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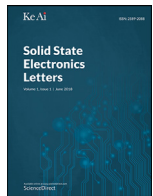
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Modeling the negative capacitance effect in dispersive organic materials using modified Drude theory

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ABSTRACT

Frequency- and mobility-dependent admittance have been observed in organic polymer light-emitting diodes. In this paper, we developed a model to describe this dispersive behavior using a modified Drude theory. In this model, a phase angle difference between the applied electric field and the average displacement of the charge carriers is introduced rather than using a complex mobility. This newly proposed model successfully describes the dispersive nature, as well as the negative capacitance effect, at low frequencies in organic polymers. The simulation results of this model also fit the negative capacitance data reported in the literature, provided that a suitable phase angle difference is given.

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1. Introduction

It has been reported that the admittance of organic polymer light-emitting diodes is frequency- and carrier mobility-dependent, and becomes negative under certain biased conditions [1,2]. This dispersive property has been attributed to carrier trapping and hopping in the polymer layer [1,3–5]. Based on Drude theory [6,7], one of our authors, H.L. Kwok, proposed a model to describe the negative capacitance effect found in dispersive organic polymers [8,9]. In his model, complex carrier mobility is assumed to account for the dispersive behavior of the medium. The model has successfully described the frequency-dependent capacitance and negative capacitance effect of organic polymers. In addition, the simulation results of Kwok's model are in good agreement with the experimental data reported in the extant literature [1]. However, the physical concept of complex carrier mobility is not simple to understand. In this work, we propose an alternative model, also based on Drude theory but with real carrier mobility. Instead of using complex carrier mobility, a phase angle difference between the applied electric field and the average displacement of charge carriers is introduced to account for the dispersive nature of the medium in our newly proposed model.

2. The model

Here, we take the organic layer as a plasma medium, in which a collection of positive and negative charges exists. These charges are assumed to oscillate with a Hook's law force constant K_0 , and an ac electric field with an angular frequency ω , $E = E_0 e^{j\omega t}$, is applied. From the Drude theory, we can obtain the following equation (by considering positive charge only for convenience) [8,9]:

$$m \frac{d^2x}{dt^2} + \frac{q}{\mu} \frac{dx}{dt} + K_0 x = qE \quad (1)$$

where m is the mass of the positive charges; x is the average position of the charges; q is the electronic charge; and μ is the carrier mobility. Note that, in Kwok's model, a complex parameter γ is used to represent the frequency of occurrence of inelastic collisions per

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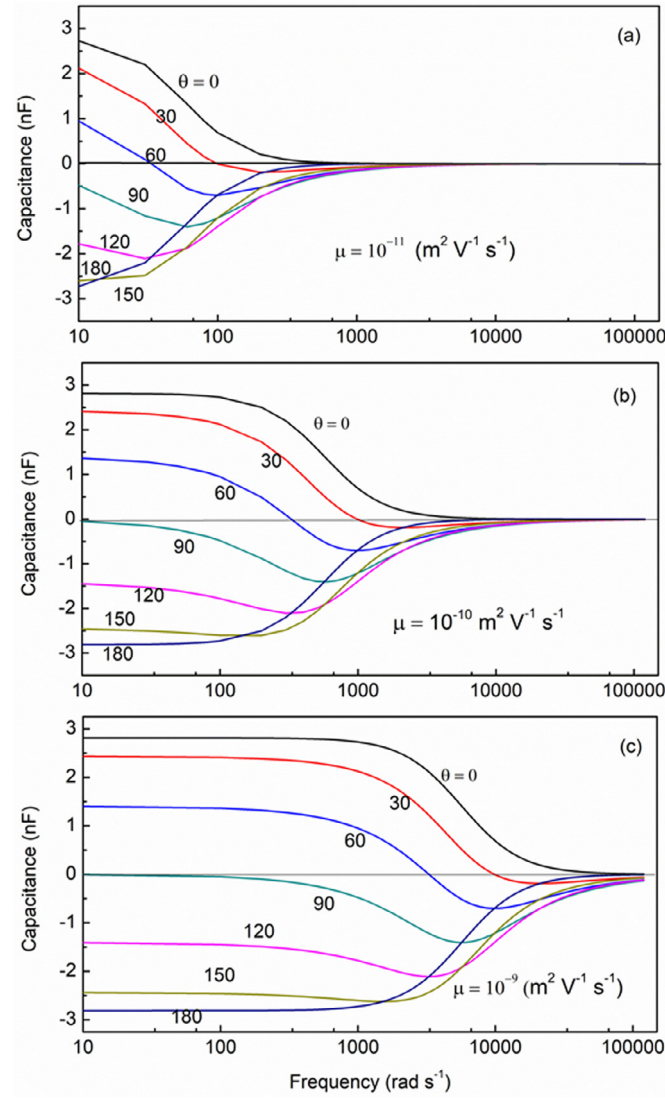


Fig. 1. Simulated capacitance versus frequency curves under various phase angle difference ($\theta = 0\text{--}180^\circ$) for mobility μ equals to (a) 10^{-11} , (b) 10^{-10} , and (c) $10^{-9} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}$.

unit charge for dispersive medium, and equals to the reciprocal of the carrier mobility μ , i.e., $\gamma = 1/\mu = \gamma_1 + j\gamma_2$ [8,9]. However, the physical concept of complex mobility is not simple to comprehend. In our model, we introduced a phase difference θ between the applied electric field and the average position of the charges x , rather than utilizing complex mobility to account for the dispersive nature of the medium. Therefore, the solution to Eq. (1) can be expressed as $x = x_0 e^{j(\omega t + \theta)}$. The phase angle difference indicates how fast the charge carriers can follow the variation of the applied field. By substituting $E = E_0 e^{j\omega t}$ and $x = x_0 e^{j(\omega t + \theta)}$ into Eq. (1), one can easily find that:

$$x_0 = \frac{(qE_0/m)e^{-j\theta}}{[(\omega_0^2 - \omega^2) + j\omega q/\mu m]} \quad (2)$$

where $\omega_0 = K_0/m$ is the characteristic frequency of the plasma. The polarization vector P can then be expressed as follows:

$$P = pqx_0 = \frac{(pq^2E_0/m)e^{-j\theta}}{[(\omega_0^2 - \omega^2) + j\omega q/\mu m]} \quad (3)$$

Assume a parallel plate capacitor of area A and plate separation L having the organic dispersive medium as the dielectric. Capacitance is then given by:

$$\begin{aligned} C &= \text{Re} \left[\left(\frac{P}{E_0} \right) \left(\frac{A}{L} \right) \right] = \text{Re} \left\{ \frac{(pq^2A/Lm)e^{-j\theta}}{[(\omega_0^2 - \omega^2) + j\omega q/\mu m]} \right\} = \text{Re} \left\{ \frac{C_0 e^{-j\theta}}{[(1 - \xi^2) + j\xi q/\mu \omega_0 m]} \right\} \\ &= \text{Re} \left\{ \frac{C_0 (\cos \theta - j \sin \theta)}{[(1 - \xi^2) + j\xi q/\mu \omega_0 m]} \right\} \end{aligned} \quad (4)$$

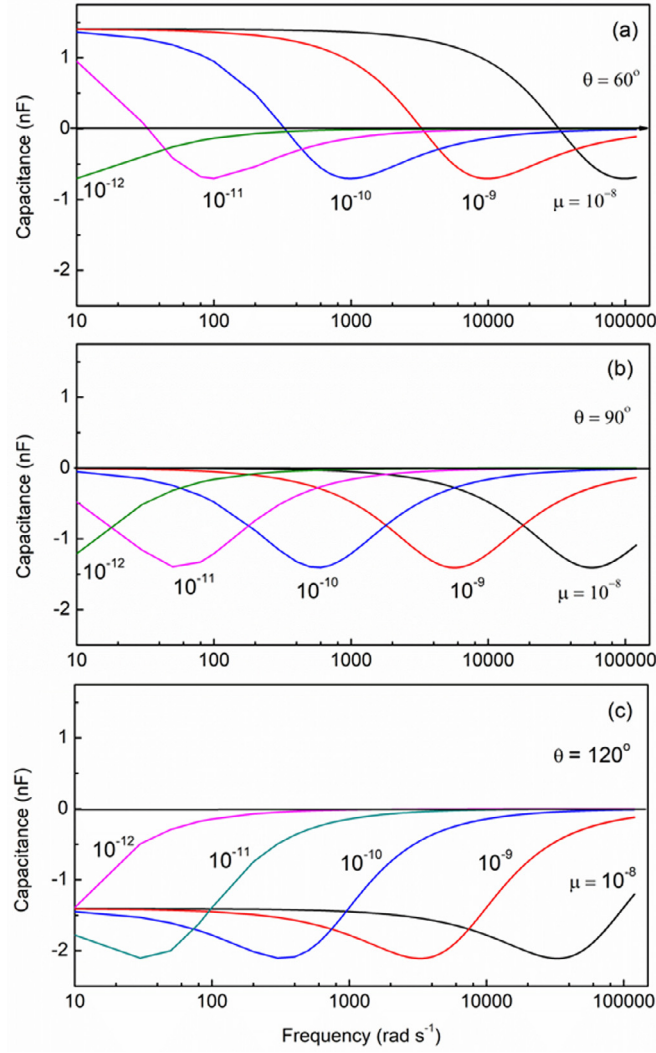


Fig. 2. Calculated capacitance versus frequency curves with carrier mobility varying from 10^{-8} to 10^{-12} $\text{m}^2 \text{V}^{-1} \text{s}^{-1}$ for phase angle difference (a) $\theta = 60^\circ$, (b) $\theta = 90^\circ$, and (c) $\theta = 120^\circ$.

where $C_0 = pq^2A/Lm$ and $\xi = \omega/\omega_0$. Note that the mobility here is real. From Eq. (4), we can easily find that:

$$C = C_0 \left\{ \frac{(1 - \xi^2) \cos \theta - (\xi q / \mu m \omega_0) \sin \theta}{(1 - \xi^2)^2 + (\xi q / \mu m \omega_0)^2} \right\} \quad (5)$$

It is obvious that C becomes negative when:

$$(1 - \xi^2) \cos \theta < (\xi q / \mu m \omega_0) \sin \theta \quad (6)$$

or

$$\theta > \tan^{-1} \frac{(1 - \xi^2)}{\xi q / \mu m \omega_0} \quad (7)$$

For the case of non-dispersive medium, $\theta = 0$, and the capacitance given in Eq. (5) reduces to:

$$C = C_0 \left\{ \frac{(1 - \xi^2)}{(1 - \xi^2)^2 + (\xi q / \mu m \omega_0)^2} \right\} \quad (8)$$

which is exactly the same as the one given in Refs. [8] and [9].

3. Results and discussion

By using Eq. (5) and the same parameter values given in references [8] and [9], i.e., $A/L = 100$, $p = 1 \times 10^{21}$ (m^{-3}), $\omega_0 = 1 \times 10^{12}$ (rad s^{-1}), $m = 0.91 \times 10^{-31}$ (kg), and $q = 1.6 \times 10^{-19}$ (C), we calculated the capacitance of the organic medium with mobility $\mu = 10^{-11}$, 10^{-10} , and 10^{-9} ($\text{m}^2 \text{V}^{-1} \text{s}^{-1}$) for various phase angle differences and for the frequency ranging from 10 to 120,000 (rad/s). In general, the carrier mobility for organic polymers is in the range of 10^{-8} – 10^{-12} $\text{m}^2 \text{V}^{-1} \text{s}^{-1}$ [10,11]. The calculated capacitance versus frequency curves are shown in Fig. 1(a), (b) and (c). As observed, the capacitance is a function of carrier mobility, frequency of the applied field,

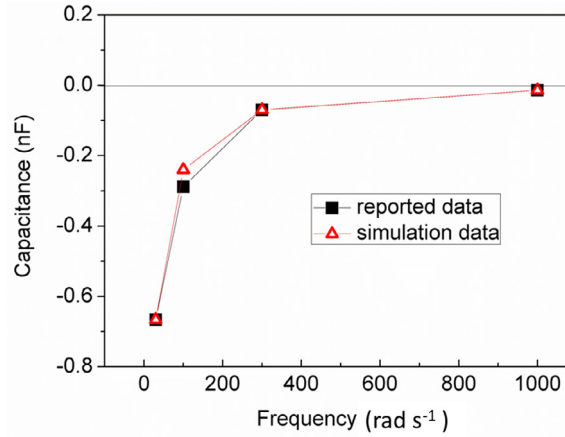


Fig. 3. Comparison of the simulated capacitance data at low frequencies using the proposed model and the reported experimental data given in Ref. [1]. The parameter values for the simulation are presented in Table 1.

Table 1

Parameter values used for the calculation of Fig. 1.

ω (rad s ⁻¹)	P (m ⁻³)	ω_0 (rad s ⁻¹)	θ (°)	μ (m ² V ⁻¹ s ⁻¹)
30	4.50×10^{21}	2.30×10^{21}	91.67	3.00×10^{-13}
100	4.50×10^{21}	2.30×10^{21}	95.68	3.35×10^{-13}
300	4.50×10^{21}	2.30×10^{21}	106.00	3.00×10^{-13}
1000	4.00×10^{21}	2.30×10^{21}	111.00	2.30×10^{-13}

Table 2

Comparison of the negative capacitance of the reported experimental data shown in Ref. [1], the previous data from Kwok's model given in Ref. [9], and the calculation results using the model proposed in this work. The numbers shown in the parentheses are the percentage error compared with the reported data.

ω (rad s ⁻¹)	C (nF)		
	Reported [1]	Kwok's Model [9]	This work
0	-66.6	-66.9 (0.45%)	-66.6 (0%)
100	-28.8	-23.0 (20.13%)	-24.0 (16.67%)
300	-69.8	-65.0 (6.88%)	-69.7 (0.14%)
1000	-14.8	-14.0 (5.41%)	-14.1 (4.73%)

and phase angle difference. For the case of non-dispersive medium, i.e., $\theta = 0$, we see that the capacitance saturates at low frequencies, and the saturation values become higher as the carrier mobility increases. It is readily understood that higher carrier mobility causes less carrier collision or trapping, and thus the capacitance is higher. It is also noticed that, except for $\theta = 0$, the capacitance becomes negative after a critical frequency is reached. This critical frequency decreases with increasing phase angle difference when the mobility is fixed, while it increases with increasing carrier mobility when the phase angle difference is fixed. For a dispersive medium with fixed mobility, a higher θ value means higher polarization charge, and a lower frequency is needed to yield the same capacitance. When the θ value is fixed, in a medium with higher carrier mobility, the polarization charge more easily follows the changes of the applied field, and hence negative capacitance occurs at higher frequency. More importantly, it is found that the capacitance becomes all negative when $\theta > 90^\circ$. These behaviors are basically in agreement with the nature of the dispersive medium. All of the arguments discussed above can be more clearly seen in Fig. 2, which shows the capacitance versus frequency curves under various carrier mobilities for $\theta = 60^\circ$, 90° , and 120° . We compare the calculation results of our model and the experimental data of the negative capacitance reported in reference [1] to verify the proposed model, as shown in Fig. 3. The values of the parameters used for the calculation are given in Table 1. It is obvious that the calculation results agree well with the experimental data. As observed in Table 1, the θ values increase with increasing frequency. This is because higher polarization charges are needed when the frequency is higher. In addition, the values of the parameters used for the calculation are all reasonable. Table 2 compares the negative capacitance values obtained from the reported experimental data given in Ref. [1], the calculated data from Kwok's previous model [9], and the simulation results of the present work. We also specified in parentheses the percentage error of Kwok's model and our model compared with the reported experimental data. It can be seen that our model gives smaller deviation error than does the previous Kwok's model. For conjugate polymers, it has been shown that carrier mobility varies in a field-dependent manner and can be written as $\mu = \mu_0(-\beta E^{1/2})$, where μ_0 is the zero field mobility, β is the field activation factor, and E is the applied field [2,12]. We utilize this correlation between the applied field (voltage) and the carrier mobility in Eq. (5) to calculate the capacitance of the dispersive medium. Fig. 4 compares the calculation results with the experimental negative capacitance data reported in Ref. [2] at low frequencies. In the calculation, the following parameters are used: $\mu_0 = 10^{-10}$ m² V⁻¹ s⁻¹; $A/L = 8$; and $p = 2 \times 10^{20}$ cm⁻³. From Fig. 4, it can be seen that our calculation results agree well with the reported data, except for the one with applied voltage of 3 V at $\omega = 16$ rad s⁻¹. This discrepancy is believed to be due to the fluctuation in the experimental data at extremely low frequencies [2].

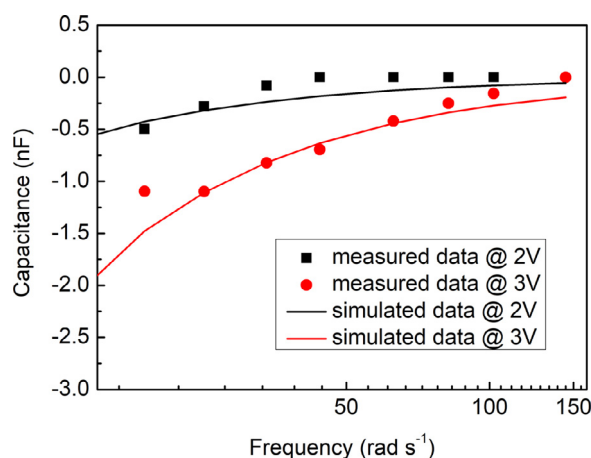


Fig. 4. Comparison of the simulated negative capacitance under different applied voltages (2 V and 3 V) using the proposed model and the reported experimental data given in Ref. [2].

4. Conclusion

In this work, we have developed a model based on Drude theory to describe negative capacitance in dispersive organic polymers. In the model, we used real carrier mobility, and introduced a phase angle difference between the average position of the charges and applied an electric field, instead of utilizing complex mobility. When a suitable phase angle difference is given, our model can accurately describe the dispersive nature, as well as the negative capacitance effect, of organic polymers with a good match in key parameters, such as carrier mobility, carrier concentration, etc. More importantly, the concept of phase angle difference used in this model provides a more direct physical insight into negative capacitance than the one using complex mobility.

Declaration of Competing Interest

None.

CRediT authorship contribution statement

You-Lin Wu: Conceptualization, Methodology, Funding acquisition, Writing - original draft. **Jing-Jenn Lin:** Data curation, Visualization, Formal analysis. **H.L. Kwok:** Supervision, Investigation, Resources.

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