CHAPTER 3

HOST MATERIALS AND DOPING PROCESS

3.1 HOST MATERIALS

In the early studies of solid-state dye lasers, problems arose due to photodegradation of the dye molecules in the solid-state host. In order to overcome these problems, host materials with high laser-damage threshold, high optical quality and high thermal conductivity have been developed. These solid matrices with embedded organic dyes can be categorized into homo-polymers, modified polymers, co-polymers, sol-gel glasses, sol-gel glass-polymer composites (polycom glasses), and organically modified silicate (ORMOSIL) glasses.

3.1.1 Homo-Polymers

Homo-polymers with high optical transparency, such as poly(methyl methacrylate) (PMMA), poly(vinyl alcohol) (PVA), polystyrene (PS), poly(vinyl chloride) (PVC) and polybutadiene (PB) \(^{[3, 39, 105, 106]}\) are employed as the host matrices. The laser dye is dissolved in the monomer of one of these host materials and is allowed to undergo a polymerization process \(^{[7, 8, 107]}\). This whole polymerization process requires several days.

3.1.2 Modified Polymers

Early reports of low laser-damage resistance of polymer matrices impregnated with organic dyes and aggregation of dye molecules \(^{[71-75]}\) lead to the development of modified polymers with higher resistance to laser radiation damage. The level of resistance achieved
is comparable to that of most laser-damage resistant inorganic glasses and crystals. For homo-polymers such as PMMA and PVA, a high laser-damage resistance was attained in the elements by eliminating the initial monomer compositions of foreign inclusions and by doping PMMA with low-molecular weight additives \(^{72, 73, 75}\) such as ethanol, ethylene glycol dimethacrylate, ether, and methacrylic acid.

### 3.1.3 Co-Polymers

The most widely used host material for solid-state dye lasers to date is PMMA. However, most of the conventional laser dyes are insoluble in PMMA and low-molecular weight additives must be added to enable dye doping. Thus, co-polymers in which a heterogeneous monomer mixture is polymerized to provide enhanced cross-linking or dye compatibility in comparison with PMMA \(^{5, 6, 77}\), have been developed. A co-polymer of methyl methacrylate (MMA) and 2-hydroxyethyl methacrylate (HEMA) are the most promising of these emerging host materials \(^{78}\). These co-polymers have been claimed to exhibit improved lasing efficiency and lifetime compared with PMMA \(^{78}\).

### 3.1.4 Sol-Gel Glasses

The sol-gel method allows the preparation of porous glasses at temperatures not more than \(100^\circ\text{C}\) \(^{87}\), below the temperatures that will decompose most organic dyes. The organic dye can be impregnated into porous silica matrix either by mixing the dye into the precursor components of sol-gel glass (predoping), or by diffusing the dye into the pores of the glass structure after synthesis (postdoping). Sol-gel glass can be further heated in a densification process (temperature between 600 and \(1000^\circ\text{C}\)), in order to produce a porous material with better mechanical properties. However, due to thermal degradation of the organic dye, densified sol-gel glass can only be doped with organic dyes via postdoping.
route. The duration of the fabrication of sol-gel glass is of the order of a few days; whereas the postdoping route usually requires a few weeks for completion.

3.1.5 Polycom Glasses

During the laser pumping process of dye-doped sol-gel glasses, the optical scattering from the largest of the pores and pore aggregations have caused the optical attenuation of sol-gel glass. This has affected the beam quality and laser efficiency of the dye-doped sol-gel glasses. Index-matching fluid has been filled into the pores to improve the optical quality of the material \(^{87, 88}\). The precursor of PMMA (i.e. MMA) is mixed with dye solution and allowed to diffuse into the pore network of sol-gel glass, with benzoyl peroxide as catalyst. The glass is then immersed in this monomer solution for several hours; after which it is taken out of the solution and allowed for a further 48 hours at 40°C to ensure more complete polymerization.

3.1.6 Organically Modified Silicate (ORMOSIL)

ORMOSIL glass is a composite of organic and inorganic materials, where the components are bonded to each other with covalent bonds\(^ {108}\). ORMOSIL glass possesses excellent mechanical strength, due to the presence of strong covalent bonds and the non-porous nature. The fabrication of dye-doped ORMOSIL is usually carried out by reacting the dye-organic monomeric mixture with an inorganic metal oxide. The reaction usually takes few weeks and the volume of the gel will be reduced by half due to the evaporation of the volatile by-products \(^ {89}\). The resulting transparent material has to be heated at 55°C for several weeks to eliminate any remaining volatile by-products.
In this work, the laser performance of organic dye doped in poly(vinyl alcohol) (PVA) is investigated. This is due to PVA possesses an optical properties similar to poly(methyl methacrylate) (PMMA), and the fabrication of dye-doped PVA films are simple and relatively low cost. At the same time, organic dye doped in PMMA is also studied. This is due to PMMA is a hard, transparent polymer with high optical clarity (> 92% light transmission in the visible spectrum), high refractive index and good resistance to light and aging. Furthermore, the impregnation of comparatively large dye concentration in this host material can be achieved without the formation of dimers. Some characteristics (such as fabrication technique, laser spectra, operating lifetime, etc.) from these dye-doped PMMA will serve as a reference in the studies of the laser performance of dye-doped PVA.

The next section will discuss the basic characteristics of PMMA and PVA. The discussions from point of view of dye-doped polymer fabrication technique of these two host materials were also presented.
3.2 POLY(METHYL METHACRYLATE)

Poly(methyl methacrylate) (PMMA) (Fig. 3.1(a)) is the most important member of the range of acrylic polymers, which may be considered structurally as derivatives of acrylic acid (Fig. 3.1(b)). The commercial names of PMMA are Perspex® (ICI), Oroglass® (Röhm & Haas, USA), Plexiglas® (Röhm & Haas GmbH, Germany) and Lucite® (Du Pont) etc. PMMA is a high transparency material with a high softening point (90–125°C), better impact strength, and better weatherability. PMMA is a linear thermoplastic, with about 70 – 75% syndiotactic. Since its lack of complete stereoregularity and its bulky side groups, it is amorphous. PMMA is resistant to many aqueous inorganic reagents, including dilute alkalis and acids.

![Diagram of poly(methyl methacrylate) and acrylic acid](image)

Fig. 3.1 Structure of (a) poly(methyl methacrylate) and (b) acrylic acid.

The tensile strength of PMMA ranges as high as $1 \times 10^4$ psi and its impact strength is about equal to that of the impact-resistant styrene co-polymers. Fabricability of PMMA is quite good, where only slightly higher temperatures (~110°C) are needed for molding PMMA than for polystyrene. A limitation to the optical uses of PMMA is its poor resistance to abrasion.

The major uses of PMMA arise from its high light transmission and good outdoor weathering properties. Injection-molded PMMA articles include automotive lenses, reflective devices, instrument and appliance covers, optical equipment, and home
furnishing. PMMA sheeting is used for signs, glazing (aircraft windows), furniture, partitions, and lighting-fixture diffusers.

3.2.1 Fabrication of Dye-Doped PMMA Slabs

The fabrication of dye-doped PMMA is usually carried out by conventional polymerization methods \(^{[7, 8, 107]}\). The monomer used for the synthesis of dye-doped polymer is methyl methacrylate (MMA). A measured quantity of dye is dissolved in a mixture of MMA and alcohol (or other organic solvents, which depends on dye). Radical initiator such as benzoyl peroxide is added to the mixture to enable the polymerization process. The mixture is then put into a glass tube or a mold and is kept in nitrogen atmosphere. The polymerization is carried out in a temperature-controlled water bath and the temperature of the water bath is maintained at 50°C for few days. The polymerized cell is subsequently removed from the glass tube or mold, and cut into the desired dimensions and polished to the optical quality (less that one wavelength flat) required.

However, with this method, the laser dye could decompose in radical polymerization \(^{[26, 109]}\) and thus reduces the laser efficiency \(^{[26]}\). Furthermore, this process takes a long period for polymerization (several days). In this work, dyes were directly doped into PMMA polymer by low-pressure-compression molding method, where fabrication process did not involve radical polymerization of MMA.

The low-pressure-compression molding method used here is adapted from an earlier work in this laboratory \(^{[34]}\). The mold (Fig. 3.2) was adapted from a concept of an industrial compression mold \(^{[110]}\) for PMMA. The mold cavity (4.5 × 1.3 × 0.9 cm³ volume) is enclosed by top and lower part of brass mold; whereas the front and back sides are covered by Pyrex® glass windows, which are held by metal clamps. Two springs were adjusted by cap screws and washers, to provide a compression pressure of about 50 Ncm⁻².
Coumarin 460 (C460) and Rhodamine 590 perchlorate (R6G(ClO₄)) (Exciton Inc., USA) were used without further purification. In a typical preparation of dye-doped PMMA preform, 2.32 mg of R6G(ClO₄) was dissolved in 50 ml chloroform (spectroscopic grade, Merck). This was followed by adding 5 g of PMMA pellets (high molecular weight $M_w > 900,000$, industrial grade, Röhm & Haas GmbH, Germany). The mixture was stirred until the PMMA pellets dissolved completely, after which the mixture was poured into a Teflon dish. The dish was then placed in a vacuum oven (Napco, model 5851) maintained at 180°C and at a pressure of less than 1 mbar for 1 hour. The dried spongy preform of dye concentration of $1 \times 10^{-3}$ M that resulted was ground into a powder form.
5 g of the preform was weighed and loaded into the mold cavity. The top part of the mold was compressed downwards by applying pressure through cap screws and springs (Fig. 3.3). The mold was then placed into a vacuum oven (pressure < 1 mbar) and heated at 180°C which was approximately 70°C above the glass transition temperature of this industrial grade PMMA pellets. After half an hour, the preform melted and filled the mold cavity. The mold was removed from the vacuum oven and the pressure on the melt was released. The mold returned to the vacuum oven again and left for annealing process at 180°C and atmospheric pressure. Fig. 3.4 and 3.5 show the C460-doped PMMA slabs and R6G(ClO₄)-doped PMMA slabs respectively.

Fig. 3.3 The top part of the mold was compressed downwards by applying pressure through cap screws and springs.

An attempt was also done with molding low molecular weight PMMA powder \( (M_w \approx 120,000, \text{ Aldrich, USA}) \) at temperature of about 130°C. However, the resulting slab is brittle and cracked easily when removed from the mold cavity.
As compared with previous work [34], this molding method was improved by increasing the spring compression pressure of the mold, in order to minimize the formation of bubbles in the dye-doped slabs. Besides, the molding temperature was increased to 180°C (5°C higher than the previous work) to reduce the viscosity of the melt, in order to have a more even appearance of the dye-doped slabs. Furthermore, the studies of the annealing effect on dye-doped PMMA slabs were also extended in detail.

![Image of C460-doped PMMA slabs (center and right) and PMMA slab (left).](image1)

**Fig. 3.4** C460-doped PMMA slabs (center and right) and PMMA slab (left).

![Image of R6G(ClO₄) doped PMMA slabs.](image2)

**Fig. 3.5** R6G(ClO₄) doped PMMA slabs.
During the molding and fabrication process, the dye-doped PMMA slab may develop internal stress. Chemicals may sensitize the dye-doped PMMA slab and cause crazing. The slab develops internal stress as a result of cooling immediately after molding, as the chemical reactions continue during complete polymerization. The internal stresses of dye-doped PMMA slab may be reduced or eliminated by annealing. Furthermore, the optical quality of the dye-doped PMMA slabs might be improved through annealing process. Annealing consists of prolonged heating of the slab at molding temperature and atmospheric pressure, with the pressures of the springs removed. The annealing process takes a few hours and the slab is then cooled down slowly. The next section described the examination on the effect of annealing on dye-doped PMMA slabs.

3.2.2 Annealing of Dye-Doped PMMA Slabs

The dye-doped PMMA slabs were prepared based on distinct annealing periods (ranging from $t_a = 0, 1, 2, 3, ..., 6, 7, 8$ hour(s)) by the method described in the previous section. These dye-doped PMMA slabs with distinct annealing periods were then examined by the method as shown in Fig. 3.6. The dye-doped PMMA slab was placed vertically at a fixed position in front of a HeNe laser. The HeNe laser beam penetrated the center of the side of dye-doped PMMA slab and exited from the opposite side of the slab. The beam then illuminated a grid translucent screen at a fixed position beyond the slab. The light spot illuminated on the screen was recorded by a CCD camera (Sony XC75), which is connected to a computer equipped with a frame grabber and image capturing software.
The effects of the scattering of light on dye-doped PMMA slabs with different annealing periods, examined by a HeNe laser, were illustrated by the size of the light spot on the screen. The images of the light spot on the screen were then digitized by the image processing software. The measurements of the vertical and horizontal height of the light spot were taken based on 90 percentile of the distribution histogram of the digitized image. The vertical and horizontal divergence of the beam illuminated on the screen are given by

\[ \theta_v = 2 \tan^{-1} \left( \frac{h}{2d} \right) \]  

and  

\[ \theta_h = 2 \tan^{-1} \left( \frac{w}{2d} \right) \]  

(3.1)  

(3.2)
where $h$ is the increment of the vertical height of the size of the spot, $w$ is the increment of the horizontal width of the size of the spot and $d$ (= 45 cm) is the distance between the dye-doped PMMA slab and the screen. Fig. 3.7(a) – (i) show a series of light spots from the dye-doped PMMA slabs with different annealing periods ($t_a = 0, 1, 2, 3, ..., 6, 7, 8$ hours), and examined by HeNe laser, whereas Fig. 3.8 shows the light spot of the undoped commercial Perspex® slab.

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**Fig. 3.7** (a) – (i) Light spots from the dye-doped PMMA slabs with different annealing periods ($t_a = 0, 1, 2, 3, ..., 6, 7, 8$ hours), under the examination of a HeNe laser.
Fig. 3.8 The light spot of the undoped commercial Perspex® slab under the examination of a HeNe laser.

From Fig. 3.7, the vertical divergence of light spots undergoes slight changes, even though the annealing period has increased. The significant changes were shown on the horizontal divergence of light spots. Generally, the horizontal divergence reduced as the annealing period increased (Fig. 3.7(a) – (h)). When the annealing period reached 7 hours, the light spot became a circular spot, which is similar to the light spot from an undoped commercial Perspex® slab (Fig. 3.8). There is no further changes observed for annealing periods longer than 7 hours (Fig. 3.7(i)).

<table>
<thead>
<tr>
<th>Intervening Medium</th>
<th>$\theta_v$ (mrad.)</th>
<th>$\theta_h$ (mrad.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>a) air</td>
<td>$(4.4 \pm 0.6)$</td>
<td>$(4.4 \pm 0.6)$</td>
</tr>
<tr>
<td>b) undoped commercial Perspex® slab</td>
<td>$(5.6 \pm 0.6)$</td>
<td>$(5.6 \pm 0.6)$</td>
</tr>
<tr>
<td>c) dye-doped PMMA slab without annealing</td>
<td>$(6.7 \pm 1.7)$</td>
<td>$(17.8 \pm 1.9)$</td>
</tr>
<tr>
<td>d) dye-doped PMMA slab with 1 hours annealing</td>
<td>$(5.6 \pm 1.7)$</td>
<td>$(15.6 \pm 1.1)$</td>
</tr>
<tr>
<td>e) dye-doped PMMA slab with 2 hours annealing</td>
<td>$(6.7 \pm 0.6)$</td>
<td>$(15.6 \pm 2.3)$</td>
</tr>
<tr>
<td>f) dye-doped PMMA slab with 3 hours annealing</td>
<td>$(5.6 \pm 3.6)$</td>
<td>$(21.1 \pm 4.8)$</td>
</tr>
<tr>
<td>g) dye-doped PMMA slab with 4 hours annealing</td>
<td>$(5.6 \pm 1.7)$</td>
<td>$(13.3 \pm 0.6)$</td>
</tr>
<tr>
<td>h) dye-doped PMMA slab with 5 hours annealing</td>
<td>$(4.4 \pm 1.3)$</td>
<td>$(10.0 \pm 1.7)$</td>
</tr>
<tr>
<td>i) dye-doped PMMA slab with 6 hours annealing</td>
<td>$(5.6 \pm 1.9)$</td>
<td>$(6.7 \pm 3.6)$</td>
</tr>
<tr>
<td>j) dye-doped PMMA slab with 7 hours annealing</td>
<td>$(5.6 \pm 1.1)$</td>
<td>$(5.6 \pm 1.1)$</td>
</tr>
<tr>
<td>k) dye-doped PMMA slab with 8 hours annealing</td>
<td>$(5.6 \pm 0.6)$</td>
<td>$(5.6 \pm 0.6)$</td>
</tr>
</tbody>
</table>

Table 3.1 The vertical and horizontal divergence of dye-doped PMMA slab with different intervening medium under the examination of HeNe laser.
From Fig. 3.7, 3.8 and Table 3.1, the minimum divergence was obtained from the dye-doped PMMA with annealing period longer than 7 hours, which is comparable with the undoped commercial Perspex® slab. However, other smaller scattering surrounding the major light spot were probably caused by the impurities that are inadvertently impregnated into the dye-doped PMMA slab during the fabrication process.

The dye-doped PMMA slabs with annealing periods less than 7 hours were found to have greater divergence in the horizontal direction (molding compression direction) than in the vertical direction. This is probably due to the melt of dye-doped PMMA, which has been relaxed towards molding compression direction during the annealing process, when the compression pressure from the mold was removed. Thus, the melt will gradually release the internal stresses to the direction which the compression pressure has been removed. The horizontal divergence was found to reduce as the annealing period increases. This indicates that the internal stresses have been released during the annealing process. The internal stresses of dye-doped PMMA were completely removed after annealing period of 7 hours.

![Fig. 3.9](image)

(a) Light spot of HeNe laser from the dye-doped PMMA slabs under vacuum condition during fabrication process, and undergo annealing process for 7 hours after fabrication; (b) Light spot of HeNe laser from the dye-doped PMMA slabs without vacuum condition during fabrication process, and undergo annealing process for 7 hours after fabrication.
Fig. 3.9(b) shows the light spot of HeNe laser from the dye-doped PMMA slabs, which vacuum was not applied during the fabrication process. Although the slab has undergone for annealing process for 7 hours, the divergence was greater and more scattering was observed compared with the dye-doped PMMA slab that was fabricated under vacuum condition, and annealed for 7 hours after fabrication process (Fig. 3.9(a)). As the dye-doped PMMA slab was fabricated under vacuum condition, the oxidation and formation of pores by residual air was reduced. Thus, the optical homogeneity of the dye-doped PMMA slab was improved and hence increases the laser efficiency.
3.3 POLY(VINYL ALCOHOL)

Vinyl alcohol is unstable and does not exist in the free state. Poly(vinyl alcohol) (PVA) is thus prepared by alcoholyis of a poly(vinyl ester) and in practice poly(vinyl acetate) is used (Fig. 3.10). Methanol or ethanol can be used to effect the alcoholyis, with either an acid or a base as catalyst. The alcoholyis is usually carried out by dissolving the poly(vinyl acetate) in alcohol. The process is accelerated by the adding of an acid or a base, and heating at the temperature of 60°C. The poly(vinyl alcohol) precipitates from the solution.

\[
\text{CH}_2\text{CH(OH)} + \text{CH}_3\text{OH} \rightarrow \text{CH}_2\text{CH(OH)} + \text{CH}_3\text{COOCH}_3
\]

Fig. 3.10 Alcoholyis of poly(vinyl acetate).

PVA is amorphous when unstretched. Despite the atactic chain structure, it can be drawn into a crystalline fibre, as the hydroxyl groups are small enough to fit into a crystal lattice. PVA does not melt into a thermoplastic, but decomposes by loss of water from two adjacent hydroxyl groups at temperature above 150°C.

PVA is water-soluble. It can usually be dissolved completely above 85°C. The tensile strength of PVA dependents greatly on humidity: The increase in water absorption as humidity increases, lowers the tensile strength. The water acts as a plasticiser, which reduces the tensile strength but increases the elongation and tear strength. Due to the high polarity of PVA, it is very resistant to hydrocarbon. It does not dissolve in pure alcohols, but dissolve in lower alcohol-water mixture. Only a few organic solvents, such as diethylenetriamine and triethylenetetramine are effective at room temperature.
Commercial PVA such as Gelvator, Elvanol, Mowiol, Rhodoviol etc. are available in a number of grades which differ in molecular weight and in the residual acetate content. PVA is employed for a variety of applications. The incompletely ‘hydrolysed’ grades of PVA have been used for water-soluble packages for bath salts, bleaches, insecticides and disinfectants. In many applications it serves as a binder and thickener in addition to an emulsifying agent. PVA is also employed in adhesives, binders, paper sizing, paper coatings, textile sizing, ceramics, cosmetics and as a steel quenchant.

3.3.1 Fabrication of Dye-Doped PVA Films

In a typical preparation of dye-doped PVA films, 4.07 mg of R6G (ClO₄) was dissolved in 15 ml ethanol (BDH Laboratory Supplies) to give a red-orange-colour solution of $1 \times 10^{-3}$ M concentration. The PVA solution was obtained from an industrial grade adhesive ($M_w = 150,000$). The PVA solution was added into the R6G (ClO₄)/ethanol solution and stirred. The mixture was left for 1 hour after stirring in order to eliminate the bubbles.

The PVA material that is employed in this work possesses high elongation and tear strength, and the dye-doped PVA film can only be fabricated by dip-coat or spin-coat methods. In this work, the dip-coat method is chosen. The dye-doped PVA film was fabricated by dipping vertically a glass substrate ($76 \times 26 \times 1$ mm) into this dye-doped PVA mixture for approximately 1 second. Half of the glass substrate was dipped into the dye-doped PVA mixture. The glass substrate was withdrawn and left to hang vertically to air dry for more than 24 hours. The thickness of the dye-doped PVA film was determined to be approximately $11 - 12$ μm. The determination of thickness and refractive index of the dye-doped PVA film by optical methods are described in Chapter 4 and Chapter 5.
Fig. 3.11 and 3.12 show the C460-doped PVA films and R6G(ClO₄⁻)-doped PVA films respectively.

Fig. 3.11 C460-doped PVA films.

Fig. 3.12 R6G(ClO₄⁻)-doped PVA films.
3.3.2 Configurations of Dye-Doped PVA Films

The dye-doped PVA films can be fabricated in different configurations (Fig. 3.13), the combinations of which is film-glass-film (Type A model), film-glass-aluminium (Type B model) and film-glass (Type C model). These configurations were used in the investigation of the factors that affected the laser performance of these dye-doped PVA films. These factors include (a) the thickness of the dye-doped PVA film, (b) the effect of aluminium coated glass substrate, and (c) the role played by the glass substrate.

![Diagram of configurations]

**Fig. 3.13** Cross-sections of dye-doped PVA sample with different configurations.

[A(n) = single glass substrate with both surface coated with PVA, where n is a number of repeated immersion and withdrawal; B(n) = single glass substrate with one of the surface coated with aluminium, both surfaces were coated with PVA; C = single glass substrate (with different thickness) with only one surface coated with PVA]

In the combination of film-glass-film (Type A model), in order to study the effect of thickness of the dye-doped film, these dye-doped films were fabricated with different thickness by repetitive immersion method. The thickness of the film increased with repetitive immersion of the glass substrate in dye-doped PVA mixture. The duration between withdrawal and immersion was set to 1 minute with the immersion time of
approximately 1 second. In this project, dye-doped PVA films were fabricated with the number of immersion ranging from 1 to 8 times \( n = 1, 2, 3, ..., 7, 8 \) and the glass substrate thickness was 1 mm.

![Diagram](image)

**Fig. 3.14** (a) The UV pump light propagated through dye-doped PVA film into glass substrate and reflected by aluminium-coated layer through glass substrate into the film; (b) the UV pump light propagated through dye-doped PVA film and directly reflected by aluminium-coated layer into the film without passing through the glass substrate.

For the combination of film-glass-aluminium (Type B model), the aluminium-coated glass substrate (glass substrate thickness = 1 mm) was also dipped into the dye-doped PVA mixture. The aluminium layer reflects the entire UV pump light into the dye-doped PVA laser medium. Two attempts had been carried out to investigate the role played by the glass substrate and aluminium-coated layer: (1) the route that UV pump light propagated through the film into the glass substrate and reflected by the aluminium-coated layer through glass substrate into the film (Fig. 3.14(a)), and (2) the route that UV pump light propagated through the film and directly reflected into the film without passing
through the glass substrate (Fig. 3.14(b)). These dye-doped PVA films on aluminium-coated glass substrates were also fabricated with various film thickness.

Besides, the dye-doped PVA films with the combination of film-glass (Type C model) had also been fabricated in order to investigate the role played by the glass substrates. This combination had been fabricated with single immersion of glass substrate into the dye-doped PVA mixture, in which one of the surfaces of the glass substrate was covered by masking tape. Thus, the dye-doped film was coated on a single surface of the glass substrate with a thickness of approximately 11 – 12 μm. In this combination, glass substrates with thicknesses varying from 1 mm to 10 mm were used.