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The Roles of Polarizabilities and Dipole Moments in Hole Transport Processes in Molecularly-Doped Polymers

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Polarizabilities and dipole moments of hole transport molecules with different mobilities were measured. The zero-field mobilities of the molecularly-doped polymers increase with increasing polarizabilities and decrease with increasing dipole moments of the dopants. Concerning molecules with similar dipole moments, mobilities correlate well with polarizabilities. Also concerning molecules whose polarizabilities are almost equal, the correlation between mobilities and dipole moments is clear. The 3-D plot of the mobility as a function of the polarizability and the dipole moment is nearly coplanar. Within a framework of the small polaron formalism, polarizabilities are good indices to evaluate the degree of electron cloud extension that affects transfer integral J. On the other hand, the magnitude of the polaron binding energy E_p is macroscopically influenced mostly by the intramolecular structural relaxation. However, we propose that the E_p has a distribution caused by intermolecular interactions, which is not considered in the small polaron formalism. Such distributions, which have recently been discussed within a framework of the disorder formalism, are due to the random internal field caused by the local charge-dipole interaction. These analyses support the experimental data that the logarithm of mobility is described as a linear combination of the polarizability and the dipole moment.

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Introduction

There is considerable interests in charge transport phenomena in molecularly doped polymers that are practically used as the charge transport layer of photoreceptors for electrophotography. Compared with other amorphous organic glass systems, one of the unique features of molecularly doped polymers is that hopping site distance can be controlled by varying the dopant concentration. Moreover, very weak van der Waals interactions between molecules lead to very narrow energy bands, and the charge carrier is localized in one molecular site. Recently, the charge carrier transport of a variety of charge transport molecules and binder polymers has been studied actively. In doped polymers, hole or electron transport occurs by the transfer of charge among states associated with the dopant molecules. The charge transfer is characterized by an electric-field-driven chain of redox processes between neutral and ion radical states of the charge transport molecules. Many studies have been preformed to describe the hopping processes in terms of the intersite distance, electric field, and temperature dependencies.

Among a variety of theories to explain the experimental data, the polaron theory [1–4] and the disorder theory[5–8] have been accepted as plausible explanations for the charge transport properties. The small polaron formalism well explains the Arrhenius-type temperature dependency of the mobilities. One of the merits to use the small polaron model for the analysis of the carrier transport process is that the physical background of the used parameters is clearer than other theories. Also its success in the case of molecular crystals[9] supports the applicability. However, some limitations have been pointed out concerning the interpretation of the experimental data observed in molecularly-doped polymer films. For example, the small polaron formalism cannot predict the $E^{1/2}$-type electric field dependency of the mobility.[2] Moreover, the small polaron formalism is not successful in explaining the negative field dependency of the mobility at low field reported in several experiments.[10–13] Such dependency can successfully be explained based on the disorder formalism. In the disorder formalism, charge transport is assumed to occur by hopping through a Gaussian distribution of localized states with superimposed positional disorder. The key parameters of the formalism are σ, the width of the energy distribution of states, and Σ, a parameter that describes the degree of positional disorder.

In early polaron arguments, polyacene crystals have been actively studied.[9] In such systems, it is believed that during the time a carrier is localized on a hopping site, the charge induces polarization of the electronic orbitals, mainly the π orbitals, of surrounding molecules. For a mobility of 10^{-4} cm^2/Vs, electronic polarization is several orders of magnitude faster than the residence time of carriers on one molecule which is approximately 10^{-10} s. Thus the charge-induced dipole interaction between a localized carrier and the polarized neighboring molecules occurs. In the small polaron formalism, a carrier moves, not as a free particle, but dressed with a deformed lattice while moving. In the early polaron

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treatment, it was generally assumed that the deformation is associated with intra- and inter-molecular vibrational modes in the case of molecular solids. In molecularly doped polymers where intermolecular interaction is so weak that a carrier is almost localized, a structural relaxation of neutral and cationic molecules can be dominant to form the polaron. However, Vannikov and co-workers have argued that the intermolecular contribution is also important.[14] In the disorder formalism, the distribution of hopping site energy is considered to be essential to the carrier transport. The permanent dipole has been suggested as an important factor affecting the transport, because random distribution of permanent dipoles generates fluctuations in electrostatic potentials that increase local variations of hopping site energy.[15]

In this study, we use two physical parameters, polarizability and the dipole moment of the hole transport molecules, for analyzing the mobility in molecularly doped polymers. Several experimental data indicate that increasing the polarity of the transport molecule has decreased the hole mobilities in doped polymers[16–18] and in vapor-deposited molecular glasses.[19,20] Borsenberger and co-workers have analyzed the effect of dipole moments based on disorder formalism and divided the energetic disorder parameter σ into a dipolar component and a van der Waals component.[15,21–23] The dipolar component relates the dipole moment through the expression proposed by Young.[24] On the other hand, there have been few arguments in molecularly doped polymers describing the roles of polarizabilities on carrier transport processes based on experimental evidence. In this article, the relationship between mobilities and the polarizabilities as well as the dipole moments of hole transport molecules are discussed.

Experimental

The molecular structures of the charge transport materials (CTMs) used in this study are illustrated in Fig. 1. These molecules are electron-donating, thus a charge carrier is a hole. Films of these materials were prepared by dissolving 1:1 weight ratios of CTMs and bisphenol-A-polycarbonate in 1,4-dioxane and then the resulting solutions were bar-coated onto Al-deposited poly(ethylene terephthalate) substrates that had previously been coated with β-oxytitanium phthalocyanine-dispersed poly(vinyl butyral) as a charge generation layer (CGL). Samples were dried at 125°C for 20 min and then 55°C for 48 h. The typical thickness of the charge transport layer (CTL) was 10 to 15 μm. Al was then vacuum-deposited onto the free surface of the CTL to form a semi-transparent counter electrode.

A standard time-of-flight technique was used to measure hole drift mobilities. After application of voltage, carriers were generated in the CGL when excited with light from a xenon flash lamp monochromated with a 700 nm interference filter. The displacement of a sheet of holes induced by the excitation of the CGL was time-resolved. To minimize distortion of the internal field by the injected charge, the exposures were filtered such that the total charge injected into the CTL was less than 0.05 CV, where C is the sample capacitance, and V the applied voltage. The transient photocurrent was measured with a current-voltage amplifier (Keithley 428) and a digital storage oscilloscope (Nicolet model 3091). All transit times were obtained on log–log current-versus-time plots, and the shoulder identifies the transit time τ which is related to the drift mobility μ through

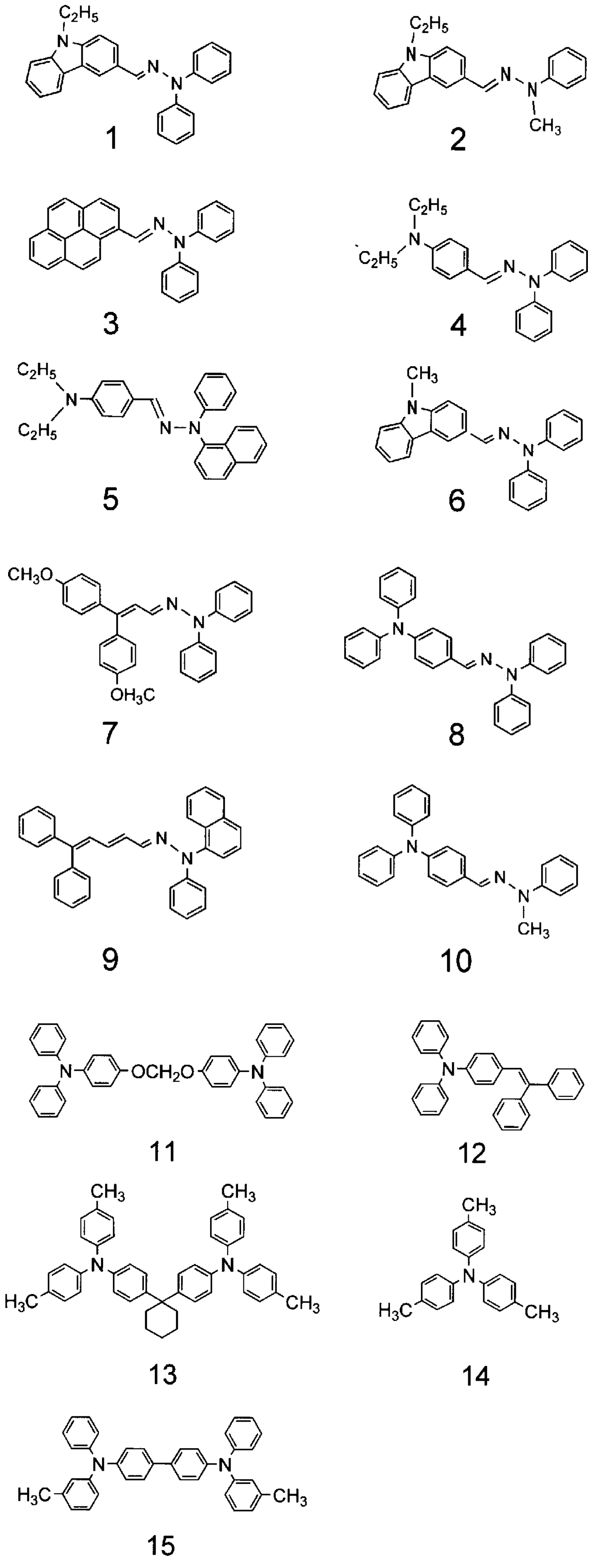

Figure 1. Molecular structures of the hole transport materials.

TABLE I. Dipole Moments, Polarizabilities, and Mobilities of Hole Transport Molecules

CTM	Dipole moment (D)	Polarizability (Å^3)	Zero-field mobility (10^{-6} cm^2/Vs) 297 K	Zero-field mobility (10^{-6} cm^2/Vs) calculated from Eq. 1	Mobility (10^{-6} cm^2/Vs) 3.0×10^5 V/cm, 297 K
1	2.67	59.1	0.94	0.91	6.2
2	2.57	48.6	0.092	0.80	0.87
3	2.48	62.3	1.4	1.3	9.6
4	3.37	49.8	0.27	0.24	2.6
5	3.34	57.7	0.26	0.31	2.4
6	2.65	57.8	0.60	0.90	4.4
7	3.48	65.9	0.58	0.31	4.1
8	2.51	65.6	2.8	1.4	1.2
9	2.57	71.6	5.5	1.5	21
10	2.35	56.1	1.9	1.4	9.1
11	1.78	74.8	1.6	5.4	5.7
12	1.25	62.3	14	8.7	30
13	1.38	86.5	11	13	19
14	0.95	38.2	17	7.2	29
15	1.39	75.7	6.7	10	15

the relation $\tau = L^2/\mu V$, where L is the thickness of the sample. Zero field mobility was obtained by plotting the mobility versus square root of electric field, followed by extrapolating to zero field.

Dipole moments and polarizabilities were determined by measuring the dielectric constant and refractive index of several concentrations of dilute 1,4-dioxane solutions of CTMs, following the procedures previously described.[25,26] A concentration extrapolation was applied to dielectric constant and refractive index measurements in order to eliminate solute–solute interactions.

Results and Discussion

Table I shows the hole mobilities, polarizabilities and dipole moments of the molecules shown in Fig. 1. Figure 2 shows the mobilities as a function of polarizabilities and dipole moments. Some corrrelations are found between mobilities and polarizabilities as well as dipole moments. The zero-field mobilities increase with increasing polarizabilities and decrease with increasing dipole moments. With regard to the relationship between the mobilities and polarizabilities, there seems to be some correlation, but not enough to explain the mobilities only by the polarizabilities. Also concerning the relationship between the mobilities and the dipole moments, the correlation is not so distinct as previously reported.[16–18] In particular, without the data for materials with dipole moments over 3 Debye and less than 1.5 Debye, the correlation becomes poorer. This is also the case with previously reported data. There, correlations seem to be distinct because of the use of molecules having relatively small (< 1.5 Debye) or large (> 3.5 Debye) dipole moments, and without such molecules, i.e., molecules whose dipole moments are about between 1.5 to 3.5, the correlations are very poor.

Then we found that for the molecules having dipoles moments between 2 and 3 Debye, the correlation between mobilities and polarizabilities is good as shown in Fig. 3, suggesting that in this series of molecules, mobilities are described better with polarizabilities than with dipole moments. Also in the previous data[16–18] indicating correlations between mobilities and dipole moments, fluctuations of data especially at dipole moments between 1.5 to 3.5 Debye are not negligible. In such a range of dipole moments, a correlation between mobilities and polarizabilities then becomes appropriate to explain the magnitude of mobilities. Similarly, for molecules with approximately equal polarizabilities, the mobilities are

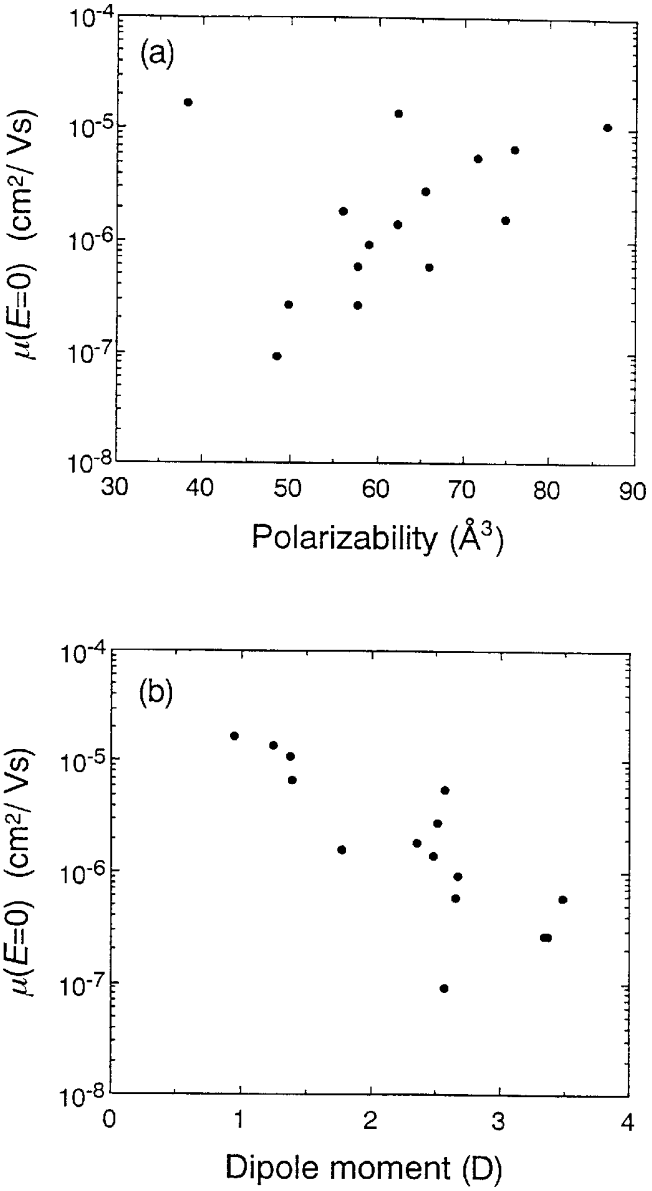

Figure 2. Zero-field hole mobility as a function of (a) polarizabilities and (b) dipole moments.

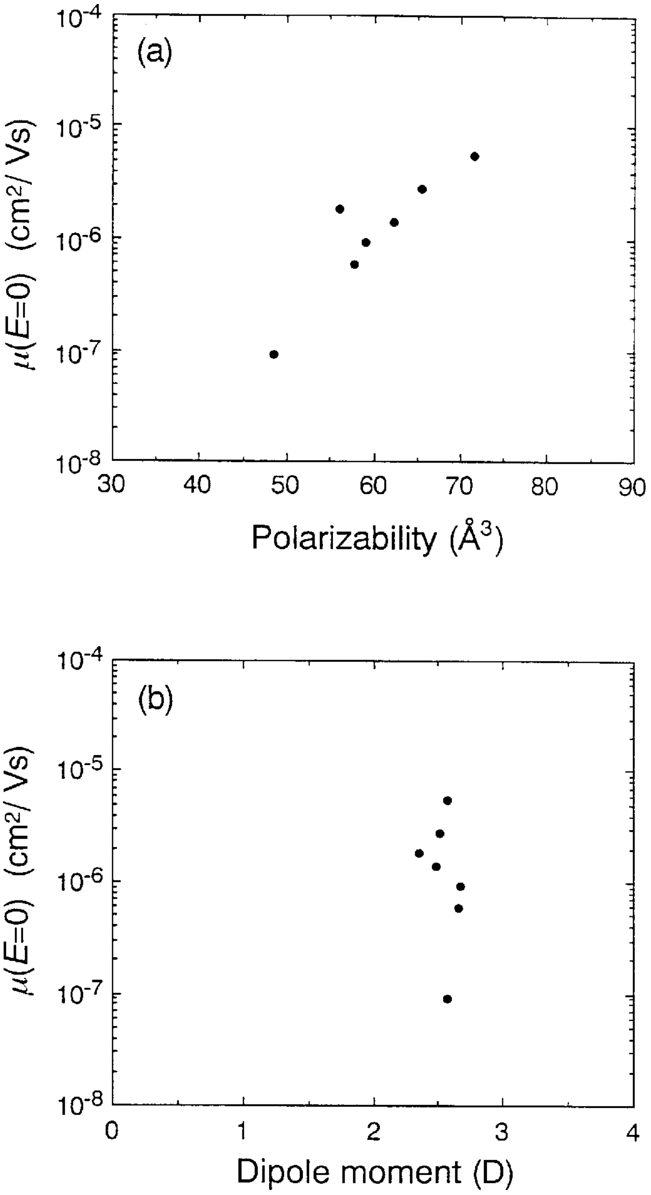

Figure 3. Zero-field hole mobility of materials with similar dipole moments plotted versus (a) polarizabilities and (b) dipole moments.

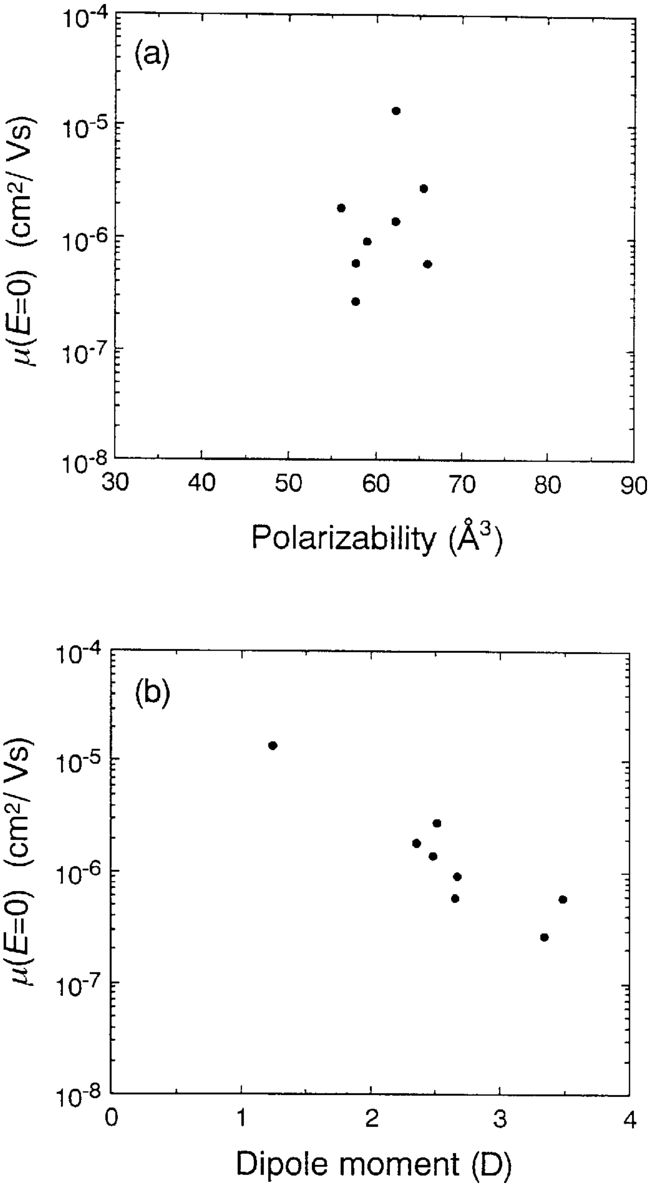

Figure 4. Zero-field hole mobility of materials with similar polarizabilities plotted versus (a) polarizabilities and (b) dipole moments.

described better with dipole moments than polarizabilities, as shown in Fig. 4. The correlation between mobilities and dipole moments is good especially when the polarizabilities of the molecules used are similar.

Therefore it is meaningful to plot the mobility data as a function of both the dipole moment and the polarizability. Figure 5 shows the 3-D plot of mobilities as a function of the polarizability and the dipole moment. The plot seems to be nearly planar, indicating that the logarithm of mobility μ can be described as a linear combination of the polarizability and the dipole moment. According to a multiple regression analysis,

$$\ln \mu(E = 0) = 0.0266\alpha - 1.53p - 11.4, \tag{1}$$

where α is the polarizability (Å³) and p is the dipole moment (D). The multiple correlation coefficient was 0.841. The statistical test indicated that $\ln \mu(E = 0)$ is more dependent on p than α. However, with regard to only the hydrazone molecules **1–10**, α is a dominant factor. The calculated values of the mobility predicted by Eq. 1 together with the measured values are shown in Table I.

Next, physical interpretation of this result is discussed. It is well known that the charge carrier mobility μ in molecularly-doped polymers depends on intermolecular distance ρ, electric field E, and temperature T. The form of drift mobility observed experimentally is

$$\begin{aligned} \mu &= a_0\rho^2 \exp f_1(\rho) \exp f_2(T,\rho) \exp f_3(E,T,\rho) \\ &= a_0\rho^2 \exp[-(2\rho/\rho_0)] \exp(-\Delta(\rho)/kT) \\ &\quad \exp(\beta E^{1/2}[1/kT - 1/kT_0(\rho)]), \end{aligned} \tag{2}$$

where a_0 is a constant, k is the Boltzmann constant, and ρ_0 and Δ represent the wave-function decay length and zero-electric-field activation energy, respectively. β and T_0 are parameters fit to the data. Procedures by Mack, Schein and Peled have enabled us to deconvolute the

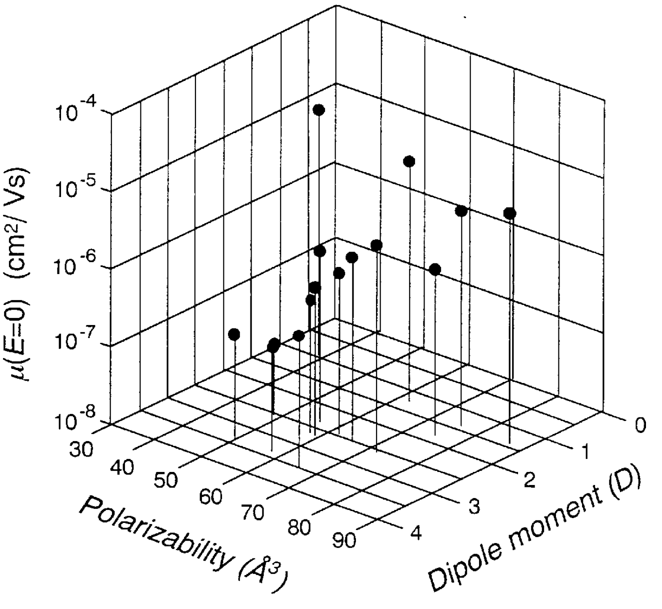

Figure 5. 3-D plot of zero-field hole mobilities as a function of polarizabilities and dipole moments.

dependence of E, T, and ρ on mobility μ.[1] They analyzed the transport process in the molecularly-doped polymers based on these procedures and interpreted the data within a framework of small-polaron hopping theory. The mobility of a small polaron in the limit of zero electric field can be expressed as

$$\mu(E = 0) = (e\rho^2/kT)P(\omega/2\pi)\exp[-(E_p/2 - J)/kT)], \quad (3)$$

where E_p is the polaron binding energy, J the transfer integral, and ω the phonon frequency associated with the deformation. In comparison with Eq. 2,

$$P \propto \exp[-(2\rho/\rho_0)]\ , \ \Delta(\rho) = E_p/2 - J(\rho). \quad (4)$$

P represents the probability that a charge carrier will hop once an energy coincidence occurs. Two regimes are predicted by this theory. The probability P and the activation energy $\Delta(\rho)$ have different forms in the two regimes. First, nonadiabatic regime is defined by P<1. In this regime, P depends on the transfer integral, $P \propto J^2 \propto \exp[-(2\rho/\rho_0)]$. Also, because J is small, the activation energy $\Delta(\rho)$ is essentially independent of intermolecular distance ρ : $\Delta(\rho) = E_p/2$. Second, in the adiabatic regime, P does not have ρ dependency, which does not obey the relation described in Eq. 4, and then alternatively defined by $P = 1$. In this regime the electron cloud overlap between each molecule is large enough that a jump is assured whenever there is an energy-level coincidence. Because $J(\rho)$ is large, the activation energy is reduced from the polaron binding energy, i.e., $\Delta(\rho) = E_p/2 - J(\rho)$. In both regimes, $J(\rho)$ and E_p are the key parameters.

First, transfer integral $J(\rho)$ is closely related to the extension of the wave function of the charge transport molecule. With increasing $J(\rho)$, an increase in P in the nonadiabatic regime and a decrease in $\Delta(\rho)$ in the adiabatic regime enhance the mobility. This implies that an increase in the electron cloud extension of a charge transport molecule, which leads to an increase in $J(\rho)$, is advantageous in view of increasing the mobility. In order to estimate the magnitude of $J(\rho)$, polarizability is a good index because the polarizability of a molecule measures the extension of the electronic distribution.[27] Also the polarizability shows how the electron cloud distorts the electric field. In the hopping process, the hole transport is more likely to occur when the distorted electron cloud of a neutral molecule comes closer to the neighboring charged molecule. Thus it is plausible that large polarizability contributes to the hole transport.

Concerning molecules used in this study, the polarizability is closely related to the extension of π-systems because the molecules as described in Fig. 1 have highly-conjugated skeletons. It might also be accompanied by stabilization of charge carriers which could occur by polarization of surrounding polarizable molecules through charge-induced dipole interactions. This effect could lead to a decreasing mobility with increasing polarizability. However, we consider such stabilization of charge carriers through charge-induced dipole interactions is unlikely because of the data shown in Fig. 2a. Therefore, we believe that large polarizability is advantageous to exhibit high mobility. With regard to the conjugated systems of compounds used in this study, it is natural that the extension of conjugated systems is also advantageous to attaining high mobility, especially because the molecules used here are highly-conjugated and do not have strongly electron-donating or electron-withdrawing substituents. However degree of the extension of conjugated systems is not a significant enough factor to estimate the mobility especially in the case of molecules with substituents such as halogen, nitro, cyano, etc.

Next, polaron binding energy E_p represents the degree of stabilization of the small polaron in the hopping process. In molecularly doped polymer systems, the possible reasons for the stabilization are:

(i) the relaxation accompanied by the structural changes of neutral and cationic molecules,
(ii) charge-permanent dipole interactions and
(iii) charge-induced dipole interactions.

Among these factors, (i) is intramolecular; (ii) and (iii) are intermolecular effects. Concerning intermolecular effects, charge-permanent dipole interactions (ii) include those between a radical cation of a charge transport molecule and surrounding polar units of charge transport molecules together with binder polymers. When the dipole moment of surrounding charge transport molecules is large, it might be expected that the probability that a charge carrier is strongly bound to the site increases. However, it has been reported[1,2,28,29] that in nonadiabatic regimes $\Delta(\rho)$ values that are equal to $E_p/2$, do not depend on the intersite distances. Also in adiabatic regimes, the decrease in $\Delta(\rho)$ with decreasing intersite distances was ascribed to an increase in $J(\rho)$, thus E_p was thought to be constant.[1,2] It was also reported that E_p is constant regardless of the binder polymers.[28] This experimental evidence indicates that charge-permanent dipole interactions between charged molecules and neutral charge-transport molecules as well as binder polymers do not affect E_p.

In addition to the charge-permanent dipole interaction, the charge-induced dipole interaction (iii) might be taken into consideration. However, this effect can be neglected also because E_p is independent of intersite distances and polymers. Moreover, as shown in Fig.2a, mobilities increase with increasing polarizabilities, which implies that the stabilization of charge carrier by large induced dipoles of surrounding molecules through charge-induced dipole interaction is not important. The experimental evidence described above infers that the magnitude of E_p is independent of intersite distances and polymers suggests that intermolecular interactions do not affect the magnitude of E_p thus E_p is intramolecular in origin.

With regard to intramolecular effects (i), Pacansky, McLean and Miller pointed out[30] that the changes in molecular structures between neutral and radical cation states are significant in hydrazone derivatives. Also Kanemitsu reported[31] that E_p is determined by the relaxation accompanied by the structural changes of neutral and cationic molecules and neglected intermolecular effects. Therefore, we consider that the effect of dipole moment of charge transport molecules is not reflected in the small polaron formalism expressed in Eq. 3, and the magnitude of E_p is determined mostly by the structural relaxation. However, it is plausible that even though the magnitude of E_p does not macroscopically seem to be influenced by the intermolecular interactions (ii) and (iii), E_p can have some distribution and be influenced microscopically by intermolecular interactions as a perturbation. This means that the mean value of E_p is determined by structural relaxation, but the distribution of E_p can be ascribed to intermolecular interactions such as the charge-permanent dipole interaction. Although there is no experimental evidence that intermolecular interactions (ii) and (iii) are ascribed to such distribution of E_p, we think the following view is possible: once a charge hops to one site, accompanied by stabilization by structural relaxation, the energy level of the charged site is influenced by the surrounding random dipoles as a perturbation.

Although the energetic fluctuation is not considered in the traditional small polaron formalism, the energetic fluctuation based on dipolar dopants has recently been discussed within a framework of the disorder formalism. In the disorder formalism, the zero-field mobility is expressed as

$$\mu(E = 0) = \mu_0 \exp\,[-(2\sigma/3kT)^2], \tag{5}$$

where μ_0 is a prefactor mobility and σ is the energy width of the distribution of states. Borsenberger and co-workers analyzed the effect of dipole moments by splitting σ into a dipolar component σ_d and a van der Waals component σ_{vdW}.[15,21–23] Assuming the two factors are independent

$$\sigma^2 = \sigma^2_d + \sigma^2_{\mathrm{vdW}}\,. \tag{6}$$

An expression for the dipolar component has been given by Young[24] as

$$\sigma_d = 7.07c^{1/2}\,p/\rho^2\varepsilon, \tag{7}$$

where c is the fraction of lattice occupied by dipoles, p the dipole moment in Debye, ρ the intersite distance in Å, ε the dielectric constant, and σ_d is in eV. Equation 7 applies only to the contribution of the dopant molecule. The polymer is polar and should also contribute, but perhaps by a constant amount,[32] thus we excluded the effect. Assuming the van der Waals component is constant, Eqs. 6 and 7 provides a means for analyzing the effect of dipole moment on σ by plotting σ^2 versus c. The reported data[22] clearly show the linear relationship between σ^2 versus c, indicating the justification for the analysis. Eqs. 5 through 7 also predict that the relationship between ln $\mu(E = 0)$ and p^2 is linear, although supporting data has not been shown in published papers. Then we speculate that the relationship between δE_p and p is analytically similar to Eq. 7. Together with the speculations described above, namely that J can be estimated by α, our view is consistent with the result given in Eq. 1 which indicates that the logarithm of mobility is described as a linear combination of the polarizability and the dipole moment.

In terms of a molecular design, polarizabilities and dipole moments are good indices to predict high mobility in CTL. For example, hydrazone molecules generally have higher dipole moments and larger relaxation energy when they change from neutral to cationic forms than do triarylamine derivatives,[31] which can be disadvantageous to attaining high mobility. This is largely true in Table I, showing that most triarylamine derivatives **11**–**15** have higher mobility than hydrazone derivatives **1**–**10**. However, even hydrazone derivatives have the potential for enhanced mobilities by increasing polarizabilities. For example, the high mobility of compound **9**, which is comparable to that of triarylamine derivatives **11**–**15**, especially at high electric field, is most likely due to its high polarizability. In view of the π-electron distribution, a large extension of π-electron and its homogeneous delocalization on a molecule have been reported to be essential to obtaining high mobility.[33] The molecules based on this guiding principle will be reported in future publications.

It should also be noted that there is a limitation in the method of analysis used in this study, especially in a case of using a large variety of molecular structures. For example, the outlier point in Fig. 2a is compound **14**. In this study we used an equal weight concentration in all doped samples, whereby **14** has approximately twice the number of molecules compared with **13**, while the polarizability of **14** is about half as large as that of **13**. Thus it might be more appropriate to use an equal molar concentration. However, we used the equal weight concentration because **13** contains two active units in one molecule. Of course, in such a case, it is necessary to consider that intramolecular charge transfer is more likely to occur than the intermolecular charge transfer. By the use of **13** and **14** in this study it appears that the molecule **13** with high polarizability has approximately the same mobility as **14** with half the polarizability even though the molar concentration is about half and the mean center-to-center distance of **13** is about twice compared with those of **14**. Additionally, in such a high concentration (50 wt%) of the dopants, the degree of molecular packing and orientation should also be taken into consideration. The extraordinary small mobility of **2** compared with the calculated value described in Table I can be ascribed to its low molecular overlap even at high concentration.[29]

Summary and Conclusions

Polarizabilities and dipole moments of hole transport molecules with different mobilities were measured. The mobilities increase with increasing polarizabilities and decrease with increasing dipole moments. The 3-D plot of the mobility, as a function of the polarizability and dipole moment is nearly planar, indicating that the logarithm of mobility is described as a linear combination of the polarizability and the dipole moment. The multiple correlation coefficient was estimated to be 0.841. These data were analyzed first within a framework of the small polaron formalism. With regard to transfer integral $J(\rho)$, a polarizability can be a good index to evaluate the degree of electron cloud extension that affects $J(\rho)$. On the other hand, the magnitude of the polaron binding energy E_p is macroscopically influenced mostly by the intramolecular relaxation energy. However, we propose the distribution of E_p, which is not considered in the small polaron formalism, caused by charge-dipole interactions as a perturbation. Such distributions, which have recently been discussed within a framework of the disorder formalism, are due to the random internal field caused by the local charge-dipole interaction. Thus we consider the linear relationship between the logarithm of mobility and polarizabilities as well as dipole moments is reasonable. This is consistent with the experimental data that the logarithm of mobility is described as a linear combination of the polarizability and the dipole moment. This relationship is applicable to the molecular design to attain high mobility. The guiding principle is to design a molecular structure with large extension of the π electron together with low dipole moment. Even hydrazone molecules having large dipole moments, the high mobility can be attained by designing the molecule such as compound **9** which has a large polarizability.

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