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Proton transport in photooxidation of water: A new perspective on photosynthesis

(uncouplers/plastoquinone/photosystems I and II/oxygenic and anoxygenic photosystems)

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ABSTRACT **The currently prevalent concept of the generation of photosynthetic reducing power in oxygen-evolving cells envisions a linear (noncyclic) electron flow from water to ferredoxin (and thence to $NADP^+$) that requires the collaboration of photosystems I and II (PSI and PSII) joined by plastoquinone and other electron carriers (the Z scheme). The essence of the Z scheme is that only PSI can reduce ferredoxin—i.e., that, after being energized to an intermediate reducing potential by PSII, electrons from water are transported via plastoquinone to PSI which energizes the electrons to their ultimate reducing potential adequate for the reduction of ferredoxin. Basic to the Z scheme is the function of plastoquinone as the obligatory link in electron transport from PSII to PSI. However, we have found that, when plastoquinone function was inhibited, ferredoxin was photoreduced by water without the collaboration of PSI. We now report evidence for an important function of plastoquinone in the translocation of protons liberated inside the thylakoid membrane by photooxidation of water. When the oxygenic photoreduction (i.e., by water) of ferredoxin was blocked by plastoquinone inhibitors, dibromothymoquinone or dinitrophenol ether of iodonitrothymol, the photoreduction of ferredoxin was restored by each of four chemically diverse uncouplers, similar only in their ability to facilitate proton movement across membranes. Similar results were obtained for the oxygenic reduction of $NADP^+$. Our results suggest that the light-induced electron flow from water cannot be maintained unless the simultaneously liberated protons are removed from inside the membrane via plastoquinone. The new evidence is embodied in a concept of an *oxygenic photosystem* for photosynthetic electron and proton transport, which we propose as an alternative to the Z scheme, to account for photoreduction of ferredoxin-$NADP^+$ by water and the coupled oxygenic (formerly noncyclic) ATP formation without involving PSI. The role of the *anoxygenic photosystem* (formerly called PSI) is ATP formation by cyclic photophosphorylation.**

Photooxidation of water, $2H_2O \rightarrow 4\,e^- + 4H^+ + O_2$, is a key reaction in plant photosynthesis. Aside from its all-important by-product, oxygen, the liberated electrons eventually reduce ferredoxin (1, 2) and then $NADP^+$ (3), thereby accounting for photosynthetic reducing power; the liberated protons contribute to the protonmotive force (4, 5) which accounts for the ("noncyclic") ATP formation that is coupled to $NADP^+$ reduction (6).

Photooxidation of water takes place inside the thylakoid membranes (7). These contain two photocenters identified with photosystems I and II (PSI and PSII) and a chain of carriers responsible for the transport of electrons and protons within and across the membrane. According to the now-prevalent concepts embodied in the so-called Z scheme (8, 9), PSII photooxidizes water but cannot reduce ferredoxin because it can energize the released electrons only to an intermediate potential; the electrons are transported from PSII to PSI where a second photoact energizes them to their ultimate reducing potential adequate for the reduction of ferredoxin and the more electronegative components of PSI. Associated with this noncyclic (linear) electron flow is depicted a transport of protons in which plastoquinone is the preeminent carrier; because oxidoreductions of plastoquinone involve transfers of hydrogen atoms, this abundant chloroplast component serves as a carrier of both electrons and protons (8–10).

$$\begin{array}{ccccccccccc} H_2O & \rightarrow\rightarrow & PSII & \rightarrow\rightarrow & PQ & \rightarrow\rightarrow & PSI & \rightarrow\rightarrow & Fd & \rightarrow & NADP^+ \\ \downarrow & & \uparrow & & & & \uparrow & & & & \\ O_2 & & h\nu & & & & h\nu & & & & \end{array}$$

As summarized by this sequence, in which PQ is plastoquinone, Fd is ferredoxin, and double arrows indicate other carriers, the Z scheme includes plastoquinone as an obligatory link in electron transport between PSII and PSI; not shown is the concurrent transmembrane shuttle of protons via plastoquinone from the outside (stroma side) to the membrane's inside aqueous space (lumen) (8, 11). Note that the Z scheme assigns no role to plastoquinone in the removal of protons released by the photooxidation of water (8, 11). These are thought to diffuse from inside the membrane to the lumen without the participation of plastoquinone (11).

Our perspective on photosynthetic electron and proton transport changed because of recent findings (12, 13), particularly those pertaining to plastoquinone (14), that were in conflict with fundamental postulates of the Z scheme. Thus, when plastoquinone function was inhibited, ferredoxin was oxygenically reduced without the collaboration of PSI (14). ("Oxygenic reduction" denotes reduction by electrons that originate from water.) These results were incompatible with the accepted role of plastoquinone as the indispensable link between PSII and PSI but provided no alternate role. That plastoquinone function is essential for the oxygenic photoreduction of ferredoxin-$NADP^+$ was evident from the well-documented sensitivity of this reaction to plastoquinone inhibitors (15, 16). The role of plastoquinone was therefore reinvestigated.

We report here evidence indicative of an important function for plastoquinone in the translocation of protons liberated inside the thylakoid membrane by photooxidation of water. These findings lend support to a concept, presented here as an alternative to the Z scheme, that envisions an oxygenic photosystem (supplanting PSII) capable of photoreducing ferredoxin-$NADP^+$ by water without the collaboration of PSI.

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Abbreviations: PSI, photosystem I; PSII, photosystem II; DBMIB, 2,5-dibromo-3-methyl-6-isopropyl-*p*-benzoquinone (dibromothymoquinone); DNP-INT, dinitrophenol ether of iodonitrothymol; EPR, electron paramagnetic resonance; FCCP, carbonyl cyanide trifluoromethoxyphenylhydrazone.

METHODS

Chloroplasts were isolated from spinach leaves (*Spinacia oleracea*, var. Marathon) grown in a greenhouse in nutrient solution culture (17) and freshly harvested before each experiment. The preparation used consisted of osmotically disrupted ("broken") chloroplasts that retained the integrity of the thylakoid membrane structure needed for complete electron transport from water to $NADP^+$ and for ferredoxin-catalyzed cyclic photophosphorylation (18). Chlorophyll was estimated (17), ferredoxin was isolated and purified (19) (by R. K. Chain), and the photoreduction of $NADP^+$ was measured (20) as described previously. Glucose oxidase (type VII), bovine catalase, and $NADP^+$ were purchased from Sigma. 2, 5-Dibromo-3-methyl-6-isopropyl-*p*-benzoquinone (DBMIB) and the dinitrophenol ether of iodonitrothymol (DNP-INT) (gifts of A. Trebst) were added as methanol solutions (DNP-INT was first dissolved in a drop of dimethyl formamide). Equal concentrations of methanol were added to the control treatments.

The photoreduction of ferredoxin was measured by electron paramagnetic resonance (EPR) spectroscopy. The chloroplasts (in their respective reaction mixtures) were placed in quartz EPR tubes (3-mm inside diameter) that had been gassed with nitrogen. The tubes were illuminated at a physiological temperature (293 K) for 30 sec and then, with illumination continued, were immersed for 30 sec in liquid nitrogen contained in a silvered Dewar flask with a window that admitted light. The frozen samples in the quartz tubes were further cooled in the EPR cavity with liquid helium to 60 K by an Oxford temperature controller (model DTC) and a cryostat (model ESR9) equipped with a quartz Dewar cell (made by J. Scanlon, Solvang, CA). First-derivative EPR spectra of the frozen samples were obtained with a Bruker Instruments (Billerica, MA) X-band spectrometer (model ER200tt) [equipped with a 20-cm (8-inch) magnet] operated at a frequency of 9.43 GHz and were recorded after processing by a digital signal averager (model 1070, Nicolet Instruments, Madison, WI).

Monochromatic illumination (650 nm) was provided by a light beam from a Quartzline lamp (type DXN, 1000 W). The light beam was passed through heat-absorbing and interference filters (Baird, Medford, MA).

RESULTS

Investigations of the role of plastoquinone in photosynthesis were greatly facilitated when Trebst and associates introduced potent and specific plastoquinone inhibitors, notably DBMIB (15, 21) and, more recently, DNP-INT (16, 21). In previous experiments we found a differential effect of these two inhibitors on the oxygenic photoreduction of ferredoxin and components of PSI. DBMIB and DNP-INT at concentrations that blocked completely the photoreduction of the bound Fe-S centers of PSI had no effect on the oxygenic photoreduction of ferredoxin (12, 14).

In the experiments just cited (12, 14), the chlorophyll concentration (1 mg/ml) was much higher than that commonly used for $NADP^+$ reduction. Given a molar plastoquinone/chlorophyll ratio in spinach chloroplasts of about 1:10 (22) and a ferredoxin/chlorophyll ratio of about 1:400 (1), the high chlorophyll (i.e., chloroplast) concentration, which was selected to facilitate the measurement of the Fe-S signals, resulted in a high concentration of plastoquinone—specifically, in a plastoquinone/ferredoxin ratio of 10:1, similar to that in intact chloroplasts (see *Discussion*). Therefore, we undertook to reinvestigate the effect of plastoquinone inhibitors on the oxygenic photoreduction of ferredoxin at a low chlorophyll concentration (50 μg/ml), similar to that often used experimentally for

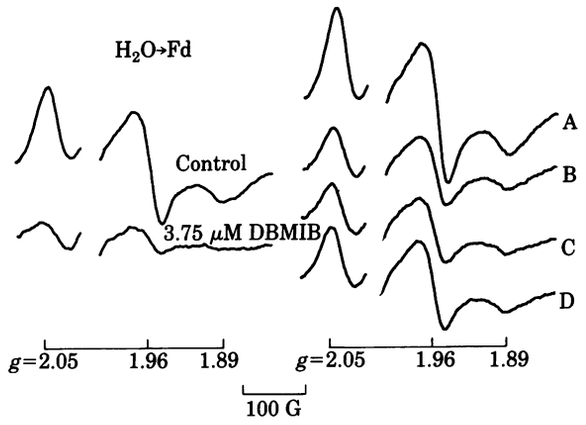

FIG. 1. Uncouplers reverse DBMIB inhibition of ferredoxin (Fd) photoreduction, measured by EPR spectroscopy. The reaction mixture, equilibrated with N_2, contained osmotically disrupted chloroplasts (equivalent to 50 μg of chlorophyll per ml), 50 mM *N*-tris(hydroxymethyl)methylglycine (Tricine) buffer (pH 7.7), 10 μM spinach ferredoxin, 5.0 mM $MgCl_2$, 50 mM KCl, 2.5 mM ADP, 2.5 mM K_2HPO_4, 10 mM glucose, glucose oxidase, catalase, and 6% methanol. The EPR tubes were illuminated for 30 sec at 293 K and immediately frozen in liquid N_2 under continuing illumination (650 nm, 5×10^5 ergs·cm^{-2}·sec^{-1}). EPR spectra were recorded at 60 K. Spectrometer field setting, 3450 ± 200 G; microwave power, 10 mW; modulation amplitude, 10 G; gain, 1×10^5. Curves A–D are for 3.75 μM DBMIB plus uncouplers: A, 10 μg of gramicidin; B, 0.5 μM nigericin; C, 5 μM FCCP; D, 0.5 μM SF 6847.

$NADP^+$ reduction. The plastoquinone/ferredoxin ratio was thus decreased by a factor of 20 to about 0.5:1.

In the EPR traces obtained, the extent of ferredoxin reduction is indicated by the amplitude of its characteristic (in the reduced state) signals at $g = 1.89$, 1.96 (main signal), and 2.05. At the low chlorophyll (and correspondingly low plastoquinone) concentrations used, the oxygenic photoreduction of ferredoxin was strongly inhibited by DBMIB (Fig. 1) and DNP-INT (Fig. 2). A novel finding was that the inhibition by DBMIB and also by DNP-INT was reversed by each of four uncouplers: gramicidin, nigericin, carbonyl cyanide trifluoromethoxyphenylhydrazone (FCCP), and SF 6847 [a ditertiary phenol derivative (23)].

Given the known mode of action of uncouplers as transmembrane proton carriers (5, 24) and the role of plastoquinone as the proton shuttle in thylakoids (reviewed in ref. 8), these results suggested that the oxygenic photoreduction of ferredoxin was linked to a translocation of liberated protons by plastoqui-

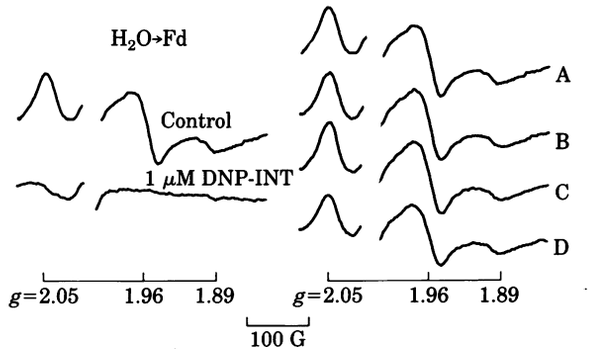

FIG. 2. Uncouplers reverse DNP-INT inhibition of ferredoxin photoreduction. Experimental conditions as in Fig. 1, except that 1 μM DNP-INT replaced DBMIB. A, 15 μg of gramicidin; B, 0.25 μM nigericin; C and D, as in Fig. 1.

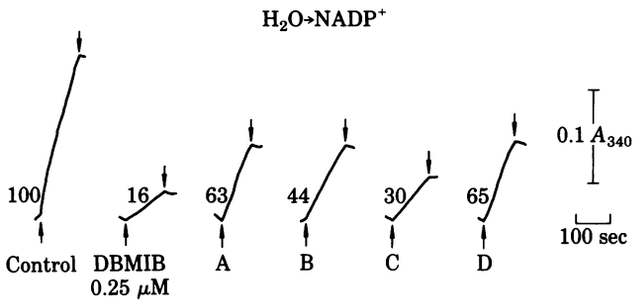

FIG. 3. Uncouplers reverse DBMIB inhibiton of $NADP^+$ photoreduction. The reaction mixtures were as in Fig. 1 except that 2 mM $NADP^+$ was present throughout and N_2/glucose, glucose oxidase, and catalase were omitted. The reaction mixtures were illuminated at room temperature in cuvettes (2.0-mm light path) open to the air. Monochromatic illumination: 650 nm; 5×10^4 ergs·cm^{-2}·sec^{-1}. Arrow up, light on; arrow down, light off. Numbers give rates of $NADP^+$ reduction (μmol/mg of chlorophyll per hr). Curves A–D are for 0.25 μM DBMIB plus uncouplers: A, 10 μM FCCP; B, 20 μg of gramicidin; C, 0.5 μM nigericin; D, 0.5 μM SF 6847.

none. When this plastoquinone function was inhibited, electron flow to ferredoxin stopped unless proton translocation was restored by the addition of an uncoupler. This interpretation was corroborated by the parallel findings of reversal, by the same uncouplers, of the inhibition by DBMIB of the oxygenic photoreduction of $NADP^+$ (Fig. 3) and, even more strikingly, by DNP-INT (Fig. 4). A reversal by gramicidin of the DNP-INT inhibition of $NADP^+$ reduction was also reported by Trebst *et al.* (16).

There remained the question of the origin of protons whose translocation by plastoquinone is coupled to the oxygenic photoreduction of ferredoxin. We have concluded that the protons originate from the intramembrane photooxidation of water (see *Discussion*).

DISCUSSION

As might be expected, the perceived role of plastoquinone, the most abundant redox component of thylakoid membranes (8, 10, 22), profoundly influences perspectives on the nature of the photosynthetic electron transport that is responsible for the reduction of ferredoxin (and hence $NADP^+$) by water. We have investigated the role of plastoquinone under two conditions: (*i*) high chlorophyll concentration that resulted in a plastoquinone/ferredoxin ratio of 10:1 (14) [compared with 40:1 in intact chloroplasts (1, 10)] and (*ii*) low chlorophyll concentration, used in this study, that resulted in a plastoquinone/ferredoxin ratio of 0.5:1. Our results in both cases were incompatible with the role ascribed to plastoquinone in the Z scheme.

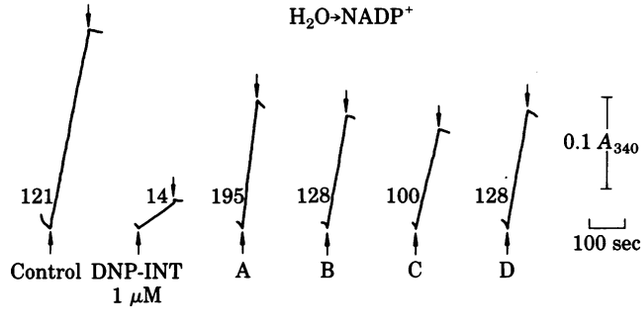

FIG. 4. Uncouplers reverse DNP-INT inhibition of $NADP^+$ photoreduction. Experimental conditions as in Fig. 3, except that 1 μM DNP-INT replaced DBMIB. A, 5 μM FCCP; B, 15 μg of gramicidin; C, 0.25 μM nigericin; D, 0.5 μM SF 6847.

At the high plastoquinone/ferredoxin ratio, electrons originating from water normally reduced both ferredoxin and the components of PSI, represented by the bound Fe-S centers. When plastoquinone function was inhibited, the PSI components were no longer reduced but there was no inhibition of the oxygenic photoreduction of ferredoxin (14). Thus, it appeared that, contrary to the Z scheme, the photoreduction of PSI components was not a precondition for ferredoxin reduction but a parallel event and one that was more sensitive to inhibitors of plastoquinone function than was the oxygenic photoreduction of ferredoxin.

At the low plastoquinone/ferredoxin ratio used here, we found that the oxygenic photoreduction of ferredoxin was highly sensitive to plastoquinone inhibition. Moreover, this inhibition was reversed by four, chemically diverse uncouplers similar only in their ability to facilitate proton movement across membranes (5, 24). We conclude therefore that the primary role of plastoquinone in the oxygenic photoreduction of ferredoxin-$NADP^+$ is to transport to the thylakoid lumen protons liberated by the photooxidation of water inside the thylakoid membrane that is impermeable to protons. The protons do not diffuse but are released into the lumen by the oxidation of plastohydroquinone.

Our results suggest that electron flow from water cannot be maintained unless the simultaneously liberated protons are removed from inside the membrane via plastoquinone or artificially via uncouplers and lipophylic oxidants. As will be discussed more extensively elsewhere, we interpret in this manner the reversal by phenylenediamines and benzoquinones of DBMIB inhibition of Hill reactions (25, 26). We attribute the varied effectiveness of these components to differences in their properties as proton carriers.

In essence then, we suggest that electron transport from water to ferredoxin is catalyzed by an *oxygenic photosystem* capable not only of photooxidizing water (like PSII) but also of generating a reducing potential sufficient for ferredoxin reduction, a function limited to PSI in the Z scheme. The designations "oxygenic" and "anoxygenic" (see below) are introduced to avoid confusion with the widespread notions that PSII photooxidizes water but cannot reduce ferredoxin whereas PSI reduces ferredoxin but cannot photooxidize water.

As shown in Fig. 5, we envision that the transfer of one electron from water to ferredoxin requires two photoacts. One photoact would account for the linear transfer of an electron to ferredoxin, possibly through the mediation of pheophytin, an intermediate (E_m *ca.* −610 mV) (27) recently identified with PSII (28, 29). The second photoact is envisaged as generating a novel "oxygenic" cyclic electron flow (distinct from the anoxygenic cyclic type below) that permits plastoquinone to trans-

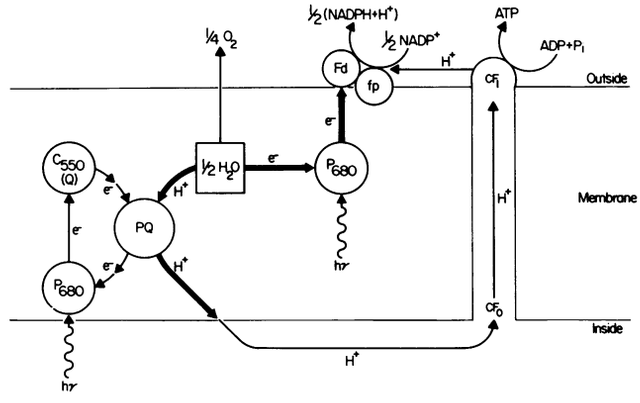

FIG. 5. The oxygenic photosystem. Details are in text.

port, to the thylakoid lumen, protons liberated inside the thylakoid membrane by the photooxidation of water. We propose that this cyclic component of the oxygenic photosystem includes as carriers C_{550} (Q), plastoquinone, cytochrome b_{559}, and plastocyanin and that it contains the site of diuron inhibition. Plastoquinone (PQ) receives the complementary electron from the reaction center chlorophyll (P_{680}) as it binds H^+ inside the membrane and it returns the electron to P_{680} as it releases H^+ into the lumen. This oxygenic proton transport generates the protonmotive force that drives, via the membrane-localized reversible ATP synthase (CF_0-CF_1) (30), the oxygenic (noncyclic) photophosphorylation which is stoichiometrically coupled to ferredoxin (2) and $NADP^+$ reduction (6).

The need for two photoacts—i.e., two quanta—for the transfer of one electron from water to ferredoxin is in good agreement with direct measurements by different laboratories of a requirement of two quanta per electron in the photoreduction of $NADP^+$ by water (20, 31–34). An expenditure of two quanta per electron also has favorable thermodynamic consequences for the postulated overall mechanism of photooxidation of water. An improbably high efficiency would be needed to bring about the photoreduction of pheophytin by water (potential span *ca.* 1.5 eV) by the energy of one quantum of 670 nm light (equivalent to 1.84 eV). However, the thermodynamic probability is greatly enhanced by coupling this event with a second photoact in which the energy demand is considerably less (potential span *ca.* 0.9 eV). Stated otherwise, the removal of protons from the membrane facilitates the photooxidation of water reaction.

Space restrictions permit only limited discussion of related matters. The *anoxygenic photosystem* is equivalent to PSI but we envision its role to be not the reduction of ferredoxin-$NADP^+$ as portrayed by the Z scheme but the production of ATP by cyclic photophosphorylation (18). Given an artificial direct donor (e.g., dichlorophenol indophenol/ascorbate), the anoxygenic photosystem can photoreduce ferredoxin (and $NADP^+$) but this reduction does not mirror any physiological pathway. As shown elsewhere (12), ferredoxin so reduced is readily distinguishable from ferredoxin reduced by the physiological donor, water.

In the anoxygenic photosystem, a single photoact transfers an electron from P_{700} to a primary acceptor (A) from which the electron returns to P_{700} via a photoredox chain that includes bound Fe-S centers (A and B), ferredoxin, and plastoquinone (Fig. 6). (Other carriers in the chain include cytochrome b_6, the "Rieske" Fe-S center, cytochrome *f*, and plastocyanin.) The protonmotive force needed for ATP synthesis is generated as plastoquinone shuttles protons across the membrane from the outside (stroma) into the thylakoid lumen; protons return to the outside via the ATP synthase (CF_0-CF_1).

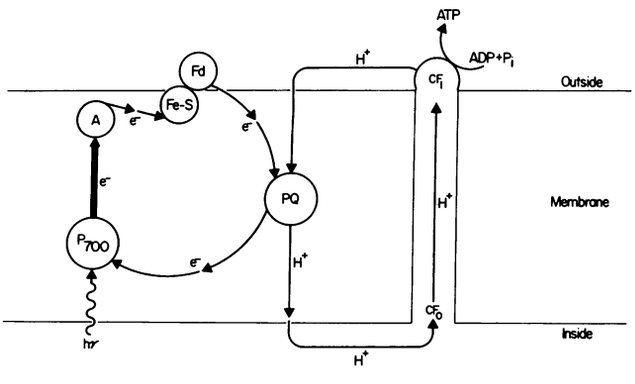

FIG. 6. The anoxygenic photosystem. Details are in text.

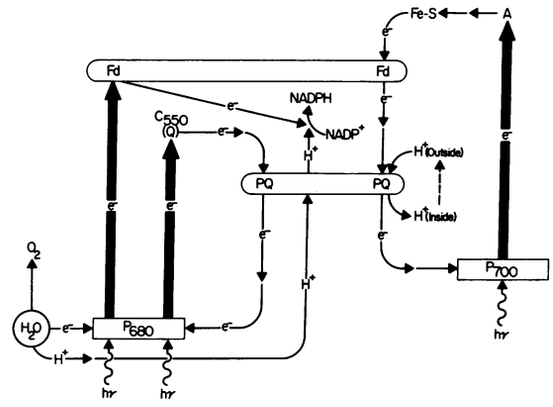

FIG. 7. Linkage between the oxygenic and anoxygenic photosystems. Details are in text.

Ferredoxin and plastoquinone appear to be regulatory links between the oxygenic and anoxygenic photosystems (Fig. 7) although other regulatory links are not excluded. Basically, the two photosystems operate not collaboratively in series but synchronously in parallel.

To recapitulate, the oxygenic photosystem effects a massive vectorial export of those electrons and protons that are liberated inside the thylakoid membrane by photooxidation of water. Electron transport results in ferredoxin reduction; proton transport results in the stoichiometrically coupled ATP formation (2). Reduced ferredoxin is the carrier of photosynthetic reducing power (35) that provides (directly or via $NADP^+$) a conduit for massive electron flow for bioreductions [of CO_2 (as phosphoglycerate), NO_2^-, SO_3^{2-}] or a trickle of electron flow for regulation, as in cyclic photophosphorylation (36, 37) or in thioredoxin-regulated chloroplast enzymes (38). By contrast, the role of the anoxygenic photosystem is limited to ATP production. By inducing a cyclic flow of *intrasystem* electrons, this photosystem generates a transmembrane protonic potential that makes ATP synthesis possible. This ATP supplements that produced by the oxygenic photosystem to give the high ATP/NADPH ratios required for CO_2 assimilation (39) or is used by chloroplasts for such ATP-dependent processes as protein synthesis (40, 41).

The new perspective, viewing the two photosystems as (except for regulatory connections) basically autonomous, excludes the need for a 1:1 stoichiometry between them which is assumed in the Z scheme but which in fact shows wide variability (42). Moreover, by permitting spatial separation of the photosystems, the new perspective provides excellent agreement between structure and function in chloroplasts. Andersson and Anderson (43) have recently shown that the two photosystems are spatially separated in spinach chloroplasts, with the appressed regions of grana (the partitions) containing predominantly PSII (i.e., the oxygenic photosystem) and the exposed regions (grana end membranes and margins), as well as the stroma thylakoids, containing predominantly PSI (i.e., the anoxygenic photosystem). Such spatial separation has been reconciled with the Z scheme by a postulation that plastoquinone serves as a lateral shuttle of reducing equivalents between the two photosystems (43, 44). No such postulation is required by the new concept.

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