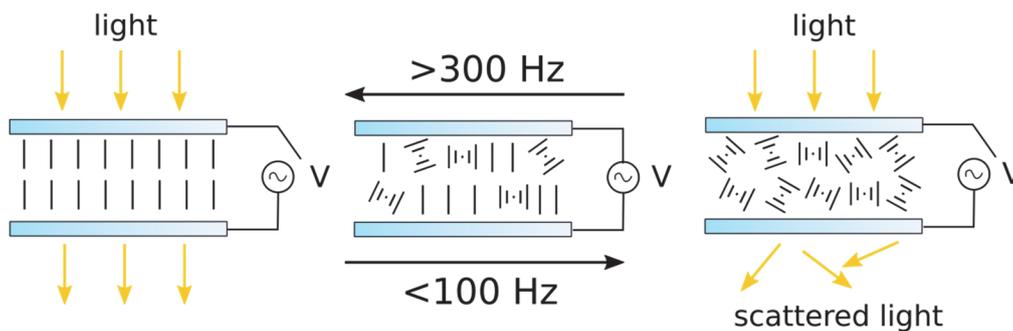


Fig. 2. Working principle of bistable liquid crystal display.

Cholesteric liquid crystal displays is one of the most promising technologies for fast switching shutters, due to fast switching speed. Cholesteric liquid crystal is a mixture of nematic LC and chiral dopant and forms a helical structure. When voltage is applied, LC takes homeotropic state and LCD becomes transparent. Upon removal of applied voltage LC rapidly switches to focal conic texture by passing through transient planar texture (see Fig. 3). Transmittance in scattering state depends on the device design and LC used and typically is $< 10\%$ [2].

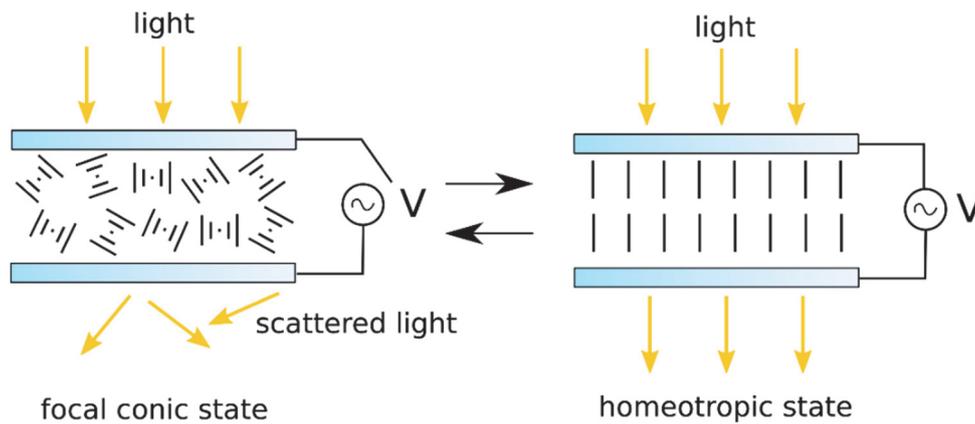


Fig. 3. Working principle of cholesteric liquid crystal

A phenomenon is generally referred to as electric breakdown or dielectric breakdown is an increase in conduction of a material by several orders of magnitude due to failure of dielectric material under applied electric stress. The terms breakdown field strength and dielectric breakdown strength describe the quality of material as dielectric from the point of view of dielectric breakdown. It is calculated by dividing applied voltage when breakdown process takes place with dielectric thickness. The magnitude of such a strength depends not only on intrinsic properties of dielectric, but also on external variables such as the uniformity of electrical field, temperature and the quality of electrodes. Dielectric breakdown is associated with an appearance of optical effects like sparks and fractals at the surface of an electrode [3].

Only one study can be found on dielectric breakdowns in liquid crystal cells, where use of polyimide thin film was found to be the most effective method to improve dielectric breakdown strength [4]. Description of dielectric breakdown mechanism cannot be found, several breakdown mechanisms and models are described in liquids: 1) model based on formation of conductive channels; 2) electron avalanche breakdown; 3) suspended solid particle mechanism; 4) bubble model; 5) electrochemical model [3], [5].

Several methods of improving dielectric breakdown strength are reviewed in literature.

1. Surface roughness reduction. Both calculations and experimental results show, that surface roughness reduction increases dielectric breakdown strength of the device [6].
2. Smoothing layer. Deposition of conductive polymer PEDOT:PSS on ITO electrode can reduce surface roughness and leakage current, increasing device lifetime [7]. Use of PVK and 10GN mixture also have been reported [8].
3. Electrode patterning. Electrode can be divided into stripes forming series of row electrodes to create high internal electrical resistance in series at any point in the liquid crystal optical shutter whilst the overall external resistance is kept at low level [9].

4. Use of dielectric thin film. This method is used in electroluminescent devices. Dielectric thin film limits current. The following properties are required for thin films: small number of pinholes and defects, high dielectric constant, and high dielectric breakdown field. Typically materials such as Al_2O_3 , SiO_2 , Si_3N_4 and BaTiO_3 are used.

In liquid crystal display industry thin films are deposited by flexographic printing, magnetron sputtering, chemical vapor deposition, screen printing and spin coating. In other industries thermal evaporation and atomic layer deposition are widely used.

EXPERIMENTAL PART

Liquid crystal display preparation process is shown Fig. 4. ITO glass substrates size 300×400 mm with sheet resistance 40–80 Ω /sq were purchased from China and Taiwan. Twelve LCDs size 80×80 mm were placed on one substrate, therefore out of two substrates 12 displays were made. After washing the substrates were patterned with laser, ablating isolation lines for active area, markers, numbers, etc. After laser patterning, substrates were washed again and thin films deposited. Three methods were used to deposit thin films: flexographic printing, screen printing, and magnetron sputtering. Flexographic printing is a traditional method in liquid crystal display industry, where material is deposited from liquid with solid content 3–6%. Polyimide and SiO₂ were used with thickness 105–485 nm. Thin film thickness was increased by depositing multiple layers. PEDOT:PSS was deposited by screen printing. Silica was deposited by reactive magnetron sputtering. Thin films of various thickness (100–300 nm) were deposited by changing conveyor speed. Thin films were deposited in FHR Anlagenbau GmbH, Germany. Spacer (10–15 μ m) suspension in organic solvent was sprayed in heated chamber, where solvent evaporates and spacers deposit on substrate. On opposite glass gasket was dispensed on the substrate. The panel was made by assembling both substrates together. Bistable liquid crystal displays were made using one drop fill method, where the assembly took place in vacuum with LC already dispensed. After assembly the gasket was cured in temperature > 150 °C. In the next step the samples were cut out of the panel and filled in a vacuum chamber. The last step in sample preparation was contact soldering with ultrasonic solder.

Resistance and current-voltage characteristics were measured for thin films. ITO under thin film was used as one electrode and liquid metal was used as the second electrode. Resistance of 154 mm² area was measured with multi-meter, but I-V characteristics with electrometer/high resistance meter Keithley 6517B. TTi TG210 Function Generator was used to generate square waveform. TREK MODEL PZD350 was used to increase the amplitude up to 2000 V pk–pk. Voltage was increased continuously until dielectric breakdown occurred.

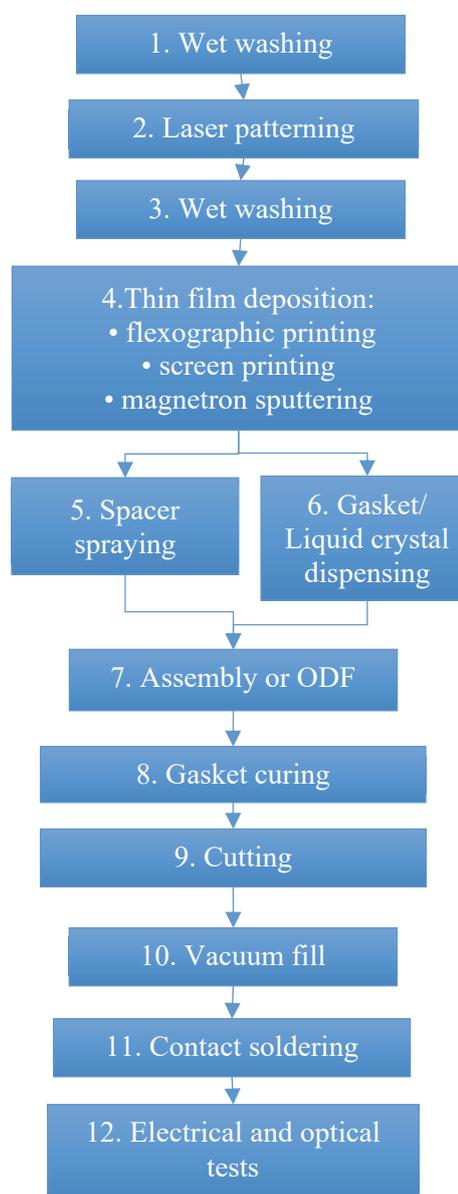


Fig. 4. Display sample preparation process steps.

Transmittance and switching speed for cholesteric and bistable LCDs was measured with a homemade test equipment at EuroLCDs. The transmittance was measured using white LED. For both types of displays four values were measured:

- 1) transmittance in open state (homeotropic state);
- 2) transmittance in closed state (scattering state);
- 3) fall time – switching speed from open to closed state;
- 4) rise time – switching speed from closed to open state.

In the case of cholesteric displays, the properties were measured in the range of 30–200 V with step 1–3 V. At every voltage values were measured 25 times and average value was calculated. Bistable liquid crystal displays were measured in the range of 60–240 V with step 10 V. To switch to open state, 1 kHz frequency was used, but 50 Hz were used to switch to a scattering state.

RESULTS AND DISCUSSION

1. Electrode roughness

In order to determine the effect of surface roughness on dielectric breakdown value, cholesteric LC displays were made out of ITO glass sourced from various manufacturers. ITO glass was purchased from Token in China, Aimcore and Gemtech from Taiwan. Surface roughness R_a measurements for each ITO glass are listed in Table 1. As one can see from Fig. 5, dielectric breakdown values have large deviation and do not agree with surface roughness measurements. The smoothest Aimcore ITO glass displays have the lowest resistance to dielectric strength ($6.8 \text{ V}/\mu\text{m}$). It can also be explained by lower sheet resistance compared to other ITO glass substrates. The samples made with Token ITO glass have the highest average breakdown value ($10.8 \text{ V}/\mu\text{m}$) and also the largest roughness deviation. It can be expected, that larger surface roughness can have more ITO defects such as hillocks and spikes leading to dielectric breakdown, but these measurements do not prove it. It also might be the case, that surface roughness does not correlate with the amount of dielectric breakdown inducing defects, since roughness is measured on a very small area ($6.6 \cdot 10^{-8} \%$ from $300 \times 400 \text{ mm}$ substrate).

Table 1

Average surface roughness and sheet resistance for various ITO glass manufacturers

Manufacturer	Average R_a , nm	Std. dev., nm	Sheet resistance, Ω/sq
Aimcore	0.5	0.03	50
Gemtech	0.9	0.14	80
Token	1.0	0.27	80

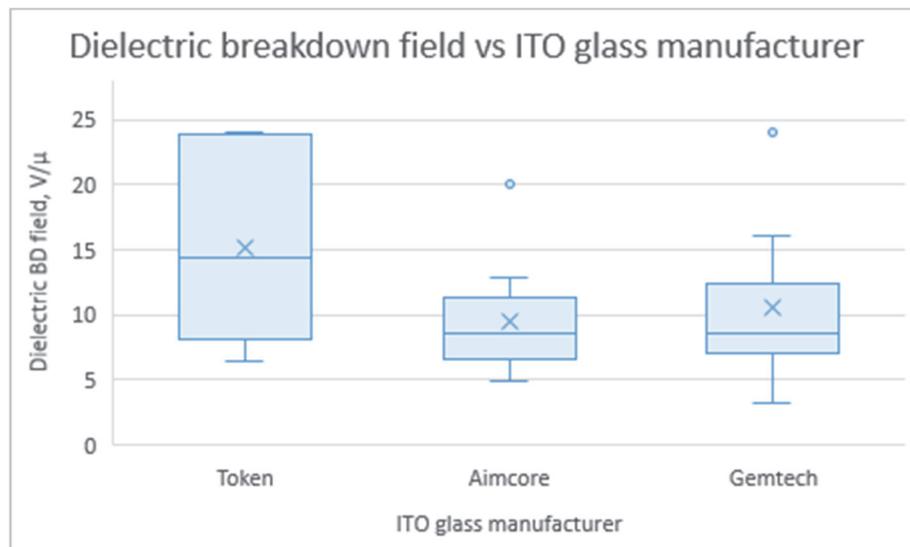


Fig. 5. Dielectric breakdown field intensity for cholesteric LCDs made by three different glass suppliers. For Token glass mean value is $10.8 \text{ V}/\mu\text{m}$. For Aimcore glass mean value is $6.8 \text{ V}/\mu\text{m}$ and for Gemtech glass it is $7.5 \text{ V}/\mu\text{m}$.

2. Electrode patterning

Bistable displays with electrode divided in various widths were tested to determine the dielectric breakdown field. As one can see in Fig. 6, the breakdown field of display with electrode divided in stripes with resistances up to 1554 is very close to the switching threshold field and a reliable device with high yield could not be made due to the large deviation of breakdown field. Optimal breakdown field should be at least twice the switching threshold field $\sim 55 \text{ V}/\mu\text{m}$, which corresponds to the display with electrodes divided in stripes with each stripe resistance $\sim 1900 \Omega$. The displays made with resistance > 7000 did not break down at all.

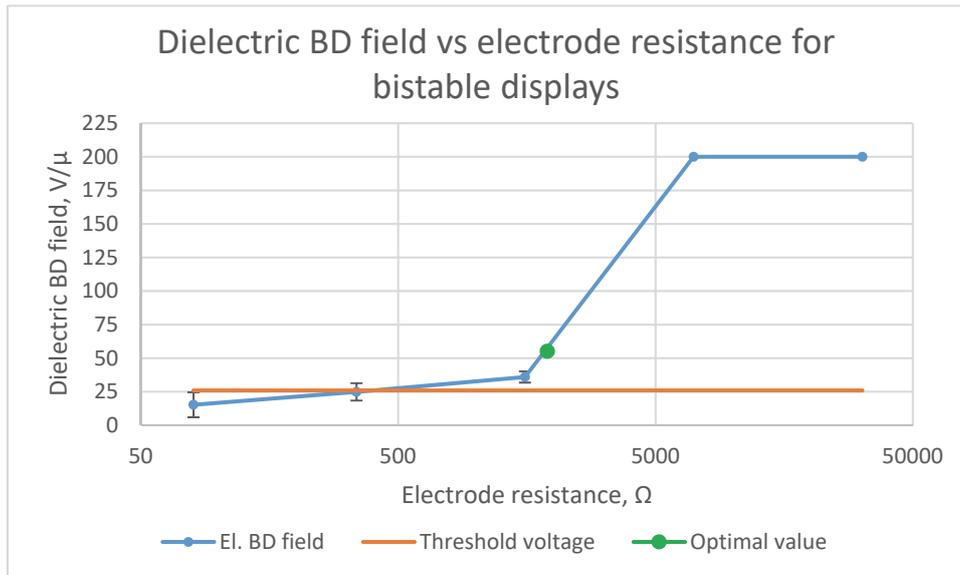


Fig. 6. Dielectric breakdown field of bistable displays with patterned electrode. Switching threshold voltage is depicted with orange line at $26 \text{ V}/\mu\text{m}$. Displays with electrode stripe with resistance $> 7000 \Omega$ did not break down.

Switching speeds and transmittance in open and closed states are shown in Fig. 7. The displays with electrode stripe resistance of 80Ω and 344Ω broke down before reaching maximum voltage. Switching threshold voltage is defined by the voltage when open transmittance reaches maximum. Switching threshold voltage for all displays is around 130 volts or $26 \text{ V}/\mu\text{m}$ (see Fig. 7 a)). Open transmittance for resistances 1554 and 7030 drops at 220 and 230 V, respectively. For the highest resistance transmittance starts to decrease already at 140 V. This effect can be explained by heating of electrode due to high resistance – LC also heats up and does not switch to clear state completely. At around 150 volts the fall time starts to increase (see Fig. 7 c)), because scattering and transparent states start to *compete*. Switching LC to transparent state with high voltage leads to overdriving – LC gets “stuck” in homeotropic alignment and it takes longer time or higher voltage to switch back to the scattering state. This is also the reason why the closed transmittance decreases at 170 V, because voltage is high enough to switch LC from “stuck” clear state to scattering state. In all cases the acceptable switching speed is $< 1 \text{ s}$.

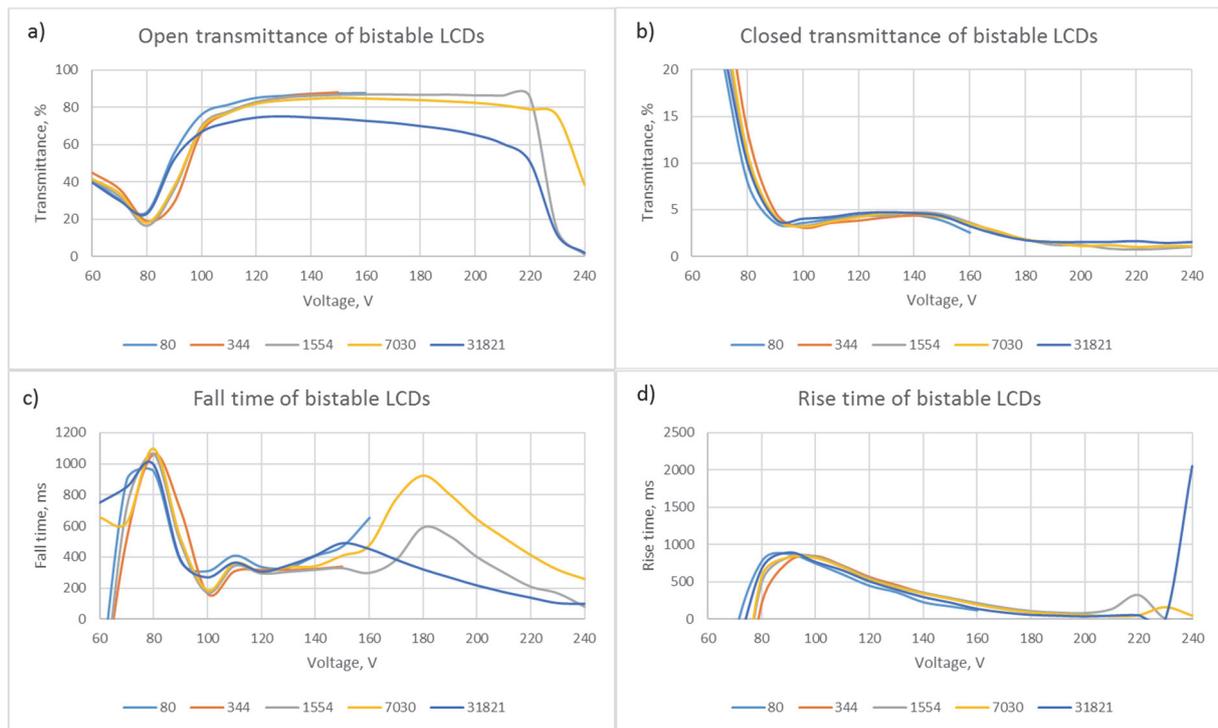


Fig. 7. Transmittance and switching speed of bistable displays with $80 \text{ } \Omega/\text{sq}$: a) open transmittance; b) closed transmittance; c) fall time of bistable LCDs; d) rise time of bistable LCDs. Displays with patterned electrodes with resistance $80 \text{ } \Omega$ and $344 \text{ } \Omega$ broke down before reaching 150 V and 160 V .

Photographs of bistable displays can be seen in Fig. 8 a)–c). The display switched to scattering state is opaque because it scatters, absorbs and reflects light. Patterned lines are well visible because LC does not switch in the areas where electrode is ablated away (see Fig. 8 d)). It can also be explained by the scattering inducing ion concentration around sharp ablated ITO edges due to high electric field. Ion concentration is too high around the ablated ITO lines and LC does not switch to clear state. For this reason, electrode patterning method is better suited for fast switching shutters.

Cholesteric displays were made of substrates with two sheet resistances of $80 \text{ } \Omega/\text{sq}$ and $40 \text{ } \Omega/\text{sq}$ to see whether the sheet resistance also influences dielectric breakdown. As one can see from the graph in Fig. 9, the displays with lower resistance electrode break down more easily – difference in BD field is $\sim 2\times$. It can be explained by faster discharging of the device due to lower sheet resistance, that leads to dielectric breakdowns. The switching threshold value for both display types is $21 \text{ V}/\mu\text{m}$, meaning that the devices should work $> 40 \text{ V}/\mu\text{m}$. For $80 \text{ } \Omega/\text{sq}$ electrodes the resistance should be $> 1554 \text{ } \Omega$, but for $40 \text{ } \Omega/\text{sq}$ electrodes the resistance should be $> 3550 \text{ } \Omega$. The Token glass samples ($80 \text{ } \Omega/\text{sq}$) also exhibit much larger deviation, which might be explained by different ITO deposition process and larger inhomogeneity than of $40 \text{ } \Omega/\text{sq}$ substrates provided by JMT.

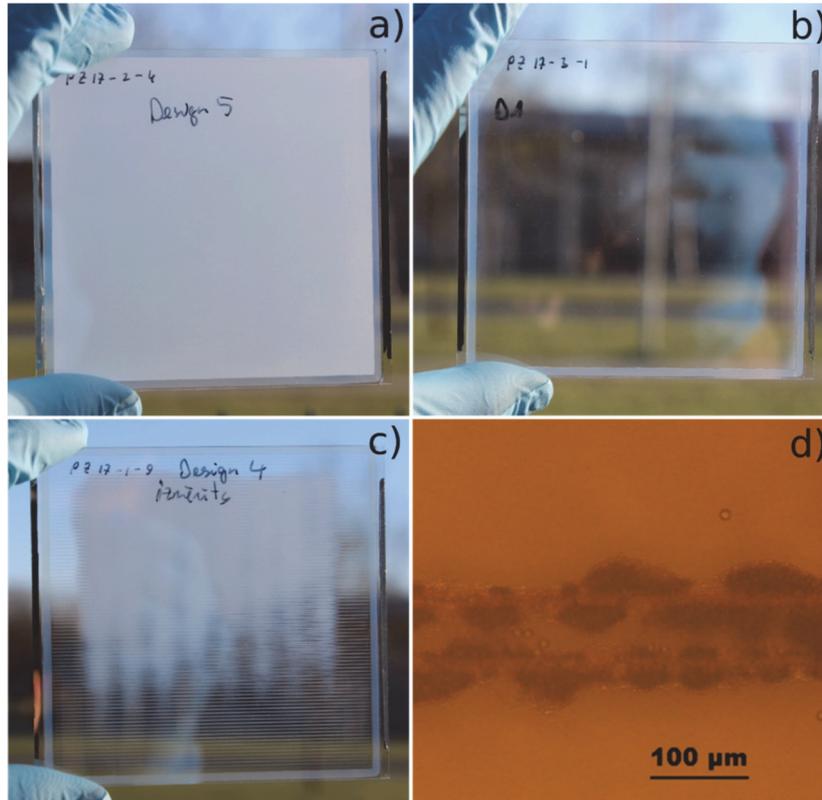


Fig. 8. Photos of bistable displays. a) Bistable display switched to scattering state. b) Display switched to clear state. c) Bistable display switched to clear state. Lines are visible due to the scattering of unswitched liquid crystal around separation. At voltage > 200 V display overheats and does not switch to clear state completely. d) Electrode separation of bistable liquid crystal display lines under microscope. One line is on the top electrode, but other is on the bottom electrode.

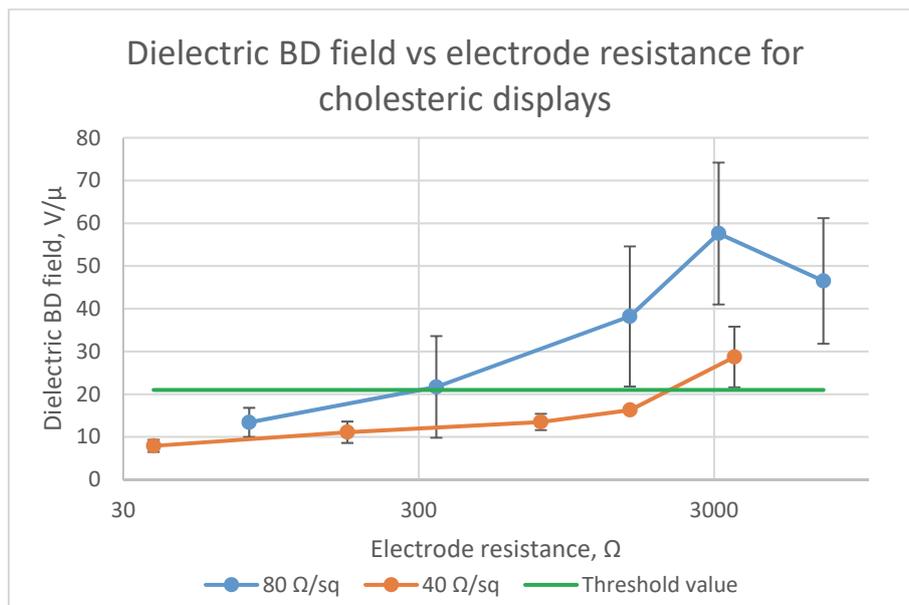


Fig. 9. Dielectric breakdown field of cholesteric displays with patterned electrode. Switching threshold is depicted by the green line at 21 $V/\mu m$.

For both type of cholesteric displays the switching speed and transmittance were measured. As one can see from the graphs in Fig. 10 and Fig. 11, open transmittance does not decrease by increasing the switching voltage of displays with high resistance patterned lines as we can see in the case of bistable displays. Since the displays with 40 Ω /sq electrodes break down more easily, there are no measurements up to 200 V for all displays. The switching threshold value for both resistances is the same \sim 108 V. Open transmittance is higher for 40 Ω /sq shutters (89.5 to 92.4 %) compared to 80 Ω /sq shutters (86.9 to 89.1 %), because 40 Ω /sq ITO glass substrates are coated with AR coating. The displays with the most separation lines have the lowest transmittance, because ablated lines reflect and scatter light. Closed transmittance is also 0.1–0.5 % higher for 40 Ω /sq displays, because of AR coating. The switching speed for both display types is the same. One could expect, that lower resistance electrode would allow to charge display faster and the switching would be faster, but these measurements show, that the switching speed is limited by LC.

For cholesteric shutters electrode patterning is much more suitable than for bistable devices, since the lines have very little effect on optical transmittance and switching speed.

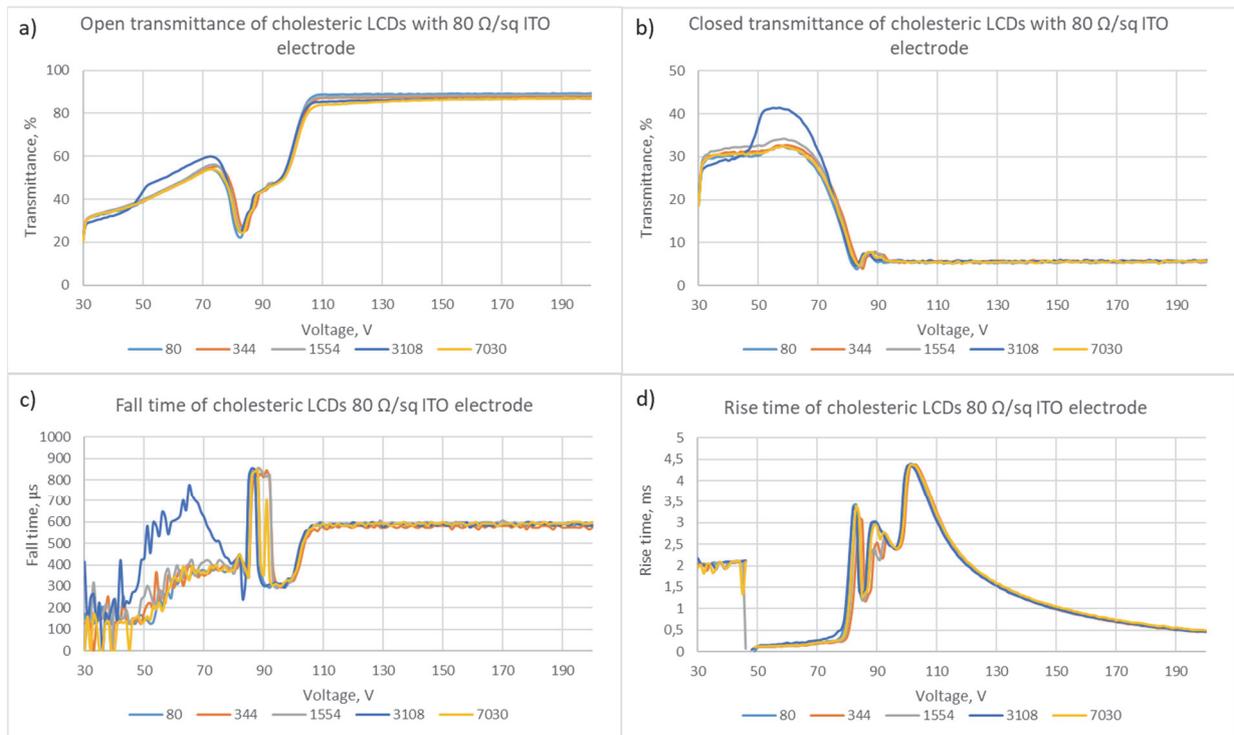


Fig. 10. Transmittance and switching speed measurements for shutters made of 80 Ω /sq electrodes. a) Open transmittance varies from 86.9 % for 7030 Ω to 89.1 % for 80 Ω unpatterned electrode. b) Closed transmittance is \sim 5.5 % for all displays. c) Fall time is \sim 580 μ s, the same for all displays. d) Rise time is 0.5 ms, almost the same for all displays.

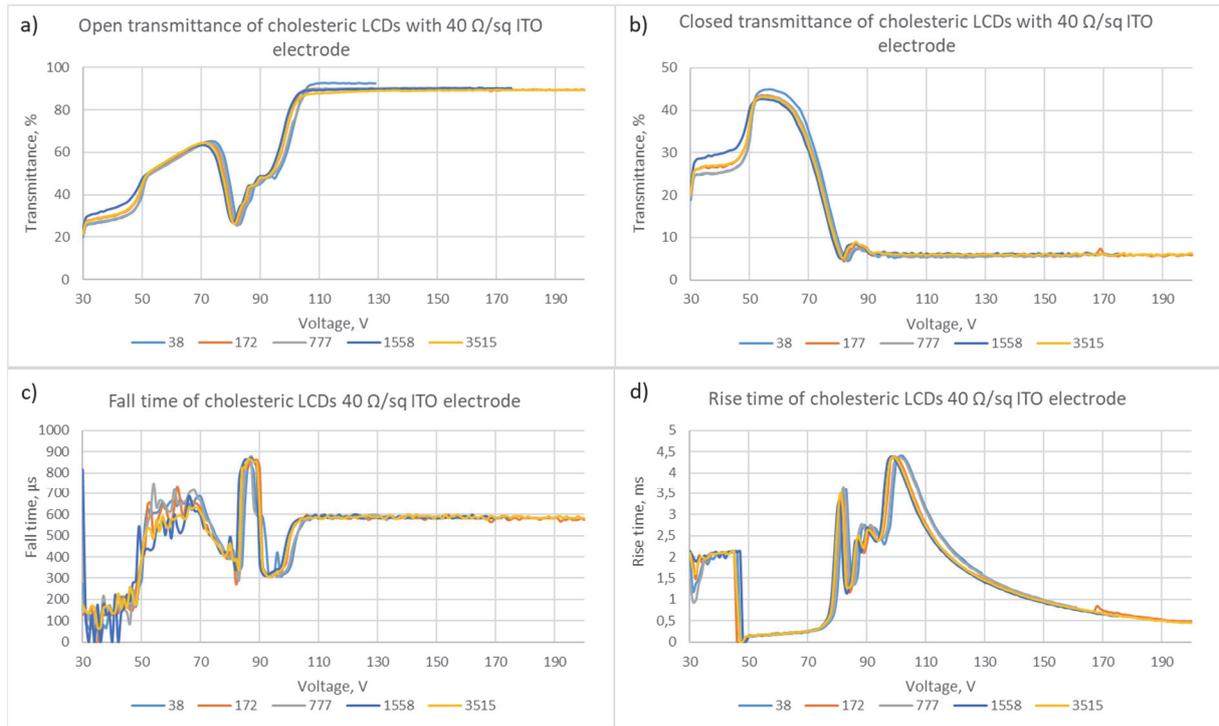


Fig. 11. Transmittance and switching speed measurements for shutters made of 40 Ω/sq ITO. a) Open transmittance varies from 89.5 % for 7030 Ω to 92.4 % for 40 Ω unpatterned electrode. b) Closed transmittance varies from 5.6 % for 3550 Ω to 6 % for 38 Ω. c) Fall time is ~ 580 μs, the same for all displays. d) Rise time is 0.5 ms, almost the same for all displays.

3. Smoothing layer

Four different sets of at least 10 samples were prepared using flexoprinting (for SiO₂) and screen-printing (PEDOT:PSS) methods. Screen-printing was the only available deposition method for PEDOT:PSS. All displays were filled with cholesteric liquid crystal and cell gap was 15 μm. As shown in Table 2, the obtained PEDOT:PSS layer was very rough. The standard deviation of layer thickness was around 52 nm or 80 % of the total thickness. For some samples the PEDOT:PSS was deposited on the SiO₂ which was prepared with flexo-printing method described in the experimental part. One set of samples had flexoprinted SiO₂ deposited on PEDOT:PSS. The measured breakdown field intensity for the prepared samples, including LC displays with no conducting PEDOT:PSS smoothing layer, are displayed in Fig. 12. As is evident from the graph, the breakdown field intensity is reduced if the PEDOT:PSS layer was employed between the ITO electrode and the LC layer. The obtained results clearly indicate that the PEDOT:PSS, if applied with the screen-printing method, does not improve the electrical properties of the LC cell. The main cause of the reduction of breakdown field intensity in the displays is the rough surface of the PEDOT:PSS.

Table 2

Thicknesses of prepared thin films with PEDOT:PSS and SiO₂.

No	Thin film abbreviation	Thickness
1	PEDOT:PSS	65±52 nm
2	SiO ₂ +PEDOT:PSS	225±60 nm
3	2x PEDOT:PSS	130±104 nm
4	PEDOT:PSS+ SiO ₂	225±60 nm

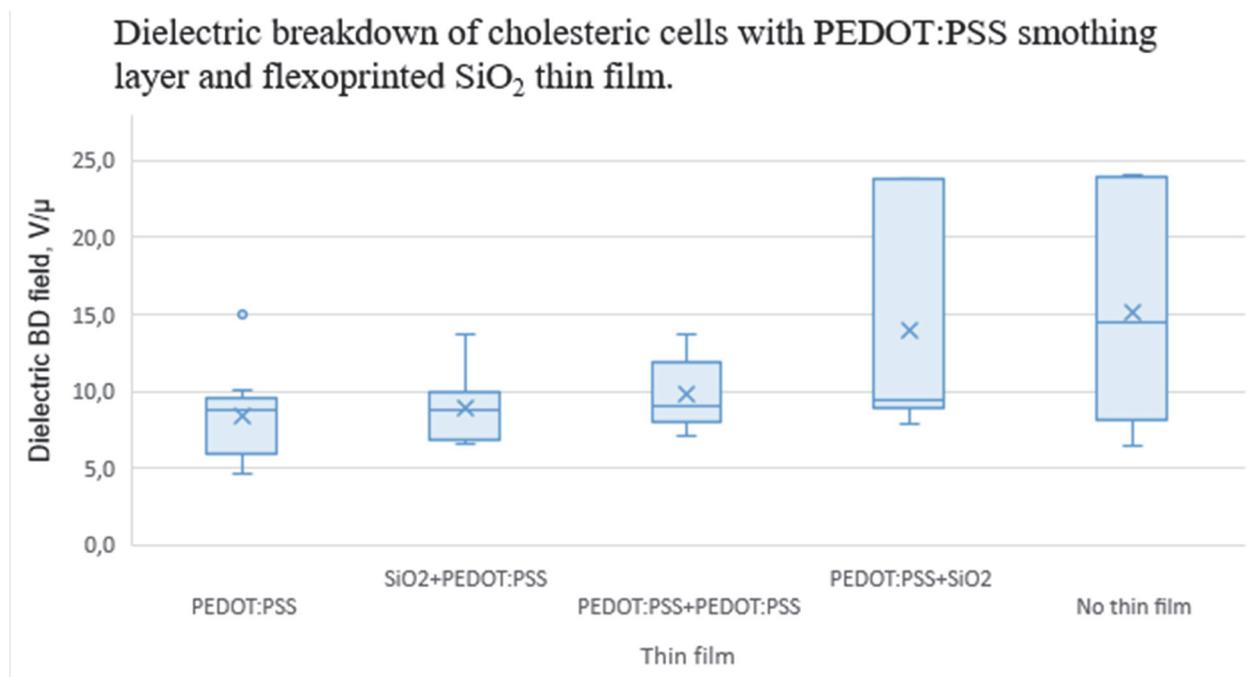


Fig. 12. Breakdown field intensity in the LC displays with various PEDOT and SiO₂ thin films between the ITO electrode and the LC layer. Crosses represent mean value and lines represent median values.

4. Dielectric thin films

Resistance and I-V curves of dielectric thin films

Dielectric thin film electrical properties were tested to understand their role in prevention of dielectric breakdown. Silica thin films were deposited by flexography and magnetron sputtering.

Immediately after filling Teflon ring with liquid metal resistance indicated by a regular multimeter is tens of mega ohms (MΩ). In a few minutes resistance decreases and reaches stable value. Liquid metal poorly wets thin film and it takes time for liquid metal to fill pinholes if such are present (see Fig. 13). Thin film resistance measurements are listed in Table 3. Resistivity is shown to be independent of the thin film thickness. On the other hand resistance of sputtered thin film differs significantly; 120 nm thick thin film's resistance is only 10 kΩ, but resistance of thin film double the thickness (240 nm) is ~200 times higher. The explanation

of this result was found after performing the AFM measurements of the thin insulating film. The surface of the layers showed to be very smooth, however at some locations of the film we found small holes which obviously had formed during the preparation of the film (see Fig. 13 c)). The width of the holes was typically on the order of 100 nm. Resistance variation can be explained by the amount and size of pinholes in thin film.

Flexographic printing is a wet coating method, where thin film is deposited from liquid. If there is any contamination on the substrate, the coating material does not wet it and forms a pinhole around it. The next layer of liquid coating material also forms a pinhole on most of the pinholes and fills some of them. Atoms and molecules in magnetron sputtering are more likely also to adsorb on contamination and form a thin film. By increasing thickness resistance per nm increases multiple times suggesting that the number of pinholes and/or their size reduces significantly.

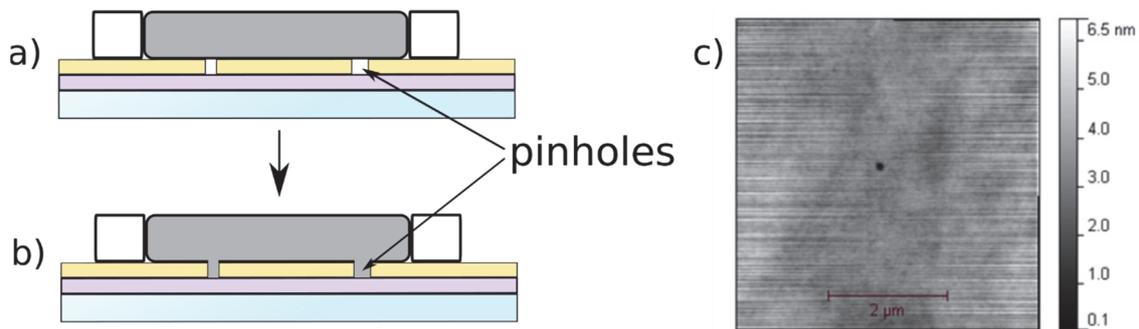


Fig. 13. a) Teflon ring is filled with liquid metal. b) Pinholes in a thin film are slowly filled with liquid metal reducing the resistance between contacts. c) The AFM image of flexoprinted SiO₂ surface.

Table 3.

Resistance measurements of SiO₂ deposited by flexography and reactive magnetron sputtering. Resistivity per nanometer is calculated by dividing the resistance of 145 mm² and thickness of thin film.

Deposition method	Thickness, nm	Resistance of 145 mm ² , MΩ	Resistivity, kΩ/nm
Flexography	185	0.55	3
	300	1.1	3.7
	465	1.45	3.1
Reactive magnetron sputtering	120	0.01	0.08
	240	2	8.3
	360	10	27.8

I-V characteristics for flexoprinted SiO₂ are shown in Fig. 14 a). Rapid increase of current means that dielectric breakdown takes place. Dielectric breakdown voltage increases with the increase of thickness of thin films from 10.8 V/μm to around 17 V/μm. I-V characteristics for sputtered thin films are shown in Fig. 14 b)–d). Thin film with thickness has very poor dielectric

resistance, it breaks down already at 0.25 V. Very low dielectric breakdown strength agrees with resistance measurements, where 120 nm thin films showed very low resistance compared to other thin films. For 240 nm and 360 nm thin films the current starts to increase at 2 V and 2.75 V, respectively, which is close to a flexoprinted one layer thin film. After reaching only 25–30 μA , the current decreases and saturates. In the second measurement loop, the current starts to increase at much higher voltage 7–8 V. It could be interpreted as a self-healing process taking place for 240 nm and 360 nm thin films. Apparently during dielectric breakdown temperature reaches high enough value for ITO to decompose at the location of breakdown forming non-conducting defect. In contrast to sputtered thin films, self-healing is not observed in flexoprinted thin films. The reason might be decomposition of thin film in high temperature, since flexoprinted thin films contain organic by-products of polysiloxane decomposition reaction.

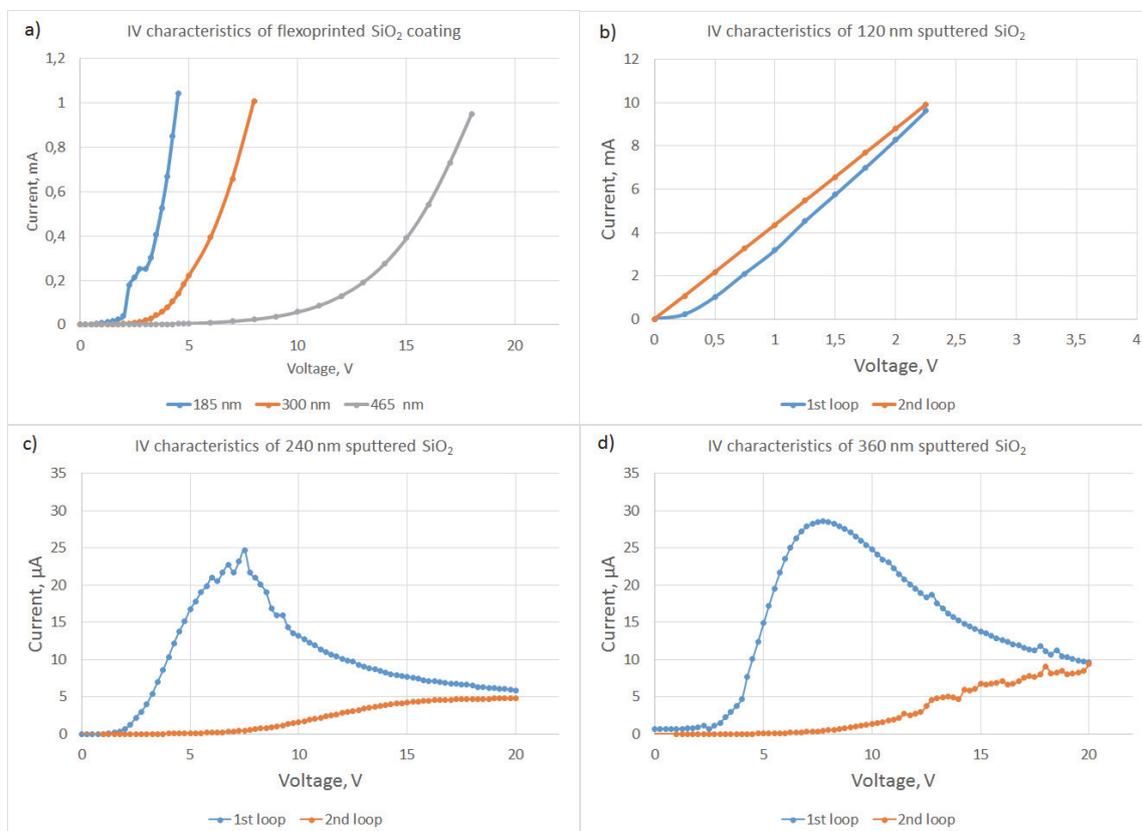


Fig. 14. Current-voltage characteristics of flexoprinted and sputtered SiO_2 thin films. a) For 185 nm thin film diel. breakdown occurs at 2 V, but for 300 nm thin film breakdown occurs at 3.5 V. For thicker thin films the increase is slower and breakdown occurs around 8 V. b) I–V characteristics for 120 nm thick sputtered coating. Current starts to linearly increase already after the first measurement point at 0.25 V. c) I–V characteristics for 240 nm thin film. Diel. breakdown starts at 2 V. Current reaches maximum 24 μA at 7.5 V and then decreases down to 5 μA . In the second loop the current starts to increase at ~ 7 volts and reaches 5 μA . d) I–V characteristics for 360 nm thin film. Current reaches maximum 28 μA at 7.75 V and then decreases down to 10 μA . In the second loop the current starts to increase at ~ 8 volts and reaches 10 μA .

Smart glass application requires high transmittance and colorless appearance. Flexoprinted SiO₂ thin film reduces the transmittance by 6–8 % compared to the uncoated substrate. It can be explained by thin film interference and by-product formation during polysiloxane decomposition, which increases absorption. It also explains yellowish-brownish sample colour. Sputtered SiO₂ increases transmittance up to 5 %. Light diffraction coefficient of sputtered SiO₂ is 1.5 and it works as antireflective layer between ITO ($n = 1.9$) and air ($n = 1$).

Dielectric breakdown of LCDs with dielectric thin films

Cholesteric shutters were made with three dielectric films: flexoprinted SiO₂, flexoprinted PI and magnetron sputtered SiO₂. Each thin film had various thickness starting from 100 up to 465 nm. Flexoprinted SiO₂ thin film displays have the highest dielectric strength and even one layer of 185 nm thin film is enough (see Fig. 15). Flexoprinted PI have lower dielectric strength than SiO₂. Sputtered SiO₂ have the lowest dielectric strength, but > 240 nm thin film is durable enough to be able to switch shutters with ± 200 V (40 V/ μ m).

Thin film breakdown versus LCD dielectric breakdown is shown in Fig. 16. Thin film breakdown was determined by I–V characteristics. As one can see from the graph, there is a very well visible correlation between the strength of thin film and liquid crystal display. It shows, that the properties of thin film determine the dielectric breakdown strength of LCD, and the dielectric breakdown is not caused by introduction of dust particles during manufacturing process.

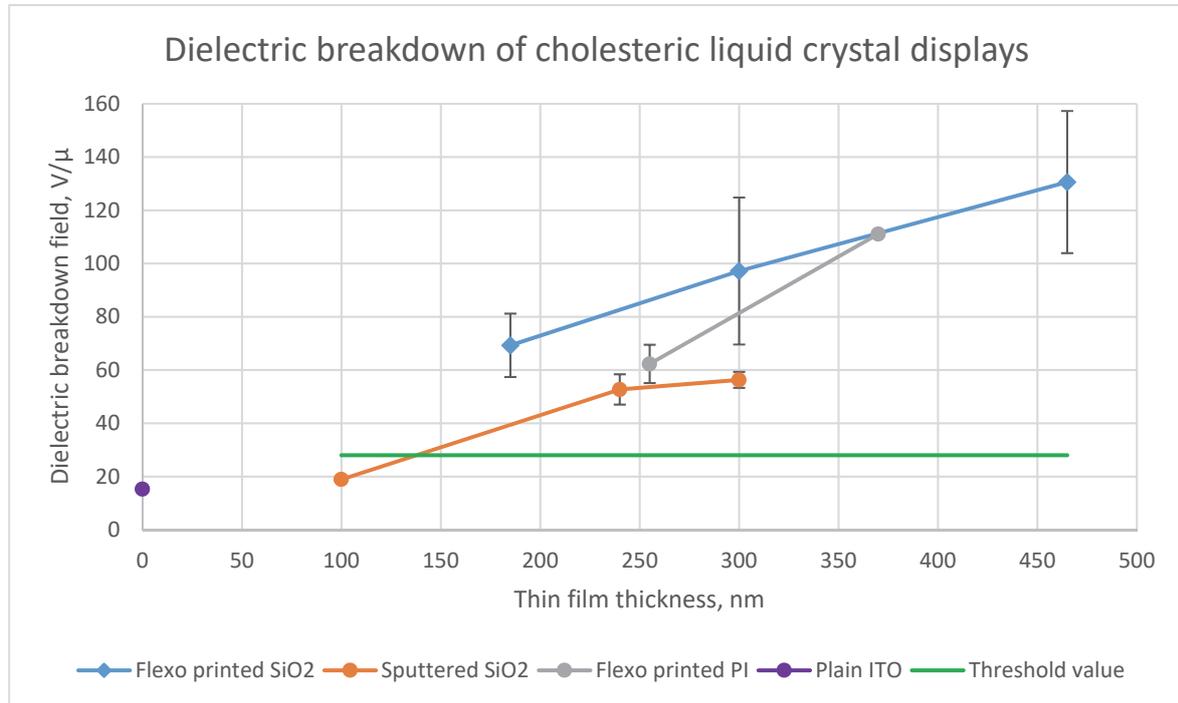


Fig. 15. Dielectric breakdown of cholesteric liquid crystal displays with various thin films: SiO₂ and PI deposited by flexography and sputtered SiO₂. Switching threshold value is depicted in green colour at 28 V.

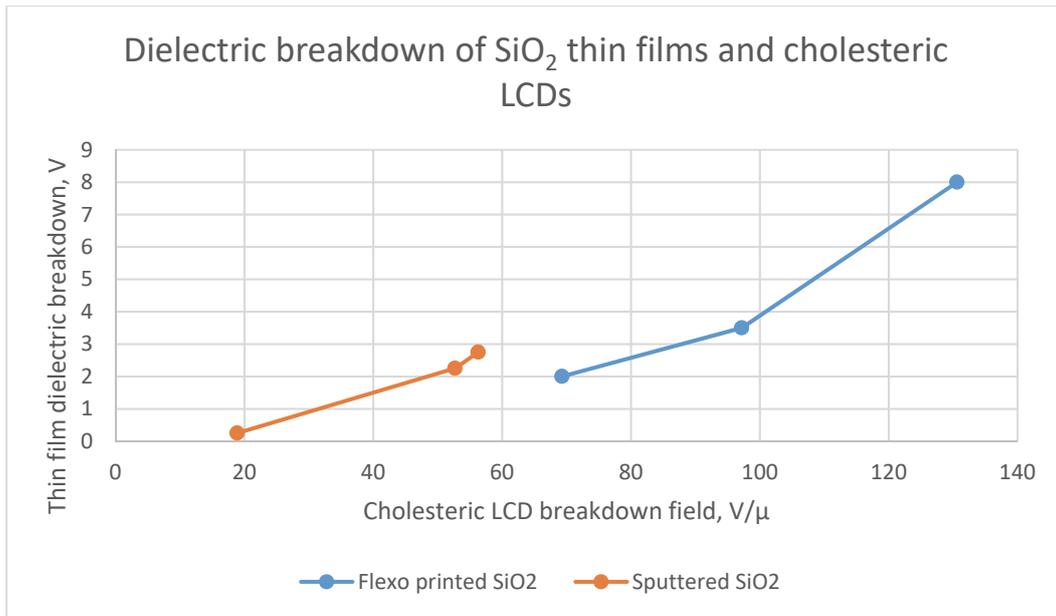


Fig. 16. Dielectric breakdown of flexoprinted SiO₂ and sputtered SiO₂ thin films are plotted in one graph with dielectric breakdown of cholesteric shutter displays.

Bistable displays were made with flexoprinted thin films and dielectric breakdown values were tested (see Fig. 17). Similar as for cholesteric shutters, the displays with flexoprinted SiO₂ have the highest dielectric strength. Displays with 300 nm and 465 nm thin films did not break down at the limit of measurement setup 200 V/μm. For a reliable device the thickness of both thin films should be > 200 nm.

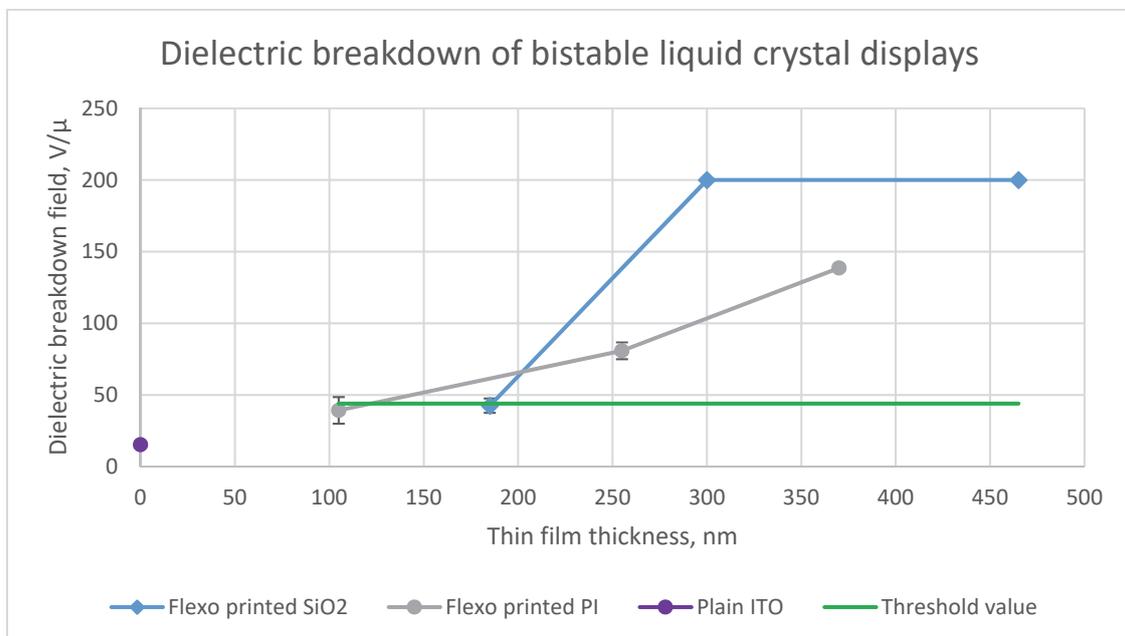


Fig. 17. Dielectric breakdown of bistable liquid crystal displays with flexoprinted SiO₂ and PI. Switching threshold is depicted for the thickest PI thin film at 44 V/μm. Measurement limit is 200 V/μm and the displays with PI > 300 nm did not break down. The breakdown value of display without dielectric thin film is added as a reference in purple.

Switching speed and transmittance of cholesteric liquid crystal displays.

For all three types of cholesteric display devices transmittance and switching speeds were measured. Results are summarized in Table 4. Transmittance and fall time varies with applied voltage, but is constant after threshold voltage, which is determined by voltage needed to switch to a fully open state. For flexoprinted SiO₂ threshold voltage is around 105 V and varies in the range of 3 V for samples with different dielectric coating. Only the rise time continuously decreases with increase of voltage. Switching to open state is compared as voltage needed to switch to open state in 1 ms and difference between various thicknesses is just a few volts. For the samples with sputtered SiO₂ the threshold voltage is higher (117 V) and the rise time voltage is more than 20 V higher. The threshold voltage difference is related to the difference in dielectric constant – for sputtered SiO₂ dielectric constant is 3.9, but for flexo printed SiO₂ dielectric constant is 14 according to material’s specification. For flexoprinted PI the threshold voltage varies from 112 V with 105 nm thin film up to 138 V for 370 nm thin film. Rise time voltage also is higher than of the SiO₂ thin films with similar thickness. Switching threshold voltage for the display with 370 nm PI thin film is ~40 V higher than for the display with 485 nm flexoprinted SiO₂. The main reason for this difference is a lower dielectric constant ($\epsilon = 2.9$) than that of SiO₂ thin films.

Open transmittance depends on the substrate transmittance. Substrates with the lowest transmittance (flexoprinted SiO₂) have also the lowest transmittance of display in open state. The main reason is the deposition method. In flexo printing method SiO₂ is formed by decomposition of polysiloxane and its byproducts that absorb light. LCD shutters with 300 and 485 nm flexo printed thin films have noticeable brownish colour. Another factor is the difference in refractive index. Sputtered SiO₂ ($n = 1.5$) and PI ($n = 1.66$) work better as an anti-reflective layer between ITO ($n = 1.9$) and air ($n = 1$) than flexoprinted SiO₂ ($n = 1.7$).

The displays with sputtered SiO₂ have the lowest closed transmittance. The reason for this effect is the different surface interaction with liquid crystal. Flexoprinted thin film is very smooth, but sputtered thin films have higher surface roughness, that induces LC scattering state.

Table 4

Summary of transmittance and switching data for cholesteric LCDs with flexoprinted and sputtered thin films. Listed results are measured at 140 V.

Deposition method	Thin film	Thin film thickness, nm	Dielectric constant, ϵ	Transmittance ITO glass + thin film at 550 nm, %	Transmittance in open state, %	Transmittance in closed state, %	Voltage to switch in 1 ms to open state, V	Fall time, ms
Flexographic printing	SiO ₂	185	14	81.7	83.5	7.3	141	0.56
		300		82.6	83.6	7.2	144	0.58
		485		79.9	82.5	7.4	144	0.57
	PI	105	2.9	92.7	89.9	5.6	159	0.64
		255		92	91.2	7.1	176	0.52
		370		91.3	89	6.3	186	0.56
Magnetron sputtering	SiO ₂	240	3.9	88.9	87.7	4.4	165	0.55
		300		93.7	87.8	6	168	0.56

For both types of bistable LCDs transmittance and switching speeds were measured. The results are summarized in Table 5. Switching threshold voltage for SiO₂ was between 140–150 V for all three thicknesses, while switching threshold voltage increases with increasing thin film thickness for PI in the range of 170–220 V. It can be explained by larger dielectric constant for SiO₂ ($\epsilon = 14$) than for PI ($\epsilon = 2.9$). Open transmittance similarly as in the case of cholesteric shutters correlated with the transmittance of substrates.

Similarly, as with displays with patterned electrode, overdriving could be observed. This is the reason why the rise time is so small, it takes a very short amount of time to switch from 50 % “closed state” to open state.

PI is more suited for smart glass application mainly because of higher transmittance in open state. Overdriving can be prevented by using lower voltage when switching to clear state than switching to the scattering state.

Table 5.

Summary of bistable display switching speed data and transmittance with SiO₂ and PI flexoprinted thin films. Listed results are measured at 200 V. For the displays with SiO₂ thin film transmittance in closed state is measured at 130V

Deposition method	Thin film	Thin film thickness, nm	Dielectric constant ϵ	Transmittance ITO glass + thin film at 550 nm, %	Transmittance in open state, %	Transmittance in closed state, %	Rise time, ms	Fall time, ms
Flexographic printing	SiO ₂	185	14	81.7	84.1	3.9	62	876
		300		82.6	82.3	3.6	56	707
		485		79.9	78.5	3.1	30	359
	PI	105	2.9	92.7	87.6	3.3	170	601
		255		92	85	2.8	194	929
		370		91.3	81	2.6	298	758

Liquid crystal displays with asymmetric design

Cholesteric LCDs with PI thin film only on one electrode were made and compared with LCDs with thin films on both electrodes. Displays with only one thin film layer would reduce manufacturing time and reduce cost of the device.

Dielectric breakdown field was tested for asymmetric cells and compared with the results of cells tested in chapter “Dielectric breakdown of LCDs with dielectric thin films”. Unfortunately, since there were no cells tested with 105 nm thin films on both electrodes, thickness does not overlap for each cell type (see Fig. 18). From the graph in Fig. 18 we can see that the displays with one 355 nm PI thin film have about the same dielectric strength as the displays with 510 (255+255) nm PI thin films. Apparently, one thick dielectric thin film has less defects that lead to dielectric breakdown.

Switching speed and transmittance are as important as dielectric strength. As cholesteric shutters are tested by applying voltage to only one of the electrodes, it needs to be tested whether it matters on which electrode dielectric thin film has been deposited. Displays were tested by applying voltage to both TOP and BOT electrodes and electrical and optical properties were compared. The results are listed in Table 6. As one can see from the values, the results are

almost identical meaning that asymmetric design does not cause asymmetric switching behaviour.

The displays with one and two layers of thin film were compared to see which design provides faster switching and higher transmittance. The display with one 255 nm thin film and displays with two 105 + 105 = 210 nm thin films were compared. The switching speed difference was negligible, but open transmittance with two layers had ~2 % higher transmittance. There are couple of reasons why the transmittance was higher for symmetric design. Firstly, symmetric design benefits from PI layers working as anti-reflective layer to increase transmittance. Secondly, asymmetric design has around 25 % thicker total thin film.

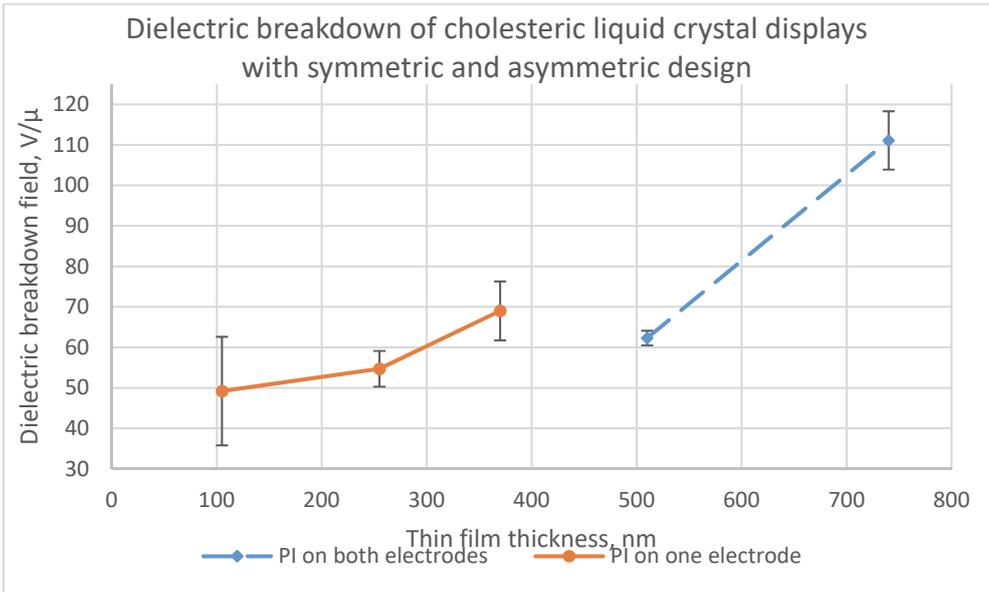


Fig. 18. Dielectric breakdown of cholesteric LCDs with dielectric thin film on both electrodes (dashed line) and only on one of the electrodes (continuous line). Thickness for all thin films inside the cells are added. LCDs with film thickness 510 nm have two dielectric thin films, each 255 nm.

Table 6

Switching and transmittance data of cholesteric LCDs with asymmetric design. Listed results are measured at 200 V

PI thickness, nm	Electrode on which voltage is applied	Transmittance in open state, %	Transmittance in closed state, %	Rise time, μs	Fall time, ms
105	TOP	88.2	6.2	463	0.58
	BOT	88.3	6.4	471	0.57
255	TOP	87.4	5.6	555	0.59
	BOT	87.5	5.6	556	0.60
370	TOP	87.2	5.9	611	0.60
	BOT	87.2	5.9	615	0.60

5. Dielectric breakdown mechanism in liquid crystal displays

Dielectric breakdown takes place in two dielectric mediums: thin film and liquid crystal. Dielectric breakdown strength of highly purified liquids is in range of 1–4 MV/cm [5]. Breakdown strength of highly purified solid dielectrics is of the order of 10 MV/cm [11]. Dielectric breakdown strength of cholesteric and bistable liquid crystal displays is much lower: 0.08–0.15 MW/cm. The values in this study are lower than the values found by Dierking (0.23–0.78 MV/cm) [4] but comparable. The reason for higher breakdown field in Dierking study most likely is related to much smaller size (1.5 cm²) of samples, where the probability of dielectric breakdown is smaller.

Dielectric constant of bistable and cholesteric liquid crystal used in experiments is not measured, but we can assume that it is in the range of 3.5–10 as measured in other studies. The values of dielectric constant are comparable with the dielectric constants of thin films. As thickness of LC is 20–100 x larger, majority of electric field drops on LC, not the dielectric thin film. For cholesteric LC threshold voltage without dielectric thin film is 108 V. By introducing dielectric thin film, threshold voltage is increased, because it reduces the electric field on liquid crystal. Depending on material the increase is up to 30 V. There is almost no increase of threshold value for flexoprinted SiO₂, whereas for polyimide the threshold value increases to ~8 V/100 nm thin film for cholesteric and to ~20 V/100 nm thin film for bistable devices. Insertion of sputtered SiO₂ increases the threshold value to ~3 V/100 nm. Threshold voltage increase is very small compared to the increase of dielectric breakdown voltage 350–1850 V. It means that the electric field reductions have a very small impact on dielectric breakdown strength.

After the dielectric breakdown of LCD, there was a black burned spot left in the middle of the breakdown spot and it was impossible to study the breakdown inducing defect. Dielectric breakdown speed was not measured, but during the switching speed measurement it was observed that dielectric breakdown takes place instantaneously after voltage has been applied, thus ruling out such breakdown mechanisms as thermal, erosion and electrochemical breakdown, since it takes seconds for them to take place.

In LCD without dielectric thin film, dielectric breakdown is most likely initiated by a defect in electrode, where electric field is enhanced and electrons are ejected in liquid crystal. If thin film is introduced, electrons in liquid crystal are ejected through pinholes. Thin film works as a current limiting layer. These electrons gain more energy due to the field, then loose during collisions and knock out other electrons thus initiating an avalanche process. When the avalanche process reaches the threshold value, the current builds up and the applied field collapses. Top and bottom electrodes are connected by conductive channel. It might be that dielectric breakdown takes place in spots were two pinholes align on top of each other. The process can be observed by spark and light emission. Due to high temperature, liquid crystal decomposes and electrode surface is damaged due to melting. There are two outcomes of dielectric breakdown:

- 1) self-healing takes place and LCD still remains operational with dielectric breakdown as a small visual defect;

- 2) dielectric breakdown forms a conductive channel, LC decomposes in large area and LCD does not switch anymore; due to high temperature the glass might heat up and crack (see Fig. 19).

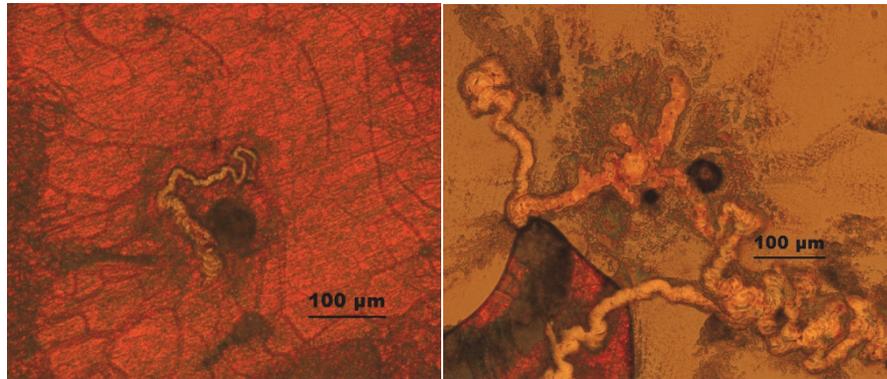


Fig. 19. Dielectric breakdown of cholesteric liquid crystal displays. a) Self-healing defect. There is a relatively small dielectric breakdown of $\sim 80 \mu\text{m}$ in the middle and a small gas formation around it. Red material around the breakdown spot is liquid crystal. b) liquid crystal has decomposed into gases around the centre of dielectric breakdown, forming a large dielectric breakdown area.

CONCLUSION

1. Liquid crystal displays without thin films or other modifications have low resistance to dielectric breakdown and large electrical field deviation. Electrode surface roughness in the range of few nm does not influence the strength of dielectric breakdown.
2. Inhomogeneous PEDOT:PSS smoothing layer decreases the liquid crystal display resistance to dielectric breakdown.
3. The possibility of liquid crystal display dielectric breakdown can be reduced by electrode patterning in stripes with resistance larger than $1.5 \text{ k}\Omega$. Unfortunately, electrode separation lines are visible in transparent state, therefore this method is suitable for fast switching cholesteric shutters.
4. LCDs with 200–450 nm SiO_2 and polyimide thin films deposited by flexographic printing have a higher dielectric breakdown field than the devices with sputtered SiO_2 thin films, but have lower optical transmittance and coloration can be observed. Colorless thin films are preferred for smart glass and shutters.
5. The main cause for dielectric breakdown in liquid crystal display is pinholes in thin films with the size of few hundred nanometers. Liquid crystal displays break down through electron avalanche mechanism.
6. The switching speed using electric field of up to $20 \text{ V}/\mu\text{m}$ of cholesteric LC with $80 \Omega/\text{sq}$ and $40 \Omega/\text{sq}$ ITO electrode is equal. It indicates that switching speed is limited by liquid crystal, not by the charging speed of the device and using lower resistance electrodes with lower sheet resistance does not increase the switching speed.

7. Asymmetric designed displays with dielectric thin film deposited only on one electrode have higher dielectric strength compared with the displays with two dielectric thin films with equally combined thickness, because one thicker thin film has higher dielectric breakdown strength compared to two thinner films. Open optical transmittance with only one thin film is 2–3 % lower, since the dielectric thin film works as an anti-reflective layer between the ITO electrode and liquid crystal.
8. Silicon dioxide thin films deposited by magnetron sputtering with thickness 200–300 nm are suited best for the smart glass and 3D volumetric display applications due to high optical transmittance and high resistance to dielectric breakdown.

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THESIS APROBATION

The main results and scientific research of the Thesis have been published in four full text papers and disseminated at two international scientific conferences:

Published papers

1. G. Mozolevskis, A. Ozols, E. Nitiss, E.Linina, A. Tokmakov, M. Rutkis, “Reduction of Electric Breakdown Voltage in LC Switching Shutters”, *Latvian Journal of Physics and Technical Sciences*, 2015.
2. J. Klavins, G. Mozolevskis, A. Ozols, E. Nitiss, M Rutkis, “Screen Printing of SU-8 Layers for Microstructure Fabrication”, *Latvian Journal of Physics and Technical Sciences*, 2015, (SCOPUS)
3. G. Mozolevskis, E. Nitiss, A. Medvids, “Electric breakdown of dielectric thin films for high-voltage display applications”, *Proceedings of SPIE – The International Society for Optical Engineering*, 2017.
4. G. Mozolevskis, I. Sekacis, E. Nitiss, A. Medvids, M. Rutkis, “Dielectric breakdown of fast switching LCD shutters”, *Proceedings of SPIE – The International Society for Optical Engineering*, 2017.

International conferences

1. G. Mozolevskis, E. Nitiss, A. Medvids, “Electric breakdown of dielectric thin films for high-voltage display applications”, *SPIE/COS Photonics Asia*, Beijing, 12–14 China, October, 2016.
2. G. Mozolevskis I. Sekacis, E. Nitiss, A. Medvids, M. Rutkis, “Dielectric breakdown of fast switching LCD shutters”, *SPIE Photonics West*, San Francisco, USA, 28 January – 2 February, 2017.