Ab-initio molecular dynamics simulation of polaron- and exciton-OLED degradation

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Organic light-emitting diodes (OLEDs) offer the potential of using both the singlet and triplet excitons in realizing 100% internal quantum efficiencies of electro-luminescence. However, the injected charge carriers (electrons and holes) may become trapped at morphological and chemical defects, and recombine non-radiatively. These phenomena, not only limit the quantum efficiency of the device, but are also responsible for the degradation of the device (leading to aging and failure), through the formation of highly reactive radical species. A complete understanding of the degradation pathways, beyond phenomenological models, based on atomistic/microscopic modeling, is still missing to date.

In this work, we performed Car-Parrinello molecular dynamics with constrained orbital occupations to simulate the fate of an electron, a hole and an electron-hole pair, electrically injected into three host OLED materials (CBP, mCP and α-NPB). We found that polaron- and exciton-trapping is associated with large conformational changes and weakening of some ancillary C-C and C-N bonds. By computing the conformational and bond-breaking barriers we estimate the polaron/exciton residence time and tendency towards chemical degradation.