Cooperative Research and Development Agreement (CRADA) Final Report

Report Date:
October 6, 2020

In accordance with Requirements set forth in the terms of the CRADA, this document is the CRADA Final Report, including a list of Subject Inventions. It is to be forwarded to the DOE Office of Scientific and Technical Information upon completion or termination of the CRADA, as part of the commitment to the public to demonstrate results of federally funded research.

Parties to the Agreement: DOW Chemical Company

CRADA number: FP00003009

CRADA Title: Modeling the Effect of Film Morphology on the Performance of an OLED Device

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Sponsoring DOE Program Office(s):
DOE Basic Energy Sciences (BES)

LBNL Report Number:
LBNL-2001362

OSTI Number:

Joint Work Statement Funding Table showing DOE funding commitment:

DOE Funding to LBNL	$37,000
Participant Funding to LBNL	$0
Participant In-Kind Contribution Value	$99,322
Total of all Contributions	$136,322

Provide a list of publications, conference papers, or other public releases of results, developed under this CRADA:
(Publications must include journal name, volume, issue, Digital Object Identifier)

None

Provide a detailed list of all subject inventions, to include patent applications, copyrights, and trademarks:

(Patents and patent applications are to include the title and inventor(s) names. When copyright is asserted, the Government license should be included on the cover page of the Final Report)

None

Executive Summary of CRADA Work:

Organic Light Emitting Diode (OLED) technology is replacing the liquid crystal displays (LCD) in cell phones and is also expected to impact television displays in the future. Dow has an active OLED research program. To complement and ultimately drive this effort, it is necessary to develop efficient computational screening tools for selection and optimization of target molecules. To this end we sought to identify and expand on existing models used by Dow that can better predict the mobility of electrons and holes in organic materials. We focused on N,N′-bis(1-naphthyl)-N,N′-diphenyl-1,1′-biphenyl-4,4′-diamine (NPD), frequently used in academic studies of organic light-emitting diodes; we also studied 4,4′-Bis(N-carbazolyl)-1,1′-biphenyl (CBP), tris(4-carbazoyl-9-ylphenyl)amine (TCTA), and bathophenanthroline (BPhen). We developed a workflow, using a combination of molecular dynamics and quantum chemistry calculations, to predict trends in the electronic structure that correlate with measured electron and hole mobilities of small molecule materials such as NPD for OLEDs, comparing with measurements in the literature and at Dow. This work comprised a first step toward predictive charge carrier mobilities in small-molecule electronic materials with complex morphologies.

Summary of Research Results:

We developed a workflow, using a combination of molecular dynamics and quantum chemistry calculations, to predict the electron and hole mobilities of NPD and other small molecule-based films of relevance to OLEDs, comparing with measurements in the literature and at Dow. Charge transport properties are known to be sensitive to NPD film morphology.

In this work, we initially examined the role of morphology on the electronic properties of the film via the following two-step approach. First, we performed two different classical MD simulations of condensed-phase NPD with periodic boundary conditions with 250 molecules/unit cell. We first generated a morphology using a constant-temperature approach with the system held at 300 K for 3 ns. Then, we generated a different morphology using a simulated annealing approach: here, we heated the same simulation box from 300 K to 1000 K over 1 ns, and then held it at high temperature for 4 ns. Subsequently, the system was cooled to 300 K for 4 ns and finally equilibrated for 1 ns at constant temperature.

Second, having generated two distinct morphologies, we computed the gas phase ionization potentials using density functional theory (DFT) and the Gaussian quantum chemistry package for each of the 250 individual constituent molecules for both morphologies. Our calculations showed that the distributions of the IPs were the same for both morphologies, indicating that both simulations lead to morphologies with similar local environments for the NPD molecules.

Having established this workflow for NPD, we repeated these calculations for three other molecular thin film systems: electronic properties of 4,4′-Bis(N-carbazolyl)-1,1′-biphenyl (CBP), tris(4-carbazoyl-9-ylphenyl)amine (TCTA), and bathophenanthroline (BPhen). These sytems are literature-based systems of relevance to OLED devices. After generating morphologies for each molecular solid, we computed IPs and nearest-neighbor coupling energies using DFT with Gaussian. The coupling energies were computed using the frontier molecular orbitals of the individual molecules as basis set in the calculation of the dimers. For these four systems, we looked at the distributions of the reorganization energies of the constituent molecules, their frontier orbital energies, and their coupling strengths. We found that the average coupling strength appears to be an indicator of measured charge carrier mobility; however, our sample size is small and that this quantity neglects the other electronic properties. To predict mobilities, we applied a commonly-used, semi-empirical phenomenological transport model based on nearest-neighbor hopping of electrons and holes, the Generalized Effective Medium Model (GEMM), finding that the trends in computed mobilities are comparable with experiments except for TCTA. GEMM neglects spatial and energetic correlations, and on-site dipole moments, and therefore more explicit models are required for more conclusive results. A next step would be to approximately include this missing physics with a kinetic approach that combines MD and DFT calculations, similar to the ones we performed, with Kinetic Monte Carlo. Our calculations with Dow provided a baseline for such future calculations.