Recent trends in retardation films

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INTRODUCTION

Liquid crystal television has quickly acquired worldwide popularity. The liquid crystal display (LCD) around which the receivers are constructed is superior to the cathode ray tube in its capacity for miniaturisation, increased screen area, and reduced power consumption, and has numerous other uses, as in computer displays, mobile telephones, games machines, and digital cameras.

A characteristic feature of LCDs is that they use many polymer materials as key components. Examples are the liquid crystal and colour filter organic materials, photoconductive sheet, and plastic optical film including diffuser, polariser and retardation films [1].

Since the quality of the LCD materials is evaluated in terms of screen image, i.e. by that most sensitive of the human senses, vision, the materials must meet stringent requirements in both performance and quality. Hence, the optical film of plastic materials used in LCDs is subject to extremely rigorous quality control of physical properties and surface parameters.

This review focuses on the retardation films used as optical film for LCDs, examining the film properties and method of manufacture. Firstly, a brief account of the operating principles and optical properties of LCDs is given; the processes used to form the base film, such as solution casting and melt extrusion, are then examined, together with the component technologies of the functionalisation process, notably stretching and coating.

1. OVERVIEW OF RETARDATION FILMS

1.1 Liquid crystal displays

The principle of the liquid crystal display (LCD) was discovered by Williams and Heilmeier at the RCA Corporation, whose device became the forerunner of today’s displays [2]. The inventors showed that when voltage is applied to a liquid crystal the molecular orientation changes, allowing control over the transmission of light. The press release by RCA in 1968 initiated competition to develop the LCD as a candidate second generation display. However, display quality was not entirely satisfactory owing to the problem of colour cast arising from orientation of the liquid crystal molecules. It was retardation film that provided a solution to this problem.

Figure 1 is a schematic diagram of the current make-up of television LCDs, which use numerous plastic optical films including retardation film to achieve high contrast and wide viewing angle.

![Figure 1. Composition of an LCD](image-url)
Figure 2 shows how LCD optical films are classified. They divide broadly into three kinds: polarisers, retardation films and backlight film. Retardation films are further divided according to the corresponding type of LCD, each of which requires different materials and manufacturing processes.

The initial development of LCDs encountered a wide range of problems, including the slow response of liquid crystals to change in the electric field, but the development of liquid crystals of rapid response and new display drivers has now made it possible to realise, amongst other things, the contemporary liquid crystal television receiver with high quality display capability.

At the development stage, the viewing angle problem and problems with contrast and colour cast encouraged the view that LCD could not provide a display good enough for television. Nowadays, however, we can watch screens at least as good as a conventional vacuum tube display. Optical compensation with retardation film is one of the technologies that made this advance possible.

1.2 Types of retardation film

A classification of the retardation films corresponding to different liquid crystal modes is shown in Table 1. Corresponding retardation films have been developed for each of the liquid crystal display modes and are now being used to enhance display quality. They may be separated by manufacturing process into films made with film stretching technology [3] and films made with liquid crystal coating technology [4, 5]. Both technologies employ the same basic concept of imparting optical anisotropy to achieve optical compensation. However, because of structural differences, etc, they have different optical characteristics: stretched film is used for LCDs operating in VA (vertical alignment) or IPS (in-plane switching) mode, while retardation film of the liquid crystal coating type is used chiefly for STN (super-twisted nematic) and TN (twisted nematic) displays.

Stretched film itself must be further selected to suit the display mode, since the optical characteristics are dependent on the film material used. The largest difference is in the intrinsic birefringence of the materials. If the refractive index is increased in the direction of stretching, the material is said to have positive intrinsic birefringence; when the opposite is true, the material is said to have negative birefringence. The respective optical characteristics are very different. In general, however, few materials have negative birefringence and the choice is limited. The same considerations apply to the selection of coated retardation film using liquid crystalline molecules.

Another important property of the materials is their wavelength dispersion characteristic. Wavelength dispersion refers to the wavelength dependence of refractive index arising when light travels through matter. Refractive index is independent of wavelength in a vacuum (much the same applies to air) but when light

<table>
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<th>Table 1. Liquid crystal modes and the corresponding retardation films</th>
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<td>STN</td>
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<tr>
<td>(1) Uniaxial retardation film</td>
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<tr>
<td>(2) Biaxial retardation film</td>
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<tr>
<td>(3) Liquid crystal film (liquid crystal twisted orientation)</td>
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<td>(4) Liquid crystal film (liquid crystal oblique orientation)</td>
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<td>(5) Half wave, quarter wave plate</td>
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travels through matter, the refractive index varies with wavelength. As a result, the state of polarisation of light transmitted through the anisotropic medium of a liquid crystal or retardation film changes during transmission. These properties are generally expressed by the wavelength dispersion characteristic of retardation, which may be divided into normal dispersion, flat dispersion or inverse dispersion according to the material, as illustrated in Figure 3. Retardation is defined as the difference in the two principal refractive indices in the film plane for light of perpendicular incidence, multiplied by the film thickness (q.v. Figure 8).

Table 2 lists the proprietary names of the main retardation film products. The examples listed represent universally well-known products.

Retardation films can be variously classified according to the method of stretching and type of liquid crystal coating. Thus, depending on the manufacturing process, the characteristics expressing anisotropy will differ, and likewise the type of LCD that can be compensated. By way of example, the retardation films corresponding to TN mode display are typified by WV film [5]. A diverse range of retardation films is available for VA displays, films such as ZeonorFilm® [6, 8] and TAC film [9] being used. The IPS films include JX Nikko-Nisseki Energy’s NV film [4].

2. PROPERTIES AND FUNCTION OF RETARDATION FILMS

A retardation film operates between the liquid crystal cell and polariser. The liquid crystal and polariser are anisotropic media and alter the polarisation of the light travelling through them. Hence, the retardation film designed to compensate for this is itself preferably an anisotropic medium. A brief account will now be given of the properties of such anisotropic media, followed by a review of the functions and properties of retardation films.

2.1 Optical properties of LCD

The light incident on the liquid crystal cell in an actual LCD is said to be linearly polarised. Its properties are a little different from the light that normally reaches our eyes. Actually, the difference in properties amounts to no more than the light having a different state of polarisation. Polarisated light denotes light with its plane of vibration confined to one direction, as in Figure 4.

A liquid crystal panel creates gradations in brightness by controlling the amount of light transmitted after linear polarisation by the polariser. A transmission LCD consists basically of a liquid crystal cell sandwiched between two polarisers. Image colour is created by passage through a colour filter layer, image cell by image cell.

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**Table 2. Main retardation film products**

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<tr>
<th>Type</th>
<th>Product</th>
<th>Manufacturer</th>
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<tr>
<td>① Uniaxial retardation film</td>
<td>Pure-Ace®</td>
<td>Teijin Corp.</td>
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<tr>
<td>② Biaxial retardation film</td>
<td>1) ZeonorFilm®</td>
<td>Zeon Corp.</td>
</tr>
<tr>
<td></td>
<td>2) N-TAC</td>
<td>Konica-Minolta</td>
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<td></td>
<td>3) V-TAC</td>
<td>Fujifilm</td>
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<td></td>
<td>4) Arton Film®</td>
<td>JSR Corp.</td>
</tr>
<tr>
<td>③ Liquid crystal film (hybrid orientation)</td>
<td>NH film</td>
<td>JX Nikko Nisseki Energy</td>
</tr>
<tr>
<td></td>
<td>NV film</td>
<td></td>
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<tr>
<td>④ Liquid crystal film (hybrid orientation)</td>
<td>WV film</td>
<td>Fujifilm</td>
</tr>
<tr>
<td>⑤ Half-wave, quarter-wave plate</td>
<td>Pure-Ace®</td>
<td>Teijin Corp.</td>
</tr>
<tr>
<td></td>
<td>ZeonorFilm®</td>
<td>Zeon Corp.</td>
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**Figure 5** is a schematic diagram of the molecular orientation in the field ON and field OFF states of the VA mode liquid crystal display most commonly used in contemporary liquid crystal television screens [10]. This arrangement makes it possible to switch the light incident on the liquid crystal display between two states, transmission and absorption.

The polarisers are arranged in the crossed Nicol configuration so that the axis of light absorption is rotated through 90°. The incident light is thus linearly polarised by the first polariser. In the OFF state on the left hand side of **Figure 5**, the linearly polarised light incident within the liquid crystal cell is transmitted intact, until impinging on the upper polariser. Since the upper polariser has its absorption axis rotated 90° with respect to lower polariser, the light from the liquid crystal is intercepted and cannot be transmitted. The display hence assumes the dark state. In the ON state, on the right, the liquid crystal molecules oriented by the electric field have the function of rotating the plane of vibration of the light, and the linearly polarised light impinges on the polariser after the plane of vibration of the field has rotated by 90°. This light can then pass through the upper polariser. The display hence assumes a bright state while the electric field is applied.

An actual display device has a complicated structure, involving various electrodes in the liquid crystal cell, colour filters, and so on, but whichever mode it uses, the device essentially utilises the liquid crystal cell and polarisers on either side of it in **Figure 5**.

The problem with this arrangement, however, is the display quality. As we have seen, liquid crystals and polarisers exhibit optical anisotropy. In other words, they change the state of polarisation of the light passing through them. While this property is precisely why they function as an LCD, it also means that, from the opposite standpoint, the way the light is substantially altered confers potential for detracting greatly from the original image quality.

Examples of the effects that arise are reduced contrast in oblique view, colour cast, and inversion of intermediate gradations [11]. **Figure 6** shows the angular distribution of luminance in the field ON and OFF states in the VA liquid crystal mode just considered. The angular distribution of luminance is a planar representation of the visual field, viewed from all possible angles, on a hemisphere placed over the LCD, where the viewpoint with the LCD screen viewed perpendicularly is located at the centre of the circle. While the display in the bright state has the expected high luminance in the absence of retardation film, the dark display itself will clearly appear fairly bright in some viewing directions. This results in loss of contrast. In the presence of retardation film, on the other hand, luminance in the dark state is reduced across all azimuth angles, giving good contrast, as in **Figure 6**. **Figure 7** shows the dependence of contrast on viewing angle with and without retardation film. Contrast is improved over all azimuth angles when a compensation film is present.

### 2.2 Properties of retardation films

Liquid crystal molecules are anisotropic substances and so have three different refractive indices. Thus, depending on the molecular structure, the refractive index is different.
in the x, y and z directions. In optics, this characteristic is represented by an index ellipsoid [12], where the three refractive indices \( n_x, n_y \) and \( n_z \) are the lengths of the mutually orthogonal axes x, y and z as in Figure 8, from which the refractive index experienced by a light ray incident on the substance can be found. Figure 8 depicts the situation where linearly polarised light is incident perpendicular to a given plane. Light travelling through an anisotropic substance will vary in its state of polarisation according to the two refractive indices \( n_o \) and \( n_e \), expressed by means of the index ellipsoid and an elliptical section constructed in the plane perpendicular to the incident beam. The respective refractive indices bear the subscripts o and e deriving from the terms ordinary ray and extraordinary ray.

In general, the linearly polarised light incident perpendicular to the plane represented by such an ellipse is transformed into elliptically polarised light. In other words, the light travelling through a liquid crystal is transformed from the linearly polarised light produced by passage through the polariser into elliptically polarised light. The role of the retardation film is to restore this light to linear polarisation.

The light transmitted through an anisotropic medium is greatly altered in its state of polarisation, and this has the effect of impairing the LCD display quality. For example, when white is displayed on the screen, even if a well-defined white can be displayed from a certain direction, the image may acquire a bluish tint or yellow cast when viewed obliquely owing to the properties of the liquid crystal. Thus, the polarisation is changed from linear to elliptical, and the light component vibrating along the absorption axis is then absorbed when the light travels through the second polariser. It is the function of the retardation film to compensate for such dependence on viewing angle.

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We have seen that the optical properties of retardation film are given by the three refractive indices \( n_x, n_y \) and \( n_z \). In the LCD field, refractive indices are defined taking the x and y directions in the film plane and the z axis in the direction of film thickness; optical characteristics taking film thickness into account are expressed by the three further definitions in Figure 9. Equation (A) is called the in-plane retardation or more simply the retardation, and expresses the degree to which linearly polarised light incident perpendicular to the film is elliptically polarised. Equation (B) has no formal name but is referred to as the retardation in the thickness direction and serves as a rough index of the conversion of linearly polarised light incident at an oblique angle to elliptically polarised light. Equation (C) is similar to (B) but is called the Nz coefficient; it has the birefringent part \( n_x - n_y \) of Equation (A) as the denominator, and since this is the difference in refractive indices of the molecules in the y- and x-directions, it may be said to express the ratio of birefringence in the film plane and the birefringence in cross-section. Figure 10 depicts the directions when the refractive index ellipsoid is expressed on the film, referred to the definition of retardation in Equation (A) of Figure 9.

Whichever retardation index is singled out, the definition is formulated as a difference in refractive indices and it is clear that birefringence is an important property.

A number of additional properties are required from retardation film. Examples are 1) a high transmission of around 90%, 2) birefringent expressibility, and 3) an appropriate wavelength dispersion characteristic. Also important are heat-resistance, mechanical strength, and humidity tolerance. High transmission indicates that incident light is transmitted with as little loss as possible, and for retardation film made from a polymer material, this function may be unachievable if the light is ultimately scattered because of crystal structure or other factor. Expressibility of birefringence is essential when making retardation film by stretching plastic film: unless the oriented molecular chain has the function of expressing anisotropy of refractive index, optical
compensation will be unattainable. The same functions are important when retardation film is made by coating plastic film with special liquid crystals and they are key factors in determining the design of retardation film.

2.3 Compensation with retardation film

Figure 7 compared the function of retardation film for VA mode in terms of contrast, where the retardation film was sandwiched between a polariser and liquid crystal cell, in line with the configuration in Figure 11. Contrast has in this case been improved by optical compensation with two sheets of retardation film of different kinds. Figure 11 also shows the index ellipsoids corresponding to each of the retardation films. The VA cell can be expressed by an index ellipsoid with its longer axis upright, compensation for the change in phase of the incoming linearly polarised light here being sought with the two retardation films. Figure 12 is a schematic representation of the function of the retardation film and shows the situation where the index ellipsoid of the liquid crystal layer is compensated with the index ellipsoid of the retardation film so that it takes the form of an isotropic sphere, giving zero difference in phase at any viewing angle.

Retardation film for IPS mode display requires retardation material of a different kind since the orientation of the liquid crystal molecules differs from the VA mode orientation. Furthermore, WV film or the like is used for compensation in TN mode. Although in principle the same compensation mechanism operates as in Figure 12, the different orientation of the liquid crystal molecules in the respective liquid crystal modes has to be taken into account in designing the optimum optical properties.

Quarter wave retarder can be used for reflection type LCD. Like other retardation film, quarter wave retarder is made by stretching plastic film. In terms of function, however, it is set at a retardation that converts linear polarisation to circular polarisation, and is specifically named quarter wave retarder to distinguish it from other retardation film. Figure 13 illustrates the use of quarter wave retarder for reflection type LCD. The light has to be circularly polarised within the liquid crystal layer in reflection LCDs. Hence, the quarter wave plate is positioned as shown in the diagram so that the light phase is twice transformed, at incidence and reflection, before display.

3. MANUFACTURE

The forming technology for retardation film can be divided into the technology for forming the base film and technologies for stretching - the process that functionalises the film - and coating with liquid crystal. The solution casting [3] and melt extrusion [3] processes for making the base film for retardation film will be looked at here, along with a review of the stretching and coating technologies. Stretching will be considered in some detail since a number of new stretching processes have recently been developed.

3.1 Solution casting

Figure 14 is a schematic diagram of the difference between manufacture by the solution casting process and melt extrusion process. Both processes have now found general application for the manufacture of optical film.
Solution casting first entails spreading a solution of resin in solvent as a thin film on a metal band or metal drum, as illustrated. The film is then hardened under drying conditions controlled to give uniform film thickness, and then peeled from the metal band and fed into a long drying zone for further removal of solvent.

Solution casting is used for polymers unsuited to extrusion forming because of their high viscosity, and for polymers of high melting point close to the thermal decomposition temperature, such as the films used as polariser protective film, \(\text{Fig. 15}\) in Table 2. The polymer solution in solvent has a low viscosity and allows casting at relatively low temperature; degradation is therefore minimal and the forming burden is small.

However, all manner of corrective measures must be devised to maintain uniformity of film thickness and prevent scratches, etc., from degrading surface quality. Film thickness, for instance, can often become uneven in the solvent drying step; processes may hence be provided to correct thickness midway through the drying zone and immediately ahead of spool take-up.

Another problem is presented by scratches due to slippage between roll and film in the course of drying across a multiplicity of rolls. One remedy is to impart resistance to slip by embossing the edges of the film once drying on the metal band has finished.

### 3.3 Stretching

Retardation film produced by stretching is usually made by either a longitudinal stretching process or transverse stretching process [13]. \(\text{Fig. 15}\) is a schematic depiction of film deformation behaviour in stretching. It will be seen from the diagram that the axis of film orientation set up by the processes lies in the direction of film flow (MD: machine direction) or the transverse direction (TD), respectively. The axis of orientation here denotes the direction in which the polymer chains have been extended by stretching.

The polarisers made of polarising sheet described earlier are manufactured by longitudinal stretching. Hence, if the polariser sheet and retardation film are welded together by highly efficient roll-to-roll processing as in \(\text{Fig. 16}\), the optical axis of the retardation film made by longitudinal or transverse stretching will be either parallel or at right angles to the absorption axis of the polariser sheet. This is extremely important in both simplifying the manufacturing process and reducing cost. Roll-to-roll production is essential to improve productivity.
and the general view is that this method of manufacture can achieve maximum competitiveness for various kinds of optical film, retardation film included.

Considered in a little more detail, longitudinal stretching is a forming process in which the heated film is stretched in the direction of film flow (MD) by stretching between rolls rotated at different speeds as in Figure 15a. When longitudinally stretched, film comprising resin of normal specific birefringence forms uniaxial retardation film of large refractive index in the stretching direction. The term specific birefringence refers to the birefringence $\Delta n = n_x - n_y$ when the molecular chains are ideally stretched (perfectly oriented), where $n_x$ is the refractive index in the stretching direction and $n_y$ is the refractive index at right angles to the stretching direction. A normal birefringence means that the refractive index is greater in the stretching direction so that the birefringence $\Delta n_x$ is positive. In other words, an orientation axis in the MD stretching direction as in Figure 17a gives retardation film popularly known as an A plate for which the two refractive indices at right angles to the orientation axis are equal. A well known example of an A plate is the retardation film made of PC used for compensation of STN liquid crystals.

Transverse stretching entails chucking at the film edges and running the chucking tool over rails as in Figure 15b. The rails slowly widen as the film is heated in an oven, stretching the film en route. The chucking tool normally has a fixed pitch and so the film is unstretched in the MD direction. The stretched state can hence be described as uniaxially constrained-uniaxially stretched. Retardation film from transverse stretching with the direction of maximum refractive index (the axis of orientation) oriented in the TD direction as in Figure 17b affords biaxial retardation film.

Reflection film from biaxial stretching in which longitudinal and transverse stretching are combined provides biaxial retardation film with the direction of maximum refractive index (the axis of orientation) oriented in the TD or MD direction. The ranges of $Re$ (retardation in-plane) and $Rth$ (retardation in thickness direction) that can be produced are thereby widened, and the range of application for optical compensation is correspondingly expanded. The biaxial retardation films listed under (a) in Table 2 may be cited as products.

As LCDs have increased in size, the need has arisen for optical film of large width, but uniform retardation is difficult to achieve across the whole film plane in the case of conventional transverse stretching because of bowing and other phenomena. Bowing refers to the phenomenon whereby, because of geometric distortion, the axis of orientation becomes uneven in the transverse direction, developing a bow-like profile. Once this happens the film acquires different optical properties in the transverse direction and is incapable of displaying a uniform image in large screen television receivers or the like. However, stretching technology has now witnessed a great many improvements, making film products of large width possible. In the context of productivity, moreover, manufacture of retardation film integrated through from extrusion to stretching is much to be desired.

3.4 Novel stretching technology [14]

Retardation film is currently being developed that has a film orientation axis in neither the machine direction nor the transverse direction, and has found practical application in the quarter wave plate under item (b) in Table 2. This is retardation film made by the so-called oblique stretching process. As noted earlier, retardation film has usually been manufactured by either a longitudinal stretching or transverse stretching process and when welded to polariser sheet roll-to-roll, the absorption axis of the polariser and the axis of orientation of the retardation film have been either parallel or at right angles.

In diagonally stretched film the in-plane optical axis can be set in any desired direction as in Figure 17c. This new technology for controlling the optical axis makes roll-to-roll manufacture as in Figure 16 possible for diverse LCD display modes, and has enabled the elimination of
batch paste-up processing and elimination of components added due to the concurrent use of retardation film as protective film.

3.5 Coating

Apart from manufacture by the above stretching processes, retardation film may be produced as liquid crystal coated film in which liquid crystalline material has been oriented to the film plane [4, 5]. The usual procedure entails that orientation film is coated on the film surface, the surface is then rubbed, and the liquid crystal is applied over it.

Depending on the LCD display mode, the refractive index structure of the retardation film may need to vary continuously in the thickness direction to realise a wide viewing angle. For instance, LCD using TN mode may deploy liquid crystal coated retardation film of this kind to improve viewing angle characteristics.

Widely different coating methods are available, but slot-die coating would appear to be preferred for optical film, in which uniformity of thin film thickness is required. Slot-die coating consists in applying the coating solution to the base film with the lip end of a die like the T-die described earlier, which is set at a clearance of several micrometres above the base film as shown in Figure 18.

Well known liquid crystal coated retardation films include DLC (discotic liquid crystal) film [5], commonly known as WV film, nematic hybrid liquid crystal film (NH film) [4], and twisted nematic retardation film (LC film) [4]. DLC film denotes film that has a hybrid structure of discotic liquid crystals; it employs a constitution in which, as shown in Figure 19, orientation film is provided on a TAC support and over-coated with discotic liquid crystal. The discotic liquid crystal layer has a perpendicular angle of orientation at the air interface, the orientation becoming horizontal at the orientation film interface; the angle of orientation thus varies continuously in the thickness direction. NH film and LC film are similarly derived by coating liquid crystal on a plastic film support to form structures suited to optical compensation.

CONCLUSIONS

While retardation film has found increasing application with the growing popularity of LCDs, a sweeping reduction in cost has been sought in recent years alongside improvement in quality. In the context of film technology, the existing range of melt extrusion and high width consecutive biaxial stretching processes has been extended with oblique stretching technology to meet different requirements. Obliquely stretched retardation film corresponding to batch-process film can be made by roll-to-roll forming, contributing to reduced cost. It is likely that in future a diverse range of retardation film will be manufactured with oblique stretching technology, cutting total cost.

Given this anticipated future expansion, further progress with LCDs requires growth in components development with a view to energy conservation in the product as a whole, and it is hoped that novel retardation film will continue to be developed.

REFERENCES