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OXIDATION-REDUCTION PROPERTIES OF THE ELECTRON ACCCEPTORS OF PHOTOSYSTEM II

II. REDOX TITRATION AT VARIOUS pH VALUES OF THE FLASH-INDUCED FORMATION OF C550 IN *CHLAMYDOMONAS* PHOTOSYSTEM II PARTICLES LACKING THE SECONDARY QUINONE ELECTRON ACCEPTOR

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Redox titrations of the flash-induced formation of C550 (a linear indicator of Q^-) were performed between pH 5.9 and 8.3 in *Chlamydomonas* Photosystem II particles lacking the secondary electron acceptor, B. One-third of the reaction centers show a pH-dependent midpoint potential ($E_{m,7.5}$) = −30 mV) for redox couple Q/Q^-, which varies by −60 mV/pH unit. Two-thirds of the centers show a pH-independent midpoint potential (E_m = +10 mV) for this couple. The elevated pH-independent E_m suggests that in the latter centers the environment of Q has been modified such as to stabilize the semiquinone anion, Q^-. The midpoint potentials of the centers having a pH-dependent E_m are within 20 mV of those observed in chloroplasts having a secondary electron acceptor. It appears therefore that the secondary electron acceptor exerts little influence on the E_m of Q/Q^-. An EPR signal at *g* 1.82 has recently been attributed to a semiquinone-iron complex which comprises Q^-. The similar redox behavior reported here for C550 and reported by others (Evans, M.C.W., Nugent, J.H.A., Tilling, L.A. and Atkinson, Y.E. (1982) FEBS Lett. 145, 176–178) for the *g* 1.82 signal in similar Photosystem II particles confirm the assignment of this EPR signal to Q^-. At below −200 mV, illumination of the Photosystem II particles produces an accumulation of reduced pheophytin (Ph^-). At −420 mV Ph^- appears with a quantum yield of 0.006–0.01 which in this material implies a lifetime of 30–100 ns for the radical pair P-680$^+Ph^-$.

Introduction

Nugent et al. [1] have recently shown that illumination at low temperature or reduction by dithionite in the dark of *Chlamydomonas* PS II particles [2] produces a broad EPR signal centered at *g* 1.82. A similar signal has been observed in bacterial reaction centers for the reduced primary electron acceptor [3,4]. The signal observed in PS II particles was attributed to Q^-, the reduced primary electron acceptor of PS II, and was suggested to arise from a plastosemiquinone-iron complex [1]. In this paper we show that the redox properties of C550 in *Chlamydomonas* PS II particles are similar to those observed by Evans et al.

Abbreviations: C550, an absorbance shift which acts as a linear indicator of Q^-; DCMU, diuron, 3-(3,4-dichlorophenyl)-1,1-dimethylurea; Hepes, 4-(2-hydroxyethyl)-1-piperazineethanesulfonic acid; Mes, 4-morpholineethanesulfonic acid; P-680, primary electron donor chlorophyll of PS II; PS, photosystem; Ph, pheophytin.

[5] for the g 1.82 EPR signal, thus confirming its assignment to the reduced primary quinone acceptor.

With but one exception [6] redox titrations of fluorescence quencher Q_H [7,8], of C550 [9] and of low-temperature oxidation of cytochrome *b*-559 [10], all indicators of the redox state of Q [11–13], show a -60 mV/pH unit dependence for the midpoint potential (E_m) of redox couple Q/Q^-. Knaff [10] has shown that this pH dependence persists up to pH 8.9, above which the E_m becomes independent of pH. A proton is thus taken up following the reduction of Q at pH values less than 8.9, the pK of the site which becomes protonated in the presence of Q^-. In this paper we show that between pH 5.9 and 8.3 in *Chlamydomonas* PS II particles the E_m of redox couple Q/Q^- is pH independent in two-thirds and pH dependent in one-third of the reaction centers. These data indicate that the environment of Q in the pH-independent centers has been modified such as to stabilize the plastosemiquinone anion, Q^-.

Wraight [14] has reported that in bacterial reaction centers the redox state of the primary acceptor, Q_A, modifies the affinity of the secondary quinone acceptor for its binding site. This conclusion is based on the decreasing E_m for Q_A/Q_A^-, observed with increased occupancy of the secondary quinone-binding site. The redox behavior of the *Chlamydomonas* PS II particles, lacking the secondary electron acceptor, and chloroplasts are compared to see if such Q-modulated binding of secondary acceptor, B, exists in PS II.

Light-induced absorbance changes were studied at redox potentials below the E_m of Q/Q^- in a search for alternate or more primary electron acceptors than Q in PS II. Measurements at -420 mV show the light-induced reduction of pheophytin (Ph), the intermediate electron acceptor of PS II. Quantum yield measurements of formation of Ph^- in PS II particles suggest that the lifetime of radical pair $P\text{-}680^+\text{-}Ph^-$ may be an order of magnitude greater than the 2–4 ns reported by Klimov et al. [15].

Materials and Methods

Intact PS II particles were prepared as described in Ref. 2. These were suspended in either 50 mM Mes-KOH for pH 5.9 and 6.6 or in 50 mM Hepes-KOH for pH 7.5 and 8.3. The final Triton X-100 concentration was approx. 0.003%.

Redox titrations were performed as described in the previous paper. The redox mediators used were: 1,2-naphthoquinone, 1,4-naphthoquinone, duroquinone, 2-hydroxy-1,4-naphthoquinone and anthraquinone-2-sulfonate, all at 20 μM; and phenazine methosulfate and pyocyanin, both at 5 μM. The reductive and oxidative titrants were degassed solutions of 40 mM $Na_2S_2O_4$ in 0.1 M Tris-HCl, pH 9.0, and 0.1 M $K_3Fe(CN)_6$, respectively.

All absorbance change measurements are reported in terms of $\Delta I/I$ for a 16 mm light path. All measurements were taken at ambient temperature (approx. 22°C). All redox potentials are reported relative to the standard hydrogen electrode.

Results

Redox titration of C550, pH dependence of E_m

We undertook the titration of Q/Q^- using intact PS II particles isolated from *Chlamydomonas reinhardii* [2] which have been shown previously to

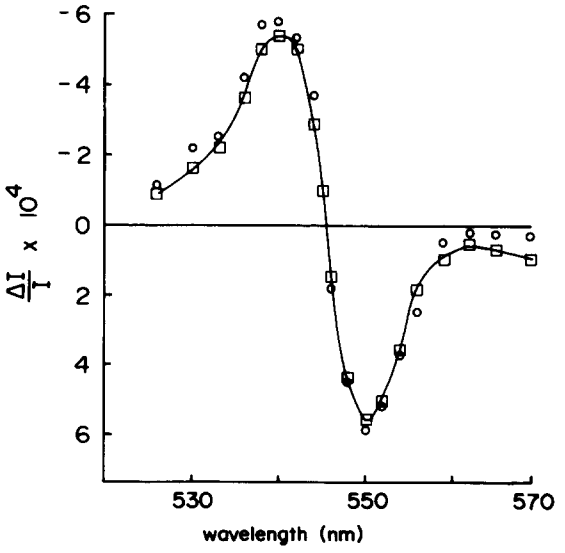

Fig. 1. Spectrum of C550 in intact *Chlamydomonas* PS II particles (8 μg Chl/ml) at approx. +300 mV. Detection at 400 μs (○) and 3 ms (□) after a 1 μs actinic flash exciting 80% of the PS II centers. The particles are suspended in 50 mM Hepes-KOH, pH 7.5, 0.1 mM $K_3Fe(CN)_6$ and 1 mM $K_4Fe(CN)_6$.

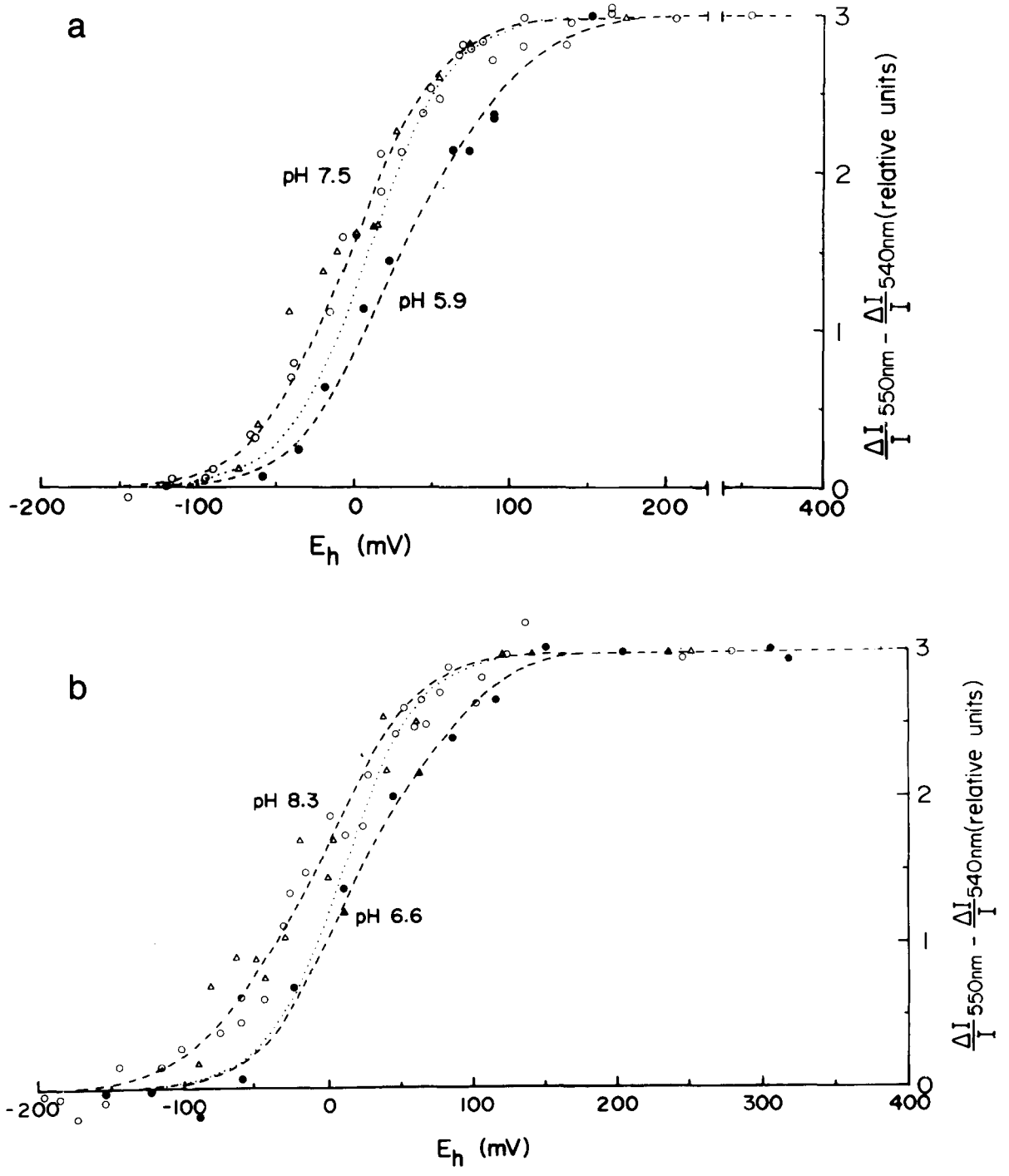

Fig. 2. Redox titration in PS II particles of saturating flash-induced formation of C550 ($\Delta I/I$(550 nm)$-\Delta I/I$(540 nm)). The C550 was detected 10 ms after the actinic flash. The reductive (○, ●) and oxidative △, ▲) titrations were performed in separate experiments and are normalized to the same maximum C550 signal. Theoretical Nernst curves ($n = 1$, $E_m = +10$ mV) are indicated by the dotted lines. Chlorophyll concentration, 12 μg/ml. (a) Titrations in 50 mM Hepes-KOH, pH 7.5 (○, △), and 50 mM Mes-KOH, pH 5.9 (●, ▲). Dashed line (pH 7.5): sum of two theoretical Nernst curves ($n = 1$) with one-third having an E_m of -30 mV and two-thirds having an E_m of $+10$ mV. Dashed line (pH 5.9): sum of two theoretical Nernst curves ($n = 1$) with one-third having an E_m of $+85$ mV and two-thirds having an E_m of $+10$ mV. (b) Titrations in 50 mM Hepes-KOH, pH 8.3 (○, △), and 50 mM Mes-KOH, pH 6.6 (●, ▲). Dashed line (pH 8.3): sum of two theoretical Nernst curves ($n = 1$) with one third having an E_m of -70 mV and two-thirds having an E_m of $+10$ mV. Dashed line (pH 6.6): sum of two theoretical Nernst curves ($n = 1$) with one-third having an E_m of $+80$ mV and two-thirds having an E_m of 0 V.

lack the secondary quinone electron acceptor, B. We chose to titrate C550 as a linear measure of $[Q^-]$ [13] rather than relying on the fluorescence yield, which we found to be subject to quenching by the quinone mediators. Fig. 1 shows the flash-induced absorbance changes detected between 520 and 570 nm in the PS II particles at positive potential and which show the characteristicc line shape described by Van Gorkom et al. [16] at room temperature.

The redox couple Q/Q^- was titrated by measuring the difference in the flash-induced $\Delta I/I$ at the maxima and minima of C550 ($\Delta I/I$(550 nm) $-\Delta I/I$(540 nm)) as a function of redox potential. These titrations (Fig. 2a and b) were performed at four different pH values between 5.9 and 8.3. Reductive (○, ●) and oxidative (△, ▲) titrations were usually performed in separate experiments. While normalized to the same maximum C550 signal in Fig. 2, the final levels of the oxidative titrations were always about 1/3 lower than the starting points of the reductive titrations. This difference was principally due to activity losses at low potentials. Nonetheless, the close agreement in the E_m indicates that the titrations were reversible and were performed under conditions close to equilibrium. The only exception was at pH 5.9 where considerable hysteresis was observed between the reductive and oxidative experiments, with the E_m of the latter appearing at potentials up to 100 mV more positive than the reductive E_m. Equilibration for the oxidative titration at pH 5.9 was extremely slow. Otherwise, equilibration times were normally 10–15 min for the reductive and about 20 min for the oxidative titrations after the addition of titrant. The latter titrations generally showed greater scatter in the experimental points. While less satisfactory as a monitor of the redox state of Q (the oxidized redox mediators quench fluorescence), the fluorescence yield at pH 7.5 did titrate with an E_m of approx. 0 V (in collaboration with Dr. J. Bowes, unpublished data, not shown) as did C550 (Fig. 2a). These titrations also showed a constant fluorescence yield between −160 and −350 mV.

As shown in Figs. 2 and 3a, the midpoint potentials show only a slight pH dependence (−15 to −20 mV/pH unit) which is linear over the pH range investigated. This small yet linear dependence (Fig. 3a) implies that the reaction centers are heterogeneous and either (a) show a continuum of pK values for the proton-binding site associated with Q reduction (an unlikely possibility), or (b) are of two types, those that are pH dependent and those that are pH independent. As the −15 to −20 mV/pH unit dependence shown in Fig. 3a corresponds to 25–33% of the −60 mV/pH unit dependence for one proton bound per Q^-, one might predict that 25–33% of the reaction centers are pH dependent and the remainder are pH independent.

The redox titration wave of C550 at pH 7.5 (Fig. 2a) is somewhat wider than the theoretical Nernst curve ($n = 1$, dotted line). At pH 5.9, 6.6 and 8.3 (Fig. 2a and b) the experimental curves are wider still, with a low-potential trailing edge at pH 8.3 and a high-potential trailing edge at pH 5.9 and 6.6. These titration curves were found to fit

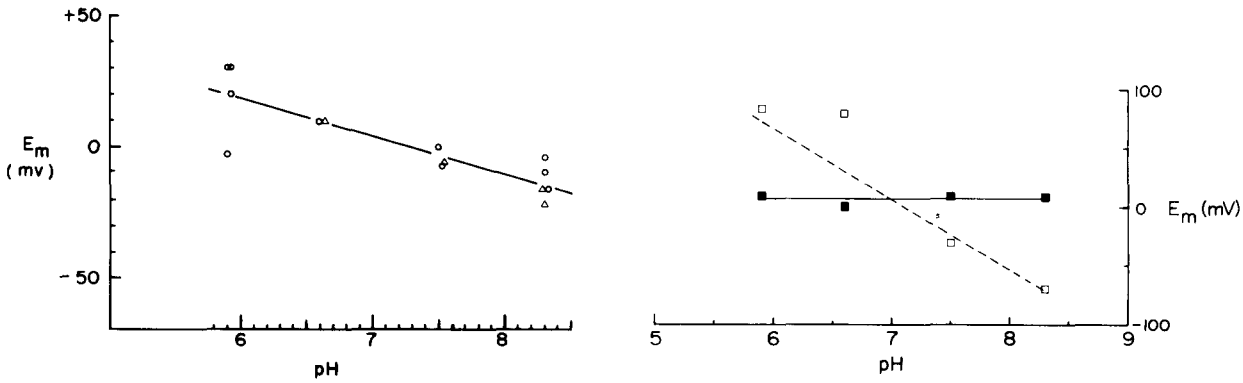

Fig. 3. (a) pH dependence of the E_m of the flash-induced formation of C550 (Fig. 2) considering the data as corresponding to homogeneous titration curves. Oxidative (△) and reductive (○). (b) pH dependence of the E_m of the component Nernst curves which contribute to the dashed lines of Fig. 2: (□) the one-third component; (■) the two-thirds component.

well (Fig. 2, dashed lines) to a sum of two $n = 1$ Nernst curves, one of which represents two-thirds of the centers with a pH-independent E_m of approx. 10 mV (Fig. 3b); the other represents one-third of the centers and shows an approx. −60 mV/pH unit dependence of the E_m (Fig. 3b). The latter centers are thus responsible for the low-potential trailing edge at pH 8.3 and the high-potential trailing edge at pH 5.9 and 6.6.

Light-induced absorbance changes at intermediate and low potentials

At −175 mV (pH 7.5), where Q is entirely reduced, flash excitation produces small absorbance changes as shown in Fig. 4. These do not accumulate during a series of flashes and their half-life is approx. 200 ms. They bear no resemblance to the blue difference spectrum of $Q^- - Q$, previously measured on this material [17], further confirming the full reduction of Q in the dark at this potential.

As the potential is lowered further, a new signal appears, the amplitude of which on a first saturating flash continues to increase until the potential reaches −250 mV, beyond which there is no further increase. At approx. −200 mV and below the signal amplitude increases with each flash in a series of flashes (e.g., 5 Hz). The half-life of this species is of the order of 20 s at −420 mV (pH 7.5). The spectrum of this species is shown in Fig. 5 and closely resembles that reported by

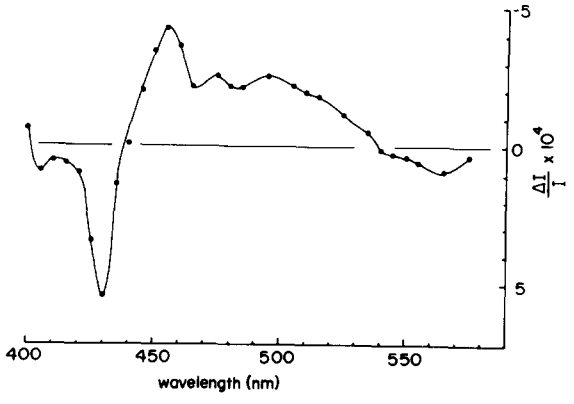

Fig. 4. Flash-induced absorbance changes in PS II particles at pH 7.5 measured at 10 ms after a 1 μs saturating flash at approx. −175 mV. Chlorophyll concentration 12 μg/ml.

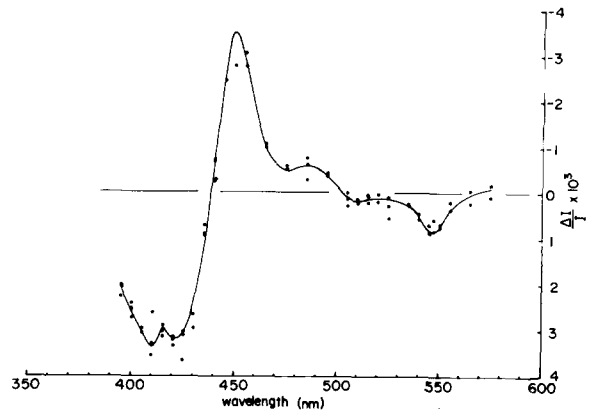

Fig. 5. Flash-induced absorbance changes in PS II particles measured 10 ms after the last of ten (5 Hz) 1 μs saturating flashes at approx. −355 mV. The first flash shows the same spectrum. Chlorophyll concentration 12 μg/ml.

Klimov et al. [15] for reduced pheophytin. We estimate the quantum yield of formation of this species at −420 mV to be 0.006–0.01 based on a difference extinction coefficient at 425 nm of 27 $mM^{-1} \cdot cm^{-1}$ obtained from the spectra of Klimov et al. [18] and Fujita et al. [19] with a difference extinction coefficient of 32 $mM^{-1} \cdot cm^{-1}$ at 685 nm. This estimation is also based on a ratio of 50 chlorophyll molecules per PS II reaction center in the particles and a quantum yield of 1 for photoreduction of Q in this material at +400 mV [2]. The reduction kinetics of $P\text{-}680^+$ by the secondary donor ($t_{1/2} = 5$–10 μs) [17] are probably the same at these low potentials as under more oxidizing conditions where the secondary donor is nonetheless reduced prior to photoexcitation [17]. Based on the quantum yield and the rate of reduction of $P\text{-}680^+$ by the the secondary donor, one would estimate the $P\text{-}680^+\text{-}Ph^-$ recombination time to be of the order of 30–100 ns at −420 mV in this material. This estimate is at least an order of magnitude greater than that reported by Klimov et al. [15] and Shuvalov et al. [20] (2–4 ns).

Discussion

Midpoint potential of Q/Q^- in PS II particles and chloroplasts. E_m of C550 and g 1.82 EPR signal

Nugent et al. [1] have reported an EPR signal at *g* 1.82 in *Chlamydomonas* PS II particles which

appeared upon either reduction with dithionite or illumination at low temperature. By analogy with bacterial reaction centers which display a similar signal, this species was attributed to the plasto-semiquinone-iron complex which comprises Q^-. Using *Chlamydomonas* PS II particles, both we [21] and Evans et al. [5] have recently reported largely pH-independent midpoints for C550 ($E_{m,7.5} = 0$ V) and for the g 1.82 EPR signal ($E_{m7} \simeq -10$ mV), respectively. The titration results for C550 are further developed here and confirm the attribution of the EPR signal to Q^-. Further support for this assignment has also been obtained through Mössbauer studies [22] where a high-spin Fe^{2+} has been detected having Mössbauer parameters similar to those observed for the primary quinone acceptor (an iron-ubiquinone complex) in bacterial reaction centers. Detection of a doublet in the EPR spectrum at 7 K for centers in state P-680Ph^-Q^- also supports the assignment of an iron-quinone complex to Q [23].

The $E_{m,7.5}$ of approx. 0 V observed here for C550 in PS II particles was also observed for the absolute and variable fluorescence yields (in collaboration with Dr. Jane Bowes, unpublished data). This titration of a fluorescence quencher, undoubtedly Q_H, indicates that Q_H and Q are the same species in agreement with other titrations performed under different conditions [11–13].

The fluorescence yield in these titrations was constant between -160 and -350 mV which would suggest the absence of fluorescence quencher, Q_L, ($E_{m,7} \simeq -250$ mV) observed in chloroplasts. This observation is similar to that of Bowes et al. [11] in particles isolated from *Phormidium laminosum*. While unlikely, fluorescence quenching by mediators could have obscured the quenching by Q_L.

pH dependence of $E_m(Q/Q^-)$

The midpoint potential for Q/Q^-, as indicated by redox titrations of Q_H [7,8], of C550 [9] and of low-temperature oxidation of cytochrome *b*-559 [10], shows, with one exception, a pH dependence of -60 mV/pH unit. Horton and Croze [6] have reported a much weaker dependence for Q_H (approx. -23 mV/pH unit) similar to what we observe here for C550 (-15 to -20 mV/pH unit, pH 5.9–8.3) if we ignore the heterogeneity of the titration curves (Figs. 2 and 3). This weak dependence cannot be explained by a pK in the pH range studied as the E_m and the pH appear to be linearly related throughout. The reaction centers thus appear to be heterogeneous in their pH dependence. A continuum of pK values for the site which becomes protonated upon formation of Q^- is unlikely. More likely is that some centers show a normal pH dependence (-60 mV/pH unit) while others show none at all. This hypothesis is borne out by a deconvolution of the redox titration curves (Figs. 2 and 3). A reasonable fit is obtained by attributing a pH dependence of -60 mV/pH unit to one-third of the centers and no pH dependence to the remaining two-thirds.

Knaff [10] has shown that the E_m of Q/Q^- in broken chloroplasts is pH dependent (-60 mV/pH unit) below pH 8.9 and pH independent above, where the E_m remains constant at -130 mV. The pK of the site which becomes protonated upon formation of Q^- is thus 8.9.

Fowler and Kok [24] showed that on the seconds time scale at pH 7, no proton uptake occurs on the acceptor side of PS II in chloroplasts following a saturating flash in the presence of DCMU. The E_m of redox couple Q/Q^- and its pH dependence are independent of the presence or absence of DCMU [6,7,9,25].

Proton uptake associated with the reduction of Q is thus a slow process (much greater than 1 s) in the presence of DCMU and probably in its absence as well. The spectroscopic measurements of Van Gorkom [26] in the presence and absence of DCMU are consistent with this conclusion in that they indicate that on the seconds time scale no direct protonation of Q^- occurs. However, there is no evidence to indicate which site (Q^- or otherwise) is protonated on the minutes time scale of the redox titrations.

The pH-independent E_m observed here for C550 in two-thirds of the centers (approx. $+10$ mV) and observed by Horton and Croze [6] for Q_H ($E_{m,7} \simeq -20$ mV) are considerably more positive than the -130 mV observed by Knaff [10] at pH > 8.9. This observation is consistent with a modification in the environment of Q such as to stabilize the semiquinone anion, Q^-. Modifications such as the appearance of a positive charge or loss of a negative charge close to Q or an increase in polarity

could all lower the pK below 5.9 for $Q^-/Q^-(H^+)$ and raise the E_m of Q/Q^- in the pH-independent region.

Several authors have proposed that inhibitors like *o*-phenanthroline which block electron transport between the primary and secondary quinone acceptors of PS II and of bacterial reaction centers may do so by displacing the secondary quinone [13,27]. Opposite effects of *o*-phenanthroline and increased occupancy of the secondary quinone-binding site on the E_m of Q_A/Q_A^- in bacterial reaction centers appear to support such a model [14]. The presence of Q_B decreases the E_m of Q_A/Q_A^-, the presence of *o*-phenanthroline increases the E_m [14]. Similarly, in PS II, Knaff [9] has reported a considerable increase in the $E_m(Q/Q^-)$ upon addition of 0.1 mM *o*-phenanthroline (+70 mV, as in bacterial reaction centers [14]). Malkin and Barber [25], however, observed no effect of this inhibitor.

DCMU, which has also been suggested [27,28] to compete with plastoquinone for binding to the secondary electron acceptor site, has no effect on the E_m of Q [6,7,9,25]. If DCMU does indeed act by the competitive mechanism, as suggested, then the lack of effect of DCMU on the E_m of Q/Q^- would suggest the independence of this E_m from B.

In the PS II particles described here which lack the secondary electron acceptor, B, we see very little difference (within 25 mV) in the overall E_m of Q/Q^- (ignoring the heterogeneity) and that of Horton and Croze [6] in chloroplasts where the secondary acceptor is present. Furthermore, the centers showing a pH-dependent $E_m(Q/Q^-)$ yet lacking B show redox properties (E_m and pH dependence) similar (within 20 mV) to that reported by Knaff [10] for $E_m(Q/Q^-)$ as detected by oxidation of cytochrome *b*-559 at 77 K. These results would suggest that the presence or absence of B has very little effect on the E_m of Q/Q^-.

Low-potential electron acceptor

At −175 mV (Q reduced) small light-induced absorbance changes are observed which reflect neither formation of plastosemiquinone nor of reduced pheophytin. If indeed pheophytin is the intermediate acceptor, one would expect its light-induced reduction in a series of flashes to appear in a manner complementary to the dark reduction of Q_1 in redox titration. That stable light-induced pheophytin reduction (lifetime $\gg$ 1 ms) is not observed at above −200 mV suggests that at intermediate potentials (approx. −175 mV) an additional endogenous and/or exogenous acceptor (e.g., one of the mediators) is able to rapidly oxidize Ph^-. Possible candidates for such an endogenous acceptor are Q_L and Q^-. The latter possibility (double reduction of Q) is, however, eliminated in the accompanying paper [12]. As light-induced accumulation of Ph^- titrates in around −250 mV (not shown) titration of Q_L or of a mediator could be occurring. These possibilities are under investigation.

We hoped that detection of light-induced absorbance changes at −175 mV might reflect the photoreduction of Q_L. While these changes bear a certain resemblance to the reduction of chlorophyll *a* [19] their amplitude is too small to expect that this species is reduced in any more than a small fraction of reaction centers. Such absorbance changes could reflect local electrochromic effects linked to the reduction of Q_L. It is certain that if Q_L is indeed still present in the particles its difference extinction coefficient ($Q_L^- - Q_L$) between 390 and 580 nm must be quite small. The absence of a −250 mV wave in the titration of the fluorescence yield in these particles would, however, argue against the presence of Q_L, though this is not an absolutely airtight argument as pointed out earlier.

A lifetime of 30–100 ns for $P\text{-}680^+\text{-}Ph^-$ was estimated from the quantum yield of Ph^- formation and the kinetics of $P\text{-}680^+$ reduction by the secondary donor. This estimate is over an order of magnitude greater than that estimated by Klimov et al. [15] and Shuvalov et al. [20] based on spectroscopic measurements, and by Haehnel et al. [29] based on fluoresccence lifetime measurements in chloroplasts and in whole algae. The explanation for this discrepancy is not immediately obvious. Comparative measurements of fluorescence and radical pair lifetimes are planned to see if $P\text{-}680^+\text{-}Ph^-$ is not more stable in this material.

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References

1 Nugent, J.H.A., Diner, B.A. and Evans, M.C.W. (1981) FEBS Lett. 124, 241–244
2 Diner, B.A. and Wollman, F.A. (1980) Eur. J. Biochem. 110, 521–526
3 Feher, G. (1971) Photochem. Photobiol. 14, 373–387
4 Dutton, P.L., Leigh, J.S. and Reed, R.W. (1973) Biochim. Biophys. Acta 292, 654–664
5 Evans, M.C.W., Nugent, J.H.A., Tilling, L.A. and Atkinson, Y.E. (1982) FEBS Lett. 145, 176–178
6 Horton, P. and Croze, E. (1979) Biochim. Biophys. Acta 545, 188–201
7 Cramer, W.A. and Butler, W.L. (1969) Biochim. Biophys. Acta 172, 503–510
8 Golbeck, J.H. and Kok, B. (1979) Biochim. Biophys. Acta 547, 347–360
9 Knaff, D.B. (1975) Biochim. Biophys. Acta 376–583–587
10 Knaff, D.B. (1975) FEBS Lett. 60, 331–335
11 Bowes, J.M., Horton, P. and Bendall, D.S. (1981) FEBS Lett. 135, 261–264
12 Diner, B.A. and Delosme, R. (1982) Biochim. Biophys. Acta 722, in the press
13 Erixon, K. and Butler, W.L. (1971) Biochim. Biophys. Acta 234, 381–389
14 Wraight, C.A. (1981) Isr. J. Chem. 21, 348–354
15 Klimov, V.V., Klevanik, A.V. and Krasnowsky, A.A. (1977) FEBS Lett. 82, 183–186
16 Van Gorkom, H.J., Tamminga, J.V., Haveman, J. and Van der Linden, I.K. (1974) Biochim. Biophys. Acta 397, 417–438
17 Diner, B.A. and Bowes, J.M. (1981) in Proceedings of the 5th International Congress on Photosynthesis (Akoyunoglou, G., ed.), pp. 875–883, Balaban International Science Services, Philadelphia
18 Klimov, V.V., Dolan, E. and Ke, B. (1980) FEBS Lett. 112, 97–100
19 Fujita, I., Davis, M.S. and Fajer, J. (1978) J. Am. Chem. Soc. 100, 6280–6282
20 Shuvalov, V.A., Klimov, V.V., Dolan, E., Parson, W.W. and Ke, B. (1980) FEBS Lett. 118, 279–282
21 Diner, B.A. and Delosme, R. (1982) Abstr. 100, Society for Experimental Biology, Leiden Conference
22 Pertrouleas, V. and Diner, B.A. (1982) FEBS Lett. 147, 111–114
23 Klimov, V.V., Dolan, E., Shaw, E.R. and Ke, B. (1980) Proc. Natl. Acad. Sci. U.S.A. 77, 7227–7231
24 Fowler, C.F. and Kok, B. (1974) Biochim. Biophys. Acta 357, 299–307
25 Malkin, R. and Barber, J. (1979) Arch. Biochem. Biophys. 193, 169–178
26 Van Gorkom, H.J. (1974) Biochim. Biophys. Acta 347, 439–442
27 Velthuys, B.R. (1982) in Quinones in Energy Conserving Systems (Trumpower, B.L., ed.), Academic Press, New York, in press
28 Lavergne, J. (1982) Biochim. Biophys. Acta 682, 345–353
29 Haehnel, W., Nairn, J.A., Reisberg, P. and Sauer, K. (1982) Biochim. Biophys. Acta 680, 161–173