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Review of the doctoral dissertation of M. Sc. Paola Zimmermann Crocomo on "The intermolecular and intramolecular donor-acceptor charge transfer behaviour as a way to control thermally activated delayed fluorescence emission"

The reviewed dissertation was prepared by M. Sc. Paola Zimmermann Crocomo at the Faculty of Chemistry under the supervision of prof. dr. hab. inż. Przemysław Data, and submitted as a doctoral dissertation to the Silesian University of Technology in Gliwice.

The subject of research described in the dissertation concerns the study of physicochemical properties of three selected series of ompounds: dibenzophenazine based TADF emitters, D-A-D iminosttilbene and iiminodibenzyl derivatives and family of emitters basedon concaved with tunable TADF/RTP properies. The investigated emitters which were the object of research of Paola Zimmermann Crocomo, are currently the subject of intensive research due to their application in optoelectronics, especially in Organic Light Emitting Diodes (OLEDs).

The doctoral thesis of Paola Zimmermann Crocomo, M.Sc., submitted for my review has a classical layout. The dissertation consists, together with the list of cited

literature, of 137 pages; the text has been divided into 7 chapters (not including literature references).

The presented research focuses on the investigation of compounds showing thermally activated delayed fluorescence and room-temperature phosphorescence. The selected compounds were properly characterized by means of electrochemical and photophysical methods. Moreover the compounds were also applied in organic light emitting diodes, and the work parameters of the devices were clearly demonstrated. The research carried out allowed the doctoral student to determine which physico-chemical properties of molecules determine their emission properties, and thus to draw conclusions on how to improve their emission properties by chemical modification.

The first chapters of the PhD thesis provide a very good description of the phenomenon of thermally activated delayed fluorescence and of the research methods that make it possible to confirm and characterise this phenomenon. Both chapters together with the description of sample preparation constitute a compendium of knowledge on how to identify and optimise thermally activated delayed fluorescence emission.

In chapter four the Author has presented the results obtained for two compounds containing the same donor and acceptor groups, but differing in the presence of an additional C-C bond, which caused more strict configuration and an increase of the electron conjugation along with the molecular structure in one of these molecules. The study of these materials allowed the author to confirm the importance of angular position between donor and acceptor moieties to detect TADF emission.

The chapter five is dedicated to the investigation of the electrochemical and spectroelectrochemical properties of dibenzophenazine based TADF emitters. The author has thoroughly analysed the effects of the distorted D-A-D structure and the movement between donor and acceptor moieties on the efficiency of TADF emission and on their mechanochromic behaviour. On page 57 the Author has compiled the values of HOMO and LUMO energy levels and electrochemical bandgap values for investigated compounds. I want to ask what is the error in determining the values of HOMO and LUMO in cyclic voltammetry method.

In the chapter 6 the Author presents her contribution in in publication published in Chemistry-A European Journal, in 2021, where the doctoral student is the first author. The presented studies are nice example of full electrochemical

characterization of the compounds which are considered as the candidates for application in OLEDs.

The chapter 7 contains an exhaustive analysis leading to the better understanding the influence of the tuning between thermally activated delayed fluorescence, room-temperature phosphorescence, or dual TADF and RTP behaviour, depending on the substituent group on the acceptor's structure.

In this doctoral dissertation, in my opinion there is lack of the final summary of the results. Although each of the chapter in the experimental part was provided with correctly formulated conclusions and remarks, the doctoral student did not present conclusions for the entirety of the research. Such final conclusions / summary would provide valuable guidance for further work to improve material performance and stability for OLED applications.

Regarding the editorial side of this doctoral dissertation, I have only one point of criticism about the presentation of figures: some figures, e. g. Fig. 57. or 61. could be larger in order to improve their readability.

Summarizing, I conclude that the submitted dissertation meets the requirements set out in the Act on academic degrees and academic title and on degrees and title in the field of art of March 14, 2003 (Journal of Laws 2003 No. 65, item 595, as amended). Therefore, in my opinion, Mrs Paola Zimmermann Crocomo can be admitted to further procedures necessary to obtain the PhD degree. Moreover taking into account: the fact that Paola Zimmermann Crocomo is a co-author of 8 publications published in highly ranked, internationally recognised journals, the number of analysis included in this doctoral dissertation and a high importance of the provided data, which are very useful and important for the organic electronic society, I recommend this thesis for being awarded.

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