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Electron donation in Photosystem II

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The electron transfer resulting from illumination and dark storage of PS II has been studied using EPR signals from several electron carriers. The recombination of D^+ (Signal II) and Q_A^- formed by illumination occurred during dark storage at 77 K and was used to deplete reaction centres of D^+. The donor D was then shown to be oxidized in the dark by the S_2 state of the oxygen-evolving complex. A slow change which occurred during dark storage of PS II samples was detected using the power saturation characteristics of D. We interpret this effect on D to be an indirect result of a rearrangement of the manganese complex during long-term dark adaptation. A role for D in the stability, protection and perhaps initial manganese binding of the oxygen-evolving complex is suggested.

Introduction

The first step of electron transfer in the Photosystem II membrane-protein complex (PS II) involves the photo-oxidation of P-680, the reaction centre chlorophyll. The displaced electron is passed through the complex to the membrane pool of plastoquinone and the $P\text{-}680^+$ is reduced by an electron from water. A number of electron carriers have been identified in the PS II complex, most of which can be studied by EPR spectrometry. The electron-acceptor chain consists of a pheophytin [1] followed by two plastoquinones, Q_A and Q_B [2–4], although recent evidence has indicated that additional carriers may be present [5]. During electron donation to $P\text{-}680^+$, four turnovers of the reaction centre are required to release an oxygen molecule, with the electrons being sequentially removed from water in the oxygen-evolving complex. The oxygen-evolving complex therefore cycles through five oxidation states termed S_0–S_4 [6]. The S_0 and S_1 states are relatively stable in the dark with the S_0 state present in 25% of centres following illumination being slowly oxidised to S_1 during dark adaptation [7–9]. The intermediate states S_2 and S_3 are rapidly reduced to lower S states in the dark by reducing equivalents from the PS II electron acceptors. Oxygen is released during the conversion of S_4 to S_0. EPR studies have shown that the S_2 state exhibits a multiline EPR signal characteristic of manganese ions in a mixed valence binuclear or tetranuclear complex [10,11]. The manganese complex is probably the site of the accumulation of oxidising equivalents. The multiline signal can be induced by illumination of dark-adapted samples at 200 K as well as at room temperature.

Electron transfer from other PS II electron donors to Q_A can also be observed by EPR. An EPR species near $g = 2$ termed Signal II was the

Abbreviations: Mes, 4-morpholineethanesulfonic acid; Chl, chlorophyll; DCMU, 3-(3,4-dichlorophenyl)-1,1-dimethylurea; PS II, Photosystem II; *b*-559Hp, cytochrome *b*-559 high-potential form.

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first to be associated with electron transfer in PS II [12,13] and has been observed in both chloroplast membranes and subchloroplast particles. A number of forms of the Signal II species were found at room temperature and named according to their decay kinetics [14–20]: (II_d (dark stable), II_s (slow decaying) and II_{vf} (very fast transient observed during flash excitation). Signal II_{vf} probably represents Z^+, an intermediate between reaction centre and oxygen-evolving complex. Signal II_{vf} is converted to a more slowly decaying signal II_f on inhibition of the oxygen-evolving complex. Signal II_s and II_d probably represents D^+, a slow electron donor capable of oxidation of S_0 to S_1, but not directly involved in electron transfer between P-680 and the oxygen-evolving complex. Signal II can also be formed by illumination at cryogenic temperatures [21,22] and this form was attributed to Z^+ but positive identification with either Z^+ or D^+ was not achieved.

Photo-oxidation of both high- ($E_m = 380$ mV) and low- ($E_m = 80$ mV) potential cytochrome *b*-559 and a 1 mT wide radical tentatively assigned to a carotenoid or chlorophyll cation have also been observed following illumination at cryogenic temperatures [23–27]. None of these components have so far been assigned a role in normal PS II functions.

Recent studies [26,27] using an oxygen-evolving PS II preparation gave a simple picture of electron transfer in PS II at cryogenic temperatures. In samples poised in S_1, illumination at 77 K photo-oxidised *b*-559Hp whilst this was replaced at higher temperatures (160 K) by electron donation from the oxygen-evolving complex to give S_2. Therefore at 200 K the photooxidation of the multiline S_2 EPR signal replaced cytochrome oxidation. No significant changes in Signal II were observed. Changes in the 1 mT radical were only observed in strongly oxidising conditions where both forms of cytochrome *b*-559 were chemically oxidised.

The importance of experiments at cryogenic temperatures in research on PS II means that electron-transfer reactions at these temperatures must be defined. We report here the electron transfer properties of an oxygen-evolving PS II preparation at a variety of temperatures on illumination and during subsequent dark adaptation. Where possible we have added no oxidants, reductants or inhibitors in order to allow the basic characteristics of the system to be observed. Our results show a more complex pattern of electron transfer than suggested previously which depends on the temperature during or following illumination. These characteristics allowed further experiments to explore the relationship between the various electron donors of PS II. These suggest an important role for donor D in PS II and show its use as a monitor of oxygen-evolving complex function.

Experimental

Oxygen-evolving PS II was prepared by the method of Ford and Evans [28] from market spinach (*Spinacia oleracea*) or greenhouse grown pea (*Pisum sativum* var Feltham First). It was resuspended and stored at 77 K in 20 mM Mes, 5 mM $MgCl_2$, 15 mM NaCl and 20% glycerol (pH 6.3) (buffer A). Preparations with oxygen evolution rates of 400–1000 μmol O_2 per mg Chl per h were used. EPR spectrometry was performed at cryogenic temperatures using a Jeol X-band spectrometer with 100 kHz field modulation and an Oxford instruments liquid helium cryostat. 0.3 ml samples in 3 mm diameter calibrated quartz tubes were used. Chlorophyll concentrations of samples and EPR conditions are described in the text. Spectra were stored and data manipulations performed using DEC PDP 11 and Tektronix microcomputers.

Samples were prepared as follows unless specified otherwise: The PS II was placed in EPR tubes and dark adapted for 4 h before freezing in the dark. This treatment produces a uniform S_1 state in the oxygen-evolving complex [8] which is in the resting state designated in [9]. DCMU where used was added to a final concentration of 100 μM (1% ethanol) in darkness and after 20 min the samples were either frozen in the dark or illuminated for 1 min with a 1000 W white light source and frozen under illumination. Duplicate samples of each treatment were made. Freezing these samples under illumination produces S_2 in high yield. S_2 was also produced by illumination at 200 K of dark-adapted samples with a 1000 W projector for 2 min using an ethanol/solid CO_2 bath. Electrons

reach Q_A at this temperature and a multiple turnover of the reaction centre is possible allowing the pheophytin to be reduced in some centres. Illumination at 77 K was achieved using the same 1000 W light source with samples irradiated in liquid nitrogen in a silvered dewar. Only photoreduction of Q_A was seen under these conditions. The length of illumination was designed to produce a maximum charge separation.

Samples were stored at 77 K in a liquid nitrogen refrigerator with the samples either in or just above the surface of the liquid therefore maintaining a temperature close to 77 K. Thawing experiments were performed by rapidly thawing the sample in a water bath (290 K) in the dark followed by storage for the required time on ice in the dark before refreezing in liquid nitrogen. The minimum time between thawing and refreezing was about 25 s.

Results

Illumination and storage at 77 K

EPR signals from low potential cytochrome *b*-559$^+$ and D^+ (signal II) were present (Figs. 1a and 2a) in 4 h dark adapted PS II samples. During 5 min illumination at 77 K, high potential *b*-559 and some 1 mT radical were photooxidised (Figs. 1b and 2b) and the iron-quinone acceptor, Q_A was photoreduced (not shown).

On storage of these illuminated samples for several days at 77 K in the dark only slight loss of the *b*-559Hp signal (less then 20%) was observed (Fig. 1c), but more than 75% of the iron-semiquinone signal was lost (Fig. 3). A further illumination period at 77 K restored both the *b*-559Hp and the iron-semiquinone signal to their maximum level (Fig. 3). This indicates that a different electron donor species was acting in many centres during the second illumination. This donor was observed near $g = 2$ where storage at 77 K following the initial illumination caused a more than 50% loss of D^+ which could be restored by the second period of 77 K illumination (Fig. 2c and d). The 1 mT radical was relatively stable to dark storage at 77 K with much less than 20% being lost (Fig. 2c).

Incubation of illuminated samples at 200 K in the dark for 3 min gave no major changes in the size of EPR signals. This suggests that the electron donor during the second illumination was D and not Z as Z^+ would have been expected to form S_2 or recombine with Q_A^-.

It was concluded therefore that in the majority of reaction centres Q_A^- formed by 77 K illumination preferentially recombines with D^+ during dark storage at 77 K. D^+ was then photooxidised on the second illumination at 77 K in place of *b*-

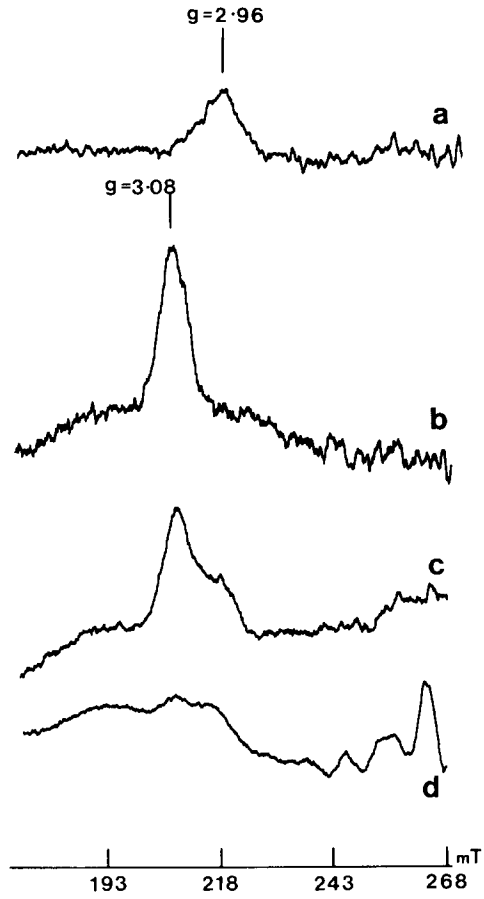

Fig. 1. EPR spectra at 14 K of the low-spin haem region of PS II samples: (a) 4 h dark-adapted sample showing low-potential cytochrome *b*-559; (b) 5 min illumination at 77 K minus 4 h dark-adapted showing high-potential cytochrome *b*-559; (c) sample kept at 77 K in the dark for 8 days following 77 K illumination; (d) 4 h dark-adapted sample illuminated at 200 K for 3 min. Chl concentration; a and b: 6 mg/ml; c and d: 5 mg/ml. EPR conditions: power, 4 mW; temperature, 14 K; modulation amplitude, 1 mT.

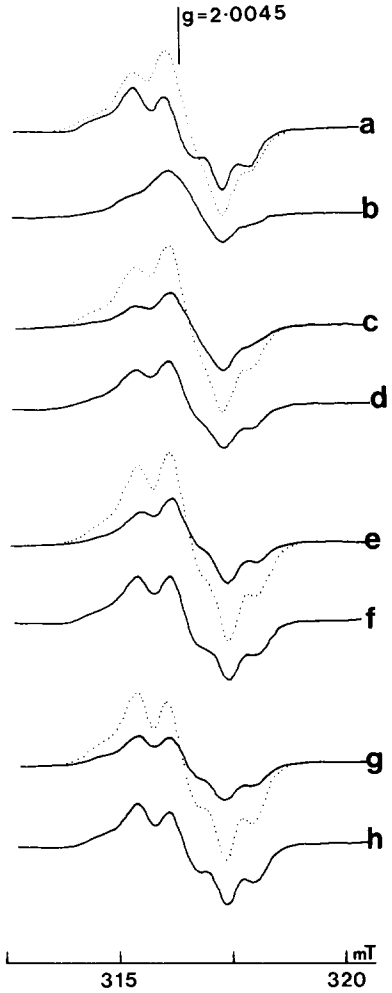

Fig. 2. EPR spectra at 15 K of the $g = 2$ radical region of PS II samples. Solid lines represent the spectrum in the dark and dotted lines represent the spectrum following 5 min illumination at 77 K. a, 4 h dark adapted; b, 77 K illuminated minus 4 h dark adapted; C, 77 K illuminated sample stored for 6 days at 77 K in the dark; d, 77 K illuminated minus c dark; e, 200 K illuminated sample stored for 6 days at 77 K in the dark; f, 77 K illuminated minus e dark; g, sample frozen under illumination and stored for 6 days at 77 K in the dark, h, 77 K illuminated minus g dark. Chl concentration, 5 mg/ml; EPR conditions, power 10 μw; modulation amplitude, 0.1 mT; and temperature, 15 K.

559Hp.

$$D^+ b\text{-}559Hp\ S_1/Q_A \xrightarrow{h\nu\ 77\ K} D^+ b\text{-}559Hp^+\ S_1/Q_A^-$$

$$\xrightarrow{77\ K\ dark} D\ b\text{-}559Hp^+\ S_1/Q_A$$

$$\xrightarrow{h\nu\ 77\ K} D^+ b\text{-}559Hp^+\ S_1/Q_A^-$$

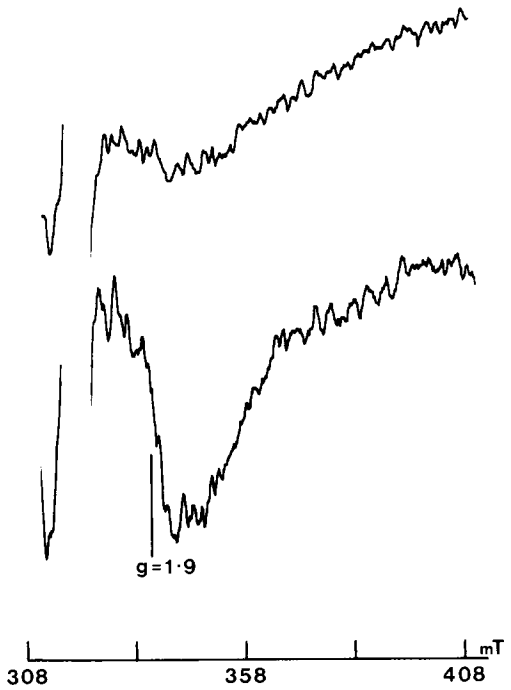

Fig. 3. EPR spectra of the iron-semiquinone signal. Upper trace: 77 K illuminated sample stored for 6 days at 77 K in the dark. Lower trace: sample after a further 5 min illumination at 77 K. Chl concentration as Fig. 2. EPR conditions power, 10 mW; temperature, 5.5 K; and modulation width, 1 mT.

Illumination at 200 K and 290 K

Illumination of 4 h dark-adapted PS II samples at 200 K or at 290 K (both with and without DCMU present) resulted in the formation of the S_2 multiline signal as the oxygen-evolving complex replaces *b*-559Hp as electron donor (Figs. 1d and 4) [26]. Some *b*-559Hp was photooxidised possibly showing that a proportion of the centres were defective in the oxygen-evolving complex. The S_2 signal was stable to dark storage at 77 K but again recombination between Q_A^- and D^+ occurred.

A second illumination at any temperature between 4.5 and 250 K would then restore both the D^+ (Fig. 2e–h) and iron-semiquinone signals. This photooxidation of D occurred despite the availability of *b*559Hp as a possible electron donor in many centres.

$$D^+ b\text{-}559Hp\ S_1/Q_A \xrightarrow{h\nu\ 200\ K} D^+ b\text{-}559Hp\ S_2/Q_A^-$$

$$\xrightarrow{77\ K\ dark} D\ b\text{-}559Hp\ S_2/Q_A$$

$$\xrightarrow{h\nu\ 4\text{–}250\ K} D^+ b\text{-}559Hp\ S_2/Q_A^-$$

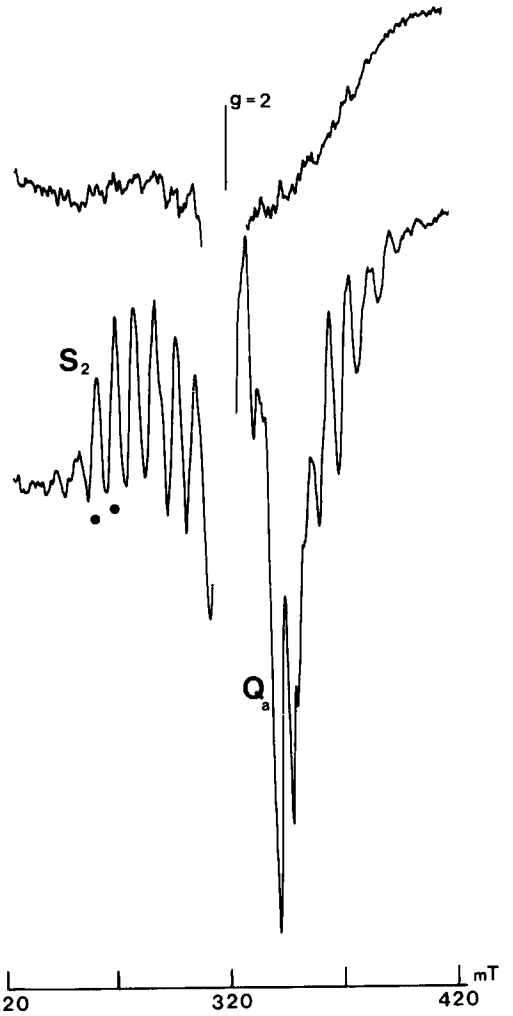

Fig. 4. EPR spectra showing the S_2 multiline and iron-semiquinone spectra. Upper trace: 4 h dark-adapted sample; lower trace; spectrum following 3 min illumination at 200 K. Chl as Fig. 2. EPR conditions: power, 50 mW; temperature, 9.5 K; and modulation width, 1 mT. ● shows the part of the signal used for size estimations.

In samples initially illuminated and stored at 77 K, photoxidation of the multiline S_2 signal occurred when the temperature of the second illumination was raised to 200 K, suggesting a competition between D and the oxygen-evolving complex in electron donation to P-680.

No loss of the S_2 multiline signal was seen when the second illumination was at 250 K indicating that D^+ photooxidation competes successfully with the S_2 to S_3 transition.

The spectra presented in Fig. 2 also show that as the illumination temperature increased so the amounts of 1 mT radical photooxidised decreased and that the 1 mT radical was usually almost absent in samples illuminated at 273 K. The amount of D^+ increased as the 1 mT radical decreased.

Effects of changes in the oxygen-evolving complex on the power saturation characteristics of D^+

The maximum level of D^+ was achieved by freezing under illumination at 273 K. The relationship between this and the lower levels seen in 4 h dark-adapted samples was difficult to quantitate because of power saturation differences between the signals. A comparison of the power saturation characteristics of D^+ in long-term

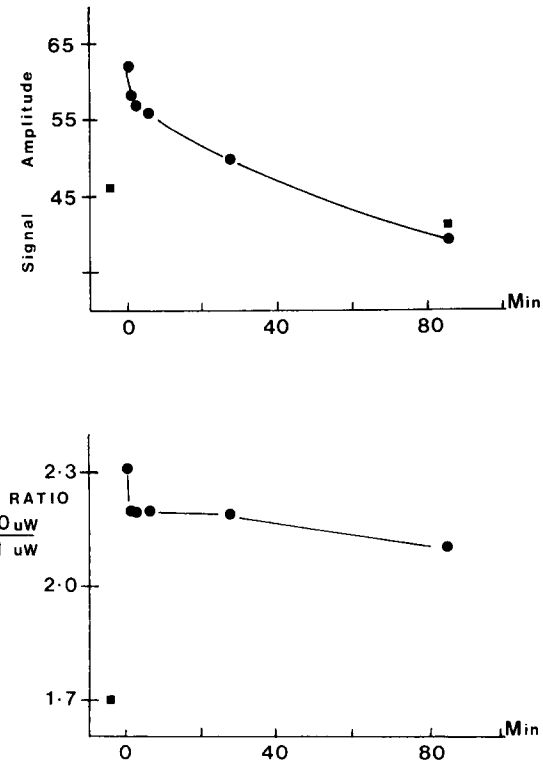

Fig. 5. Power saturation and size characteristics of D^+ on thawing an illuminated sample. Upper graph; signal II sizes measured at 10 μW (■) in a 4 h dark-adapted sample and (●) showing the rate of decay following illumination at 290 K. Lower graph: changes in the power saturation characteristics of D^+ during and following illumination at 290 K. (■) dark-adapted sample (●) ratio at times shown following 290 K illumination.

dark-dapted samples and samples frozen under illumination shows that the latter has a slightly higher power saturation profile indicating a faster relaxation time. This was as a result of interaction with manganese in S_2 or higher S states of the oxygen-evolving complex as shown by electron spin echo spectrometry [29]. A faster D^+ relaxation time was also indicated on production of S_2 by 200 K illumination. The link between higher S states and the power saturation effects on D were confirmed by the absence of an effect following 77 K illumination.

The decay of D^+ in samples frozen under illumination and then thawed was biphasic (Fig. 5). An initial rapid loss was perhaps due to reduction of any Z^+ present or recombination of Q_A^- and D^+. A slow reduction of D^+ was then observed with also a slow return towards the power saturation characteristics of a dark-adapted sample. The power saturation changes are illustrated by using the ratio of signal sizes at two power levels (Fig. 5 lower). The time-course of the slow reduction which occurred after Q_A^-, higher S states and *b*-559Hp$^+$ had decayed was similar to the well-documented loss of signal II_s [14,15]. The decay was also observed when the room-temperature illumination was done in the presence of DCMU to restrict electron flow and prevent randomisation of the S states. A difference was seen between the original 4 h dark-dapted samples and one illuminated and thawed for 4 h but mainly in the power saturation characteristics rather than in signal sizes. This suggests that the decrease in signal size and changes in power saturation reflect different processes. The power saturation ratio in Fig. 5 for the original dark-adapted sample could result from the low light conditions employed throughout the preparation of the PS II particles prior to the 4 h dark adaptation. This would greatly lengthen the effective dark adaptation period.

Effect of thawing after illumination at cryogenic temperatures

Fig. 6a and b show the maximum signal II (D^+) size obtained by illumination at 290 K compared to that in a 4 h dark-adapted sample. Samples thawed immediately following 77 K illumination and then refrozen in the dark returned rapidly to the 4 h dark adapted D^+ level. *b*559Hp$^+$, 1 mT

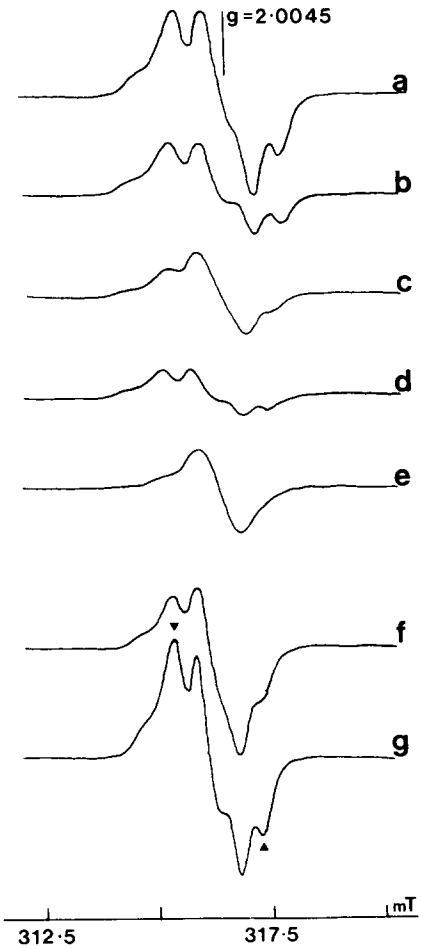

Fig. 6. EPR spectra of the $g = 2$ region at 9 K showing depletion of signal II on dark storage at 77 K. (a) 4 h dark-adapted sample illuminated for 1 min at 290 K and frozen under illumination showing the maximum D^+ signal; (b) 4 h dark-adapted sample; (c) sample illuminated at 77 K for 5 min and stored at 77 K for 1 week in the dark; (d) sample in (c) thawed in the dark and refrozen showing depletion of D^+; (e) sample in (d) illuminated at 77 K and stored for 1 week at 77 K in the dark; (f) 4 h dark-adapted sample illuminated at 200 K and stored for 1 week in the dark; (g) sample in (f) after thawing in the dark for 5 min and refreezing, showing increased D^+. ▲ and ■ show the peaks used in size measurements. Chl concentration, 4 mg/ml; EPR conditions, power, 1 μW; temperature, 9 K; and modulation width, 0.1 mT. Signals f and g are expanded 3 times.

radical and Q_A^- all decayed rapidly with a half life of less than 30 s.

Samples illuminated at 77 K and given 8 days dark adaptation at 77 K (Fig. 6c) to allow Q_A^-/D^+ recombination showed the 1 mT radical and reduced amounts of D^+ compared to the original

sample. *b*-559Hp$^+$ was also present in this sample together with a small amount of Q_A^-. On thawing at 273 K the 1 mT, Q_A^- and *b*-559Hp$^+$ signals were rapidly lost leaving the lower level of D^+ shown in Fig. 6d. This experiment confirms that following the illumination at 77 K, *b*-559Hp$^+$ does not restore D^+ lost by recombination with Q_A^-.

The rapid reduction of 1 mT radical and *b*559Hp$^+$ occurs when most centres have no Q_A^-. This suggests that this reduction occurs by rapid equilibrium between these and endogenous reductants. No marked changes in low-potential *b*-559 were observed.

When the sample shown in Fig. 6d was illuminated at 77 K the D^+ was not restored but *b*-559Hp and 1 mT radical were photooxidised to the same extent as in the original dark sample. A period of dark adaptation at 77 K then produced after thawing a sample with an even greater depletion of D^+ (Fig. 6e). This shows that D is not available for oxidation at 77 K after thawing, the preferred electron donor being *b*-559Hp. As shown earlier, illumination at 77 K or 200 K following dark storage but before thawing restored the D^+ lost by recombination with Q_A^-. This D^+ was retained on thawing the sample.

Illumination at 290 K of samples depleted of most of their D^+ restored all the D^+ giving identical levels to illuminated non-depleted samples.

Oxidation of D by S_2

Fig. 6f shows the $g = 2$ region of a 4 h dark-adapted sample illuminated at 200 K to photooxidise the multiline S_2 signal and then stored at 77 K for 1 week in the dark to deplete the D^+ by recombination with Q_A^-. Fig. 6g shows the sample following 1 min thawing and incubation at 273 K in the dark followed by freezing to 77 K. A clear increase in D^+ was observed which occurred in parallel with loss of the S_2 multiline signal. This oxidation of D^+ also occurred in samples illuminated at 290 K and then stored at 77 K prior to thawing.

Fig. 7 shows the formation of D^+ and loss of S_2 on thawing of depleted samples and the time-course for this reaction. In Fig. 7 the decay of the S_2 signal does not exactly match the time-course of appearance of the D^+ signal. It

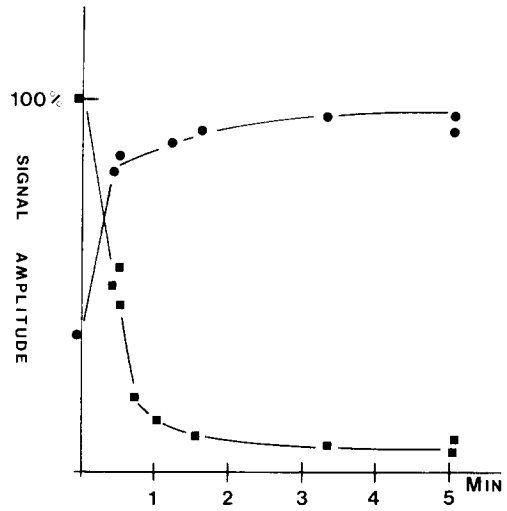

Fig. 7 Rate of formation of Signal II (D^+) (●) and loss of the S_2 multiline signal (■) on thawing Signal II-depleted samples. The samples were depleted of D^+ by illumination at at 77 K, storage for 1 week at 77 K and then thawing at 273 K. A second illumination was then given at 200 K and the samples stored at 77 K for 2 weeks in the dark. The sample was then thawed in the dark at 273 K and refrozen at the times shown. D^+ measured at 1 μW, S_2 measured at 10 mW.

suggests that the fast phase of reduction occurs by reaction with D and that a slower reduction occurs in centres still containing D^+ and Q_A^-. A slower reduction of the S_2 state was observed in samples with higher levels of D^+ obtained by illumination at 290 K than in samples depleted of D^+.

Discussion

The results show a complex pattern for both the forward and recombination reactions of PS II in which the components involved depend on the experimental temperature. Many experiments on PS II use cryogenic temperatures in order to restrict or slow reactions. In addition many EPR signals are only observed at less than 20 K and some laboratories also use PS II preparations which are made and then stored at cryogenic temperatures before use. Therefore some of the electron-transfer properties found in the present study have important implications for experiments on the PS II reaction centre. In particular the data showing that storage at 77 K after illumination

produces a changed redox state in some PS II components is important. This means that sample condition prior to experiment should be more carefully measured as illumination especially of frozen preparations does induce changes which are not reversed by dark adaptation at any temperature.

The relationship between *b*-559Hp and D as electron donors at cryogenic temperatures was complex. The results can be explained if during oxidation D undergoes a protonation/deprotonation or other similar exchange reaction. *b*-559Hp could then control the availability of D by for example affecting deprotonation.

Cytochrome *b*-559Hp is only photooxidised at cryogenic temperatures or under special conditions such as in the presence of oxygen-evolving complex deactivating (ADRY) reagents [30]. These and the present results suggest that an equilibrium exists between *b*-559Hp and mobile redox components in the membrane such as plastoquinone. *b*-559Hp could oxidise plastoquinol and then the low potential cytochrome could oxidise the semiquinone in a cycle which would operate around PS II when irreversible electron donation from the oxygen-evolving complex was not functional. This could be a protective mechanism which gives a much weaker oxidant than P-680$^+$ or the intermediates in the oxygen-evolving complex, thereby eliminating dangerous reactions between plastoquinone radicals and oxygen which could damage the membrane.

D has been linked with the oxidation of S_0 to S_1 [8] and the deactivation of S_2 and S_3 [14,31]. We have now confirmed the role in deactivation by monitoring the rise of D^+ and the loss of S_2 EPR signals. The oxidation of D appears to be the preferred route of deactivation as loss of S_2 was slower in centres containing D^+. Whilst this manuscript was in preparation two groups [32,33] have reported similar results.

The power saturation of D^+ is affected by the higher S states [29]. However, we have observed a power saturation change which occurred on a long time scale. This change was probably caused by a structural rearrangement or change in oxidation state of manganese in the oxygen-evolving complex. It is possible that generation of S_0 could occur under the illumination conditions used. To explain the results S_0 would have to increase the relaxation rate of D^+ and the return to lower power saturation levels would then accompany the oxidation of S_0 to S_1 and the reduction of D^+. The D^+ remaining would be in centres in the S_1 state. If S_0 was not generated by the illumination conditions then a slow rearrangement of the oxygen-evolving complex must occur when it is in the S_1 state in the dark.

The decay of D^+ also displayed a time-course similar to the slow oxygen consumption by the oxygen-evolving complex observed by Beck et al. [9]. This was explained as a conversion of the oxygen-evolving complex from an active to a resting state. In a previous paper we observed that there was a difference between the multiline signal formed by 200 K illumination of dark-adapted samples and that formed by room temperature illumination [34]. These phenomena could be linked to a slow change of the S_1 state in the dark. This could occur by reactions such as protonation and deprotonation or perhaps by disproportionation of the proposed Mn^{III} atoms in the dimeric or tetrameric complex [35] to give Mn^{II} and Mn^{IV}. In the models of manganese oxidation states during oxygen-evolving complex function [35] this would still result in the normal S-state turnover on illumination. D^+ was previously observed on the first illumination at cryogenic temperatures in some PS II preparations [23,36]. This may indicate that the isolation procedure can affect the stability of the D state.

The properties of D are therefore consistent with it being oxidised via either P-680$^+$ or decay of S states and being reduced by S_0. This role suggests that the oxygen-evolving complex has an efficient mechanism to avoid the long term presence of S_0 perhaps because of a loss of manganese which may occur. D^+ could also be involved in the initial poising of the oxygen-evolving complex by oxidising the Mn^{II} to Mn^{III} and stabilising the complex.

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