

Electro-Optical Properties of a Polymer Dispersed and Stabilized Cholesteric Liquid Crystals System Constructed by a Stepwise UV-Initiated Radical/Cationic Polymerization

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Abstract: Polymer-dispersed liquid crystal (PDLC) and polymer-stabilized liquid crystal (PSLC) are two typical liquid crystal (LC)/polymer composites. PDLCs are usually prepared by dispersing LC droplets in a polymer matrix, while PSLC is a system in which the alignment of LC molecules is stabilized by interactions between the polymer network and the LC molecules. In this study, a new material system is promoted to construct a coexistence system of PDLC and PSLC, namely PD&SchLC. In this new material system, a liquid-crystalline vinyl-ether monomer (LVM) was introduced to a mixture containing cholesteric liquid crystal (ChLC) and isotropic acrylate monomer (IAM). Based on the different reaction rates between LVM and IAM, the PD&SchLC architecture was built using a stepwise UV-initiated polymerization. During the preparation of the PDS&ChLC films, first, the mixture was irradiated with UV light for a short period of time to induce the free radical polymerization of IAMs, forming a phase-separated microstructure, PDLC. Subsequently, an electric field was applied to the sample for long enough to induce the cationic polymerization of LVMs, forming the homeotropically-aligned polymer fibers within the ChLC domains, which are similar to those in a PSLC. Based on this stepwise UV-initiated radical/cationic polymerization, a PD&SchLC film with the advantages of a relatively low driving voltage, a fast response time, and a large-area processability is successfully prepared. The film can be widely used in flexible displays, smart windows, and other optical devices.

Keywords: cholesteric liquid crystals; polymer; radical polymerization; cationic polymerization; electro-optical property; microstructure

1. Introduction

Polymer-dispersed cholesteric liquid crystal (PDLC) and polymer-stabilized cholesteric liquid crystal (PSChLC) systems are two important classes of liquid crystal (LC)/polymer composite materials [1–9]. The PDLC films exhibit a micro phase separation structure when LC droplets

are uniformly dispersed into the porous polymer matrix. In normal mode PDLC films scatter light, while on application of an external electric field they can be switched into a transparent state [10–14]. Owing to their advantages of facile formation, good film formability, and stable optical properties, PDLCs are used in a wide variety of applications, such as curtain free windows, displays, micro lenses, and light shutters, etc. [15]. However, the polymer network in the PDLCs lack directional orientation due to the weaker molecular interactions between the ChLCs and the polymer. The reorientation of LCs in the polymer network requires a high driving voltage.

On the other hand, an orientated liquid crystalline polymer network of LCs in a PSChLC system can facilitate cholesteric liquid crystal molecule re-orientation from a focal conic texture to homeotropic alignments upon the application of an electric field [16,17]. Thus, PSChLC requires a low driving voltage and has a prompt response rate. However, the low polymer content in PSChLC is usually less than 10%, causing poor mechanical strength [18–20]. Recently, a coexistent system combining the advantages of both polymer-dispersed and polymer-stabilized liquid crystals (PD&SLCs) has been developed by our group. This hybrid system is garnering substantial popularity in both academia and the industry due to its unique microstructure [21]. The produced electrically switchable PD&SChLC film not only shows a more than 50% decrease in driving voltage compared with conventional PDLC films but also possess high mechanical strength.

Generally, a two-step polymerization strategy is pursued to accomplish a PD&SChLC system. As reported in the literature, in the first step a porous polymer network is obtained by UV irradiation of isotropic acrylate monomers (IAMs). Secondly, an electric field is applied simultaneously with UV-light exposure, the liquid crystal acrylate monomers (LAMs) within the ChLC domains being crosslinked to form homeotropically aligned polymer fibers in the porous matrix [22–25]. During the first stage some of the LAMs are consumed by radical polymerization under UV light. To leave sufficient LAMs for forming the oriented polymer fibers in the second UV polymerization step, the curing time of UV irradiation in the first stage should be strictly controlled, which is not beneficial for the polymer morphology control or further optimization of the electro-optical properties of the PD&SChLC film [22–25]. To date, liquid-crystalline vinyl-ether monomers (LVMs) have not been investigated in constructing a PD&SChLC system.

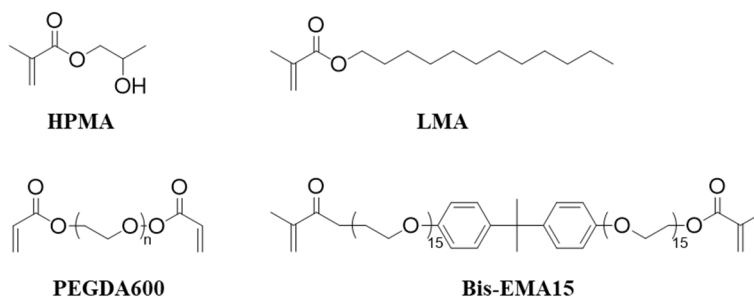
In this study, we propose a new materials system containing LVMs, ChLCs, and IAMs, and a PD&SChLC architecture is built stepwise by first consuming the IAMs via free radical polymerization, followed by crosslinking the LVMs within the ChLC domains via cationic polymerization. Due to the different rates between the radical reaction and the cationic one, the two polymerization steps are completely separated, and a coexist structure with homeotropically aligned polymer fibers inside the porous polymer matrix is formed.

2. Materials and Methods

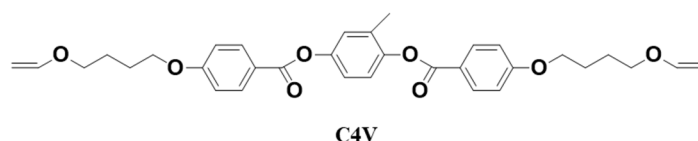
2.1. Materials

Figure 1 shows the chemical structures and physical parameters of the materials used in this study. Unless otherwise specified, the reagents were purchased from Sigma-Aldrich (St. Louis, USA). Transparent and conductive polyester substrates were gifted from Dalian Jingcai Smart Film Technology Co., Ltd. (Dalian, China). IAMs composed of lauryl methacrylate (LMA), polyethylene glycol diacrylate (PEGDA 600), and hydroxypropyl methacrylate (HPMA) were purchased from Tokyo Chemical Industry Co., Ltd. (Tokyo, Japan). LVMs and C4V were purchased from Beijing Green Innov-Tech Co., Ltd. (Beijing, China). Photo-initiators (PI), UVI-6976 and Irgacure 651 were bought, respectively, from Synasia (Suzhou) Co., Ltd. and TCI Co., Ltd. (Suzhou, China). Nematic LCs and SLC-1717 were purchased from Shijiazhuang Yongsheng Huatsing Liquid Crystal Co., Ltd. (Shijiazhuang, China). The chiral dopant, S811, was obtained from Beijing Gerui Technology Co., Ltd. (Beijing, China). Glass spacers with a diameter of 20 μm were obtained from Sekisui Chemical Co., Ltd. (Osaka, Japan) to control the thickness of the film.

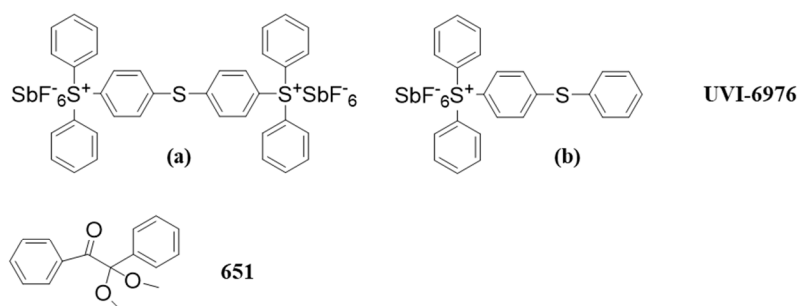
(1) Isotropic Acrylate Monomers (IAMs)



(2) Liquid Crystalline vinyl-ether Monmer (LVM)



(3) Photo-initiator (PI)



(4) Nematic Liquid Crystal

SLC1717, mixture of LCs with positive dielectric anisotropy

Cr 233.0 N 365.0 I

(5) Chiral dopant

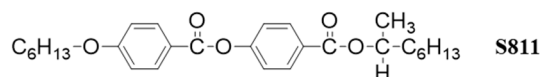


Figure 1. Chemical structures and physical properties of the materials used.

The composition of the IAMs is shown in Table S1. The ChLC comprises SLC177 and S811 and the corresponding composition is listed in Table S2. Meanwhile, the polarizing optical microscope (POM) figures of the ChLC are shown in Figure S1. The PI used in this study are UVI-6976 and Irgacure 651; their composition is listed in Table S3. All these materials were used without further purification.

2.2. Preparations of the Samples

Films A, B, C, and D, whose compositions are listed in Table 1, were prepared according to the procedures schematically illustrated in Figure 2. Firstly, a mixture of LVMs, ChLC, IAMs, photo-initiators, and glass spacers was stirred evenly until it became transparent. Then, the mixture was filled into two transparent and conductive polyester substrates via capillary action. Following this, the samples were cured under a UV light (365 nm, 3.0 mW/cm²) for about 80 s to form the PDLC-like

porous polymer structure. Next, a 100V, 1000Hz square wave electric field was applied across the samples to switch them into a transparent state. Simultaneously, a second-step UV light (365 nm) curing with the aforementioned intensity was employed for 10 min to fully consume the LVMs within the ChLC domains to form homeotropically aligned polymer fibers in the porous polymer structure. It is worth mentioning that all the films were prepared at room temperature.

Table 1. Compositions of the films with different LVM contents.

Sample	ChLC (wt%)	LVMs (wt%)	IAMs (wt%)	PI (wt%)
A	64.5	0	35.0	0.5
B	64.0	0.5	35.0	0.5
C	63.5	1.0	35.0	0.5
D	63.0	1.5	35.0	0.5

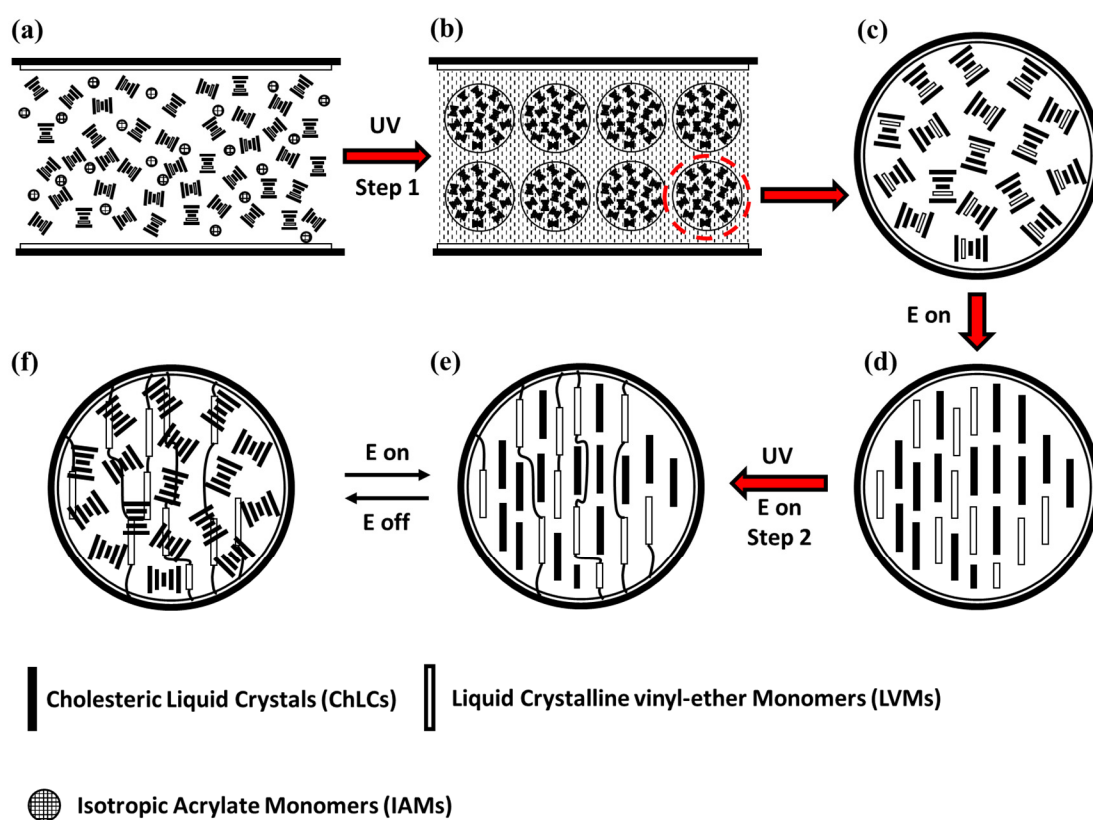


Figure 2. Schematic illustration for the preparation of the PD&SChLC film.

2.3. Measurements

The E-O properties of the samples were measured using a liquid crystal device parameter tester (LCT-5016C, Changchun Liancheng Instrument Co. Ltd., Changchun, China). Each film was applied with a square wave-modulated electric field with a frequency of 100 Hz during the measurement. The transmittance of conductive polyester substrates was normalized as 100%.

The morphologies of the polymer structure were observed via scanning electron microscopy (SEM, HITACHI S-4800, Tokyo, Japan). Cross-sectional SEM images of the samples were obtained by firstly breaking off the samples in liquid nitrogen and then eliminating the LC molecules by immersing the samples in a cyclohexane solvent for about seven days. After that, the samples were dried in an oven at 60 °C for about 3 h and coated with a thin layer of gold before SEM observation.

3. Results and Discussion

Figure 2 shows a schematic illustration of tailoring the PD&SChLC architecture in the film. First, a well-stirred mixture of ChLC, IAMs, and LVMs is sandwiched between two transparent and conductive polyester substrates (Figure 2a). The film shows a transparent state which is attributed to the isotropic nature of the mixture. After the film is irradiated, the porous polymer structure built by the IAMs is roughly formed, as shown in Figure 2b, in the first 80 s. The SEM photograph of the porous polymer morphology is also shown in Figure S2. Meanwhile, a preliminary phase separation between the cross-linked polymer and ChLCs (including LVMs) also occurs with the consumption of the IAMs. In this phase separation process, the optical appearance of the film turns from a transparent into a light scattering state due to the focal conic textures of the ChLCs, as shown in Figure 2c. It is worth mentioning that the polymerization rate of the acrylic monomer (which belongs to the radical reaction) is much faster than that of the vinyl ether monomers. Thus, the LVMs remain completely unpolymerized after this first step of UV irradiation [26–28].

Subsequently, an electric field is applied to homeotropically orient the ChLCs and the LVMs, and, accordingly, the film turns back to a transparent state, as shown in Figure 2d. Simultaneously, a second stage of UV polymerization is carried out using a high light intensity to consume the LVMs via cationic polymerization in order to form homeotropically oriented polymer fibers within the ChLC droplets, as shown in Figure 2e. During the second step the films are kept transparent via an applied electric field to ensure the homeotropic alignment of the LVMs. After the removal of the electric field, the film turns back to a light scattering state because the ChLC molecules return from the homeotropic alignments to the focal conic textures, as shown in Figure 2f.

To verify the coexistent PD&SChLC structure, SEM photographs of the polymer microstructures of the as-made films, from horizontal and cross-sectional perspectives, were taken. As shown in Figure 3a,c, the microstructures of the PDChLC film have a polymer matrix with a uniformed porous structure, with the size of each pore being about 3–5 μm , which is in agreement with typical PDLC materials. By comparing Figure 3a,b, we can see that with the introduction of LVMs, the homeotropically oriented polymer fibers can be successfully formed in the new materials system. Figure 3b shows the enlarged view of the coexistent porous polymer structure and the oriented polymer fibers. As shown in Figure 3d, the morphologies of the polymer configuration from the horizontal perspective are also distinct from those of the PD&SChLC film, compared with Figure 3c. From the above characterization, we can confirm that the coexistent PD&SChLC structure can be successfully achieved by a promoted stepwise UV-initiated radical/cationic polymerization strategy.

According to the above investigation, we deduced that the homeotropically-oriented polymer fibers formed by LVMs may bring new electro-optical properties to the as-made PD&SChLC film. Herein, a series of the PD&SChLC films were prepared by varying the LVMs contents, with the compositions of the films listed in Table 1. The results of the electro-optic properties of these films are summarized in Figure 4a,b. As the LVM content increases from 0 wt% to 1.5 wt%, the threshold voltage (V_{th}) and the saturation voltage (V_{sat}) of the films, which are defined as the voltage required when the film transmittance reaches 10% and 90% of maximum transmittance, respectively, are greatly reduced. Specifically, the V_{th} and V_{sat} of the films decreases from 43.3 V/88.7 V to 32.2 V/58.8 V, 25.5 V/44.6 V, and 21.2 V/35.5 V, as shown in Figure 4b. This electro-optical improvement can be attributed to the homeotropically oriented polymer fibers, which may reduce the anchoring force of the porous network while rotating the ChLC molecules through the applied electric field. In addition, obviously, the anchoring force can be further reduced with more polymer fibers formed via increasing the LVM content. If the LVM content is increased up to 2.0 wt%, the transmittance of the film under normal conditions will be greatly increased, which results from the stabilization effects of the high densities of the homeotropically-oriented polymer fibers, which reduce the contrast ratios (CR) of the films (Figure S3). Hence, the electro-optic properties of the films reached their best when the content of the LVMs was 1.5 wt% in our experiments.

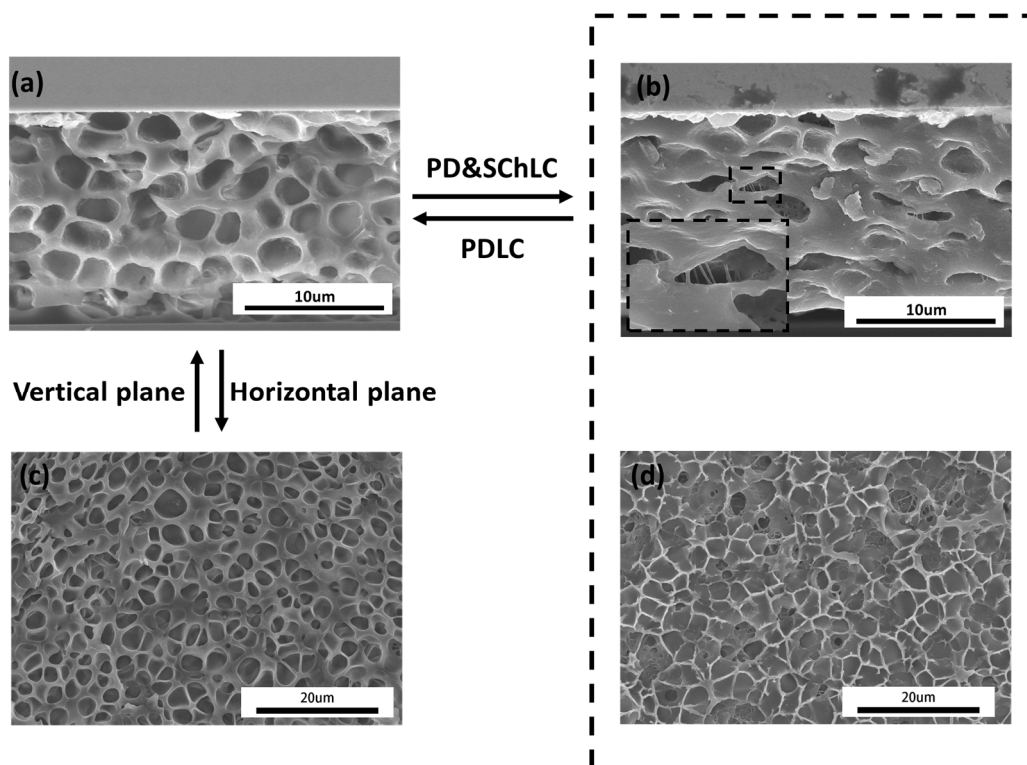


Figure 3. Scanning electron microscopy (SEM) photographs of the polymer microstructures for the as-made films from horizontal and cross-sectional perspectives: (a,c) are from the PDLC film And (b,d) are from the PD&SchLC film.

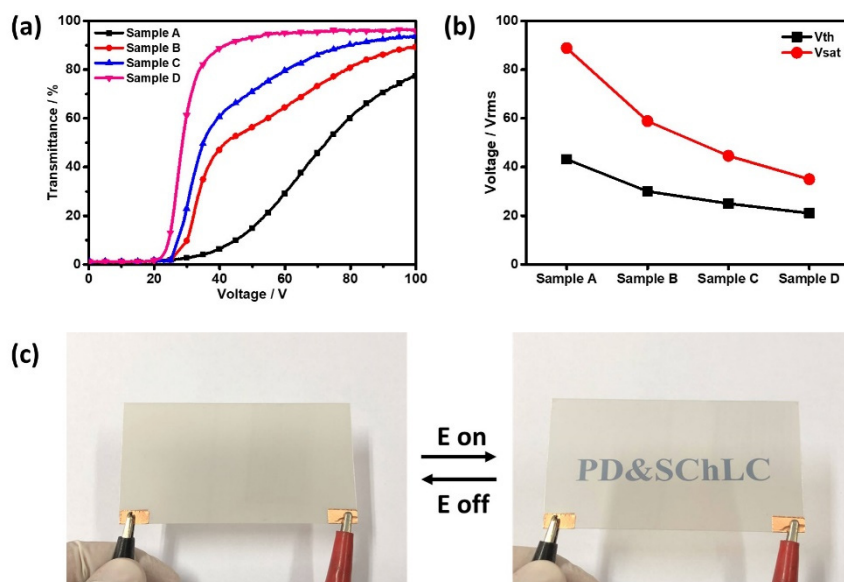


Figure 4. (a) Applied voltage dependence of transmittance of the as-made films containing A 0 wt%, B 0.5 wt%, C 1.0 wt%, and D 1.5 wt% LVMs, respectively. (b) The threshold voltage (V_{th}) and saturation voltage (V_{sat}) of the films. (c) Digital photographs showing the arbitrary transformation of Sample D from an intense light scattering state (left) to a highly transparent state (right) under electric field control.

Moreover, compared to the previously promoted PD&SchLC system, which had 3.0 wt% liquid-crystalline acrylate monomer content [21–25], having 1.5% LVM content in the new PD&SchLC system is the result of the two completely separated polymerizations steps. This is what we envisioned by introducing two means of aggregation, these being free radical and cationic.

Ultimately, the prepared films show a strong light scattering state under normal conditions, as shown in Figure 4c, and can achieve a fast transition to a highly transparent state under an electric field lower than 40 V. In addition, the response time for the electro-optical transition between the two states is fast, and the total time of the roundtrip is less than 30 ms, which is proved in Figure S4.

4. Conclusions

In conclusion, in this work a novel strategy for the preparation of PD&SChLC films by a stepwise UV-initiated free radical/cationic polymerization is proposed and validated. The new material system, containing LVMs, ChLCs and IAMs, is promoted to prepare a PD&SChLC film which has the advantages of low-driving voltage, easy preparation, good mechanical properties, and a fast response rate. Due to the reaction rate of the cationic polymerization being far slower than the radical polymerization, the two UV polymerization processes in the new system are able to be truly separated. Based on this, moving from 3.0 wt% to 1.5 wt%, the content of the LVMs can be decreased exponentially compared to the IAMs in the previous material system. Meanwhile, the homeotropically oriented polymer fibers constructed by the LVMs can effectively decrease the V_{th} and V_{sat} of the films from 43.3 V/88.7 V to 21.2 V/35.5 V. With these advantages, the new PD&SChLC system can be widely applied in flexible displays, smart windows, and other optical devices.

Supplementary Materials: The following are available online at <http://www.mdpi.com/2073-4352/9/6/282/s1>, Table S1. Weight ratio of the ChLC. Table S2. Molar/weight ratio of the IAMs we used in this work. Table S3. Weight ratio of the photo-initiator. Figure S1. POM textures of SLC 1717 and ChLC (97 wt% SLC1717 + 3 wt% S811). Figure S2. SEM photograph of the porous network in 80s. Figure S3. The contrast ratio of the films with different LVM contents. Figure S4. Response time of switching state conversion of the film.

Author Contributions: C.-Y.L. and X.W. contributed equally to this work. Conceptualization, C.-Y.L. and X.W.; methodology, X.L.; validation, C.-Y.L., X.W., and J.S.; formal analysis, C.-X.L. and S.-F.Z.; investigation, L.-Y.Z.; writing—original draft preparation, X.W.; writing—review and editing, C.-Y.L., X.W., and X.L.; project administration, H.-Q.Z. and H.Y.; funding acquisition, H.Y.

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Conflicts of Interest: The authors declare no conflict of interest.

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