Molecular Dynamics Insights into Photosystem I–Platinum Biohybrids for Photocatalytic Hydrogen Production

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Photosystem I (PSI) is a protein-pigment complex embedded in the thylakoid membrane of photosynthetic organisms that captures sunlight and delivers high-energy electrons to charge carriers like ferredoxin and flavodoxin to power downstream metabolism and carbon fixation. However, electrons captured by PSI have been demonstrated experimentally to transfer from iron-sulfur clusters within PSI to bound platinum nanoparticles (PtNPs). Once the electrons are transferred to the PtNP, they act as catalysts for hydrogen gas production, thus making these PtNPs attractive as biohybrids. Cryo-electron microscopy structures are available for these PSI-Pt biohybrids and indicate that these PtNPs occupy the ferredoxin and flavodoxin binding sites on PSI. However, cryoEM offers only a static picture over a population ensemble of PtNPs. So, to investigate the dynamics, we performed classical molecular dynamics simulations on PSI–Pt biohybrids from three organisms: *Synechococcus lividus* and *Thermosynechococcus vestitus* (each with two PtNPs) and *Synechococcus leopoliensis* (with one PtNP). Our simulations revealed that the interactions between the PSI and PtNPs are stable with minimal displacement throughout the simulation. The structural analysis from our study revealed that Lys27 and Lys30 of PsaA for NP1, while Lys314 of PsaB, Lys155 of PsaF for NP2, are the key residues involved in stabilizing the PtNPs on the stromal surface of the PSI monomer. The NP1 is positioned closer to the 4Fe-4S clusters in all three systems; however, in *S. leopoliensis*, where only one PtNP is bound, NP1 resides much closer to the 4Fe-4S cluster. This positioning highlights NP1 as the most promising site for efficient electron transfer and H_2 evolution. Therefore, this study provides the molecular insights that can further refine nanoparticle position on the PSI surface, which can pave the path for enhanced electron transfer efficiency and hydrogen production.