Photosynthesis Research **73:** 157–164, 2002.

Minireview

Changing concepts about the distribution of Photosystems I and II between grana-appressed and stroma-exposed thylakoid membranes

Jan M. Anderson
Photobioenergetics, Research School of Biological Sciences, The Australian National University, G.P.O. Box 475, Canberra ACT 2601, Australia (e-mail: anderson@rsbs.anu.edu.au; fax: +61-2-6125-8056)

Received 4 July 2001; accepted in revised form 1 January 2002

Key words: Jan Anderson, Bertil Andersson, Keith Boardman, Robin Hill, lateral heterogeneity, molecular organization, Photosystem I, Photosystem II, Philip Thornber, thylakoid stacking

Abstract

Thylakoid membranes of higher plants and some green algae, which house the light-harvesting and energy transducing functions of the chloroplast, are structurally unique. The concept of the photosynthetic unit of the 1930s (Robert Emerson, William Arnold and Hans Gaffron), needing one reaction center per hundreds of antenna molecules, was modified by the discovery of the Enhancement effect in oxygen evolution in two different wavelengths of light (Robert Emerson and his coworkers) in the late 1950s, followed by the 1960 Z scheme of Robin Hill and Fay Bendall. It was realized that two light reactions and two pigment systems were needed for oxygenic photosynthesis. Changing ideas about the distribution of Photosystem II (PS II) and PS I between the grana-appressed and stroma-exposed thylakoid membrane domains, which led to the concept of lateral heterogeneity, are discussed.

Abbreviations: Chl – chlorophyll; PS I – Photosystem I; PS II – Photosystem II

Harvesting the sun: from the photosynthetic unit to the Z scheme

Crucial observations by Robert Emerson and William Arnold (1932) led Hans Gaffron and Kurt Wohl (1936) to the concept of the photosynthetic unit as an assembly of chlorophyll molecules in which most chlorophyll molecules are not directly concerned with oxygen evolution but rather in capturing and transferring energy to the few that are engaged in photochemistry. Following the concept of the photosynthetic unit, Emerson and Charleton Lewis (1943) demonstrated a decline in the quantum yield of oxygen at the red end of the spectrum, the 'red drop.' This decline could be avoided and the quantum yield increased in the far-red region if supplementary light (absorbed by an 'accessory pigment,' including the short-wave form of chlorophyll *a*) is added to the far-red light, the Emerson enhancement effect (Emerson et al. 1957; Emerson and Eugene Rabinowitch 1960; Govindjee and Rabinowitch 1960). These and other discoveries (see Louis N.M. Duysens 1989) suggested that two separate one-quantum processes are needed for steady-state oxygen evolution.

The modern concept that oxygenic photosynthesis involves not one but two light reactions, carried out by two pigment systems acting in series to transport electrons from water to nicotinamide adenine dinucleotide phosphate ($NADP^+$) with cytochromes acting as intermediate electron carriers, was proposed by R. Hill and F. Bendall (1960) (Figures 1 and 2). Robin Hill and his PhD student R. Scarisbrick discovered cytochrome *f* in 1939 (see Hill and Scarisbrick 1951), and Harold (F or Fearless) Davenport discovered the cytochromes *b* (Davenport 1952). The two decades leading to recognition of the importance of the cytochromes for the Hill and Bendall hypothesis are recounted in Hill's 'green mansions' article (Hill 1956). Their famous 'hypothesis in the barest outline' (see Figure 3) is summarized thus: 'If the cytochrome in the chloroplasts

Figure 1. Robin and Priscilla Hill in Canberra in 1973.

is directly involved in hydrogen (or electron) transfer, the system must require more than one light-driven reaction to act in opposition to the thermochemical gradient' (Hill and Bendall 1960).

In the following year, 1961, two different light reactions were demonstrated by light-induced absorbance changes in both algal cells and isolated chloroplasts (Duysens et al. 1961; Horst Witt et al. 1961) and by electron transport studies with inhibitors (Daniel Arnon et al. 1961). Evidence for two light reactions was provided by Bessel Kok and George Hoch (1961) using light-induced absorbance changes in the reaction center chlorophyll (Chl) P700; they suggested that two quanta were successively trapped by the same photochemical reaction center P700. Govindjee et al. (1960) and Hans Kautsky et al. (1960) confirmed the two-light effect through chlorophyll fluorescence measurements. Recognition of two light reactions meant that the hypothesis of Hill and Bendall (1960) became immortalized in the zigzag, then Z scheme, which was further refined annually as additional components of the photosynthetic apparatus were discovered and characterized (Duysens and Amesz 1962). The Z scheme or its precursor, the photosynthetic unit, however, defines neither the structural nor the spatial organization of the photosystems within thylakoid membranes.

Two discrete photosystems

My initial attempt to determine if two discrete photosystems exist as implied by the Z scheme was to follow the development of photosynthesis during the greening of etiolated beans. It was believed then that Photosystem I (PS I) would have evolved prior to the more complicated PS II; naively, Keith Boardman and I had hoped this might also happen during chloroplast development (see a photograph of myself with Keith in Figure 4). However, the photochemical activities of the greening bean chloroplasts were vanishingly low, and an adequate description of the photosystems was impossible (Anderson and Boardman 1964).

We then used the non-ionic detergent digitonin to fragment isolated spinach chloroplasts. J.S.C. Wessels (1962) first used digitonin to inhibit PS II activity, and a light centrifugal fraction was able to reduce NADP with suitable electron donors, thereby allowing an operational separation of the photosystems like in Arnon et al. (1961). Digitonin fragmentation of spinach thylakoids, followed by differential centrifugation, yielded a heavier membrane fraction enriched in Chl *b* and PS II activity, whereas the lighter fraction with higher Chl *a/b* ratios was highly enriched in P700, PS I activity, and cytochromes *b6* and *f* (Boardman and Anderson 1964; Anderson and Boardman 1966). We characterized a third 'enigmatic' chloroplast cytochrome, Cyt b-559, observed by spectroscopy by Hal Lundegärdh (1961), which being tightly bound in the PS II-enriched fraction was clearly an integral component of PS II (Boardman and Anderson 1967). John Thorne, a former naval radio engineer who headed a small electronics section in the Council of Scientific and Industrial Research Organization (CSIRO), Division of Plant Industry, built a special spectrofluorometer. We characterized the fluorescence properties of enriched-PS II and -PS I thylakoid membrane fractions. At room temperature, most of the chlorophyll fluorescence was emitted by PS II and Chl *b* contributed to both the PS II- and PS I-enriched fractions (Boardman et al. 1966). Thus, this first partial physical separation and characterization of the pho-

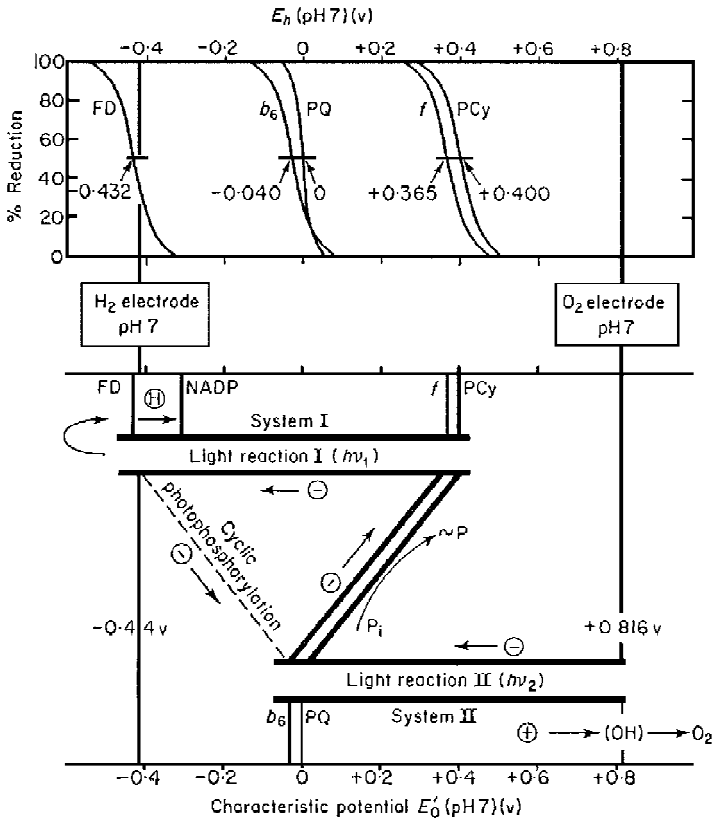

Figure 2. The complete Z scheme (see Hill 1965; the scheme is self-explanatory).

tosystems proved that there were indeed two discrete photosystems.

The harsher detergent Triton X-100 was also used to isolate a light PS I-enriched fraction (Leo Vernon et al. 1966). (Incidentally, Govindjee, who uses only one name, jokingly adopted Triton X-100 as his first name at a Brookhaven Symposium in 1966.) Mechanical means rather than detergents were also introduced to fragment chloroplasts. In 1968, Günter Jacobi used sonication while the Michels (Jean-Marie and Marie-Rose) used the French press, named after its discoverer C. Stacy French (see Sane et al. 1970 for references).

Initially, from freeze-fracture microscopy that splits the membrane longitudinally through its hydrophobic interior, it was believed by some researchers that PS I might be located in the outer half and the less accessible PS II in the inner half of the membrane (Charles Arntzen et al. 1969). Later, using fragmentation of thylakoids by the French press in combination with electron microscopy, P.V. (Raj) Sane et al. (1970) suggested for the first time that PS II and PS I were located together in appressed granal thylakoids, whereas PS I was also present in the stroma-exposed thylakoids. Paul Armond and Arntzen (1977), however, demonstrated that stroma thylakoids had about 20% of PS II.

Lively membrane structure: the fluid mosaic model is perfect for thylakoid membranes

Although there was almost immediate recognition of the two sequential light reactions and general acceptance of the Z scheme, there was little understanding of the organization of the components of the photosynthetic apparatus both across and along chloroplast thylakoids. This was due to ignorance of not only the detailed composition and molecular structure of thylakoid components, but, more importantly, of a credible generalized model for membrane architecture. In contrast, electron and freeze-fracture microscopy had wondrously revealed the unique structural organization of the continuous thylakoid membrane system of higher plants and some green algae, which

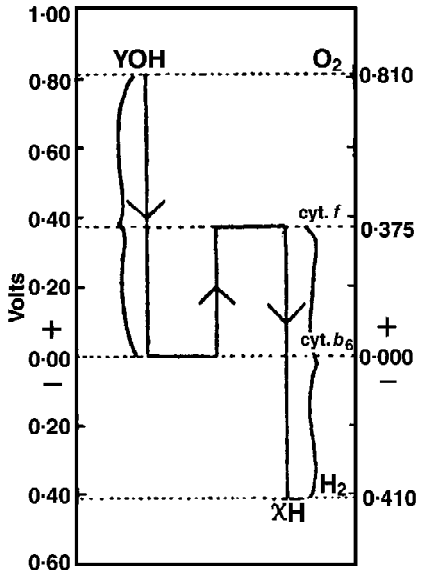

Figure 3. The Z scheme of Hill and Bendall in its barest form (Hill and Bendall 1960).

Figure 4. Keith Boardman and Jan Anderson outside the Biological Laboratories at Harvard University in 1973.

is differentiated into the closely appressed membranes of granal stacks interlinked by stroma-exposed thylakoids.

The generalized membrane model widely accepted before 1970 was originally proposed in 1925 by Gorter and Grende, with further elaborations in 1952 by Davson and Jim Danielli, and in 1964 by Sir Bob Roberston (see Jon Singer 1971 for history and references). In this model, the phospholipids are arranged in a continuous bilayer with their acyl chains occupying the interior membrane core, while the proteins are spread out on both sides of the membrane blanketing the polar phospholipid head groups. However, this membrane model is mainly stabilized by electrostatic forces, which is thermodynamically unlikely because membrane proteins are highly hydrophobic (Singer 1971). Elliot Weier and Andrew Benson (1966) proposed a specific model for thylakoid membranes composed entirely of globular lipid-proteins without any lipid bilayer; instead, globular proteins crossed the membrane with lipids and chlorophylls individually intercalated into hydrophobic folds of the protein.

The concepts of protein arrangement within lipid bilayers, introduced by Singer (1971), led Singer and Garth Nicolson (1972) to formulate a generalized fluid protein–lipid mosaic model, where the membrane consisted of a lipid bilayer into which membrane-spanning intrinsic protein complexes are embedded and to which extrinsic proteins are attached. This model rapidly gained acceptance in the animal field. A serendipitous opportunity coincided with my arrival in Cambridge (UK) in 1973; due to the Coalminers strike, electricity was severely rationed and research in Derek Bendall's laboratory became impossible. Luckily, I was able to retire to a perfect ivory tower. Inspired by the brilliant fluid mosaic model of cell membrane structure (Singer and Nicolson 1972, see above), I dreamt that multiprotein complexes spanned the entire thylakoid membrane and danced in the fluid lipid bilayer. With the luxury of time to think, I was the first to discuss thylakoid membranes in terms of the fluid mosaic model. 'This organization allows for rapid changes in the conformation and the distribution of the macromolecular complexes which are essential for the function and structure of the intricate chloroplast membrane' (Anderson 1975). Although knowledge of the structure of thylakoid proteins was still very limited, a new era for molecular organization had arrived. Achim Trebst (1974) focussed on the need for vectorial electron transport across the membrane, while Anderson (1975) assigned chloroplast components within the lipid–protein mosaic membrane model, emphasizing an asymmetry of function along thylakoid membranes and the dynamic and molecular aspects of thylakoid membrane stacking.

Figure 5. Bertil Andersson in the cold-room as usual in 1979.

Green gels and two-phase partition

Not surprisingly, photosynthesis researchers have always been intrigued by how the chlorophylls are arranged within photosynthetic membranes (cf. Kupke and French 1960; Philip Thornber 1975). Although investigators, beginning with Emil Smith in 1941, had used detergents or solvents to disrupt thylakoids and isolate pigment-proteins, the idea still persisted that not all the chlorophyll and carotenoids would be non-covalently bound to proteins. In 1966, resolution of two chlorophyll-proteins (Chl-proteins) was achieved by sodium dodecyl sulfate polyacrylamide gel electrophoresis (SDS PAGE); by 1975 at least 60% to 75% of chlorophyll had been resolved as Chl-proteins on green gels (cf. Thornber 1975). Finally, my extensive struggles to improve green non-denaturing gel procedures were rewarded, and I found that 90% of both chlorophylls and carotenoids were indeed associated with six different Chl-protein complexes (Anderson et al. 1978). The stage was set: departing from the Reading railway station after the 4th International Congress of Photosynthesis (UK), a young Swede bounded up and declared that he was coming to Australia to work with me when he had finished his PhD thesis! Bertil Andersson, having first convinced John Kendrew that he deserved a European Molecular Biology Organization (EMBO) Fellowship, arrived in Canberra in 1978 (see Figure 5 for a photograph of Bertil Andersson).

The earlier fractionation studies using differential centrifugation following detergent or mechanical fragmentation of thylakoid membranes had not allowed precise localization of the photosystems with respect to the appressed granal membrane domain. The elegant technique of aqueous polymer two-phase partition devised by Per-Åke Albertsson (1971), however, separated membrane fragments according to differences in membrane surface properties rather than size. Significantly, Bertil Andersson (1978) partitioned a granal fraction into right-side-out and inside-out vesicles derived from appressed and non-appressed membranes, respectively. Although PS II activity without the addition of artificial donors was greatly enriched in the appressed granal fraction, quantitation of PS II and PS I was still hampered by inactivation of photochemical activities incurred by thylakoid fragmentation (Hans-Erik Åckerlund et al. 1976).

Lateral heterogeneity of the photosystems along thylakoid membranes

Our joint aim was to compare the content of the main chlorophyll-protein complexes of the photosynthetic apparatus resolved by my improved ‘green gel’ method in Bertil’s membrane fractions derived from appressed and non-appressed membrane domains. Following Yeda press fragmentation of thylakoids, stroma thylakoids (Y-100) were separated from granal stacks (Y-40) by differential centrifugation, and enriched inside-out vesicles (B3) were isolated by aqueous polymer two-phase partition of the granal fraction (Y-40 fraction). Stroma thylakoid fractions were highly enriched in PS I complex together with some 10–20% of PS II and light-harvesting complex II (LHC II) complexes. By contrast, the grana-appressed vesicles were substantially depleted in PS I complex and enriched in PS II and LHC II (see color version of Figure 6 in section in the front of the issue; An-

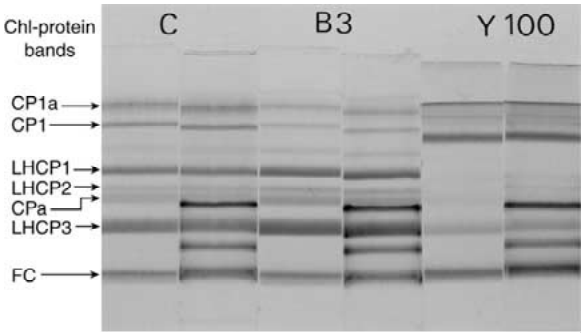

Figure 6. Chlorophyll-proteins of chloroplasts (C), appressed granal fraction (B3), and stromal fraction (Y-100) resolved by non-denaturing green gels (see text). CP1a (PS I complex with PS I Chl *a/b*-proteins), CP1 (PS I complex), Cpa (core PS II complex) and LHCP$^{1-3}$ with LHC II. The green gels were subsequently stained for haem groups to show a uniform distribution of 2 haem proteins Cyt *f* (33 kDa) and Cyt *b6* (18 kDa) (Anderson 1982). For a color version of this figure, see section in the front of the issue.

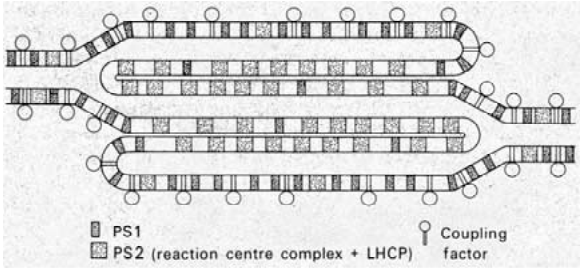

Figure 7. The original schematic model of the lateral heterogeneity in the distribution of PS II and PS I between appressed and nonappressed thylakoid membranes of higher plant chloroplasts which contain grana (Andersson and Anderson 1980). The unusual pattern of membrane folding was chosen as it seemed to explain how thylakoid membranes could be easily unstacked. Many textbooks reproduced this pattern for the following two decades, although I went back to a conventional pattern in 1984.

derson 1982). Allowing for some contamination of right-side-out vesicles in the appressed inside-out vesicles, we proposed that PS I is exclusively restricted to non-appressed granal domains, that is grana end-membranes, grana margins, and stroma thylakoids. Just to be safe, we did not entirely exclude PS I from the appressed region in our original model (Andersson and Anderson 1980) (Figure 7).

Thus, a lateral heterogeneity of distribution of the photosystems exists with PS II mainly, but not exclusively located in appressed membrane domains, while PS I is located in non-appressed stroma thylakoids, grana margins, and end membranes, together with ATP synthase as demonstrated by Kenneth Miller and Andrew Staehelin (1976; Staehelin et al. 1977). Our lateral heterogeneous distribution of the photosystems was different from that earlier proposed by Sane et al. (1970), where PS II and PS I were present together in the appressed granal membranes and PS I was also in the nonappressed membrane regions. We were delighted that our paper was speedily accepted in the Bioenergetics section for *Biochimica et Biophysica Acta*, then the prestigious journal for research in photosynthesis. Also, molecular organization was included for the first time at the splendid Vth International Photosynthesis Congress at Halkadiki (Greece), organized by dear George Akoyunoglou; the session attracted over 60 posters. To our astonishment and frustration, however, our concept of lateral heterogeneity presented by a symposium lecture and poster was largely ignored or met with horror. Neither Bertil nor I were allowed to speak at the two relevant Discussion series. An English chairman introduced one session by stating that there were two crazy ideas, fit only for the waste paper basket, which would not be discussed: lateral heterogeneity and Dan Arnon's concept of the photosystems operating in parallel rather than in series (see Arnon et al. 1981). Arnon continued until his death with his alternative scheme that PS II alone drives linear electron transport from water to ferredoxin and NADP$^+$, without the collaboration of PS I, whose role is limited to cyclic electron transport and cyclic photophosphorylation. The consequences of the exclusion of PS I from appressed granal domains focussed attention on grana stacks as a means to limit light excitation energy from PS II to PS I in higher plants (Anderson 1981).

Cytochrome *bf* complex was not placed in the original model (Figure 7). Our separate proof of the uniform distribution of cytochrome *bf* complex in both stacked and unstacked regions following extensive quantitation of cytochrome *f* and *b6* (Raymond Cox and Andersson 1981; Anderson 1982) is easily seen in haem-stained green gels (Anderson 1982) (Figure 6). Since cytochrome *bf* complex is located in both stacked and unstacked regions, the task of long-range transport of electrons between PS II and PS I is assigned to plastocyanin rather than plastoquinone (Anderson and Andersson 1982).

Why was lateral heterogeneity a heretical idea?

While it had been generally agreed that most PS II and LHC II were located in grana stacks, why was evidence of PS I exclusion from the grana partitions

initially such a heresy? This was due mainly to the two extreme models for the regulation of light energy distribution between the photosystems that were in vogue in the 1970s: continuous array (the lake model, see G. Wilse Robinson 1967) *versus* separate packages (the isolated puddle or the pond model). Continuous array models were strongly favored with a common light-harvesting antenna (LHC II) being shared between the photosystems (Gil Seely 1973; Ken Sauer 1975; Warren Butler 1977; Thornber et al. 1977; Thornber and Jim Barber 1979). Due to the ease of separation of most PS I from PS II, Boardman et al. (1978) went 'out on a limb' with a separate package model and a tentative proposal that Chl *b* had its own specific PS I antenna. Part of the difficulty of accepting lateral heterogeneity of the photosystems was the notion that prevailed throughout the 1970s, that PS I did not have its own Chl *b*. However, noting that the LHCP/core PS II ratios were very similar in appressed and non-appressed subchloroplast fragments, Andersson and Anderson (1980) suggested it was more reasonable to assume a close structural linkage of LHC II with PS II than to assume it was shared between the photosystems. John Mullet et al. (1980) were the first to show that 'native' PS I had its own antenna Chl-proteins, although they thought that the low amount of Chl *b* present was due to contamination with LHC II! Later Chl *b* was shown to be an intergral component of PS I, when Chl *a/b*-proteins were isolated from *Chlamydomonas* (Francis-André Wollman and Pierre Bennoun 1982) and spinach (Anderson et al. 1983).

The structural separation of linear and non-linear electron transport *in vivo* had long been considered (Sane et al. 1970; Arnon 1981). With the demonstration that the stroma-exposed grana margins indeed contain proteins (Andrew Webber et al. 1988; Albertsson 1995), linear electron transport is confined to both the appressed regions and margins of granal stacks thereby allowing limited redistribution of excitation energy, while stroma thylakoids perform cyclic photophosphorylation (Sane et al. 1970; Anderson 1982).

Epilogue

'Research is to see what everybody has seen and think what nobody has thought' (Albert Szent-Györgi, 1957). The significance of grana stacking for the function and temporary storage of non-functional PS IIs under stress, whereby most PS II dimers are located in the appressed membrane domain, separated from PS I in the stroma-exposed grana margins, has been and remains a major source for fascinating speculation.

Acknowledgment

I am very grateful to our incomparable photosynthesis historian Govindjee for his invitation and extensive help including the publication of green gels in color.

References

Åckerlund H-E, Andersson B and Albertsson P-Å (1976) Isolation of Photosystem II enriched membrane vesicles from spinach chloroplasts by phase partition. Biochim Biophys Acta 449: 525–535

Albertsson P-Å (1971) Partition of Cell Particles and Macromolecules (2nd edn). Wiley-Interscience, New York

Albertsson P-Å (1995) The structure and function of the chloroplast photosynthetic membrane – a model for domain organization. Photosynth Res 46: 141–149

Anderson JM (1975) The molecular organization of chloroplast thylakoids. Biochim Biophys Acta 416: 191–235

Anderson JM (1981) Consequences of spatial separation of Photosystems 1 and 2 in thylakoid membranes of higher plant chloroplasts. FEBS Lett 124: 1–10

Anderson JM (1982) Distribution of the cytochromes between the appressed membranes of grana stacks and stroma-exposed thylakoid regions. FEBS Lett 138: 62–66

Anderson JM and Andersson B (1982) The architecture of photosynthetic membranes: lateral and transverse organization. Trends Biochem Sci 7: 288–292

Anderson JM and Boardman NK (1964) Studies on the greening of dark-grown bean plants II. Development of photochemical activity. Aust J Biol Sci 17: 93–101

Anderson JM and Boardman NK (1966) Fractionation of the photochemical systems of photosynthesis. I. Chlorophyll contents and photochemical activities of particles isolated from spinach chloroplasts. Biochim Biophys Acta 112: 403–421

Anderson JM, Waldron JC and Thorne SW (1978) Chlorophyll-protein complexes of spinach and barley thylakoids: spectral characterization of six complexes resolved by an improved electrophoretic procedure. FEBS Lett 92: 227–233

Anderson JM, Brown JS, Lam E and Malkin R (1983) Chlorophyll *b*: an integral component of Photosystem I of higher plant chloroplasts. Photochem Photobiol 38: 205–210

Andersson B (1978) Separation of spinach chloroplast lamellae fragments by phase partition including the isolation of inside-out thylakoids. PhD Thesis, Lund University, Sweden

Andersson B and Anderson JM (1980) Lateral heterogeneity in the distribution of chlorophyll-protein complexes of the thylakoid membranes of spinach chloroplasts. Biochim Biophys Acta 593: 427–440.

Armond PA and Arntzen CJ (1977) Localization and characterization of Photosystem II in grana and stroma lamellae. Plant Physiol 59: 398–404

Arnon DI, Losada M, Whatley FR, Tsujimoto HY, Hall DO and Horton AA (1961) Photosynthetic phosphorylation and molecular oxygen. Proc Natl Acad Sci USA 47: 1314–1334

Arnon DI, Tsujimoto HY and Tang MS (1981) Proton transport in photo-oxidation of water: a new perspective on photosynthesis. Proc Natl Acad Sci USA 5: 2942–2946

Arntzen CJ, Dilley RA and Crane FL (1969) A comparison of chloroplast membrane surfaces visualized by freeze-etch and negative staining techniques; and ultrastructural characterization of membrane fractions obtained from digitonin-treated spinach chloroplasts. J Cell Biol 43: 16–31

Boardman NK and Anderson JM (1964) Isolation from spinach chloroplasts of particles containing different proportions of chlorophyll *a* and chlorophyll *b* and their possible role in the light reactions of photosynthesis. Nature 203: 166–167

Boardman NK Thorne SW and Anderson JM (1966) Fluorescence properties of particles obtained by digitonin fragmentation of spinach chloroplasts. Proc Natl Acad Sci USA 56: 586–593

Boardman NK, Anderson JM and Goodchild DJ (1978) Chlorophyll-protein complexes and structure of mature and developing chloroplasts. Cur Top Bioenerg 8: 35–109

Butler WL (1997) Energy distribution in the photosynthetic apparatus of plants. Brookhaven Symp Biol 28: 338–346

Butler WL and Kitajima M (1975) Energy transfer between Photosystem II and Photosystem I in chloroplasts. Biochim Biophys Acta 396: 72–85

Cox RP and Andersson B (1981) Lateral and transverse organisation of the cytochromes in the chloroplast thylakoid membranes. Biochem Biophys Res Commun 103: 1336–1342

Davenport HE (1952) Cytochrome components in chloroplasts. Nature 170: 1112–1114

Duysens LNM (1989) The discovery of the two photosynthetic systems: a personal account. Photosynth Res 21: 61–79

Duysens LMN and Amesz J (1962) Function and identification of two photochemical systems in photosynthesis. Biochim Biophys Acta 64: 243–260

Duysens LMN, Amesz J and Kamp BM (1961) Two photochemical systems in photosynthesis. Nature 190: 510–511

Emerson R and Arnold WA (1932) The photochemical reaction in photosynthesis. J Gen Physiol 16: 191–205

Emerson R and Lewis CM (1943) The dependence of quantum yield on *Chlorella* photosynthesis on wavelength of light. Am J Bot 30: 165–178

Emerson R and Rabinowitch E (1960) Red drop and role of auxiliary pigments in photosynthesis. Plant Physiol 35: 477–485

Emerson R, Chalmers RV and Cederstrand CN (1957) Some factors limiting the long-wavelength limit of photosynthesis. Proc Natl Acad Sci USA 43: 133–143

Gaffron H and Wohl K (1936) The theory of assimilation. Naturwissenschaften 24: 81–90; 103–107

Govindjee and Rabinowitch E (1960) Two forms of chlorophyll a with distinct photochemical functions. Science 132: 355–356

Govindjee, Ichimura S, Cederstrand C and Rabinowitch E (1960) Effect of combining far-red light with shorter wave light on the excitation of fluorescence in *Chlorella*. Arch Biochem Biophys 89: 322–323

Hill R (1965) The biochemists' green mansions: the photosynthetic electron-transport chain in plants. Essays Biochem 1: 121–151

Hill R and Bendall F (1960) Function of two cytochrome components in chloroplasts: a working hypothesis. Nature 186: 136–140

Hill R and Scarisbrick R (1951) The haemitin compounds of leaves. New Phytol 50: 98–111

Kautsky H, Appel W and Amman H (1960) Chlorophyll Fluorescenz und Kohlensäure Assimilation. Die Fluorescenz Kurve and die Photochemie der Pflanze. Biochem Z 332: 227–292

Kok B and Hoch G (1961) Spectral changes in photosynthesis. In: McElroy WD and Glass B (eds) Light and Life, pp 397–423. The John Hopkins Press, Baltimore, Maryland

Kupke DW and French CS (1960) Relationship of chlorophyll to protein and lipoids; molecular and colloidal solutions. Chlorophyll units. Encycl Plant Physiol V: 298–322

Lundegårdh H (1961) Response of chloroplast cytochromes to light and substrates. Nature 192: 243–248

Miller KR and Staehelin LA (1976) Analysis of the thylakoid outer surface: coupling factor is limited to unstacked membrane regions. J Cell Biol 68: 30–47

Mullet JE, Burke JJ and Arntzen CJ (1980) Chlorophyll proteins of Photosytem I. Plant Physiol 65: 814–822

Robinson GW (1967) Excitation transfer and trapping in photosynthesis. Brookhaven Symp Biol 19: 16–48

Sane PV, Goodchild DJ and Park RB (1970) Characterisation of chloroplast Photosystems 1 and 2 separated by a non-detergent method. Biochim Biophys Acta 216: 162–178

Sauer K (1975) Primary events and trapping In: Govindjee (ed) Bioenergetics of Photosynthesis, pp 116–175. Academic Press, New York

Seely GR (1973) Energy transfer in a model of the photosynthetic unit of green plants. J Theor Biol 40: 189–199

Singer SJ (1971) The molecular organization of biological membranes. In: Rothfield LI (ed) Structure and Function of Biological Membranes, pp 145–222. Academic Press, New York

Singer SJ and Nicolson GL (1972) The fluid mosaic model of the structure of cell membranes. Science 175: 720–731

Staehelin LA, Armond PA and Miller KR (1977) Chloroplast membrane organization at the supramolecular level and its functional implications. Brookhaven Symp Biol 28: 278–315

Szent-Györgi A (1957) Bioenergetics. Academic Press, New York

Thornber JP (1975) Chlorophyll-proteins: light-harvesting and reaction center components of plants. Annu Rev Plant Physiol 26: 127–158

Thornber JP and Barber J (1979) Photosynthetic pigments and models for their organization *in vivo*. Top Photosynth 3: 27–70

Thornber JP, Alberte RS, Hunter FA, Shiozawa JA and Kan KS (1977) The organization of chlorophyll in the plant photosynthetic unit. Brookhaven Symp Biol 28: 132–148

Trebst A (1974) Energy conservation in photosynthetic electron transport of chloroplasts. Annu Rev Plant Physiol 25: 423–458

Vernon LP, Shaw ER and Ke B (1966) A photochemically active particle derived from chloroplasts by the action of Triton X-100. J Cell Biol 241: 4101–4109

Webber AN, Platt-Aloia KA, Heath R and Thomson WW (1988) The marginal region of thylakoid membranes: a partial characterization by Tween 20 solubilization of spinach thylakoids. Physiol Plant 72: 288–297

Weier TE and Benson AA (1966) The molecular nature of chloroplast membranes. In: Goodwin TE (ed) Biochemistry of Chloroplasts, pp 91–113. Academic Press, London

Wessels JSC (1962) Separation of the two photochemical systems of photosynthesis by digitonin fragmentation. Biochim Biophys Acta 65: 501–504

Witt HT, Müller A and Rumberg B (1961) Experimental evidence for the mechanism of photosynthesis. Nature 191: 194–196

Wollman FA and Bennoun P (1982) A new chlorophyll-protein complex related to Photosystem I in *Chlamydomonas reinhardii*. Biochim Biophys Acta 680: 352–360