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Exciton-mediated electrochemical degradation of blue-phosphorescent organic light-emitting devices

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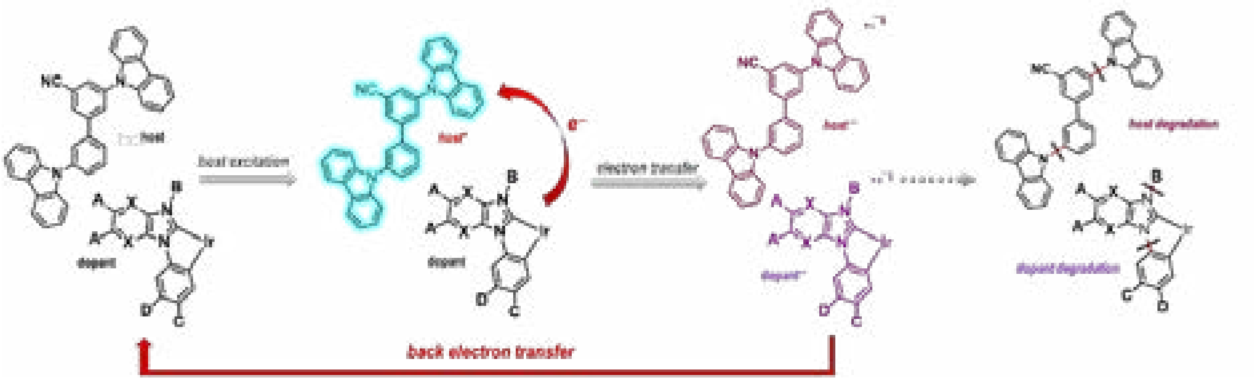
Implementation of blue electro-phosphorescence devices into displays has been retarded by short operation times. The poor device longevity originates from inherent degradation of materials in emitting layers. Despite significant research efforts, molecular mechanisms underlying the degradation have yet to be fully understood. We propose an exciton-mediated degradation mechanism which involves generation of a radical ion pair of an Ir(III) complex dopant and an organic host molecule. To examine our hypothesis, we investigated electron-transfer properties of a new series of triscyclometalated Ir(III) complexes having N-heterocyclic carbene (NHC) ligands based on benzimidazole and imidazopyrazine scaffolds. We determined the redox potentials of the complexes by cyclic and differential pulse voltammetry. Comparison of the values with the excited-state electrochemical potentials of a 3,3'-biscabazolyl-5-cyanobiphenyl revealed formation of a radical ion pair, through reductive quenching of host exciton by dopants. We directly monitored the radical ion pairs, employing laser flash photolysis. The spectral assigned were supported by spectro-electrochemical measurements and quantum chemical calculations based on the time-dependent density functional theory. We also determined the rate constants for charge recombination within the radical ion pair, and analyzed the values with driving forces based on the classical Marcus theory of adiabatic electron transfer. Finally, blue-electrophosphorescent devices incorporating the Ir(III) complexes and the host were fabricated. Evaluation of device performances revealed that the devices lifetimes (T_{95}) was linearly proportional to the rate constants for charge recombination of the radical ion pair. These findings suggested the importance of control over electron transfer within emitting layers for long device lifetimes.



Biography

Sinheui Kim is currently a graduate student at Ewha Womans University, Korea. She has experienced with a range of chemical techniques, including synthetic chemistry, electrochemistry, and photo-physics of photo-functional molecules. Her projects include low-energy photo-redox catalysis for tri-fluoro-methylation of un-activated arenes and alkenes, mechanistic studies for degradation of blue-electrophosphorescence devices, and the development of novel fluorescent materials exhibiting through-space π interactions.

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