

Photoalignment - an alternative aligning technique for Liquid Crystal Displays

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Materials

ABSTRACT

Purpose: The main aim of this work is to present our investigation in the field of photoalignment. In this work we introduce useful photoalignment techniques and discuss in detail the results of photocrosslinking and photodegradation in photosensitive polymer materials.

Design/methodology/approach: We describe the new procedure investigated in our laboratory for the following materials: HD-4100, HD-4110 and HD-8820 (DuPont Microsystems) as well as the characteristics obtained for the thin polyimide layers: optical homogeneity, thickness, surface tension.

Findings: Polyimide layers obtained on the glass/ITO substrates which were irradiated using linearly polarised ultraviolet light show the aligning properties for liquid crystal layers. The proposed new technology is fairly fine to obtain 2-3 cm² layers.

Research limitations/implications: The liquid crystal material alignment quality is not perfect, but the works are still in progress. In the next step we will try to use silan-derivative adhesion promoters to lower the aligning layers thickness. This solution will probably also have a big influence on the aligning layers homogeneity.

Practical implications: We discuss an alternative technique of the rubbing method for composition of liquid crystal displays. Liquid crystal displays are widely used for many industrial and commercial applications. Multimedia and graphics and medical-imaging systems require top quality images. Such an image can be obtained under the condition of good liquid crystal alignment. Rubbing is a method conventionally used for these purposes. Unfortunately this technique has some disadvantages that are particularly visible in new generation displays. The usage of photodegradable materials opens the route to obtain an appropriate molecular tilt and controlled anchoring energy as well as preparing layers that are suitable to providing diversified optical axes in one liquid crystal cell.

Originality/value: Agents investigated in the paper hasn't been used as the alignment materials in liquid crystal cells preparation so far. The used technology as well as HD-4100, HD-8820 and HD-4110 materials are suitable as the alternative substances for common used aligning chemical substances and technologies.

Keywords: Engineering polymers; Surface treatment; Photoalignment

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1. Introduction

Devices with flat panel liquid crystal displays are widely used for diverse purposes in many aspects of our life, including manufacturing of cellular phones, notebook computers and wide screen televisions. They are light weight, thin and have low power consumption. A typical liquid crystal display is made of two glass substrates filled with liquid crystal (Fig. 1).

The substrates are covered with a substance that ensures the existence of a monodomain within the liquid crystal. Such a liquid crystal cell can be used in a display device only if it has good transition of visible light, wide viewing angle, good contrast and fast response time. These parameters depend on the quality of LC alignment which is obtained by the interaction between the organic (e.g. polyamide or polyimide) layer and the liquid crystal.

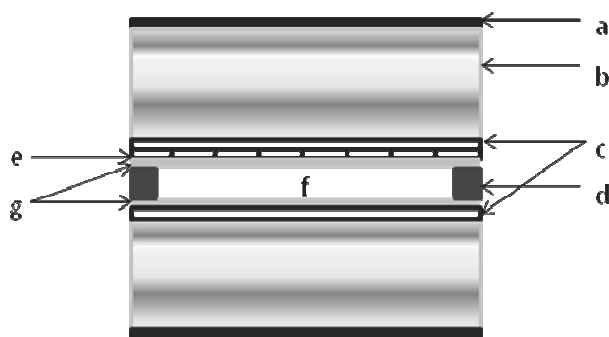


Fig. 1. Schematic representation of a liquid crystal cell. a - polariser, b - glass substrate, ITO electrode, d - spacer, e - RGB filter, f - liquid crystal, g - orienting layer

During the process the organic resin is coated on the glass substrate and dried. Afterwards the received layer is usually unidirectionally buffed (rubbed) with special cloth. The procedure is known as 'rubbing'. Despite rubbing technology being widely used in LCD manufacturing, it has some shortcomings such as generation of dust particles and fibers and charging the layer with static charges. This is the reason why new solutions and new technologies are desired. There are some alternative technologies for rubbing: laser writing, microrubbing, ion beam irradiation and a very promising one - photoalignment.

Because of its non-contact nature photoalignment eliminates the drawbacks associated with rubbing technology. Light affects the matter. It is typical of all photoalignment technologies that light (especially polarised UV light) generates anisotropy in photosensible material of the layer. There are four main photoalignment mechanisms: (1) topochemical crosslinking in polymer structure, (2) photodegradation in polyimides, (3) *cis-trans* isomerisation in azo-structures: pure-dye films, polymers containing azo- groups and monolayers of azodyes and eventually (4) pure reorientation of photo-chemically stable azo-dyes molecules.

From our point of view a good alignment technique ought to be: as fast as possible, not too vulnerable to processing procedures that have to be used, including temperature changes, presence of water *et cetera*. Further features that need to be taken into account

are: inexpensiveness and efficiency, easiness to perform and finally adequacy to use in big-scale substrates. The obtained aligning layers should be uniform and thin as well as give thermally, electrochemically and photochemically stable anchoring for liquid crystal.

2. Materials

The first polyimides were synthesised in 1908 by Bogera and Renshow [1]. Because of their considerable thermal, mechanical and chemical stability [2] they are used on a great scale in industry [3]. Polyimides are polymer materials containing an imide functional group. These groups can be organised linearly or as rings [4] (Fig. 2).

It is known that all polyimides are able to provide planar orientation ordering to the overlaying liquid crystal layer, which was an indication to conduct our study on photodefinable polyimide materials. In the present work liquid crystal alignment ability for two types of photosensitive polymer materials (Du Pont Microsystems) has been investigated: positive acting UV photosensitive polyimide HD-8820 and negative acting UV photosensitive materials: HD-4100 and HD-4110. These materials are the latest technologies for semiconductors stress buffer and packaging applications [5,6].

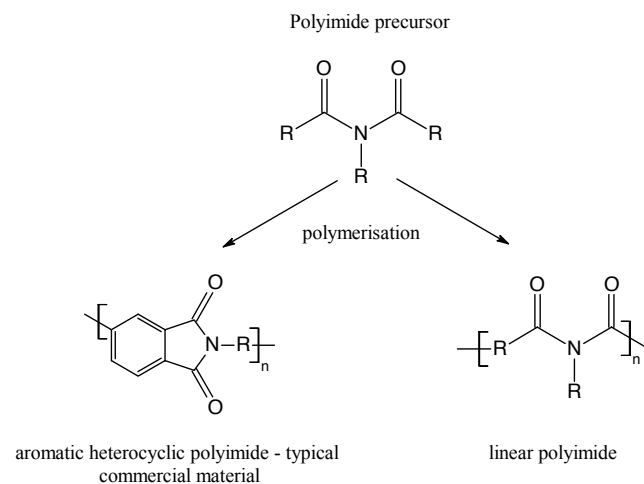


Fig. 2. Schematic representation of polymerisation reaction in polyimides

3. Mechanisms of photoalignment in polyimides

Exposure of negative acting UV photosensitive polyimide to linearly polarised UV light (LPUV) causes photoinduced crosslinking. However, in case of positive acting UV photosensitive materials LPUV causes selective decomposition of polyimide chains. These processes change the distribution in polymer chains and give anisotropy to the layer. The arrangement

on the layer is passed to the liquid crystal through dispersion and dipole-dipole interactions.

3.1. Photocrosslinking in negative acting materials

Liquid crystal molecules can be aligned through interaction with the side chains of polyimides previously irradiated with UV. The UV radiation ensures a topochemical reaction between the side chains of prepolymer containing chalcone [7], coumarin [8] or cinnamate [9,10] moieties (Fig. 3).

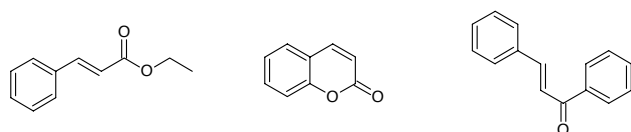


Fig. 3. On the left - cinnamate, in the middle - coumarin, on the right - chalcone

Under linear photopolymerisation (LPP) the prepolymer undergoes cycloaddition [2+2]. Substrates of cycloaddition have double bonding. After the delocalisation of π electrons there is a ring formed in the product (Fig. 4).



Fig. 4. Cycloaddition [2+2]

Poly(vinyl 4-methoxy-cinnamate) (PVMC) is a material usually used as a negative agent in photolithography [11,12]. The UV irradiation and the subsequent chemical reaction lead to anisotropic arrangement of cyclobutane molecules with their long axis lying perpendicular to the polarisation direction of the LPUV. The liquid crystal molecules will also align likewise because of Van der Waals forces acting between them and side chains with cyclobutane rings being present.

This kind of effect can also be observed in polyimides with coumarins in the side chains. It is said [11] that alignment provided by this material is more efficient in comparison to the alignment obtained for the PVMC layers.

It is worth remarking that in some photocrosslinking materials, such as chalcones UV causes *cis-trans* isomerisation which leads to homeotropic, or homogenous LC alignment depending on the UV exposition time [13].

3.2. Photodegradation in positive acting materials

UV radiation, especially short-wave UV, is able to initiate chemical reactions which sometimes leads to the degradation of the material. These processes are known as 'photoageing of the material'. There is a possibility to use photodegradation to generate anisotropy in photosensitive polyimide organic layers.

Aligning agents can be similar to those used in rubbing. During technological process the polyimide layer gets irradiated by high intensity beam of the LPUV which causes excitation of the imide bonding in polyimide molecule. The excitation leads to homolytic decomposition of the chains. When the decomposition takes place, arise free radicals which take part in the fotooxidation processes. Illumination leads to selective decomposition in the aligning material. Before the process commences, the chains are randomly oriented in the layer. During the irradiation the UV excites selectively these chains whose long axis has similar or identical direction with the polarisation direction of UV light. The direction perpendicular to above-mentioned is then the direction with the highest density of unbroken polyimide chains [14]. The liquid crystals molecules will then be oriented on such a layer thanks to the Van der Waals forces (Fig. 5).

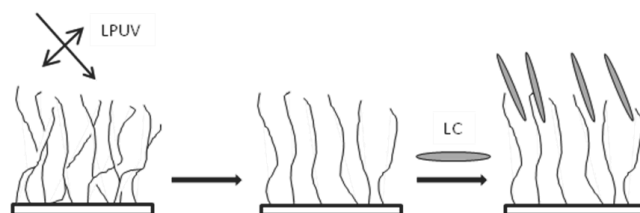


Fig. 5. Schematic diagram of the orientational photodegradation in positive acting polyimides [15]

It should be pointed out that there are some other mechanisms of photoalignment in polyimides, for example selective photoisomerisation in PI backbone structure upon UV irradiation. It was demonstrated that homogeneous (uniform and parallel), inclined homogeneous [16,17,18] and homeotropic LC alignment can be realised by using photo-aligned films of main-chain-type Azo-PI [19]. Homeotropic alignment was produced in poly(amic acid) derivatives, for example Azo-PI (Fig. 6).

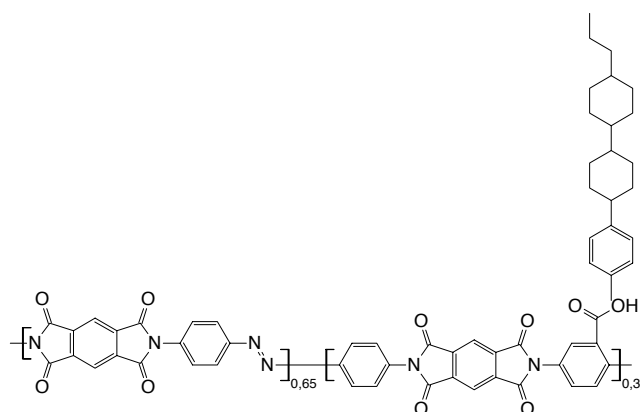


Fig. 6. Poly(amic acid) derivative used in Usami work [19]

Such side-chain molecule has the azobenzene moiety in its backbone structure. The layer got irradiated in two steps. The first irradiation was carried out by the linearly polarised light (LP-L) which caused the average orientation of the backbone perpendicular to the polarisation direction of the LP-L.

The second oblique angle irradiation - with unpolarized light (UP-L) caused a sloping of the backbone towards the direction of UP-L [19].

4. Preparation of aligning layers - technology

The procedure of obtaining an uniform and thin polyimide layer on a substrate (made of glass or an organic material) involves nine steps; steps 10-13 represent the preparation of the liquid crystal cell:

- step 1: cleaning and drying the substrates,
- step 2: preparation of the polyimide solutions,
- step 3: aligning agent (PI) deposition - spin-coating,
- step 4: soft bake - evaporation of the diluent,
- step 5: UV exposure (by polariser),
- step 6: temperature deposition (cure),
- step 7: development,
- step 8: post development bake (optional),
- step 9: temperature cure in N₂ atmosphere,
- step 10: deposition of the spacers on the layer,
- step 11: cell formation by the bonding two layers using a special glue,
- step 12: filling the cell with the LC,
- step 13: the wiring solder (optional).

The process, which involves every step from the aforesaid can be named 'photorubbing'. It should be stressed that one can carry it out without the development and following steps.

The process steps no: 2, 3, 5, 6, 7 and 9 were studied in detail and optimised in our work. These processes have utmost influence on the homogeneity and thickness of the alignment layer.

The homogeneity and thickness of the PI layers are very important to obtaining properly working liquid crystal cell. They depend on such factors as: PI precursor solution viscosity, dry matter content, thinner type, chemical structure of the PI precursor, dosage rate, and rotational speed during spin-coating process. The higher the solution concentration (and the solution viscosity) is, the thicker the PI layer will be. The amount of the solution and the coating rate have no influence on the layer thickness, but they affect its homogeneity. After the deposition on the substrate's surface the layer is dried (soft bake) with the use of a hot plate or a convector. This step is of crucial importance for the resolution of the obtained strips on the layer (if the layer is to be developed) because the PI precursor solution contains high-boiling thinners, for example N-Methyl-2-pyrrolidone (NMP). If this thinner will be present in the layer during the development then even the irradiated areas will be removed (for the negative-type of PI) and the layer will be viscous. There is a possibility to optimize this step by varying the temperature and the time of baking.

4.1. Thickness measurements

Polyimides that were used in our work are commonly applied as photoresists in industrial photolithography processes. For those applications average thickness of the layers are [5,6]: 4-13 μm for

HD-4100, 8-20 μm for HD-4110 and 4-10 μm for HD-8820 (after development). For our applications lower layer thicknesses are required. To measure the thickness of the layers after deposition to U-PL and LPUV 192 samples of three different polyimides were prepared. Four solutions of three polyimides (6%, 15%, 20% and 30%) in NMP (for HD-4100 and HD-4110) and in *N,N*-dimethylformamide (DMF) for HD-8820 were made. For each concentration 48 layers were spin-coated onto glass/ITO substrate.

Table 1.
Preparation of the samples - description

% V/V	THE NUMBER OF SAMPLES					
	HD-4100		HD-4110		HD-8820	
	UP-L	LPUV	UP-L	LPUV	UP-L	LPUV
6%, 15%, 20% or 30%	32	32	32	32	32	32
exposure energy [mJ/cm ²]	752	134	940	168	220	994

Samples were baked in the convector, divided half to half and subsequently the first one was irradiated (high-pressure Hg-Xe lamp, Oriel) with UP-L and the second one with LPUV using the HNPB/plastic (3M) foil as the polariser. The thickness was measured by spectrophotometer SPM 9001 (Prema) and microinterferometer MII-4 (Linnik). The description of the series exposure is given in Table 1.

The measurements' results are shown in the Figures 7, 8 and 9.

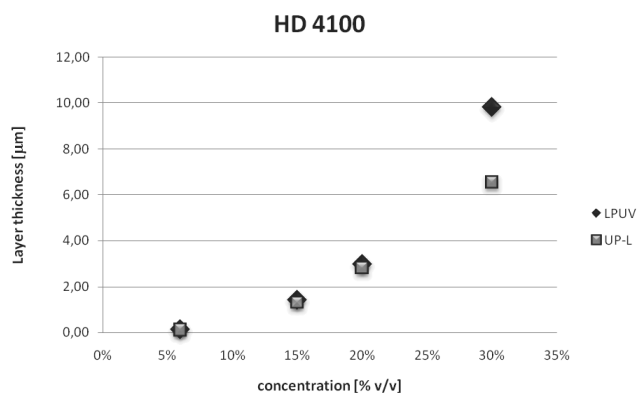


Fig. 7. Thicknesses measured for negative-type PI: HD-4100

Figures 7, 8 and 9 show that the lower the PI concentration, the lower the thickness of the layers. It can be clearly seen that there is an unambiguous dependence between the layer thickness and the PI solution concentration. The layers' homogeneity was investigated with the use of the Scanning Electron Microscope (Phenom, FEI Company) (Fig. 10).

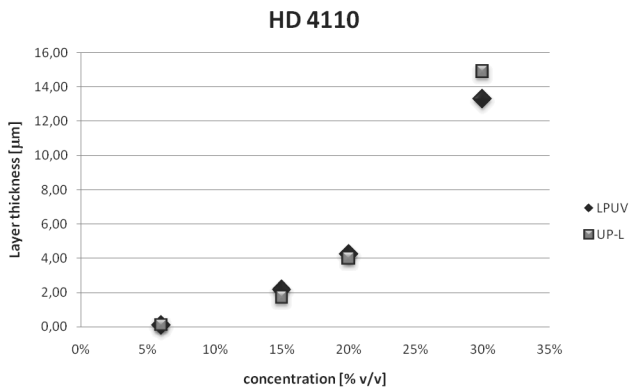


Fig. 8. Thicknesses measured for negative-type PI: HD-4110

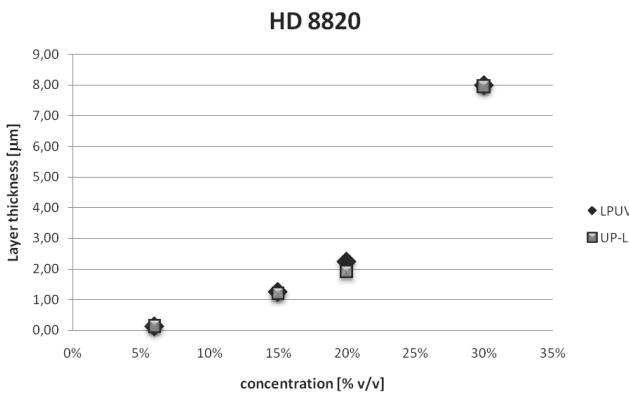


Fig. 9. Thicknesses measured for positive-type PI: HD-8820

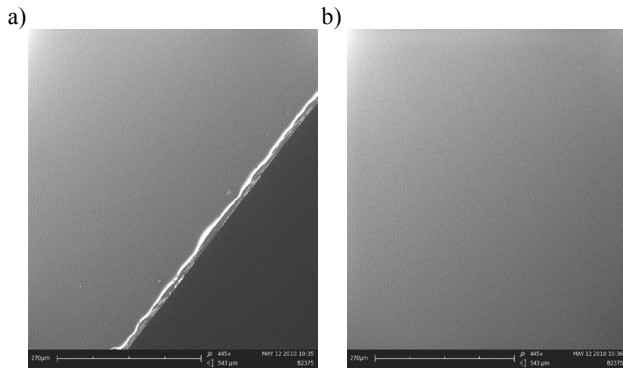


Fig. 10 (a, b). Photographs of the surface layer of the HD-4110 PI (magnitude: 445x)

4.2. UV irradiation

Different UV sources were used to attain anisotropy in PI layers. Polyimide precursors usually absorb wavelengths shorter than 384 nanometres, thus lasers, high-pressure and superhigh-pressure mercury UV lamps with intensive emission lines at 365, 405 and 436 nanometers are used. The energy dose can be modified by different illumination times. Polyimides HD-4100

and HD-4110 are sensitive on i-line (365 nm); polyimide HD-8820 is sensitive on i-line and g-line (436 nm), but they can be also irradiated with broadband radiation.

The UV light causes changes in physicochemical properties of aligning agents, for example: solubility, adhesion strength or surface tension. Linearly polarised UV light can furthermore induce anisotropy in the layer's surface.

To polarise the UV light two types of polarisers were proposed: HNPB/plastic (3M) and wire-grid PPL04A/aluminium (MOXTEK). To calculate the polarisation ratio of the polarisers their transmittance spectra were measured (Figures 11 and 12). On each figure one can see the spectra measured for the single polariser, for two crossed polarisers and for two parallel polarisers. For each polarizer the polarisation ratio (for $\lambda=365$ nm) has been counted using the following equation [20]:

$$PR = \left(\frac{\tau_{\parallel} - \tau_{\perp}}{\tau_{\parallel} + \tau_{\perp}} \right)^{\frac{1}{2}}, \quad (1)$$

where PR is the polarisation ratio, τ_{\parallel} is the transmittance measured for parallel polarisers, τ_{\perp} is the transmittance measured for crossed polarisers. A single PPL04A polarizer has the higher transmittance for 365 nm (near 38%) in comparison with HNPB/plastic (15.5%) polariser but its polarisation ratio is smaller: 0.9901 and 0.9979 respectively.

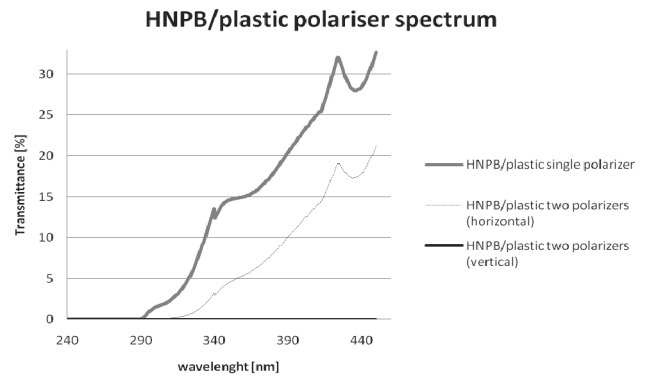


Fig. 11. Transition spectra for HNPB/plastic polariser

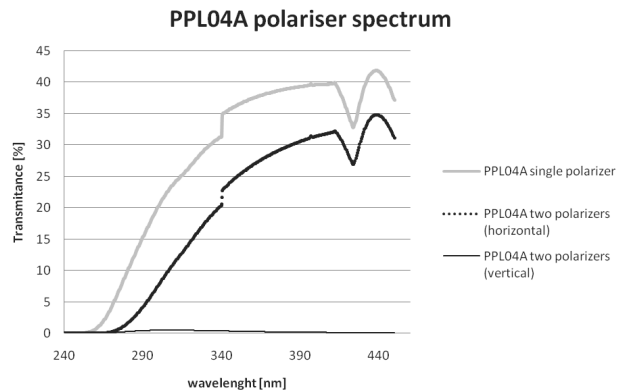


Fig. 12. Transition spectra for PPL04A polariser

Table 2.
The SFE values obtained for HD-4110

Sample - HD-4100 layer	Irradiation time [s]	Energy dose [mJ/cm ²]	SFE [mJ/m ²]	Polar component of the SFE [mJ/m ²]
glass/ITO	0	0	31.67	8.66
non irradiated	0	0	46.57	26.5
irradiated without a polariser	300	940	42.22	8.59
irradiated with HNPB/plastic polariser	300	168	45.1	9.49
irradiated by HNPB/plastic polariser - sample developed	300	168	43.58	6.84
irradiated by HNPB/plastic polariser	900	504	43.31	6.46
irradiated by HNPB/plastic polariser	3600	2016	44.43	8.69
irradiated by PPL04A polariser	300	348	44.28	9.21

4.3. Surface free energy measurements

Surface free energy (SFE) is an important parameter in predicting adhesive qualities of the materials. It can also offer information about changes in material subject to technological treatment. One can calculate the surface free energy on the basis of contact angle measurements [21] with various measurement liquids using Owens-Wendt method [22] also referred to as the Kaelble-Owens-Wendt method [23]. Two types of measurement liquids were used: deionised water (polar liquid) and non-polar liquid: diiodomethane. Eight types of samples made with polyimide HD-4100 layers were investigated. The samples differed mainly in the time of UV irradiation and in the usage of different kinds of polarisers. The values of surface free energy and its polar component can be found in the Table 2. We can see that there is a big decrease in the polar component of the SFE values between non-irradiated and irradiated polyimide layers. The differences between values obtained for layers irradiated by the polariser and without the polariser are much smaller. It seems that the exposure dose of about 168 mJ/cm² (300 s) is sufficient to cause photocrosslinking in the material. Irradiation with higher values of UV energy provides no significant changes in the SFE of the layers. The polar component of the SFE decrease during the irradiation. This effect may be caused by the decrease in the number of the polar groups in the surface layer.

4.4. Twisted nematic (TN) cells

Substrates covered by layers prepared with the use of 15% solutions of HD-4100, HD-4110 and HD-8820 were assembled to form liquid crystals cells 3 µm thick. The cells were mounted in a classical way by means of spacers dispersed in UV-sensible glue and spacers diffused on the whole surface of the layers. Then they were filled with nematic LC 1223A from the Institute of Chemistry, MUT, Warsaw Poland ($T_{CI}=77,7^{\circ}C$; $n_e=1.7020$; $n_o=1.5190$) using the capillary action. The cells were observed through the crossed polarisers.

Twisted nematic cells 10 µm thick were prepared and filled

with nematic LC 1223A. The dynamic electrooptic characteristics as well as numerical simulations of dynamic characteristics one can find in [24].

5. Application perspectives

Because of its' non-contact nature and easiness in multiple domain establishing, the photoalignment technology seems to be a good successor for the rubbing technology, especially for large display panels with high resolution with wide viewing angle. Not only it opens the route to obtain some new solutions in liquid crystal displays, but also some other devices which base on highly ordered organic layers, such as: holographic memory devices, optical shutters, or even solar cells or organic light-emitting diodes.

One of the devices considered in further application in our lab may be some liquid crystal cell with diffraction grating embedded in its' structure.

6. Conclusions

It was shown that polyimide layers which were adequately irradiated using linearly polarised ultraviolet light and were not mechanically treated, show the aligning properties for liquid crystal layers. Such an observation is very important since it fulfilled the main aim of this work - investigation concerning non-contact methods.

Obtained results show that the quality of the orienting polyimide layer depends on numerous technological factors. In this work it has been shown that the critical factors mentioned above are:

- polyimide precursor deposition method and its conditions: polyimide solution concentration and its purity, diluent type, rotational speed during the coating process,
- temperature and soft bake duration,
- polariser selection, especially transmittance spectra of the polarisers and their polarisation ratios and
- adequate dose of LPUV.

The proposed new technology is fairly fine to obtain 2-3 cm² layers. The alignment quality is not perfect, but the works are still in progress.

The deposition process can be modified to allow the control of quality of the aligning layers and their interact with liquid crystal. One of the procedure modifications could be the usage of the adhesion promoters. This modification will probably enable to lower the layer thickness and give an effect of their better homogeneity.

Acknowledgements

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