Chloride-depletion of photosynthetic water oxidase

No proton release during the second oxidation step, $S_2^* \Rightarrow S_3^*$, and a transmembrane radical pair recombination from the third on

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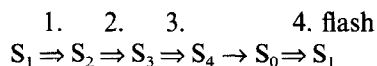*Biophysik, Universität Osnabrück, D-49069 Osnabrück, Germany*

Chloride depletion blocks the normal four-step progress of photosynthetic water oxidation. We studied proton release in chloride-depleted thylakoids which were dark-adapted and excited by flashing light. Proton release was blocked from the second flash on, possibly leaving an uncompensated positive charge in the catalytic centre. The reduction of P_{680}^+ by Tyr_Z was still very rapid ($\ll 10\,\mu s$). From the third flash on, P_{680}^+ was reduced more slowly ($70\,\mu s/200\,\mu s$), and by an electrogenic back-reaction. The uncompensated positive charge may be the reason why the rapid reduction of P_{680}^+ by Tyr_Z is prevented and the transmembrane charge-pair recombination is facilitated.

Photosynthesis; Water oxidation; Chloride; Proton; Electrochromism; P_{680} reduction

1. INTRODUCTION

Photosynthetic water oxidation in cyanobacteria and plants is driven by four light quanta. According to the Kok-model [1] (see [2] for a recent review) the catalytic centre, i.e. the manganese containing centre, undergoes four univalent oxidations from state S_0 to S_4. S_4 spontaneously decays back to S_0 under release of dioxygen. As state S_1 is most stable in the dark the progress as function of flash number is as follows:

$$
\begin{array}{cccc}
1. & 2. & 3. & 4.\ \text{flash} \\
S_1 \Rightarrow S_2 \Rightarrow S_3 \Rightarrow S_4 \to S_0 \Rightarrow S_1
\end{array}
$$

In addition to dioxygen four protons are released in this cycle. Unlike the release of dioxygen which bursts out after the third flash, proton release is distributed over all transitions. The relative H^+/e^--stoichiometries vary as function of the pH and of the protein periphery of photosystem II. The latter has become evident from different stoichiometric patterns between BBY-membranes [3], unperturbed thylakoids [4,5], thylakoids which are depleted of the light harvesting complexes [6] and PS II-core preparations [4,7,8]. The primary electron donor of PS II, P_{680}^+, oxidises the manganese cluster via an intermediate carrier, Tyr_Z (for review see [2]). Depending on the redox state the electron transfer times are in the range of 23–260 ns for $Tyr_Z \to P680^+$ [9] and 30–1300 μs for $Mn \to Tyr_Z^+$ (see [2,10,11]). We recently found that the major portion of proton release can occur in 10 μs and therefore at the level of Tyr_Z^+ ([5] – preliminary report in [12]). All results are compatible with the notion that *rapid proton release* is primary due to an electrostatic response of amino acids and, possibly, ligands to manganese, which are reprotonated only during the single (or two) steps where the oxidation of water takes place [13]. Proton release is believed to relieve the electrostatic constraints which would arise if positive charges were accumulated in the catalytic centre [14,15].

The chloride anion is an essential cofactor of water oxidation. In its absence the normal progress towards higher oxidation states, $S_i \Rightarrow S_{i+1}$, is blocked, but it is controversial where and how [2,16]. To describe the perturbation by Cl^--deficiency of the normal progression of the manganese cluster through its different oxidation states we use the following nomenclature:

S_i: i oxidising equivalents stored on the Mn-cluster or ligands to Mn *beyond Tyr_Z, unperturbed system*.
S_i^*: i oxidising equivalents stored in the perturbed Mn-Tyr_Z-system, with at least one stored as Tyr_Z^+ or on

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Abbreviations: BBY, PS II enriched membrane fragments; BSA, bovine serum albumin; Chl, chlorophyll; DMBQ, 2,6 dimethyl-*p*-benzoquinone; DNP-INT, dinitrophenylether of iodonitrothymol; EDTA, ethylene diamine tetraacetic acid; EPR, electron paramagnetic resonance; NR, neutral red; PS I, II, photosystem I, II; P_{680}, primary electron donor of PS II; P_{700}, primary electron donor of PS I; S_i, i[th] oxidation state of water oxidase according to the Kok-model in controls and, S_i^*, in thylakoids perturbed by chloride deficiency; XANES, X-ray absorption near edge structure.

another component which is not normally involved in control thylakoids.

The effects of Cl^--depletion have been studied by various techniques and through different observables, namely PS II-fluorescence [17,18], luminescence [18], a disturbed EPR-signal which, after Cl^--repletion reversed into the EPR-'multiline'-signal of the normal S_2-state [19–21] and P_{680}^+-reduction [21]. The results are interpreted as follows. (1) The first flash produces a modified S_2^* [19–21]. (2) The $S_2^* \Rightarrow S_3^*$ transition is inhibited [17,18], although the oxidation of Tyr_Z by P_{680}^+ proceeds normally [21]. On the contrary, data which resulted from other measurements of thermoluminescence [22], oxygen evolution [23], EPR ($g = 2.004$ and $g = 1.98$) and P_{680}^+-reduction [24], have been interpreted to show that the transition $S_2^* \Rightarrow S_3^*$ is still possible but that the transition $S_3^* \Rightarrow S_4 \rightarrow S_0$ is blocked.

In Ca^{2+}-depleted PS II-enriched membranes a new EPR-signal ($g = 2.004$), of then unknown chemical nature, has been detected and attributed to a formal S_3-state [25]. In Cl^--depleted and F^--substituted PS II-enriched membranes and reaction centre core preparations this signal has also been observed and by its properties attributed to an organic radical within 1 nm distance from the Mn-cluster [26]. By a combination of EPR- and UV-spectroscopy evidence for a histidine radical has been presented [27]. It has been speculated that the oxidation of a histidine (possibly His-190 of the D1 protein [2] which probably is linked by a hydrogen bond to Tyr_Z^+) might be involved in the normal progress from S_2 to S_3 in unperturbed material [27]. In Cl^--depleted PS II-enriched membranes, when the EPR- signal which is attributable to a formal S_3-state has appeared in about one half of the centres and probably $S_2Tyr_Z^+$ in the remainder, any further electron transfer from Tyr_Z to P_{680} is inhibited [24]. This is compatible with previous data on the decay time of P_{680}^+ as a function of the flash number. Whereas being in the sub- microsecond time range up to the second flash, it is much longer ($t_{1/2} = 40$–50 μs) from the third flash on [21].

We studied the extent of proton release as function of flash number in dark adapted thylakoids in order to characterise the net charge of the catalytic centre. We also studied the relaxation of the flash-induced transmembrane voltage to establish whether the comparatively slow reduction of P_{680}^+ after the third flash originates from a transmembrane charge-pair recombination. Both observables have not previously been touched in chloride-depleted material.

2. MATERIALS AND METHODS

As starting material for the preparation we used 12-day-old pea seedlings. Stacked thylakoids were prepared according to Förster and Junge [28]. Chloride deficient samples were obtained by mild EDTA-treatment according to Yachandra et al. [29]. All preparations were

stored frozen at $-80°C$ and thawed immediately before the experiment. The chlorophyll content was determined photometrically using the extinction coefficients published by Porra et al. [30]. For measurement of oxygen yield under continuous illumination a Clark-type electrode was used. Measuring conditions: 20 μM chlorophyll in 10 mM Na_2SO_4 or 20 mM NaCl, 2.5 mM $MgSO_4$, 20 mM HEPES pH 7, $\pm$ 1 μM nigericin. Proton release, electrochromism and P_{680}^+-reduction under flashing light were measured photometrically [31] in cuvettes with 2 cm optical path. pH-transients in the thylakoid lumen (Chl-conc.: 20 μM) were detected as absorption changes at 548 nm of the amphiphilic, membrane adsorbed dye Neutral red ($13 \mu M$) [32–36]. The procedures of extracting luminal pH-transients due to water oxidation and to handle dark adapted thylakoids have been described elsewhere [28,32]. pH-transients in the suspending medium (10 mM $NaSO_4$ or 20 mM NaCl, 2.5 mM $MgSO_4$) were quenched by the non-permeating buffer bovine serum albumin (2 g/l). The pH was adjusted to 7.2. Ferricyanide (2 mM) and DMBQ (5 μM) were added as electron acceptors to avoid acceptor limitation and to decrease proton uptake at the acceptor side of photosystem II by rapid oxidation of the reduced plastoquinone. 10 μM DNP-INT was added to inhibit the oxidation of plastoquinol by the cytochrome b_6/f complex and the concomitant proton release into the thylakoid lumen. Signals attributed to pH changes in the thylakoid lumen were differences of signals obtained in the presence and absence of Neutral red. Measurements of electrochromism at 522 nm [37] and P_{680}^+-reduction at 820 nm [9] were performed under the same conditions as for proton release except that 10 mM HEPES was used instead of BSA. Up to 1200 transient signals were averaged to improve signal-to-noise. Saturating light flashes were provided either by a Xenon-lamp or a frequency doubled, and Q-switched Nd:YAG-Laser (Spektrum). Transient signals were fitted with three or two exponentials plus offset by a simplex-type algorithm ('Grafit', DOS or 'Plotdata', VAX-VMS).

3. RESULTS

3.1. Controls

Measurements of oxygen yield under saturating, continuous light were used as control for the degree of chloride deficiency and membrane integrity. A typical
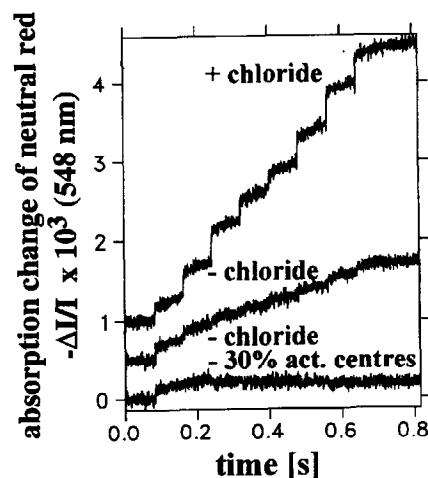

Fig. 1. pH transients in the thylakoid lumen induced by firing a series of light flashes to dark adapted thylakoids. Upper trace: chloride-repleted thylakoids; 20 μM Chl in 20 mM NaCl, 2.5 mM $MgSO_4$, 2 g/l BSA pH 7.2, 2 mM ferricyanide, 5 μM DMBQ, 10 μM DNP-INT, $\pm$ 13 μM NR; middle trace: chloride-depleted thylakoids; instead of 20 mM NaCl, 10 mM $NaSO_4$ was added; lower trace: middle trace minus 30% of the upper trace. Electric bandwidth: 3 kHz, flashes spaced 80 ms apart, 20 μs per cycle, 40 signals averaged, 30 min dark adaptation.

rate of oxygen evolution was: + chloride: 104 μM O_2/(mg Chl h), − chloride: 36 μM O_2/(mg Chl h). The reversibility of the inhibition by exchange of 10 mM Na_2SO_4 against 20 mM NaCl showed that the effect was only chloride- and not calcium-related. Under chloride deficient conditions about 35% of the centres were still active or not depleted. Electron transport in chloride deficient thylakoids was still coupled; the ratio of rates with and without nigericin was at least 2.9. The residual extent of PS I turnover under the given electron acceptors and blocking agents of electron transfer from PS II to PS I (2 mM ferricyanide, 5 μM DMBQ, 10 μM DNP-INT) was measured by absorption changes at 700 nm. It was less than 10% of the extent in Cl^--repleted thylakoids with 10 μM methyl viologen and without the above electron acceptors and blocking agents.

3.2. Luminal pH-transients in dark-adapted thylakoids

In dark-adapted, chloride-repleted thylakoids protons were released after each exciting flash (upwardly directed jumps in Fig. 1, upper trace). The extent oscillated with period of four as function of flash number. This resembled the behaviour in untreated thylakoids [5,8,28,32]. In chloride-depleted thylakoids, proton release was detected mainly after the first flash (Fig. 1, middle trace). Upon subsequent flashes the extent was much smaller and most probably due to the ≈ 30% portion of not depleted centres. When the trace recorded in the presence of chloride was multiplied by a factor of 0.3 and then subtracted from the trace without chloride a positive transient (indicating proton release) was discernible solely on the first flash (Fig. 1, lower trace). The absence of even rapid transient proton release from the second flash on was corroborated in further experiments at 10 μs time resolution (not documented).

3.3. Decay of the transmembrane electric field in dark adapted thylakoids

We measured transients of the transmembrane electric field by electrochromic absorption changes at 522 nm with a time resolution of 5 μs. After the first flash (Fig. 2) the largest portion of the electric field (60%)

Table I

The extent and the half-decay time of the transmembrane voltage in chloride-depleted and dark-adapted thylakoids as function of flash number

	1st flash	3rd flash
A_1	18%	61%
$t_{1/2}$	101 μs	71 μs
A_2	22%	12%
$t_{1/2}$	1.07 ms	436 μs
A_3	60%	27%
$t_{1/2}$	25.4 ms	10.9 ms

Data from electrochromic transients as documented in Fig. 2 were fitted by three exponentials.
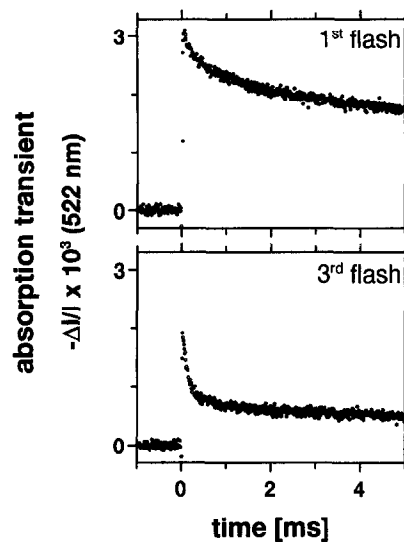

Fig. 2. Transients of the transmembrane voltage in chloride-depleted and dark adapted thylakoids. Top: first flash, bottom: third flash. 20 μM Chl in 10 mM NaSO$_4$, 2.5 mM MgSO$_4$, 10 mM HEPES pH 7.2, 2 mM ferricyanide, 5 μM DMBQ, 10 μM DNP-INT, electric bandwidth: 100 kHz, 5 μs time resolution, flashes spaced 80 ms apart, 20 μs per cycle, 45 signals averaged, 30 min dark adaptation.

decayed slowly with 25 ms (Table I) indicating a mainly normal discharge of the voltage by ionic leak currents across the thylakoid membrane. The same hold true after the second flash (not documented). After the third flash a fast decay ($t_{1/2} = 71$ μs) became predominant (Table I). The loss of the initial extent was not due to chloride deficiency as it was also present in chloride-repleted samples. The relative extent of the remaining slow phase was 27%. This was compatible with the notion that the rapid phase was not due to a general increase of the membrane conductance, but rather to a specific electrogenic back reaction in about 61% of the centres. The percentage was again compatible with the above figure of about 30% not Cl^--depleted centres. The nature of the phase which decayed with ≈ 0.4/1.1 ms remained to be clarified.

3.4. The rate of reduction of P_{680}^+ in dark adapted thylakoids

The reduction of P_{680}^+ was studied by monitoring absorption transients at 820 nm in dark adapted chloride-depleted thylakoids under conditions where the PS I portion was less than 10%. After the first two flashes the reduction occurred in the sub microsecond time range. After the third flash a slower phase in the microsecond range appeared (data not shown). This confirmed the results of Ono et al. [21], who studied chloride-deficient membrane fragments (BBY) and reported an unresolved fast reduction of P_{680}^+ after the first two flashes and a slower one ($t_{1/2} = 40$–50 μs) after the third flash. The half-decay time of P_{680}^+ which we observed in thylakoids (70/210 μs) was similar to the half-decay time of the

transmembrane electric field in this material (see above).

We questioned whether the rapid decay of the electric field and the concomitant reduction of P_{680}^+ persisted even under repetitive excitation, where improvement of signal-to-noise by averaging was facilitated. Up to 1200 transient signals were averaged, of electrochromism at 522 nm and P_{680}^+ at 820 nm with a time resolution of 2 μs. The results are given in Fig. 3. The decay at 522 nm was multiphasic (Fig. 3, upper trace). It was fitted by 3 exponentials plus (apparent) offset. The most rapid phase was attributable to chlorophyll-/carotenoid-triplett states [38,39]. It was ignored. The other fit parameters were as given in Table II. 54% of the remaining signal decayed with a half-time of 66 μs, 18% with a half-time of 255 μs. The apparent offset (truly decaying with $t_{1/2} > 10$ ms) accounted for $\approx 28\%$ of the extent. It was attributed to the remaining fully active centres. The decay of P_{680}^+ at 820 nm as shown in Fig. 3 (lower trace) was analysed in similar way. 65% of the extent decayed with 74 μs, 25% with 205 μs. The offset ($t_{1/2} > 10$ ms, $\approx 10\%$ of the whole amplitude) was attributed to a PS I contribution in agreement with the explanation by Brettel et al. [9]. The contribution of still active centres was expected in the nanosecond time-range and therefore unresolved in our experiments. When an unresolved extent of, say, 30% was added to the observed one, the relative extents of the observed kinetic phases were smaller as noted in Table II (column 2, in parentheses).

The half-decay times (66 versus 74 μs) and the relative proportions of the fast phases (54 versus 65/45.5%) agreed fairly well between electrochromism and reduction of P_{680}^+. The same holds true for the slower phase (205 versus 255 μs, 18% versus 25/17.5%).

4. SUMMARY AND DISCUSSION

According to the results by other authors which are cited in the introduction chloride-depletion alters the

Table II

The extent and the half-decay time of the transmembrane voltage and of P_{680}^+-reduction under repetitive excitation

	Field decay	Apparent P_{680}^+-reduction (corrected for only 70% Cl^--depleted centres)
A_1	54%	65% (45.5%)
$t_{1/2}$	66 μs	74 μs
A_2	18%	25% (17.5%)
$t_{1/2}$	255 μs	205 μs
offset	28%	10% (7%)

In the second column the values in parentheses represent the corrected values under the assumption, that only 70% of the centres were chloride-depleted and 30% were still chloride containing (see text). Data fitted by two exponentials plus offset.
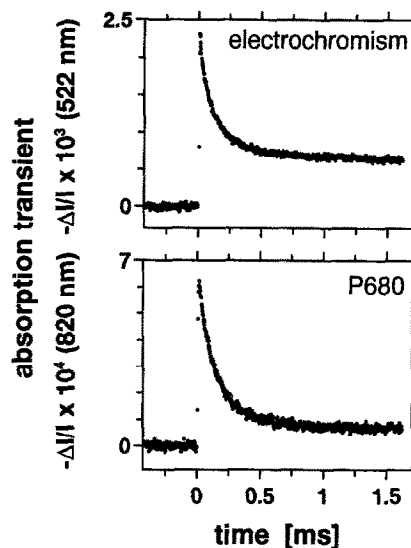

Fig. 3. Transients of the transmembrane voltage and of P_{680} in chloride-depleted thylakoids under repetitive excitation. Medium composition as in Fig. 2. Electric bandwidth: 100 kHz, 2 μs time resolution, flashes spaced 200 ms apart, 100 flashes per fresh sample, $\approx$ 1200 signals averaged.

S_3-state of the water oxidase. A histidine radical cation and/or a relatively stable Tyr_Z-radical appear when a second flash, which creates the altered state S_3^*, is applied to dark adapted samples [24–27]. From the third flash on the normally very rapid reduction of P_{680}^+ is greatly slowed down [21]. Our experiments have shown that proton release, which is normally distributed over all transitions of the water oxidase, is already impaired from the second transition, namely $S_2^* \Rightarrow S_3^*$, on. This leaves an uncompensated positive charge at the donor side of PS II in its oxidation state, S_3^*. From the next, the third transition on, the normally very rapid (23/260 ns) reduction of P_{680}^+ by Tyr_Z is blocked, as known from previous work [21]. Instead, we found a rapid transmembrane charge-pair recombination. The rate spectra of the P_{680}^+-reduction and of the voltage decay were coincident within the error limits. This excludes the existence of a long lasting intermediate state on the path of the reaction $P^+Q^- \rightarrow PQ$. The observed rise-times of the reduction of P_{680}^+, namely about 70 and 210 μs in this work, are similar to those previously reported when water oxidation was inhibited by calcium-depletion Tris-washing, low pH-treatment or acetate addition [24,40–44].

It is known from EPR-studies that already the *first flash* gives rise to a perturbed oxidation state of the manganese cluster, S_2^*, which can be reversed into the normal S_2 state by addition of chloride. The latter shows the multiline signal of a mixed-valence Mn-cluster [19–21]. It is also known that Tyr_Z is in its reduced form in this state, S_2^*. We found that proton release associated with transition $S_1^* \Rightarrow S_2^*$ was rather normal.

Proton release, however, was blocked after the *second flash* (and from there on), although the charge separation and subsequent reduction of P_{680}^+ seemed to proceed normally (i.e. in sub-μs). This implies the deposition of one uncompensated positive charge in the vicinity of the system [Mn_4-Tyr_Z]. In unmodified thylakoids we have previously shown that proton release which is caused by the transition $S_2 \Rightarrow S_3$ can occur already at the level of Tyr_Z^+ [28] with a half-rise time of $12\,\mu$s [5,12]. The dissociation of one proton persists when the positive charge on Tyr_Z^+ is displaced into the Mn-cluster. The absence of proton release in chloride-depleted material is then to be understood in two alternative ways (note that P_{680}^+ is still rapidly reduced by Tyr_Z to yield Tyr_Z^+):

1. Tyr_Z^+ is so rapidly ($\ll 10\,\mu$s) reduced by an unknown and normally not involved component, that the deprotonation associated with Tyr_Z^+ was prevented. In addition, the unknown component, which then carries the positive charge, is located such as to avoid an electrostatic response by deprotonation. In this model the effect of chloride-depletion is to create a new, much faster pathway for the reduction of Tyr_Z^+ than by the manganese cluster proper.

2. Tyr_Z^+ is long-lived since its reduction by the manganese cluster is blocked. In addition, the deprotonation by an electrostatic response of amino acids is prevented.

In the first model, the absence of a very rapid (i.e. ns) reduction of P_{680}^+, when formed by the *third flash,* might be plausibly explained by the retention caused by coulombic force exerted by the uncompensated positive charge. In the second model the absence of the very rapid reduction is trivial since the original electron donor is absent (i.e. Tyr_Z is oxidised). Both cases make it plausible that the transmembrane charge-pair recombination is favoured by the uncompensated positive charge in the catalytic centre.

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