

Towards a new horizon of optoelectronic devices with liquid crystals

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The liquid crystal having high viscosity is attracting increasing attention as a new type of quality organic semiconductor, i.e., self-organizing molecular semiconductor. In this article, its state-of-the-art in materials, properties related to charge carrier transport, and device applications, is reviewed briefly. In addition, the further researches needed towards its practical applications are discussed.

Keywords: liquid crystal, organic semiconductors, charge carrier transport, device applications.

1. Introduction

Recent revived attention to organic crystals for organic field effect transistors (OFETs) reminds us of the historical studies on the electrical properties of organic single crystals in the 1950's [1]. Those lead to a new paradigm of organic semiconductor [2].

Because of a crystal-like molecular alignment in liquid crystals, it was very natural that a new curiosity was extended to the liquid crystal in the late 1960's [3]. However, this did not result in showing up another aspect of liquid crystals as a self-organizing molecular semiconductor. This is probably because of an accidental coincidence with Heilmier's attractive invention of the first display device utilizing a liquid crystal [4]. The electronic conduction in liquid crystals might be discovered at that time, if Heilmier's display device had not been proposed: his invention attracted considerable attention indeed and triggered a new study on the ionic conduction in liquid crystals [5], because this display device was based on so-called "dynamic scattering mode", where the drift of ions in a liquid crystal cell played a major role for display performance; in fact, many reports described the ionic conduction in various liquid crystals including smectics in the 1970's [6]. As a result, it was very natural that "a wrong recognition" that the intrinsic electrical properties in liquid crystals were governed by ionic conduction had been accepted since then. Thus, the exploration of electronic conduction in liquid crystals was succeeded in discotics having a large disc-shape core moiety and more-like molecular crystal [7,8], after Chandraschal's discovery of a discotic liquid crystal in 1977 [9].

It is indeed in a discotic liquid crystal, i.e., hexahexyloxytriphenylene (H6T) that the first electronic conduction in the liquid crystal was established in 1993 [10]. A few years later, the electronic conduction was discovered in

a calamitic liquid crystal, i.e., a 2-phenylbenzothiazole derivative (7O-PBT-S12) as well [11]. Their chemical structures are shown in Fig. 1. Since then, the electronic conduction was established in various liquid crystals, and it has become recognized to be a common feature in liquid crystals having an aromatic π -conjugate system. Thus, the continuous efforts to find a new aspect of electrical properties in liquid crystals and to understand their unique properties of charge carrier transport have been made in this decade. Nowadays, the liquid crystal is being recognized as a new class of quality organic semiconductors, which is characterized by a high mobility exceeding 10^{-3} cm²/Vs originating from self-organization and electrically inactive structural defects thanks to a soft molecular aggregate.

In this article, the present state of liquid crystal researches in a framework of organic semiconductors is summarized briefly, and what must be done further towards the practical optoelectronic applications with liquid crystalline semiconductors are discussed.

2. Charge carrier transport properties

2.1. Electronic and ionic conductions

The electrical conduction in liquid crystals had been thought to be ionic for a long time as described above, which was easily acceptable due to their liquid-like fluidity. This discrepancy with the present understanding on charge carrier transport in liquid crystals is explained well by purity and viscosity of a given liquid crystal. The electronic conduction is extremely sensitive to chemical impurities that form energy levels in an energy gap of the host liquid crystal, and trace amounts of impurities, e.g., a few ppm, is high enough to terminate it [12]. Impurity molecules ionized by trapping electron or hole can move easily as ions in less viscous mesophases such as the nematic, smectic A, and smectic C phases.

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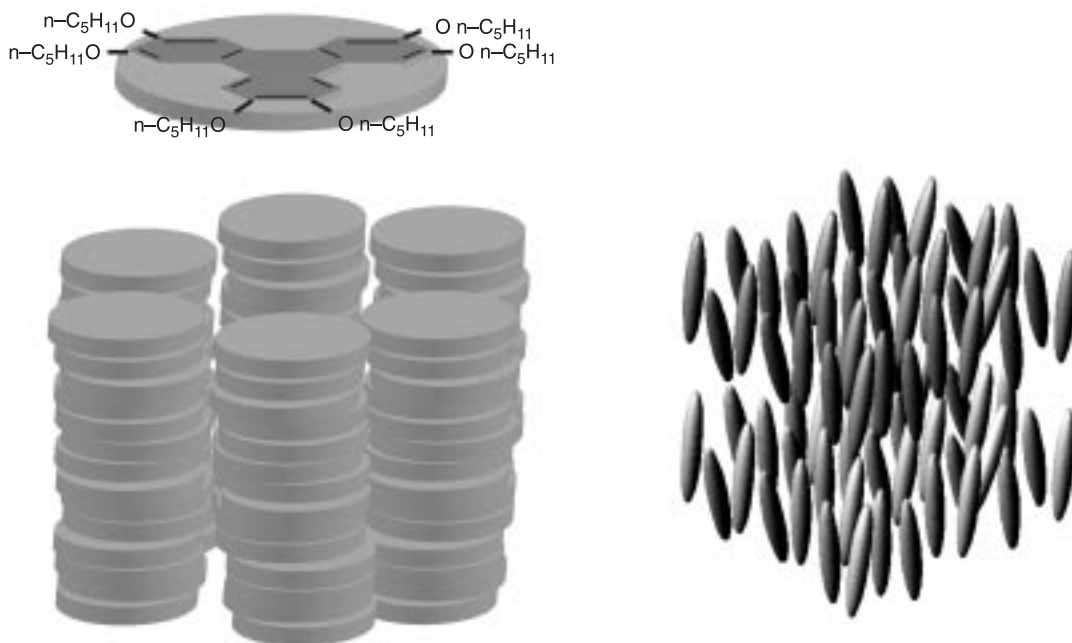


Fig. 1. The first liquid crystals in which electronic conduction was discovered. 2,3,6,7,10,11-hexapentyloxytriphenylene and 2-(4'-heptyloxyphenyl)-6-dodecylthiobenzothiazole.

In fact, a fast transit of electrons and holes characterized by high mobility exceeding 10^{-3} cm²/Vs is always observed in highly purified smectic and discotic liquid crystals, although it often accompanies the slow transit induced by ionized impurities because of insufficient purification, which is characterized by a low mobility less than 10^{-5} cm²/Vs [13], as for the nematics with a small molecular weight, it is very likely that the intrinsic conduction mechanism is ionic.

According to accumulated experimental facts, the electronic mobility in a given mesophase of highly purified materials does not depend on temperature in a temperature range over ambient temperatures [14–16], which can be explained by a narrowly distributed density of states (DOS) as described later on. Therefore, this can be the quick sign of the electronic conduction. One of the convenient ways to distinguish the electronic conduction from the ionic conduction in liquid crystals is to examine how the mobility changes in mixing a less viscous organic solvent such as a hydrocarbon, n-dodecane. For example, the mobility is decreased when the conduction is electronic, while it is increased when the conduction is ionic [13]. Recently, these conduction regimes are discussed in a relation to the microphase-separated structure in mesophases on the basis of experimental results, indicating the existence of two distinctive conduction channels for electronic and ionic charges [17].

2.2. Conduction mechanism

The electronic charge carrier transport in smectic and discotic mesophases can be described by the hopping transport in one-dimensionally and two-dimensionally localized states, respectively [19–21]. The unique charge carrier

transport properties in these mesophases, i.e., independence of both temperature and electric field, are well explained by a narrow distribution of the density of states. According to simulated charge carrier transport characteristics based on a two-dimensional disordered model for smectic mesophases, a Gaussian width of the density of states where charge carriers hop, σ , is estimated to be 50~60 meV as schematically shown in Fig. 2 [20], which is comparable to kT at that temperature range, where k is the Boltzman constant. In fact, this simulated result well reproduces the field-and-temperature independence of mobility. This small value of σ was estimated by analysis of experimental data as well [22]: indeed, at a lower temperature range the mobility depends on both temperature and electric field in the mesophases; the typical value of is 40~50 meV in terthiophene derivatives, which is in a good agreement with the value described above. The origin of small value of σ in mesophases, however, still remains to be explained, while a typical value of σ , 100~120 meV in amorphous materials [23] is attributed to a charge carrier-dipole interaction in the bulk [24–26].

2.3. Purity of materials

Organic materials are synthesized through chemical reactions of starting materials in a step-by-step manner. Therefore the purity of products is always a serious problem. It is well known that charge carrier transport properties in organic crystals degrade seriously in the presence of trace amounts of impurities, e.g., less than a few ppm, that cause trap states in an energy gap of the host crystalline material. However, it is not easy to detect such a small amount of the impurities in conjunction with the charge carrier transport

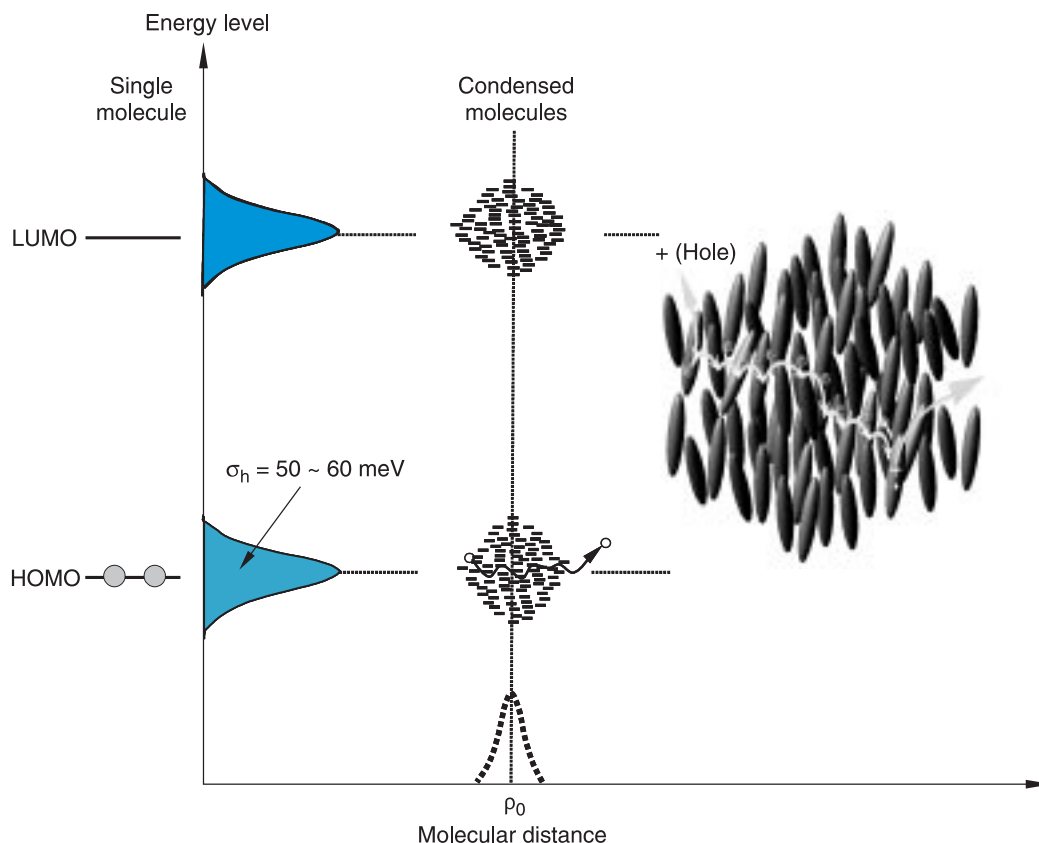


Fig. 2. Density of states model for smectic liquid crystal based on 2-dimensional Gaussian disorder model.

properties, although the gas chromatography (GC) and high performance liquid chromatography techniques and the photoluminescence spectroscopy may help to some extent [27]. As a matter of facts, the purity is assured only by checking how the charge carrier transport properties are affected when a given amount of impurities is intentionally added to a material [28,29]. Because of a narrow distribution in density of states where charge carriers hop in mesophase materials, the charge carrier transport is very sensitive to the impurities as is the case of organic crystals. In fact, the impurities of a few ppm terminate the electronic conduction in the low-viscous mesophases as described above. Therefore, we have to pay much attention to the purity in the mesophase materials.

Fortunately, it is often possible in mesophase materials to detect such a small amount of the impurities by time-resolved transient photocurrent measurements [12,13,30]. The ionic transient photocurrent induced by photo-ionized impurity molecules appears in a slow time range because of a slow mobility, so that it can be distinguished easily from the electronic transient photocurrent in time-of-flight experiments. This is another great advantage over conventional organic crystalline materials.

For purification of mesophase materials, conventional techniques including recrystallization, column chromatography, and vacuum sublimation are available, in which high solubility of mesophases materials in common organic solvent is very helpful.

2.4. Structural defects

In a molecular aggregate having an ordered molecular alignment, the misorientation of molecules always causes structural defects. Typical examples are dislocations and grain boundaries in crystalline materials. These defects are always electrically active and cause trapping and/or recombination centres for charge carriers, resulting in low performance of devices. To make the matter worse, they are often associated with degradation of materials as well. Therefore, we have to take care of the grain boundaries in organic polycrystals. The structural defects in liquid crystals such as disclinations and domain boundaries, however, are electrically inactive and hardly cause trap states for the carriers [31–33]. This is a great advantage of mesophase materials over the conventional crystalline materials. This can be a good basis for the mesophase materials in optoelectronic devices such as imaging devices requiring a large-area.

2.5. What is the highest mobility?

From materials point of view for device applications, it is one of the most interesting questions that what the highest mobility can be in mesophase materials. Of course, it cannot be higher than that of organic single crystal. According to the disorder formalism for charge carrier transport in smectic phases, which is based on a two-dimensional hopping mechanism as described above, the mobility at tem-

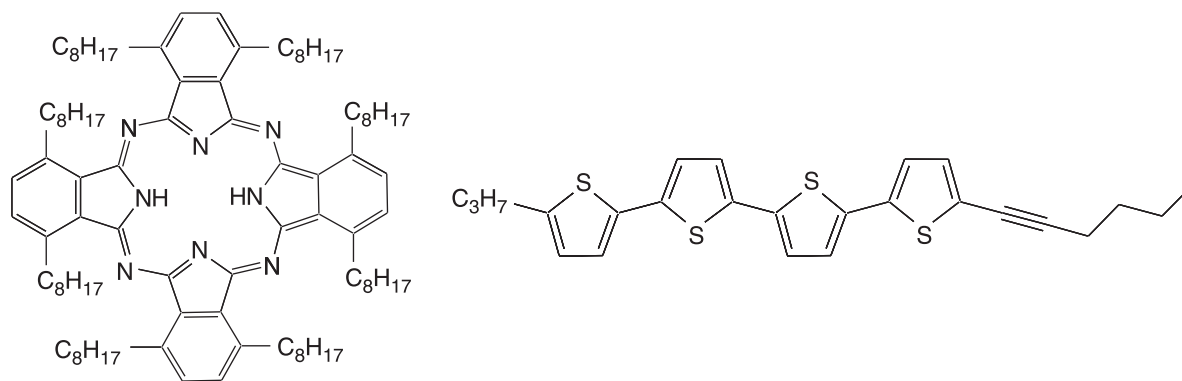


Fig. 3. Liquid crystals exhibiting the highest bulk mobility phthalocyanine derivative $\mu_e = 0.3 \text{ cm}^2/\text{Vs}$, $\mu_h = 0.2 \text{ cm}^2/\text{Vs}$, and quaterthiophene derivative $\mu_h = 0.1 \text{ cm}^2/\text{Vs}$.

temperatures where the charge carrier mobility depends on neither temperature nor electric field can be described by the following equation [20]

$$\mu = \mu_0 \exp \left[- \left(a \frac{\sigma}{kT} \right)^2 \right],$$

where a is the Gaussian width of density of states, k is the Boltzmann constant, and T is the given temperature.

Taking into account the value for σ , i.e., 40~60 meV in the mesophases materials, the mobility at the ambient temperatures is calculated to be 0.1~0.2 μ_0 , where μ_0 is a mobility for a system without any disorder. Therefore, with a mobility of 1~10 cm^2/Vs in organic single crystals for μ_0 , the highest mobility is estimated to be 0.1~1 cm^2/Vs in mesophase materials. In fact, the mobility exceeding 0.1 cm^2/Vs has been reported for the highest mobility not only in discotic but also in smectics. Typical examples are shown in Fig. 3. This value is comparable to those of organic polycrystals. Therefore, taking account of electrically inactive structural defects in mesophase materials, they can be a good competitor against the organic polycrystals for organic FETs.

3. Materials design and synthesis

Up to now, it has been reported that many different types of liquid crystals exhibited a high mobility ranging from $10^{-4} \text{ cm}^2/\text{Vs}$ up to $\sim 1 \text{ cm}^2/\text{Vs}$, including triphenylenes [34,35], phthalocyanines [36,37], porphyrines [38], perylenes [39], and hexabenzocoronenes [40] of discotics, and phenylbenzothiazoles [11], phenyl-naphthalenes [14,15], origothiophenes [16,41], benzothienobenzothiophenes [42] and polymeric liquid crystals. [43~47] as shown in Fig. 4.

Judging from the experimental results reported on the charge carrier transport in the mesophase materials so far, it is clear that a high mobility and its independence of electric field and temperature are not a specific characteristic in a particular material but the common characteristics in the mesophase materials. In addition, it is very clear that the mobility basically depends on how molecules are aligned

in a mesophase. The mobility is always upgraded abruptly when the molecular alignment is sophisticated at the phase transition temperature. For these reasons, we can be optimistic to find out a practical material suitable for optoelectronic applications, in addition to a lot of room for materials design.

For achieving high mobility, it may be true that mesophases materials consisting of large molecules with an extended π -conjugate system such as phthalocyanines and benzocoronenes are beneficial indeed, but it should be noted that the mobility exceeds $10^{-3} \text{ cm}^2/\text{Vs}$ even in a small molecule having a benzene unit such as biphenyls [48]. This mobility is comparable to the highest mobility in amorphous organic semiconductors and higher than that of H6T.

4. Device applications

The organic semiconductors have been utilized in xerographic photoreceptors and light emitting diodes (LEDs). Since these devices require a uniform large-area for imaging applications, the organic semiconductors in these applications have been used in a form of amorphous thin films. Because of no molecular alignment in amorphous films, an average molecular distance is not less than 6 Å, leading to a small mobility on the order of 10^{-6} ~ $10^{-5} \text{ cm}^2/\text{Vs}$ and $10^{-3} \text{ cm}^2/\text{Vs}$ at best. These are two to three orders of magnitude smaller than those of organic crystalline materials. Therefore, in order to make a mean intermolecular distance smaller without crystallization, bulky π -conjugated molecules having multiple chromophores for hopping sites are designed for practical use in these devices. Typical examples are triphenylamine derivatives including TPD and NDNPD [23].

From the device performance point of view, there are neither xerographic photoreceptors nor organic LEDs without making allowance for a slow process speed of tens copies/min in copiers, and without various devices for compensating the poor charge carrier transport properties in the amorphous films, for example, a thin device structure of 0.1 μm in organic LEDs.

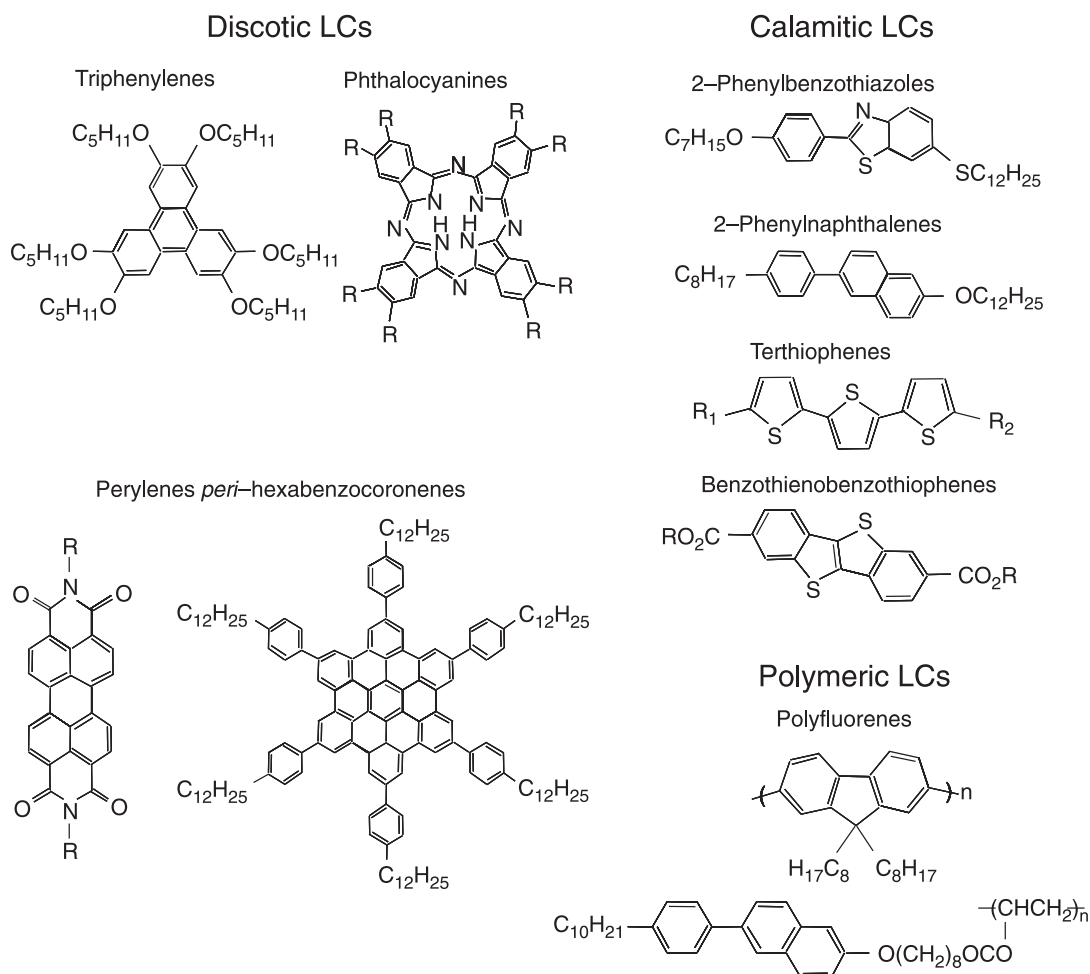


Fig. 4. Typical example of liquid crystals exhibiting electronic conduction.

Since the electronic conduction was established in mesophase materials in the 1990's, various optoelectronic devices have been fabricated by using smectic and discotic materials in order to demonstrate their availability in device applications. These include a high-speed photosensor [49], light emitting diodes [50–58], solar cells [59–61], a xerographic photoreceptor [62,63], a position sensor [64], and field effect transistors [65,66]. These still remain in a very primitive stage, but have high potential for future development because of the allowance for polydomain structure in the devices thanks to electrically inactive domain boundaries, in addition to a high mobility comparable to those of organic polycrystals.

All these devices demonstrated are on the extension of existing devices for higher performance, whose upgraded performance is thanks to the liquid crystals as a quality organic semiconductor. Considering another aspect of liquid crystalline semiconductors as “a liquid crystal”, which is characterized by liquid-like fluidity and easy orientation and its dynamic control, there exist more room for novel functionality optoelectronic devices that cannot be achieved with conventional materials. The position sensor with a wedged cell could be one of the examples for this trend. In addition, the liquid crystals have a high solubility

in common organic solvents, so that a wet-process is available for fabrication of devices. Recent fabrication of a solar cell with a discotic liquid crystal and a pigment is very interesting from this point of view.

5. Feature works towards practical device applications

The electrical properties and specific features of liquid crystals clarified in this decade promise their applications as organic semiconductors for optoelectronic devices, as briefly reviewed in the previous sections. However, there remain several issues to be investigated before practical device applications. They contain materials design, purification technique of materials, liquid crystal/electrode interface, device fabrication processes, and device structure and new devices, which are discussed as follows.

5.1. Materials design

For device applications, the organic semiconductor satisfies several requirements, e.g., a high mobility, appropriate HOMO and LUMO levels relative to electrode materials, and a long-term stability against ambient atmosphere. In

addition, a wide temperature range for a mesophase, e.g., from -20 to -80°C , is required in the liquid crystal. The conventional liquid crystals available for display applications are not suitable for new applications in optoelectronic devices, even though they may exhibit a considerably high mobility. After ten years for establishing the scientific basis to prove the promising future of liquid crystals as organic semiconductors, now it is the time to focus on the material design for practical use and synthesis of liquid crystals satisfying the requirements described above.

5.2. Purification technique

For display applications of liquid crystals, the high attention has been paid to ionic impurities in liquid crystals that cause a short holding time and “burning effect” in the displays. On the other hand, for optoelectronic applications another attention has to be pay to a trace amount of chemical impurities having their HOMO and/or LUMO levels in an energy gap of a host liquid crystal, e.g., less than a few ppm, because of its harmful effect on charge carrier transport [12,30]. Accompanying with the materials design and synthesis of liquid crystals, therefore, we have to develop an efficient method for evaluating chemical impurities in a liquid crystal and for purifying a liquid crystal.

5.3. Liquid crystal/electrode interface

In device operation, the blocking contact at the electrode interface is easily achieved in general, but the ohmic contact is often very difficult to achieve. In fact, many new approaches including doping for an ohmic layer have been tried to achieve high performance organic LEDs, in addition

to continuous efforts in the materials design for appropriate HOMO and LUMO levels in organic semiconductors. These are needed in the mesophases material also, where the unique features in the mesophases material could be utilized to give a unique answer.

5.4. Device fabrication processes

Liquid crystalline molecules have anisotropic molecular structure, which is similar to pentacene and 6T often used in polycrystalline form for organic TFTs. However, the liquid crystalline molecules have lower melting point and higher solubility in common organic solvents because of long alkyl chains. These are a great advantage over the conventional polycrystalline materials from a device fabrication point of view, a wet process is available for fabricating devices basically; in addition, the well-established cell technologies in liquid crystal displays could be utilized as well.

In addition, in order to make the best use of liquid crystals as organic semiconductors, we have to develop the new device fabrication processes that the conventional materials are not available. In this point, the distinctive features in the liquid crystals described above are very helpful and may provide us with an unconventional process for device fabrication.

5.5. New devices and device structure

It is no doubt that the improvement of device performance in existing devices would be achieved with a quality organic semiconductor of liquid crystals indeed, but the creation of unconventional functionality devices with liquid

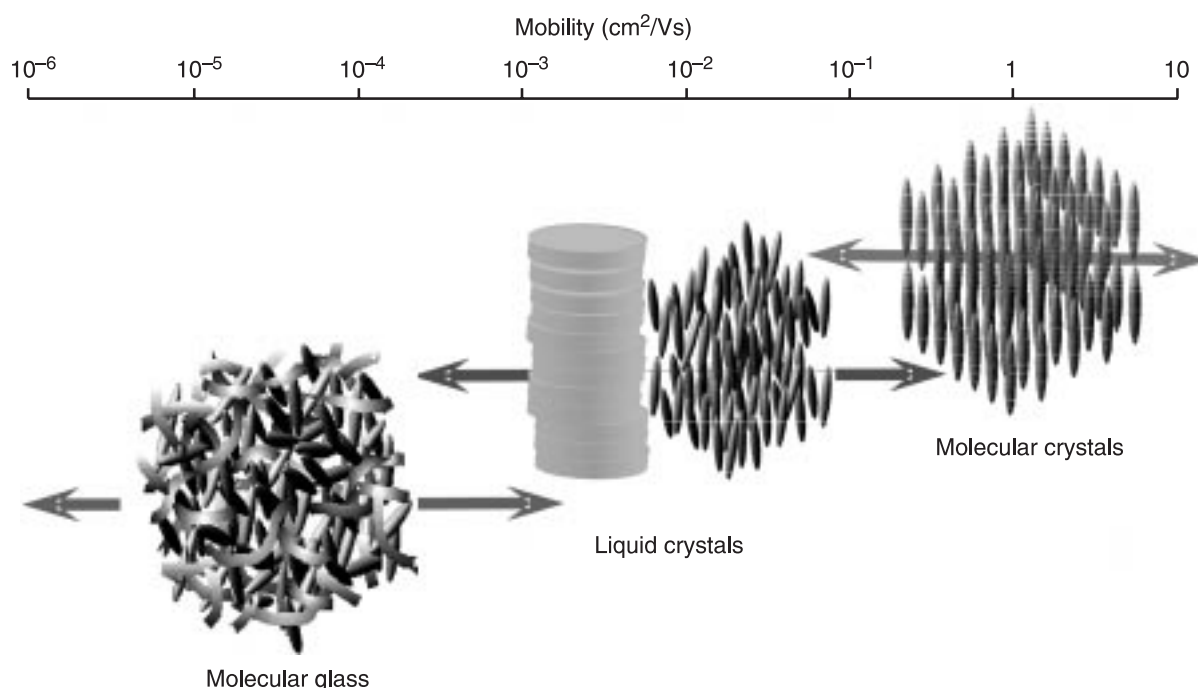


Fig. 5. Molecular aggregates of organic materials and typical mobility.

crystals is more beneficial. Taking account of unique properties of liquid crystals such as liquid-like fluidity, self-organization, easy and/or dynamic control of molecular alignment, and the anisotropy in optoelectronic properties including optical absorption and charge carrier transport, there is a large room for designing novel functionality devices and device structure that can never achieve with the conventional materials including amorphous and polycrystalline materials. These would distinguish the liquid crystals from the conventional organic semiconductors.

6. Conclusions

Since the electronic conduction in liquid crystals was discovered in the 1990's, ten years have passed. With continuous interests in exploring its new edge, a good basis for the second step towards device applications has been well established, including general features in charge carrier transport properties and their theoretical understandings in liquid crystals, which provide us with the scope and limitation of their properties, and variation of materials having high mobility. Thus, we can give a right position to the liquid crystalline materials in organic materials as shown in Fig. 5.

Now, we need another effort to open the door looking a new horizon of organic optoelectronic devices through, with a step by step approach to have the answers to the issues described in the future works, in which we have to co-operate with materials scientists, physicists, chemists, and electrical engineers as it happened to liquid crystal displays.

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