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Opponent's review of doctoral thesis
„Transient optical and electrical phenomena in organic semiconductors: Insights into fundamental photophysics, resistive switching, and emulation of synaptic plasticity“
submitted by Yadu Ram Panthi

The introduction logically links the content of the dissertation. The experimental methods for characterizing the selected materials were well chosen and applied precisely, resulting in a compelling overall interpretation of the results. The author exhibited high standards at both the theoretical and methodological levels. The conclusions, which have significant implications for understanding photophysical and electrical phenomena in organic semiconductors, were formulated with precision and conviction.

The author's work has not only been published in three works registered in WOS, with the doctoral student as the first author but has also led to the protection of a Short-Term Patent. Additionally, the author's active participation in international conferences, including a second-place win in the Young Scientist Award, further demonstrates his significant contributions to the field.

The work is well conceived and organized, with a clear presentation and vision. The thesis is written in a high-quality scientific language with minimum typos and errors (e.g., reference [12] on page 15 appears to be faulty, fig. 1.10 B) and does not represent the Nyquist plot for the case of the presence of inductance). I appreciate that the author also includes links to the publisher's versions of the cited articles.

The most significant results are related to the optical study of thiophene-diketopyrrolopyrrole (TDPP) derivatives with extended conjugation and different terminal substitutions of acceptor groups with different electron-withdrawing abilities. The study, represented by two publications in journals with high impact factors, offered insight into the role of end groups in conformationally induced excited state relaxation processes and their application to achieve long-lived phenomena desired in photophysics. The presented findings are essential in the study of triplet formation, which is interesting for the photophysics of many organic materials. The identification of the triplet state in the solution phase and its absence in

the thin film provided an opportunity to design improved DPP derivatives for applications involving the triplet state.

Of great importance are the results obtained on the poly[N-(3-(9H-carbazol-9-yl)propyl)methacrylamide] (PCaPMA) polymer, whose memristive behavior was compared to commercially available poly(N-vinyl carbazole) (PVC). The study contributed to a better understanding of the role of the side groups in polymers. The trapping and de-trapping of charges were identified as the main switching resistance mechanism, and the hydrogen bonds forming crosslinked structures were found to be probably responsible for the memory state's long persistence time. The possibility of creating a memristor with a PCaPMA as an active layer sandwiched between ITO and Au or Al electrodes that emulates the functionality of biological synapses has been proved, and the underlying mechanism has been revealed.

Memristors were also fabricated as sandwich structures with an active layer from the composite system of DPP-DTT mixed with PDI in various proportions. The device was proved to function as an electronic memory, emulating the functionality of a biological synapse. The influence of various solution-casting methods on the OFET performance with PVP dielectrics was investigated and the importance of interface engineering for a proper device performance was proved.

The following questions should be discussed:

The explanation of the resistance switching mechanism is based on space charge limited currents. From the analysis it is not clear how much the electric field dependence of the charge carrier mobility, common for organic semiconductor, is involved. Could you comment it?

I also have a question for the doctoral student above the scope of his work, even if it is related to it. In what time horizons does he see the perspective of using organic memristors and neuromorphic computing? Another hypothetical question concerning the faster aging of organic semiconductors is whether organic electronics based on materials with evidently less photochemical stability can sometimes be put into production for the market.

Yadu Ram Panthi has demonstrated the ability to do analytical and synthetic creative work in the field of research. Therefore, I recommend the dissertation thesis for defense. After a successful defense, I recommend conferring PhD title on Yadu Ram Panthi.

Bratislava, August 30, 2024

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