Functional films using reactive mesogens for display applications

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ABSTRACT

Functional films made of reactive mesogens (RMs) are widely used in display devices such as liquid crystal displays (LCDs) and organic light-emitting diode displays (OLEDs). While functional RM films have been intensively developed for commercial application in the industrial sector, the fundamental studies on it in the academic sector are relatively limited. Here, functional RM films are reviewed in terms of their materials and fabrication processes as well as their applications in display devices. Also briefly reviewed is the recent research on them that may potentially be used in future display devices. This review was intended to provide an overall summary of RM applications in display devices and of the recent trends of the relevant studies to non-experts as well as experts in display applications.

1. Introduction

A reactive mesogen (RM) is a liquid crystalline mesogen containing a reactive end group that is mutually polymerizable under ultraviolet (UV) light illumination [1,2]. RMs have the intrinsic features of liquid crystals (LCs), such as self-assembly, anisotropic optical and dielectric properties, and controlled alignment on a treated alignment layer; at the same time, they can be polymerized to solid materials with their liquid crystalline alignments and anisotropic properties retained. A wide range of applications for RM materials has been proposed and developed based on two features: the mesogenic property and polymerization, particularly in display applications.

The applications of RM materials in display devices can be roughly classified into three types. The first type is the use of RM as an additive material for host LCs. Here, the RM is used to stabilize or solidify the host LCs. A well-known example is the polymer-sustained vertical alignment (PSA) mode [3–5], in which a small amount of additive RM forms a polymerized network on the surface of the substrates and memorizes the tilting direction of the host LCs. As a result, the PSA mode exhibits improved performance in terms of optical transmittance and response time. Another typical example is the polymerized blue-phase mode. The blue-phase mode liquid crystal display (LCD) has various advantages over other nematic LCDs, such as a fast response time (in the order of submilliseconds), and a wide-viewing angle. It has a critical limitation; however, the blue phase is stable only within a narrow range of temperatures. This problem can be overcome by stabilizing the blue-phase structure using a small amount of RM [6,7]. A polymerized blue-phase mode LCD with excellent thermal stability over a wide range of temperatures has been successfully demonstrated [8].

The second type of RM application is that of solidified RM films without using host LCs, in which the RM is the main material and photoinitiators and additives can be added. Unlike the polymer films that are fabricated using mechanical elongation, the optical properties of RM films can be more diverse depending on the alignment of the RM molecules. The alignment of the RM molecules can be arbitrarily controlled in the fluidic LC state before the RM molecules are polymerized to fix their optical properties. Hence, pre-designed optical anisotropy and spatial sectionalization can provide numerous different types of RM films that have different functions and uses, such as optical compensation films, selective color reflectors, anisotropic microlenses, patterned retarders for three-dimensional (3D) displays, optical communication devices, and holographic films.

The third type of application is for the improvement of organic electronics. Aligned reactive mesogenic luminescent materials were developed to improve the outcoupling efficiency of organic light-emitting diode (OLED) devices [9]. An aligned RM polymer was used as an electrode with anisotropic conductivity in organic thin-film transistor (TFT) devices [10]. RM is also widely
used in non-display applications such as photosensitive elastomers, holographic films, and topological studies.

In this paper, the applications of RMs as functional films are reviewed. In the academic sector, RM-added LC materials and organic electronics using RM materials have been relatively intensively studied and are well documented. Functional RM films have been mostly studied in the industrial sector, however, and their fundamental research results have not been documented well despite their applications. RM films can be used not only in liquid crystal displays (LCDs) but also in other displays, such as OLED displays, quantum dot light-emitting diode (QLED) displays, visual reality displays, and projectors. Hence, a review of RM films will be helpful for the researchers in all display fields.

2. Chemical structure of RM monomers and RM film fabrication

The chemical structures of RM monomers are mostly composed of a mesogenic core group and photopolymerizable end groups [1]. The core group can be either a calamitic or a discotic liquid crystalline monomer, which can have chirality to induce a helical deformation in the RM alignment. The end groups are mostly acrylic or methacrylic groups that are easily polymerized, but any type of polymerizable group can be used. The polymerization process of the acrylic or methacrylic groups is usually conducted under an N₂ atmosphere to minimize the action of O₂, which can disturb the polymerization reaction. Diacrylate-type RM monomers with acrylate functional groups at either end (Figure 1(a)) are used to form a solid RM network, as illustrated in Figure 1(b). Similar to the LC mixtures used in LCD devices, a mixture of multiple RM compounds is commonly used in actual applications [11,12]. An RM solution is prepared by dissolving a mixture of RM monomers in an organic solvent, such as dichloroethane or dichloromethane [13].

The roll-to-roll process is commonly used to fabricate an RM film for commercial products. The roll-to-roll process is commonly used for the photoalignment layer, but a surface-treated polymer layer can be used as well. After exposing the photoalignment layer to UV using polarized UV light, the substrate is coated with an RM solution using the roll-coating method in mass production, or using the spin-coating method for research purposes. As the solvent evaporates, the RM solution becomes a liquid crystalline phase. Owing to the alignment layer beneath the RM layer, the RM monomers align along the easy direction of the alignment layer. Depending on the type of alignment layer used, a planar, patterned, tilted, or homeotropic alignment can be obtained, as shown in Figure 2. The chirality of the RM compound or the addition of a chiral dopant can induce additional structures (e.g., a twisted or cholesteric alignment) in the RM. In addition, a certain type of RM mixture is likely to vertically align on the interface with air; a splayed alignment can be obtained on a planar alignment layer owing to the competition between the planar (or tilted) alignment near the bottom surface and the vertical alignment near the top surface (the last image in Figure 2(b)). At this stage, the RM film is in the liquid crystalline phase at room temperature. When the aligned RM layer is exposed to unpolarized UV light, it becomes a polymerized rigid film. If the film is heated above a certain temperature before UV exposure, the RM can transit to the isotropic phase. Once the film is polymerized through UV exposure, however, the alignment is fixed and becomes thermally stable [14].

Thus, RM films have unique advantages compared to conventional polymer films manufactured using the mechanical elongation process. The manufacturing process of RM films is comparably simple, and the roll-to-roll process reduces the fabrication costs. Moreover, the thermal, mechanical, and chemical stabilities of RM films are usually excellent, and the patterning of the films is easily achievable. Various types of RM alignments enable a wide range of optical functions to be devised. A conventional polymer film has a ~100 μm thickness, but RM films are usually about 5 μm thick. Therefore, the RM can be used inside a display as an in-cell-type layer by directly coating the substrate with an RM solution [15].

3. RM films for optical compensation and retarder

The LC layer in an LCD has varying optical retardation values depending on the viewing direction, and it
Figure 2. (a) Optical compensation films using calamitic LCs. From left to right, planar, homeotropic, and patterned alignments are applied. Each corresponds to a positive A plate, a positive C plate, and a patterned A plate film, respectively. (b) Optical compensation films using discotic LCs. Planar, homeotropic, and tilted splay alignments are applied, respectively. Each corresponds to a negative C plate, a negative A plate, and a wide-viewing film for TN LCDs, respectively. (c) Films using cholesteric (CLC) RM, negative C type, and biaxial films can be obtained.

can cause the viewing angle performance to deteriorate. Image quality deterioration takes the form of low contrast ratio, color purity decrease, or color shift in the off-axis directions. To overcome the poor off-axis image quality issues, optical compensation films that are fabricated by elongating polymers along one or two directions are typically used. Due to the aforementioned advantages of RM films over the conventional polymer films, there have been many attempts to replace polymer compensation films with RM films [14–16]. The main advantages of RM retardation films for optical compensation are (i) a wide range of birefringence structures, (ii) micropatterning of retardation films, and (iii) in-cell-type fabrication using coating processes.

Figure 2 shows various types of compensation films obtained by combining RM type (calamitic or discotic), alignment type (planar, homeotropic, or tilted), and chirality (with or without a helical structure). Using calamitic RM compounds, a positive A-type film can be obtained on a planar alignment (Figure 2(a)). A positive A-type film has $n_x > n_y = n_z$, where $n_x$, $n_y$, and $n_z$ are the refractive indices of the two orthogonal axes within the film and along the normal direction to the film, respectively. On the other hand, a film of calamitic RM on a homeotropic alignment layer can be a positive C-type film, which has $n_z > n_x = n_y$. In addition, patterned retarders are frequently fabricated using a calamitic RM alignment. Using a discotic RM, negative C-type and negative A-type films can be fabricated on planar and homeotropic alignment layers, respectively, as shown in Figure 2(b). Here, negative C-type and negative A-type films have $n_x = n_y > n_z$ and $n_x < n_y = n_z$, respectively. A certain type of discotic RM mixture is likely to vertically align on the interface with air, and a splayed discotic alignment, as shown in the last image in Figure 2(b), is obtained, which is often used to compensate for twisted nematic (TN) LCDs [17].

In the case of in-plane switching (IPS) LCD modes, a combination of positive C-type and positive A-type compensation films is used [18,19]. In particular, the positive C-type film is difficult to fabricate in the conventional polymer films, but it can be easily obtained in RM films by vertically aligning the calamitic RM monomers on the substrate, as shown in the second image in Figure 2(a).

In vertical alignment (VA) LCDs, two approaches are used. First, the combination of negative C-type and positive A-type films can be used. In the second case, biaxial-type films are used; this is more common. Here, the biaxial film has $n_x > n_y > n_z$. The negative C-type RM film can be obtained using cholesteric liquid crystal
CLC RM compounds with high helical twist power (Figure 2(c)), in which the optical helical pitch does not overlap with the wavelengths of visible light [20]. A technology for fabricating biaxial RM films using CLC RM materials containing dichroic photoinitiators was also developed through exposure to polarized UV light instead of unpolarized UV light [16,21]. As shown in Figure 2(c), the refractive index along the direction of the polarization of light is greater than that normal to it within the film plane. By using a compensation film in VA LCDs, the viewing angle performance can be dramatically improved, and this is regarded as a sine qua non in VA LCDs [16].

A quarter waveplate (QWP) film is a positive A-type compensation film that has the birefringence of a quarter of the wavelength of visible light, and is widely used in displays, such as LCDs and OLEDs [22]. In particular, the antireflection (AR) film of an OLED is composed of a linear polarizer and a QWP film with a 45° rotation, in which the QWP changes linearly polarized light into circularly polarized light for incident light, and vice versa for reflected light, as illustrated in Figure 3(a). To obtain low reflectance over the entire range of visible wavelengths, the polarization conversion in the QWP should span the entire range of visible wavelengths; hence, the wavelength dispersion of birefringence is the determinant factor for obtaining low reflectance. For this purpose, various types of RM films, such as multiple layers of twisted RMs [23], a single RM layer using a biaxial RM material [24], an RM layer using a guest-host mixture [25], and a combination of an RM layer and a polymer layer [26], have been proposed.

4. Patterned retarder film using RM

A patterned compensation film that has multiple microdomains with either different retardation values or different optical axes is difficult to obtain in the conventional fabrication process by elongating polymer films, but a patterned film is achievable using the RM process. The fabrication of patterned retardation films can be done using two approaches: thermal treatment [27] and patterned photoalignment [28,29]. In the thermal treatment method, a film containing isotropic and nematic RM phases is fabricated following the process below. A substrate with an alignment layer is coated with an RM solution, and a uniform nematic alignment is obtained by evaporating the solvent. Then UV light is introduced through a patterned photomask to polymerize the domains exposed to UV light. In the next step, the film is heated for the RM in the unexposed regions to transit to the isotropic phase. By introducing the UV light for a second time at a high temperature, the isotropic RM alignments are polymerized. In this way, the nematic domains with optical birefringence and isotropic domains with zero birefringence are separately polymerized. The process is illustrated in Figure 4(a).

The second approach is the patterned photoalignment method introduced by Shannon et al. [30]. A micropatterned photoalignment layer can induce patterned RM alignment on it, whose process is illustrated in Figure 4(b). In this case, all the microdomains are in the nematic phase, but the alignment direction of the nematic RM varies depending on the domain. Various photoalignment methods have been suggested to induce patterned RM films [28]. Another approach to fabricating a micropatterned isotropic-nematic RM film was proposed, using the dielectrophoretic method in the isotropic-nematic mixture state [31].

A typical application of patterned retarders is the film-type patterned retarder (FPR) for 3D displays, which are fabricated via patterned photoalignment. Figure 5(a) shows the working mechanism of a spectroscopic 3D display using an FPR film, which is composed of interdigitated horizontal stripes of two types of retarders. The retardation values of the two retarders are the same as quarter light wavelengths, but the directions of the optical
Figure 4. (a) Manufacturing process of a patterned RM film via the thermal process. (b) Photoalignment method using multiple photomasking illuminations. The actual manufacturing process is conducted on a roll-to-roll system.

Figure 5. (a) Schematic of a stereoscopic 3D display using an FPR film. (b) Schematic illustration of the images seen through the left and right sides of 3D glasses.

axes are 45° and 135°. That is, the FPR film is a patterned QWP film with two different optical axes. The FPR film converts the linearly polarized light from odd and even horizontal lines into right- and left-handed circular polarizations, respectively. The 3D glasses have left- and right-handed circular polarizers for the left and right sides of the glasses, respectively. Therefore, the left and right eyes can see odd and even lines through the 3D glasses, respectively. In this way, stereoscopic 3D imaging is accomplished, as shown in Figure 5(b). The vertical resolution is reduced to half in the FPR-type 3D display, and the FPR film is expensive. The FPR-type 3D display, however, has several important advantages compared to the shutter-glasses-type 3D display (another well-known stereoscopic 3D technology). Unlike the shutter-glasses-type 3D display, flickering does not appear in the FPR 3D display; the 3D crosstalk is relatively weak [32]; and lightweight, batteryless 3D glasses are used. As such, the FPR 3D display is regarded as the most competitive stereoscopic 3D display technology at present [33].

Another application of the patterned retarder can be found in the transflective LCD, which reflects and transmits light simultaneously [15]. The pixel in a usual transflective LCD consists of two areas: transmission and reflection subpixels. Transmissive LCDs usually do not require QWP films, but reflective LCDs require QWP films to produce a good black state with low reflectance. Hence, an in-cell-type patterned retarder using RM can provide an excellent solution to the foregoing. The QWP is located in the reflection area, and the isotropic film is located in the transmission region [15].

5. Morphological shaping for microlens systems

Stereoscopic 3D displays that do not use glasses mostly use either the lenticular lens technology or the parallax barrier technology [34]. A lenticular lens is a film with a microcylindrical lens array, and it refracts the light from each pixel to two different directions to implement
Figure 6. 2D-3D conversion film for lenticular-lens-type stereoscopic 3D displays. (a) Tunable-LC-lens-type lenticular lens system. The concave lens could be an RM material. (b) Polarization-switching-type lenticular lens system. The lenticular lens itself is an RM material.

stereoscopic imaging [35]. The parallax barrier screens different pixels depending on the viewing direction, and hence, the left and right eyes see different pixels, perceiving a stereoscopic image. The 3D displays with no lenticular-lens-type glasses have advantages over the parallax-barrier-type 3D displays in terms of resolution and optical efficiency.

In the lenticular-lens-type 3D displays, switching between the 2D and 3D modes is a challenging task, and a tunable lenticular lens can provide a solution for this [36,37]. Several types of tunable lenticular lens have been proposed, and one well-known method involves using a tunable LC lens within a concave lens cavity, as shown in Figure 6(a). In this method, the LC layer is switched by applying electric fields. At zero field, the LC directors align along the long axis of the lenticular lens, and polarized light is focused using the refractive index difference between the LC layer and the solid concave lens, which is now in the 3D display mode. When an electric field is applied, the LC directors stand vertically, and the refractive index difference disappears along the light polarization direction. Hence, the light is not focused, and the display shifts to the 2D mode. The tunable LC system has a simple structure but has several limitations. To obtain a short focal length for the LC lens, the LC lens should be sufficiently thick. If the thickness increases, however, the applied voltage and the response time increase as well. To address this issue, a tunable lenticular lens using an anisotropic RM lens and a polarization-controllable LC cell has been proposed [36,38]. The structure, as shown in Figure 6(b), is more complicated than the tunable lens system in Figure 6(a). The 2D-3D switchable lenticular lens has a fixed RM lens with an anisotropic RM alignment that can focus only one type of polarized light. An LC layer that can control the polarization direction of light is placed below the lenticular lens. In the initial stage, the LC layer, which is usually a TN phase, polarizes the light parallel to the lenticular long axis. Then the light is focused by the RM lenticular lens, which converts the display to the 3D mode. On the other hand, when an electric field is applied in the LC cell, the polarization of light is perpendicular to the lenticular lens long axis, and the light is not focused by the RM lens, which is in the 2D mode. A polarization-dependent microlens array using RM has also been proposed [37].

6. Linear and circular polarizers using RM and a brightness enhancement film

Dichroic polarizers using an elongated polyvinyl alcohol (PVA) film are widely used in displays, but PVA-type polarizers cannot be used within a cell in LCDs. An in-cell-type polarizer was proposed when using RM materials with added guest dichroic dyes [39]. Guest-host-type RM polarizers have relatively low degree-of-polarization values due to the low-order parameters of the dichroic dye molecules. It has been reported that the use of smectic (or lamellar) RM compounds increases the degree of polarization [40]. Using guest-host-type RM materials, a micropatterned polarizer was demonstrated [41]. Figure 7(a) shows the dichroic transmittance and polarization efficiency of a guest-host-type RM film, and Figure 7(b) shows a micropatterned polarizer using the material.

A CLC RM film with a left- (or right-) handed helix reflects the left- (or right-) handed circular polarized light and transmits the other circular polarized light due to the photonic crystalline feature of the CLC RM film, as illustrated in Figure 8(a) [14]. The wavelength band of reflected light is from \((n_o \times p)\) to \((n_e \times p)\), where \(n_o\), \(n_e\), and \(p\) are the ordinary and extraordinary refractive indices of LC and the helical pitch, respectively. The bandwidth is usually 50–100 nm. Hence, an ordinary CLC RM film reflects only a certain color of light in the visible wavelengths. Using multiple RM layers with different helical pitches or a CLC RM film with a gradually varying helical pitch, however, an RM film with a broad reflection band that fully covers the visible wavelengths was developed (Figure 8(b)) [42,43]. The CLC RM films can reflect one type of circular polarization and transmit the other type of circular polarization – i.e. they work as circular polarizers. Similar to the guest-host-type linear RM polarizers, however, the degree of polarization of the CLC RM films is not high enough for them to replace the PVA-type polarizer. On the other hand, a CLC RM
film can be used as a brightness enhancement film on the backlight unit of an LCD, instead of the Brewster’s-angle-type brightness enhancement film called ‘DBEF’ [14]. To use the brightness enhancement film, a QWP is used together with a CLC RM film. The circularly polarized light transmitted by the CLC film is turned into linearly polarized light by the QWP, and the linearly polarized light can pass the polarizer in the bottom of an LC cell with only a slight optical loss. The mechanism is illustrated in Figure 9; by repeating the reflections between the CLC film and the reflector in BLU the brightness is significantly enhanced. The CLC RM film can enhance the optical efficiency by 160–180% [43].

It was proposed that a patterned CLC RM film could be used as a color reflector for color-reflective LCDs without a color filter [44]. As the CLC RM film selectively reflects light in the reflection band, patterned red, green, and blue RM films can function as both reflectors and color filters. In addition, a photoluminescent CLC RM film can enhance the reflection efficiency and can be used as a smart reflector for a reflective-emissive display device, in which the photoluminescent CLC RM film can reflect the external light and can emit light simultaneously [45]. Figure 10 shows a reflective-emissive electrowetting display using a smart reflector.

7. Recent academic studies using photoresponsive RM

In the academic sector, the research using RM materials is diverse. One recent frequent research topic is the photoresponsive elastomer and 4D printing technology [46,47]. Polymerized LC films containing azo compounds can shrink or stretch depending on the conformational change of the embedded azo compounds. By properly designing the initial alignment of the RM layer, the azo-RM film can actuate, change its shape, roll, and lift loads, responding to external light irradiation, as shown in Figure 11(a). Although most of the related studies aim to develop soft robotics and microrobotics, the morphological changes of films may be applied to a future tactile display technology [48].

Another interesting research topic is the drawing of arbitrarily shaped 3D microparticles using a confocal...
Figure 8. (a) Schematic structure of a circular polarizer using a CLC RM film. (b) Reflection band measured as transmission loss for a usual CLC film and a broadband CLC film with gradually varying pitches. (Reproduced from [43] with permission. Copyright 2002 Elsevier.)

Figure 9. Schematic mechanism for brightness enhancement using a CLC RM film. A normal polarizer ideally transmits 50% of the light from the BLU (left side). On the other hand, the polarizer with a CLC RM film recycles the reflected light, and the transmitted light intensity increases dramatically.

two-photon laser scanning photopolymerization setup. Martinez et al. demonstrated that microphotopolymerized knotted particles can be fabricated using the said method [49]. The method can be useful for the study of topological nematics and even for designing metamaterials for display applications (Figure 11(b)).

Figure 10. Reflective-emissive electrowetting display using a photoluminescent CLC RM reflector. The brightness is controlled in either the reflection or emission mode. (Reproduced from [45])

Holographic films can be fabricated using RM films containing azo compounds, which allow the photowritable alignment of RM [50]. The written holographic patterns
can be used as optical information storage devices, security cards, and holographic imaging films.

8. Conclusions

Functional RM films were reviewed herein, including their fabrication processes and applications. Despite their wide range of applicability and the intensive development in industry, the academic research on them has been relatively less intensive. The use of RM materials can improve the productivity and applicability of various functional films and microlens systems. In addition, the various optical functionalities of RM films, such as patterned optical retardation, selective photonic crystalline reflection, and microstructured optical lensing, can be highly useful even in the next-generation display technologies, including the QLED and micro-LED (light-emitting diode) displays. Three-dimensional (3D) RM particles produced using confocal laser scanning have attracted interest of late owing to the unique 3D geometry of optically anisotropic materials and their interaction with the nematic fields. Thus, both the applications of and the fundamental studies on RMs are expected to increase, and the advances in nano and microscopic fabrication methods along with an understanding of the synthesis and optical properties can play a key role in developing future technologies of RM applications.

Disclosure statement

No potential conflict of interest was reported by the authors.

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