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Rozprawa doktorska pt. ***Analiza transferu energii i elektronów w wieloskładnikowych układach bionanohybrydowych***

Abstract

The bionanohybrid systems are molecular constructs composed of nanomaterials (materials with size in nanometer scale at least in one dimension) and biological elements: proteins, lipid membranes, or biomolecular complexes. When the system components are redox-active or fluorescent, the energy or electron transfer can take place. Due to diverse structure and stoichiometry of the components, the energy and electron transfer in the bionanohybrid systems occurs often by complex mechanisms that involve simultaneous and competitive processes. This competition may be significant especially after delivery of the bionanohybrid systems to living cells or cell-mimicking environments.

Within this dissertation, the system that was composed of the semiconductor quantum dots (CdTe nanocrystals) and cytochrome *c* protein was used as a model to analyze the competition between two processes: the photoinduced electron transfer and the transfer of electronic excitation energy. The luminescent and redox properties of cytochrome *c* were modified by the removal of the central iron ion of the porphyrin ring and its substitution by zinc or tin atoms – such derivatives lose the redox activity and gain the fluorescent properties. The contribution of each of the occurring processes was determined using spectrophotometric and fluorimetric methods, by stationary and time-resolved measurements. The experiments showed that the energy transfer from quantum dots to cytochrome *c* dominates in comparison to the electron transfer. In the long time-scale the reduction products accumulate due to irreversibility of this reaction in the given system. However, the ratio of both processes may be modified – in the presence of various cytochrome *c* derivatives as alternative acceptors, the photoreduction rate was increased or decreased. Transient absorption measurement allowed to determine the early (picoseconds-nanoseconds after excitation) photodynamics within the bionanohybrid.

The supplemental part of this work was the construction of the system composed of quantum dots and natural antenna complexes – phycobilisomes and plant light-harvesting complexes of type II – organized on the surface of carbon nanotube scaffold. The energy transfer within the bionanohybrid and to photosystems localized in the isolated thylakoids was confirmed by fluorescence lifetime imaging microscopy. Oxygen consumption measured by Clark electrode showed approximately two-fold increase of the photosystem activity in 510-530 nm illumination.

The obtained data gives substantial insight into the understanding of the energy transfer processes occurring in the bionanohybrid systems. This work reveals the potential offered by the assemblies of nanomaterials and biomolecules used as antenna complexes, biosensors and photocatalysts.