Molecular Engineering of DSSC Sensitizers: A DFT Study

Margarita Bužančić Milosavljević,[a] Antonija Mravak[b] and Martina Perić Bakulić[b]

margarita@stim.unist.hr

[a]*Center of Excellence for Science and Technology-Integration of Mediterranean region (STIM), University of Split, Ruđera Boškovića 31, 21000 Split, Croatia*
[b]*Faculty of Chemistry and Technology, University of Split, Ruđera Boškovića 35, 21000 Split, Croatia*

Efficient harvesting of solar energy represents one of the central challenges in the transition toward sustainable energy systems. The solar energy reaching the Earth in one hour exceeds the total annual global energy consumption [1], highlighting the potential of solar technologies. Among various approaches, dye-sensitized solar cells (DSSCs) remain one of the most promising and extensively investigated systems for direct solar energy conversion. Inspired by natural photosynthesis, DSSCs represent a low-cost, structurally versatile, and sustainable alternative to traditional photovoltaic devices [2]. However, despite significant experimental and technological progress, further improvements in efficiency and stability are still required. One of the most important components governing DSSC performance is the sensitizer [3].

Here we present the theoretical design and characterization of DSSC sensitizers, a key component that can be tuned for improved electron injection into the semiconductor band and faster electrolyte regeneration. Different organic, natural, and hybrid (natural dye–noble metal nanocluster) sensitizers, as well as their doped derivatives in combination with various semiconductors and electrolytes, have been investigated in the context of DSSC applications [4].

The prediction of their structural, optical, and photovoltaic properties has been carried out within the framework of density functional theory (DFT) and time-dependent DFT (TD-DFT). These theoretical investigations enable systematic evaluation of electronic structure, absorption characteristics, and energy level alignment relevant for efficient charge injection and regeneration processes. The obtained results contribute to a better understanding of structure–property relationships and support further optimization of sensitizer design. They demonstrate the value of theoretical modeling in guiding the development of improved DSSC materials.

References:

[1] (a) E. Hossain, *The Sun, Energy, and Climate Change*, pp. 1–67, Springer Cham, 2023; (b) M. G. Norton, *A Modern History of Materials. From Stability to Sustainability*, pp. 127-146, Springer Cham, 2023.

[2] R. Sasikumar, S. Thirumalaisamy, B. Kim, B. Hwang, *Renew. Sustain. Energy Rev.* **199** (2024) 114549.

[3] W. Fan, D. Tan, W.-Q. Deng, *ChemPhysChem* **13** (2012) 2051–2060.

[4] (a) M. Bužančić Milosavljević, A. Mravak, M. Perić Bakulić, V. Bonačić-Koutecký, *RSC Advances* **13** (2023) 6010-6016; (b) M. Bužančić Milosavljević, M. Perić Bakulić, Ž. Sanader Maršić, A. Mravak, V. Bonačić-Koutecký, *Nanomaterials* **14** (2024) 1034.