

Alignment of molecules in Langmuir and Langmuir-Blodgett films of binary mixtures of 3,4,9,10-tetra-(*n*-alkoxy-carbonyl)-perylene with a liquid crystal

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Abstract

Langmuir (L) and Langmuir-Blodgett (LB) films of 3,4,9,10-tetra-(*n*-alkoxy-carbonyl)-perylene ($n=4-8$) and their binary mixtures with 4-octyl-4'-cyano-biphenyl (8CB) and 4-pentyl-4''-cyano-*p*-terphenyl (5CT) have been studied. The surface pressure-mean molecular area isotherms of L films have been recorded, and the alignment of the molecules at the air-water interface has been estimated. The L films were transferred onto the quartz plates at surface pressures below the collapse point. The absorption and fluorescence studies of LB films by using unpolarized and linearly polarized light have been performed. The results obtained have allowed conclusions to be drawn about the arrangement of the dye and liquid crystal molecules at the air-solid substrate interface. The influence of the alkyl chain length substituted to the perylene core of the dye molecules on the molecular organization in L and LB films has been determined. The fluorescence spectra of LB films have revealed the formation of excimers.

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1. Introduction

Liquid crystal displays (LCDs) are today used practically on all sides: in telephones, palmtops, television sets, computers and so on. Up to now, they require, however, a flat, relatively heavy and easily-breaking glass as well as an independent light source. The organic light emitting diodes (OLEDs) secure for the same view as LCDs but they are able to produce light by themselves and therefore are thinner and need less energy. Moreover, just now the research is carried out in order to produce OLEDs from a flexible plastic.

Generally, OLEDs are thin film multilayer structures composed of a hole transport layer, an emission layer and an electron transport layer [1–5]. The most simple OLED consists of just one organic layer located between the hole and the electron injecting electrodes. In this case

the requirements for the organic layer are that it is an efficient luminescer and a good charge transporter, and it must be capable of undergoing efficient charge transfer to an electrode. The transport of charge carriers in organic compounds will be favoured by increasing order of molecules. Liquid crystalline materials possess the ability to self-organize spontaneously into highly ordered structures. Therefore, it is not surprising that already in 1990's Adam et al. [6,7] found that some discotic liquid crystals, which are able to form a columnar phase, display an unusually high charge carrier mobility of the order of $10^{-2} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. However, in initial applications of these liquid crystals in OLEDs the incorporation of the additional organic layer of a dye with electroluminescent properties was required [8,9]. In [10] Benning et al. showed that some perylene-like dyes, namely 3,4,9,10-tetra-(*n*-alkoxy-carbonyl)-perylene, exhibit not only excellent fluorescent properties, but may also form columnar hexagonal liquid crystalline phase in certain temperature range; therefore they can be successfully used as organic layer in OLEDs [11]. The 3,4,9,10-tetra-(*n*-alkoxy-carbonyl)-perylene were first synthesized by Stolarski and Fiksiński [12] and it was ascertained [13] that these dyes have a brilliant

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yellow colour and emit fluorescent light in a spectral region advantageous for the human eye with a significantly high quantum yield. Furthermore, they are able to orient well in calamitic liquid crystals, are stable to the sunlight and do not significantly destabilize the mesomorphic phase of the host. Such features make these perylene-derivatives very interesting for use in LCD devices using the ‘guest-host’ effect (GH LCDs) [14].

In both above mentioned applications the interfacial molecular orientation plays an important role. The Langmuir-Blodgett (LB) technique [15–17] offers a unique method for fabricating ultrathin ordered layers, in which the molecular organization and interfacial interactions can be identified with ease. Moreover, some years ago Stapff et al. [8] proposed to use this technique in order to obtain active layers in OLEDs.

The first required step for the successful LB film fabrication is creation of the compressible monolayer at the air-water interface (Langmuir film) [15–18]. Previously, we showed [19,20] that it is possible to obtain a stable and compressible monolayer at the air-water interface from pure 3,4,9,10-tetra-(*n*-alkoxy-carbonyl)-perylene and to deposit this layer onto a quartz slide.

It is also well established that many of calamitic liquid crystal molecules possess a hydrophobic alkyl chain and a hydrophilic polar terminal group, and are insoluble in water; thus they are able to form mono- or multilayers at air-water and air-solid substrate interfaces [21–29]. In our previous papers [19,29] we mixed 3,4,9,10-tetra-(heptyloxy-carbonyl)-perylene with two liquid crystals: 4-octyl-4'-cyanobiphenyl (8CB) and 4-pentyl-4'-cyano-*p*-terphenyl (5CT) and obtained information about the interfacial organization of molecules in mixed L and LB films.

In this work, we have studied in detail L and LB films formed of binary mixtures of 3,4,9,10-tetra-(*n*-alkoxy-carbonyl)-perylene ($n=4-8$) with 8CB or 5CT. The analysis of the run of the surface pressure-mean molecular area isotherms of L films has allowed to estimate the molecular organization in mixed films at the air-water interface. The optical spectroscopy methods using natural and linearly polarized light have been used to determine the spectral characteristics of LB films and to obtain information about the alignment of molecules with respect to the solid substrate in ultrathin ordered layers of perylene derivatives and of binary dye-liquid crystal mixtures.

2. Materials and methods

The compounds 3,4,9,10-tetra-(*n*-alkoxy-carbonyl)-perylene (**1-5**) (see Fig. 1) were synthesized and chromatographically purified in the Institute of Dyes at Łódź University of Technology. The details of the synthesis are described in [12].

The liquid crystal materials 4-octyl-4'-cyanobiphenyl (8CB) and 4-pentyl-4'-cyano-*p*-terphenyl (5CT) were

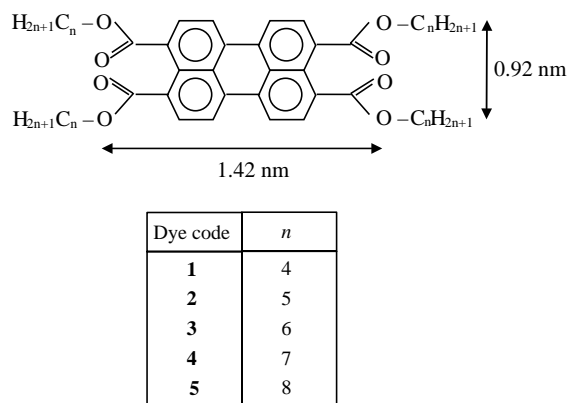


Fig. 1. Molecular structure of 3,4,9,10-tetra-(*n*-alkoxy-carbonyl)-perylene.

purchased from E. Merck (Germany) and were used without further purification: their phase transition temperatures were in substantial agreement with the data given in Merck Catalogue.

The L and LB films were formed in a Minitrough 2 Langmuir-Blodgett system (KSV Instruments Ltd., Finland). Pure compounds **1-5** and their mixtures with 8CB or 5CT were spread from chloroform solutions onto the deionized water subphase and a Wilhelmy plate balance measured the surface pressure as a function of the average molecular area at both the compression and expansion processes. The LB films fabrication was done by using vertical deposition [15–17] and keeping the floating monolayer under constant pressure. The solid substrates with a hydrophilic surface were polished quartz plates. The monolayers were transferred onto these substrates with a transfer ratio of around 1. Successful deposition of 8CB took place only on first raising the substrate. Therefore, for the dyes and their mixtures with both liquid crystals one dipping and one raising were made. Further experimental details about L and LB film preparation are given elsewhere [19,20].

The absorption spectra of LB films were recorded in the UV-Vis region by means of a spectrophotometer CARY

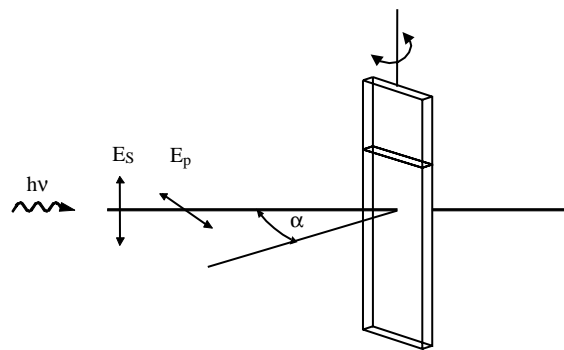


Fig. 2. Geometry for the polarized absorption and fluorescence measurements. E_p and E_s are the electric vectors of the incident (absorption measurement) or emitted (fluorescence measurement) light polarized, respectively, parallel and perpendicularly to the plane of incidence or emission. α is the incidence angle.

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