

Energy transfer pathways revealed from structural analysis of the plant PSI-LHCI supercomplex

Photosystem I (PSI) is one of the two photosystems functioning in oxygenic photosynthesis by various photosynthetic organisms ranging from cyanobacteria to eukaryotic algae and higher plants. It absorbs light energy from the sun to initiate a series of electron transfer reactions starting from electron donors plastocyanin or cytochrome c_6 to iron-sulfur centers, which is finally utilized to reduce NADP^+ to NADPH required for the reduction of carbon dioxide into sugars. The electrons used by PSI come from water through the water-splitting reaction catalyzed by photosystem II (PSII). In this way, the two photosystems work in series to extract electrons from water to reduce carbon dioxide, thereby converting light energy into chemical energy indispensable for sustaining life on the earth.

The reaction center of PSI performing light-induced charge separation and electron transfer reactions is largely conserved from prokaryotic cyanobacteria to higher plants, and is designated the PSI core. The PSI core from cyanobacteria contains 9 transmembrane and 3 peripheral subunits together with 127 cofactors (including 96 chlorophylls (Chls)), and exists as a trimer with a total molecular mass of around 1068 kDa. The structure of cyanobacterial PSI core has been analyzed at 2.5 Å resolution in 2001 [1]. On the other hand, PSI core from higher plants exists as a monomer and is surrounded by 4 trans-membrane light-harvesting complex I (LHCI) subunits Lhca1-Lhca4, forming a PSI-LHCI supercomplex with a total molecular mass over 600 kDa. One of the distinct features of this supercomplex is that the energy absorbed by LHCI is transferred to the PSI core with an extremely high efficiency (over 95%). The

structure of higher plant PSI-LHCI supercomplex has been solved up to 3.3 Å resolution [2-4]. However, this resolution was not high enough to reveal the exact location and detailed organization of a vast number of cofactors including more than 150 Chls, which hampered an unambiguous elucidation of the principles underlying the highly efficient energy transfer pathways.

In order to improve the crystal resolution, we purified the PSI-LHCI supercomplex from pea leaves with a modified procedure, and screened the crystallization conditions extensively. As a result, we were able to solve the structure of PSI-LHCI at a 2.8 Å resolution with X-rays from beamlines **BL41XU** and **BL44XU** [5]. Our structure showed that the whole complex contained 16 subunits, among which, 12 subunits belong to the PSI core whereas 4 are LHCI subunits (Lhca1-Lhca4) (Fig. 1(a) and 1(b)). Within the 12 core subunits, PsaM and PsaX found in the cyanobacterial PSI core were not present whereas 2 subunits (PsaG and PsaH) unique to higher plant PSI were found [5].

In addition to the protein subunits, a total of 205 cofactors were identified in the PSI-LHCI supercomplex. These included 155 Chls (143 Chls a and 12 Chls b), 35 carotenoids [26 β -carotenes (BCRs), 5 luteins (Luts), and 4 violaxanthins (Vios)], 10 lipids [6 phosphatidylglycerols (PGs), 3 monogalactosyldiacylglycerols (MGDGs), and 1 digalactosyldiacylglycerol (DGDG)], 3 Fe_4S_4 clusters, 2 phyloquinones, and several water molecules (Fig. 2). Among these cofactors, 98 Chls a, 22 BCRs, 5 lipids (3 PGs, 1 MGDG, and 1 DGDG), 3 Fe_4S_4 clusters, and 2 phyloquinones are located in the PSI core, which

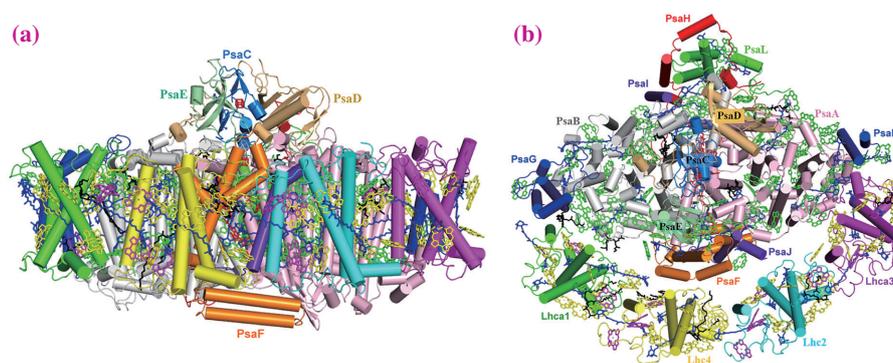


Fig. 1. Overall structure of the plant PSI-LHCI supercomplex. (a) Side view; (b) Top view from the stromal side.

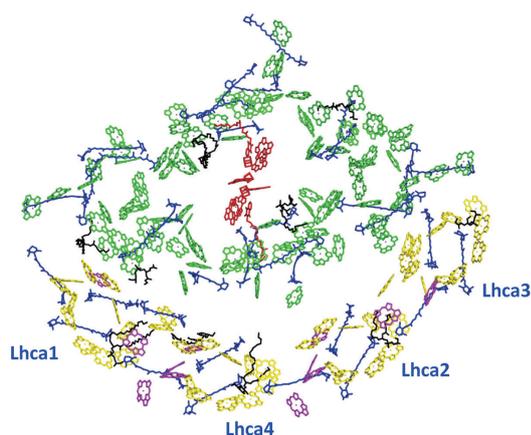


Fig. 2. Arrangement of pigments and other cofactors in the PSI-LHCI supercomplex with the same top view as in Fig. 1(b). Color codes: green, Chls a of PSI core complex; yellow, Chls a of LHCI; magenta, Chls b of LHCI; blue, carotenoids; black, lipids; red, cofactors of the electron transfer chain (Chls a, phyloquinones, and Fe₄S₄ clusters).

is largely comparable to the 96 Chls, 22 BCRs and 4 lipids found in the cyanobacterial PSI core structure, although several Chla were found to be in slightly different positions or orientations.

The most significant feature of the plant PSI-LHCI supercomplex is the presence of 4 LHCI subunits that function to harvest light energy efficiently. Our study revealed the detailed structures of these subunits and the organization of pigments they bind for the first time. Thus, we identified the location of 45 Chl a, 12 Chl b, and 13 carotenoids in these 4 subunits, and the interaction sites within the Lhca subunits and between each of the Lhca subunit and PSI core. We found that each of the LHCI subunits binds 1 BCR, 1 Lut and 1 Vio, with an additional Lut located between Lhca1 and Lhca4. The binding of Vio to LHCI was demonstrated for the first time, as was the difference between Chl a and b within LHCI. The presence of Vio in LHCI suggested its role in photoprotection, since it is well known that this carotenoid is involved in the “xanthophyll-cycle” in LHCII functioning in photoprotection. The “gap region” between the LHCI and PSI core are filled with “red Chls”, which is a dimer of Chl a with a lower energy absorption level due to the strong interaction between the 2 Chl a monomers. Each of the 4 Lhca subunits binds one red Chl dimer, and these red Chls may have important roles in the energy transfer from LHCI to the PSI core, as well as the binding of LHCI to the PSI core.

Based on our structure, 4 plausible pathways for the energy transfer from LHCI to the PSI core were deduced (Fig. 3(a)). Among these 4 pathways, Chls in Lhca1 and Lhca3 in the 2 sides of the Lhca belt have

short distances to the PSI core Chls, offering 2 efficient energy transfer pathways (Fig. 3(b) and 3(c)). Chls in Lhca2 interact with PSI core Chls with a slightly longer distance, providing another energy transfer pathway (Fig. 3(c)). Chls in Lhca4 have a much longer distance to the PSI core, and therefore may not be able to transfer their energy directly but have to pass through Lhca1 in the luminal side to transfer their energy to the core.

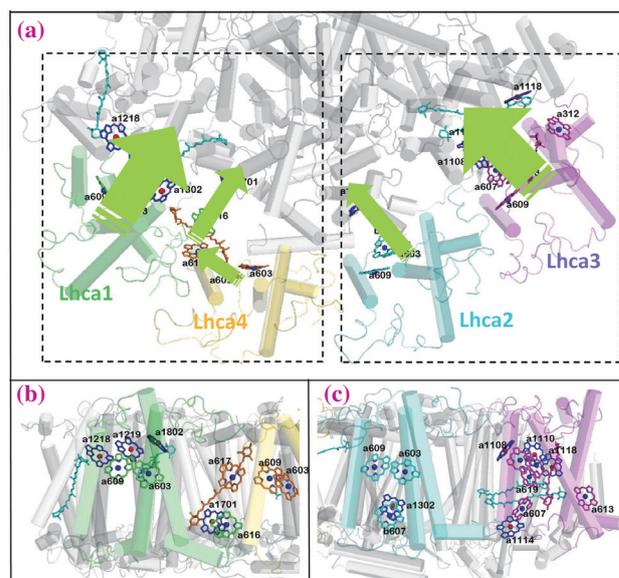


Fig. 3. Plausible energy transfer pathways from LHCI to the PSI core. (a) Overall view of the 4 energy transfer pathways from LHCI to the PSI core, with a top view from the stromal side. The green, thick arrows indicate efficient energy transfer pathways from Lhca1 and Lhca3 to the PSI core, whereas thin arrows indicate less efficient pathways from Lhca2 to PSI core, and from Lhca4 through Lhca1 to the core. (b) and (c). Detailed interactions among the pigments involved in the energy transfer pathways indicated in panel (a). Side view of the boxed areas with dashed lines in the left (b) and right (c) sides of panel (a).

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