Instantaneous determination of the electronic transport properties of polymer light emitting diodes from their complex impedance spectra using a neural network

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ABSTRACT

Electronic transport properties of polymer light-emitting diodes (PLEDs) were determined using a machine learning model developed with complex impedance spectra generated by device simulation. The mobilities and bimolecular recombination coefficients were instantaneously determined by the machine learning model using experimentally-obtained complex impedance spectra of PLEDs as inputs.

1 Introduction

Electronic transport properties (drift mobilities, bimolecular recombination coefficients and localized tail state distributions) in polymer light-emitting diodes (PLEDs) have been determined from their complex impedance spectra measured using a conventional frequency response analyzer [1-2]. The information is fundamentally important for the synthesis of highly efficient light-emitting polymers and further optimization of the device structure of PLEDs, and short data acquisition and analysis time is essential to the electronic characterization of a number of PLEDs.

We have developed a measurement setup based on time-stretched pulses and fast Fourier transform [3], which enables us to acquire complex impedance spectra in a few seconds, much faster than a conventional frequency response analyzer.

Subsequent analysis of complex impedance spectra for obtaining the electronic transport properties needs considerable experience. The development of automatic analysis of complex impedance spectra was therefore rather difficult.

In this presentation, we show that the electronic transport properties of PLEDs can be instantaneously determined from complex impedance spectra using neural networks. A machine-learning approach is the best way to develop full automatic analysis of complex impedance spectra.

2 Device simulation

A number of complex impedance spectra were generated by device simulator (SILVACO: ATLAS) to build a machine learning model. The reason for this is that sufficient number of data were easily obtained; it is rather difficult to experimentally collect a large number of data with wide range of the electric transport properties for machine learning. In the machine learning, capacitance and complex impedance spectra were used as explanatory variables and input physical quantities (mobilities and bimolecular recombination coefficients) in device simulations were used as objective variables.

The physical quantities and their ranges used in the device simulations are shown in Table 1. Bimolecular recombination coefficients were given by the product of Langevin recombination prefactors and Langevin recombination coefficients.

Table 1 Physical quantities used in device simulation

physical quantities	range
hole mobility (cm^2/Vs)	10^{-9}-10^{-5}
hole injection barrier (eV)	0.0 – 0.1
electron mobility (cm^2/Vs)	10^{-9}-10^{-5}
electron injection barrier (eV)	0.0 – 0.1
Langevin recombination prefactor	10^{-3}-10^{-1}
thickness (nm)	50-200

We generated 10 000 complex impedance spectra with input values randomly selected in the range shown in Table 1. 80% of the spectra were used for training and validation data to build a machine learning model, and the remaining 20% of the spectra were used for test data.

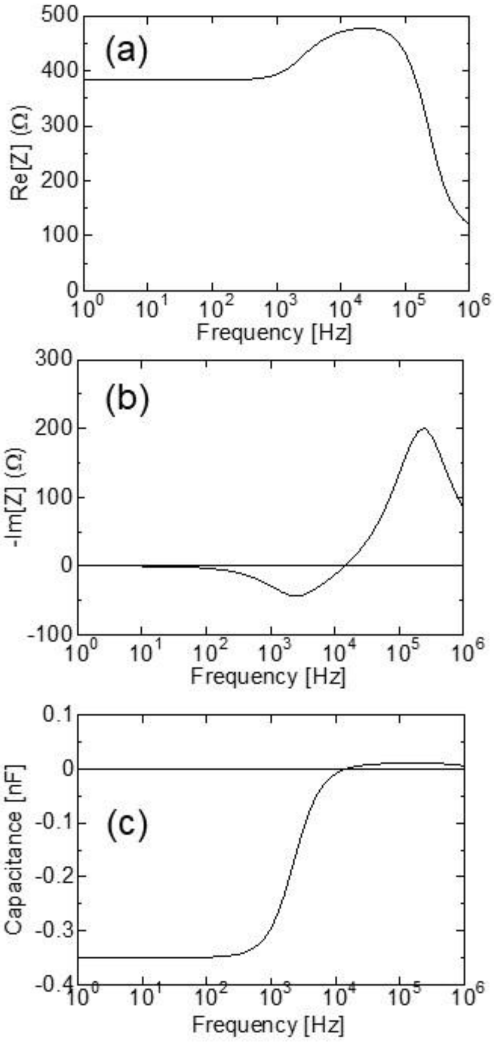

Fig. 1 Complex impedance spectra of a PLED obtained by device simulation. (a) Re[Z], (b) -Im[Z], and (c) capacitance spectra.

3 Prediction

The complex impedance and capacitance spectra, as shown in Fig. 1, were used as input to a fully connected neural network, whose outputs were electronic transport properties. The machine learning model developed in this way cannot distinguish electron and hole mobilities, and hence we use the notation of higher and lower mobilities instead of electron and hole mobilities in Fig. 2.

Scatter plots of the mobilities and bimolecular recombination coefficients input to the device simulation versus mobilities and bimolecular recombination coefficients output from the machine learning model is shown in Fig.2.

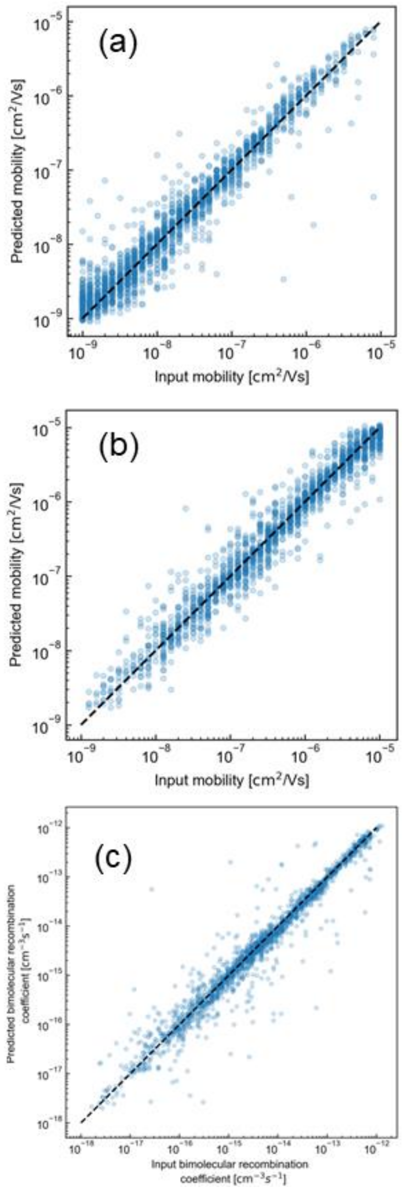

Fig. 2 Scatter plots of (a) predicted higher mobility vs input higher mobility, (b) predicted lower mobility vs input lower mobility, and (c) predicted bimolecular recombination coefficient vs bimolecular recombination coefficient

The machine learning model predicted the coefficients of the determination of 0.945 for the bimolecular recombination coefficients, 0.959 for higher mobilities, and 0.939 for lower mobilities. The coefficient of the determination was used to evaluate the accuracy of the machine learning model. The coefficient of the determination is a measure of how well a machine learning model explains the objective variable. The values of the coefficients of the determination are reasonably good for the characterization of the electronic transport properties of PLEDs.

4 Experiment

A device to be measured is an inverted PLED with Super Yellow (SY) as the emissive layer. The PLED structure was AZO/PEI/SY (Sigma Aldrich)/MoO_3/Al (the active area of the device was 4 mm^2); AZO is Al-doped ZnO, PEI is poly(ethyleneimine), and Super Yellow is a commercially available fluorescent polymer. A patterned AZO glass used for cathode was cleaned using acetone, 2-propanol, and an ultraviolet (UV)-ozone treatment. Then a thin PEI layer, working as an electron injection layer, was spun onto the AZO glass surface from an ethanol solution (30 s, 2000 rpm). The substrate was then annealed in the ambient atmosphere. A 100-nm-thick SY layer was spun onto the PEI layer from a chlorobenzene solution (60 s, 800 rpm). After spin-coating of the SY layer, the substrate was dried at 80 °C for 15 min. 10-nm-thick MoO_3 and 50-nm-thick Al layers were then thermally evaporated successively onto the SY layer in a vacuum chamber. Finally, SY PLED was encapsulated using a seal material.

5 Experiment results

Figure 3 shows experimental results for complex impedance spectra of inverted SY PLED.

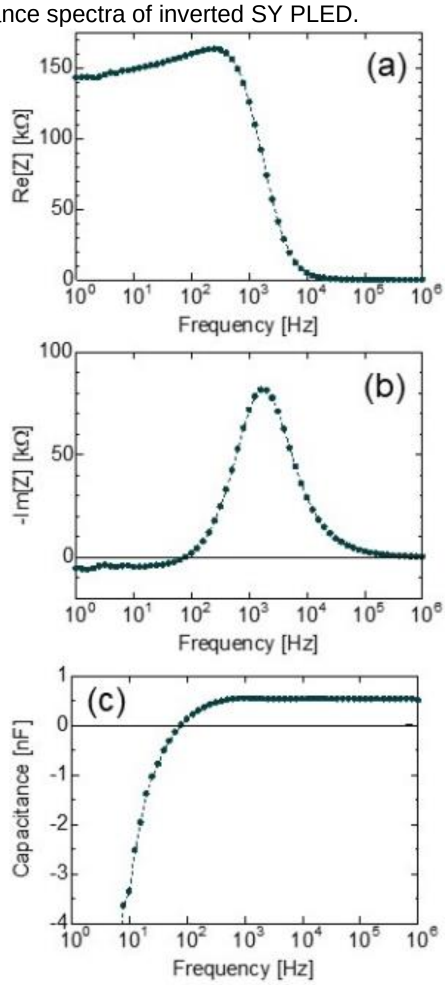

Fig. 3 Experimentally obtained complex impedance spectra of SY PLED. (a) Re[Z], (b) -Im[Z], and (c) capacitance spectra

To determine the electronic transport properties, experimentally obtained complex impedance spectra of the SY PLED were input into the machine learning model. The results are shown in Table 2.

Table 2 Electric transport properties obtained by the machine learning model and by manual analysis

Electric properties	Machine Learning	Manual analysis
higher (hole) mobility (cm^2/Vs)	4.4×10^{-7}	8.3×10^{-7}
lower (electron) mobility (cm^2/Vs)	3.5×10^{-9}	6.6×10^{-9}
bimolecular recombination coefficients ($cm^{-3}s^{-1}$)	2.6×10^{-15}	5.9×10^{-15}

There were no significant differences between the values determined by the machine learning model and the manual analysis. It takes about 600 s for analyzing manually the complex impedance spectra, while the present machine learning model gives the values of the mobilities and the bimolecular recombination coefficient in 10 ms (Core i5, 2.90 GHz).

6. Conclusions

We have developed a machine learning model based on a neural network using complex impedance spectra generated by device simulation for the determination of electronic transport properties (bimolecular recombination coefficients and drift mobilities) of PLEDs. We have demonstrated that the electron and hole mobilities and the bimolecular recombination coefficients were instantaneously determined by the machine learning model from experimentally-obtained complex impedance spectra of SY PLEDs. The time for the determination is 10 ms (Core i5, 2.90 GHz), much faster than conventional manual analysis (about 600 s).

Acknowledgements

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Reference

[1] M. Takada, T. Nagase, T. Kobayashi, and H. Naito, “Full characterization of electronic transport properties in working polymer light-emitting diodes via impedance spectroscopy,” *J. Appl. Phys.*, vol. 125, no. 11, 115501, Mar. 2019.

[2] M. Takada, T. Mayumi, T. Nagase, T. Kobayashi, and H. Naito, “Determination of bimolecular recombination constants in organic double-injection devices using impedance spectroscopy,” *Appl. Phys. Lett.*, vol. 114, no. 12, 123301, Mar. 2019.

[3] A. Okada, T. Nagase, T. Kobayashi, and H. Naito, “Fast data acquisition of complex impedance spectra using time-stretched pulses: Measurement of drift mobility of polymer light emitting diode,” The 82nd JSAP Autumn Meeting 2021, 11-075.