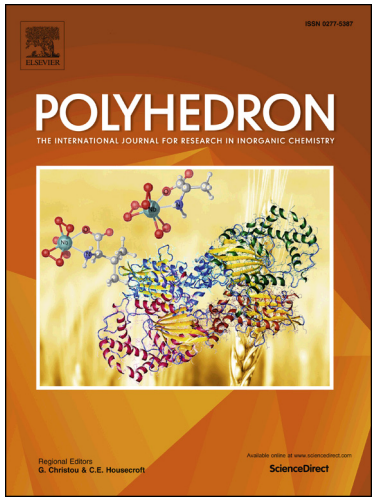

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Contextualizing yellow light-emitting electrochemical cells based on a blue-emitting imidazo-pyridine emitter

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ABSTRACT

This work provides the synthesis, structural characterization, electrochemical and photophysical features, as well as the application in light-emitting electrochemical cells (LECs) of a novel small molecule belonging to the pyridilimidazo[1,5-a]pyridine family, namely 3-(2-methoxyphenyl)-5-methyl-1-(6-methylpyridin-2-yl)H-imidazo[1,5-a]pyridine (Me-impy). This compound shows a low-cost and facile synthesis, excellent redox properties, and a high photoluminescence quantum yield (Φ) of 0.4 associated to a blue emission (~436 nm) in solution. Despite these appealing features, the emission in solid state is red-shifted to the yellow region, owing to its prone aggregation character. Additionally, the electroluminescence response shows a broader and red-shifted emission even upon dilution of Me-impy thin films with polymethyl methacrylate (PMMA). Herein, we report an in-depth study on the aggregation features and its impact on the electroluminescence response of Me-impy, providing relevant information to design small-molecule based LECs.

1. Introduction

Light-emitting electrochemical cells (LECs) are regarded as one of the most promising thin-film lighting sources, as they combine high efficiency, a simple architecture with air stable electrodes, and a low-cost, solution based up-scalable fabrication process.[1–4] In short, LECs are a single-layer electroluminescent device consisting of a mixture of a luminescent material and an ionic electrolyte. As such, the main differences from the well-established organic light-emitting diode (OLED) technology is the control of the charge injection using mobile ions.[5] Upon biasing the device, the ion redistribution towards the electrode interface assists the electrochemical doping forming a p-i-n junction structure.[3,6–13] This particular operational mode leads to a high tolerance towards i) working function of the electrodes paving the way of using air-stable cathodes, such as Ag or Al, ii) different thicknesses and minor defects of the active layer, iii) using low-cost and up-scalable solution based techniques, and iv) applying a huge palette of emitters, such as polymers, ionic transition metal complexes, small molecules, quantum dots, and perovskites.[2,14–22]

The future advances in the field head into two directions, namely i) the development of new device architectures, such as flexible and/or 3D shaped substrates based on metallic and/or semiconducting materials and ii) new low-cost, easily synthetisable, and sustainable luminescent materials spanning the whole visible range.[1–3,23] As far as the first aspect is concerned, LECs fabricated with industry relevant based techniques, such as inkjet printing,[24] roll-to-roll-coating,[25] spray-coating,[26] and slot-die coating,[25] have been reported, as well as wearable fiber-shaped devices.[27,28] Regarding the new generation of emitters, small molecules have recently attracted much attention, due to i) their wide variety using easily modifiable scaffolds, ii) their emission covering the whole visible range with high Φ values that are not subjected to ambient quenching, iii) their stable electrochemical and thermal features, iv) their easy processability and high stability in solution, v) their good carrier mobilities, and vi) the presence of a thermally activated delayed fluorescence (TADF) behavior.[3,29–34]

Numerous works have been reported in the last few years regarding various families of small molecules for LECs (hereafter abbreviated as SM-LECs), such as ionic fluorine derivatives, phenanthroimidazoles, borazines, carbazoles, benzodithiazoles, pyrene derivatives, porphyrins, pentacenes, and cyanines;[16,29,35–47] covering all the visible spectrum including white emission.[14,15] Concerning blue SM-LECs, only three main families, namely ionic fluorine derivatives, imidazoles and borazines, have been tested.[29,31,35–37,39,40,48,49] Moderate

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