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ABSORBANCE DIFFERENCE SPECTRA OF THE SUCCESSIVE REDOX STATES OF THE OXYGEN-EVOLVING APPARATUS OF PHOTOSYNTHESIS *

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The spectra of the absorbance changes due to the turnover of the so-called S-states of the oxygen-evolving apparatus were determined. The changes were induced by a series of saturating flashes in dark-adapted Photosystem II preparations, isolated from spinach chloroplasts. The electron acceptor was 2,5-dichloro-*p*-benzoquinone. The fraction of System II centers involved in each S-state transition on each flash was calculated from the oscillation pattern of the 1 ms absorbance transient which accompanies oxygen release. The difference spectrum associated with each S-state transition was then calculated from the observed flash-induced difference spectra. The spectra were found to contain a contribution by electron transfer at the acceptor side, which oscillated during the flash series approximately with a periodicity of two and was apparently modulated to some extent by the redox state of the donor side. At the donor side, the $S_0 \rightarrow S_1$, $S_1 \rightarrow S_2$ and $S_2 \rightarrow S_3$ transitions were all three accompanied by the same absorbance difference spectrum, attributed previously to an oxidation of Mn(III) to Mn(IV) (Dekker, J.P., Van Gorkom, H.J., Brok, M. and Ouwehand, L. (1984) Biochim. Biophys. Acta 764, 301–309). It is concluded that each of these S-state transitions involves the oxidation of an Mn(III) to Mn(IV). The spectrum and amplitude of the millisecond transient were in agreement with its assignment to the reduction of the oxidized secondary donor Z^+ and the three Mn(IV) ions.

Introduction

Photosynthetic oxygen evolution is brought about by the reaction centers of PS II, which operate independently in this respect and release one oxygen molecule after every four photoreactions [1]. Five successive redox states can therefore be assigned to the oxygen-evolving structure [2]:

$$S_0 \xrightarrow{h\nu} S_1 \xrightarrow{h\nu} S_2 \xrightarrow{h\nu} S_3 \xrightarrow{h\nu} S_4 \longrightarrow S_0 \quad (2\,H_2O \rightarrow O_2 + 4\,H^+)$$

Of these so-called S-states, S_0 and S_1 are stable, S_2 and S_3 revert to S_1 in minutes, and S_4 is reduced to S_0 in a millisecond, releasing oxygen. As a result, the system relaxes largely to S_1 in darkness and subsequent illumination by a series of single-turnover flashes induces an oscillation with a periodicity of 4, which is damped due to misses and double hits. The kinetics of the system have been discussed in detail by Bouges-Bocquet [3].

A damped period-4 oscillation has been detected both in ultraviolet absorbance [4] and in electron spin resonance [5]. These properties may allow an identification of the chemical species being oxidized in the successive S-state transitions. The EPR signal is apparently specific for the S_2-state and consists of a broad multiline spectrum, which can be observed only at very low tempera-

* We dedicate this paper to the memory of Bernadette Bouges-Bocquet.

Abbreviations: Chl, chlorophyll; DCBQ, 2,5-dichloro-*p*-benzoquinone; DCMU, 3(3′,4′-dichlorophenyl)-1,1-dimethylurea; Mes, 4-morpholineethanesulfonic acid; PS II, Photosystem II.

tures [5–7]. It is ascribed to a pair or more magnetically interacting manganese ions of uncertain valency [8,9]. The ultraviolet absorbance consists of a broad asymmetric band around 300 nm, which may be due to an oxidation of Mn(III) to Mn(IV) [10]. The absorbance increased on the first flash after dark adaptation and decreased in a millisecond after the third [11,12]. Therefore, it was assumed to indicate an oxidation on the $S_1 \rightarrow S_2$ transition and a reduction accompanying oxygen release [11]. The oxidant produced in the $S_0 \rightarrow S_1$ and the $S_2 \rightarrow S_3$ transitions remained undetected.

The oxidant produced by the third flash is accompanied by another EPR signal, known as Signal II_{vf} [13,14]. It may appear as a short-lived intermediate in the other S-states as well [14]. This intermediate is supposedly identical to the terminal oxidant generated in PS II after removal of the bound manganese (e.g., by Tris-washing) [15]. Both the lineshape of the EPR signal [16] and the absorbance difference spectrum [10] suggest that this oxidant, called Z^+, is a plastosemiquinone cation. The optical spectrum of the millisecond transient after the third flash should then consist of a superposition of the difference spectrum of Z and the broad band around 300 nm. The shape of the only published spectrum of this transient [17], however, is rather different, suggesting other contributions.

We have carried out a quantitative analysis of the oscillating ultraviolet absorbance changes in oxygen-evolving PS II preparations, and report here the difference spectrum of each S-state transition. The results suggest that three Mn(III) ions are successively oxidized to Mn(IV) and are reduced simultaneously with Z^+ after the next photoreaction.

Materials and Methods

PS II particles were prepared from spinach chloroplasts according to the method of Berthold et al. [18], with the exception that the first Triton X-100 incubation step was carried out at pH 6.0 and that the second Triton X-100 incubation step was omitted. Absorbance changes were measured with an optical path length of 1.2 mm and a Chl concentration of 200 μg/ml, in 20 mM Mes-NaOH/5 mM $MgCl_2$ (pH 6.0), in a single beam apparatus as described before [10]. The time constant of the apparatus was about 0.3 ms. Illumination was provided by saturating, 10 μs xenon flashes, spaced at 300 ms, unless stated otherwise. The absorbance changes were corrected for particle flattening as described before [19,10].

Results

Absorbance changes of the oxygen-evolving complex upon illumination by a series of saturating flashes were studied in the PS II preparation of Berthold et al. [18]. It was verified that our preparations did not exhibit other absorbance changes than those of PS II [10]. In order to avoid limitations of secondary electron transfer at the acceptor side, we added the lipophilic acceptor DCBQ (100 μM). With this addition the decay of the absorbance changes induced by repetitive flashes, spaced at 1 second, and measured with a 300 μs response time, was triphasic, with halftimes of 1–1.5, 20 and 120 ms. The spectrum of the 1–1.5 ms phase consisted of contributions both from the donor and the acceptor side, and will be discussed below. Fig. 1 shows the spectra of the 20 ms phase

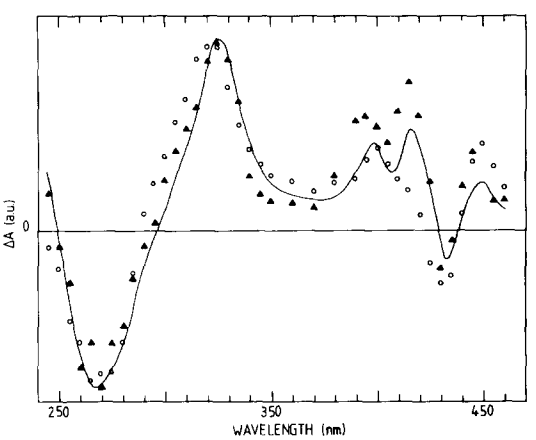

Fig. 1. Spectra of the 20 ms (solid triangles) and 120 ms (open circles) decay phases of absorbance changes induced by repetitive flashes in oxygen-evolving PS II preparations, suspended at a Chl concentration of 200 μg/ml in 20 mM Mes-NaOH (pH 6.0) with 5 mM $MgCl_2$ and 100 μM DCBQ. The data are the average of 50 flashes spaced at 1 s. The line is the difference spectrum calculated for electron transfer from Q^- to DCBQ. The $Q^- - Q$ spectrum was taken from Ref. 10, while the spectrum of reduction of DCBQ (to the quinol form) was measured in 50% ethanol. The spectra were normalized at 325 nm.

(triangles) and the 120 ms phase (circles) together with the spectrum expected for electron transfer from Q^- to DCBQ (line). The three spectra are very similar. However, in the 120 ms phase not only the 415 nm band is absent, but also C550, a band shift of pheophytin *a* around 545 nm due to reduction of Q [20] (not shown). It is concluded that Q^- is involved in the 20 ms phase, and that a differently situated semiquinone is involved in the 120 ms phase.

The spectra in Fig. 1 have been normalized at 325 nm. The actual amplitudes indicated that 40–45 and 30–35% of the centers were involved in the 20 ms and 120 ms phase, respectively, based on 280 chlorophylls per reaction center [10]. The remaining centers contribute to the 1–1.5 ms phase. In subsequent experiments a flash spacing of 300 ms was chosen to allow for a nearly complete reoxidation of the acceptor side between the flashes.

The preparations were dark-adapted for at least 15 min, after which DCBQ was added, and a series of saturating flashes, spaced at 300 ms, was fired. The flash-induced absorbance changes were measured in successive 50 ms sweeps, the offset being adjusted before each flash. Fig. 2 shows a typical trace, measured at 300 nm, where the donor causes relatively large absorbance changes. It appears that, superimposed on the changes of the acceptor side, a fast absorption increase occurs on the first flash,

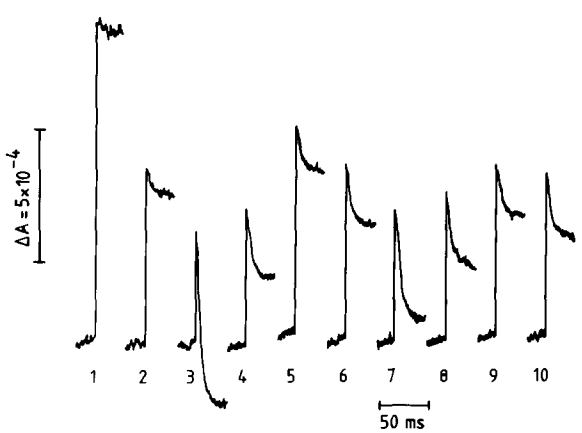

Fig. 2. Absorbance changes at 300 nm induced by ten flashes, spaced at 300 ms, in dark-adapted PS II preparations, suspended as in Fig. 1. The recordings are the average of 25 measurements.

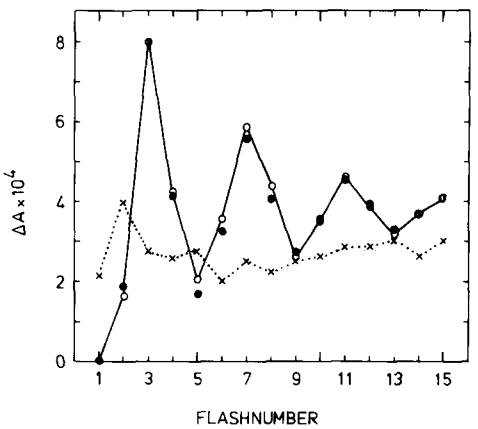

Fig. 3. Amplitudes of the 1–1.5 ms phase after the first 15 flashes in dark-adapted PS II preparations as in Fig. 1, recorded at 300 nm (open circles, full line) and at 400 nm (crosses, dotted line). The measurements at 300 nm are compared to the amplitudes of the $S_3 \rightarrow S_0$ transition calculated for a dark S-states distribution of 75% S_1 and 25% S_0, and 9% misses and 9% double hits on all transitions (solid circles). The values at 300 and 400 nm were the average of 75 and 10 measurements, respectively.

which is reversed on the third flash with a 1–1.5 ms halftime. This result is in accordance with the measurements of Velthuys on dark-adapted chloroplasts [11]. The same pattern is repeated every four flashes, with decreasing amplitude. In Fig. 3, the open circles indicate the extent of the 1–1.5 ms phase at 300 nm. This phase supposedly accompanies oxygen release and could be fitted with the Kok model (solid circles), assuming a dark distribution of 25% S_0 and 75% S_1, and 9% misses and 9% double hits on each turnover. At 400 nm (crosses) no such oscillation was observed, indicating a different contribution to the 1–1.5 ms phase. This second contribution is approximately the same on all flashes.

Fig. 4A, circles, shows the spectrum of the difference between the third and the fifth flash, presumably caused by the oxygen-releasing reaction only. (The dashed line will be discussed later). The initial amplitude was determined by extrapolation, assuming exponential kinetics. The $\Delta\epsilon$ scale is based on the amount of contributing reaction centers calculated from the fit of Fig. 3. The spectrum is very different from that attributed to the same reaction in Ref. 17. The spectrum of the

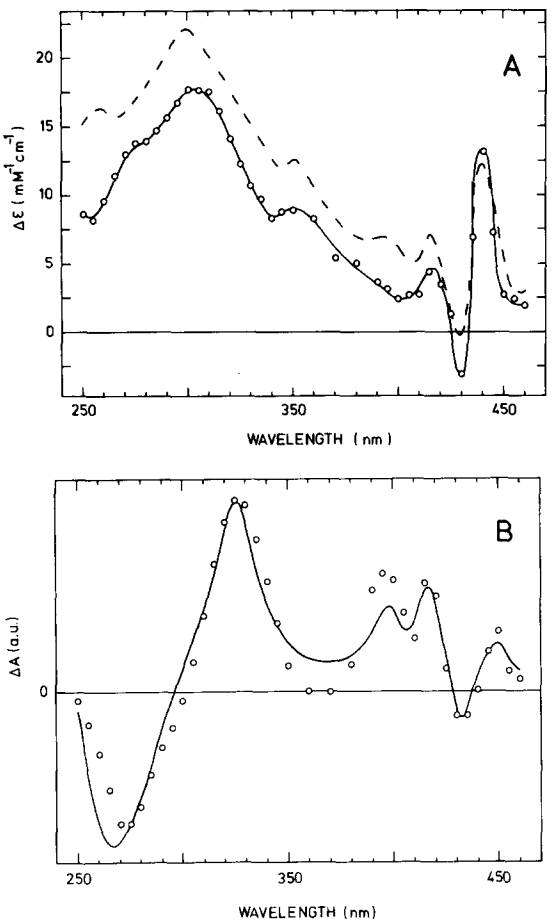

Fig. 4. (A) Spectrum of the 1–1.5 ms phase due to the donor side of PS II (circles, full line), calculated as described in the text. The dashed line is the sum of three times the spectrum of Fig. 7A (solid circles) plus the $Z^+ - Z$ spectrum at pH 6.0, taken from Ref. 10. (B) Spectrum of the 1–1.5 ms phase due to the acceptor side of PS II (circles), determined as described in the text. It is compared to the spectrum of reduction of DCBQ by Q^- (full line), taken from Fig. 1. Both spectra are normalized at 325 nm. The data are, depending on the wavelength, the average of 30–100 experiments.

other contribution to the 1–1.5 ms phase was obtained from the fifth flash by subtracting the spectrum of Fig. 4A to the extent indicated by the amplitude of the 300 nm change at the fifth flash (Fig. 3, open circles), and is shown in Fig. 4B. The spectral shape is similar to that of the 20 ms phase (Fig. 1, triangles) and the changes may be ascribed to electron transfer from Q^- to DCBQ (line). The amplitude indicated that about one-third of the reaction centers was involved.

The absorbance changes remaining at the end of the sweep, i.e., 30 ms after the flash (see Fig. 2) are shown in Fig. 5. After the oscillations are damped out the changes shown in Fig. 5B remain. The spectrum is only partially due to DCBQ re-

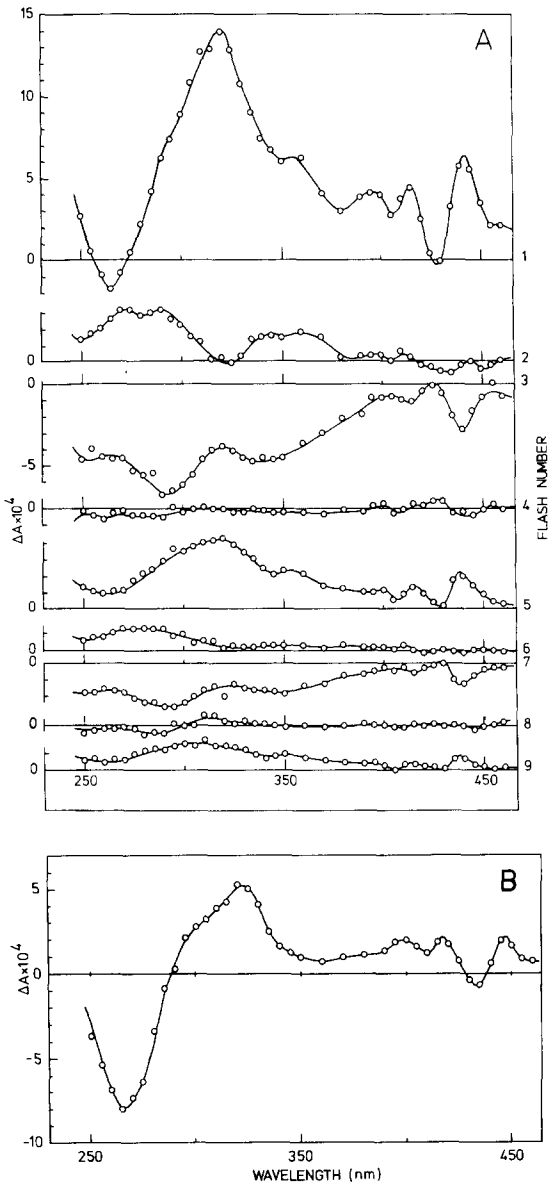

Fig. 5. (A) Spectra of the absorbance changes at 30 ms after each of the first nine flashes, measured in dark-adapted PS II preparations as in Fig. 2, from which the spectrum shown in (B) is subtracted. The data are, depending on the wavelength, the averages of 10–100 measurements. (B) The spectrum obtained by averaging flash numbers 16 to 25.

duction, since at 30 ms after the flash about half of the centers still contained a semiquinone (cf. Fig. 1). Fig. 5A shows the difference spectra after the first nine flashes, from which the spectrum of Fig. 5B has been subtracted. This subtraction is necessary for a quantitative analysis of the oscillating changes.

We assumed that each spectrum in Fig. 5A was composed only of absorbance changes related to the four successive S-state transitions and that the amount of each transition on each flash was determined correctly by the fit of Fig. 3. On this basis, the four S-state transition spectra were computed from all sets of four successive flashes (from Fig. 5A) and, using the fact that the four transition spectra should add up to zero, from all sets of three successive flashes as well. Except when the first flash was included, the different sets produced nearly the same results, with increasing variation for higher flash numbers. The averaged results of all sets beginning with flash numbers 2, 3, 4 and 5 are shown in Fig. 6.

It is obvious at first sight that more than one component contributes to these spectra, and the spectrum calculated for the $S_1 \rightarrow S_2$ transition (Fig. 6B) is clearly different from that observed in the presence of DCMU [10]. The shape of the spectra, especially around 320 nm, strongly suggests the alternating appearance and disappearance of a semiquinone anion. This phenomenon most likely reflects the two electron gating mechanism normally observed during reduction of the plastoquinone pool [4].

The amplitudes of the spectra of Fig. 6A, B and C are equal near 295 nm, where no contribution of the acceptor side is expected. Therefore, when the spectrum in Fig. 6D is multiplied by $-1/3$, the four spectra have a common isosbestic point, as illustrated in Fig. 6E. In the ultraviolet, the curves appear to differ only in the amplitude of a contribution by the acceptor side, with a difference spectrum similar to those of Fig. 1. The donor side would then contribute a single spectral component, occurring in a sequence $+1$, $+1$, $+1$, -3 during the $S_0 \rightarrow S_1$, $S_1 \rightarrow S_2$, $S_2 \rightarrow S_3$ and $S_3 \rightarrow S_0$ transitions. To verify this hypothesis, the ratio of absorbance changes at 320 and 295 nm due to the donor side was assumed to be the same as determined previously in the presence of DCMU

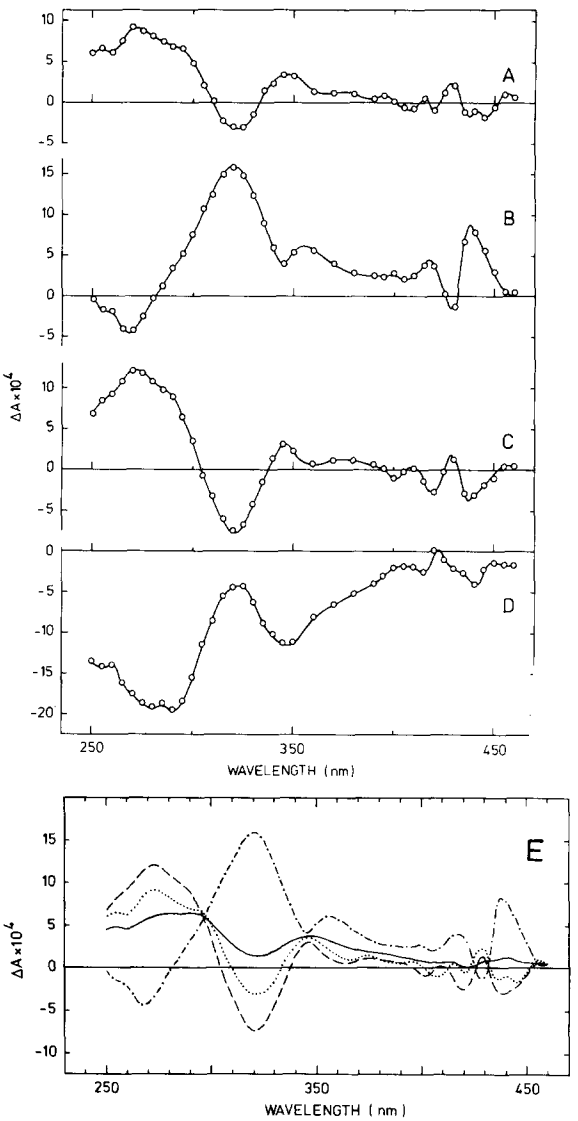

Fig. 6. Absorbance difference spectra associated with $S_0 \rightarrow S_1$ (A), $S_1 \rightarrow S_2$ (B), $S_2 \rightarrow S_3$ (C) and $S_3 \rightarrow S_0$ (D) transitions, calculated as described in the text from the spectra shown in Fig. 5A and the parameters of the period-4 oscillation as determined in Fig. 3. (E) The same spectra (A dotted, B dash-dot, C dashed line) shown in one plot, after mutliplying D by $-1/3$ (full line).

[10]. With this assumption, the relative amplitudes of the acceptor side contribution at 320 nm were calculated to be -0.77, $+0.97$, -1.19 and $+1.07$ during the successive S-state transitions. Subsequently, the amplitude of the donor change which

would agree best with this sequence of the acceptor change was determined at all wavelengths.

The donor spectrum thus obtained is shown in Fig. 7A (solid circles), together with that determined previously in the presence of DCMU (open circles). Note that only the ratio of changes at 320 and 295 nm in the two spectra has been normalized. At wavelengths above 260 nm, the spectra are very similar, both in shape and amplitude, except for the amplitudes of the chlorophyll band shift around 435 nm. Fig. 7B shows the additional absorbance changes contained in each of the spectra in Fig. 6, normalized at 320 nm. Apparently the four calculated amplitudes are very similar at all wavelengths, so there is no need to postulate more than one component to account for these spectra. The ultraviolet spectrum resembles those of Fig. 1. Its lower values near 250 and 345 nm correspond to differences between Q and the secondary acceptor in chloroplasts [21], suggesting that the period-2 oscillation is due to plastoquinone. For wavelengths above 370 nm a difference spectrum of the period-2 oscillation has not yet been reported, but is probably different from that of Q because of a smaller pheophytin contribution.

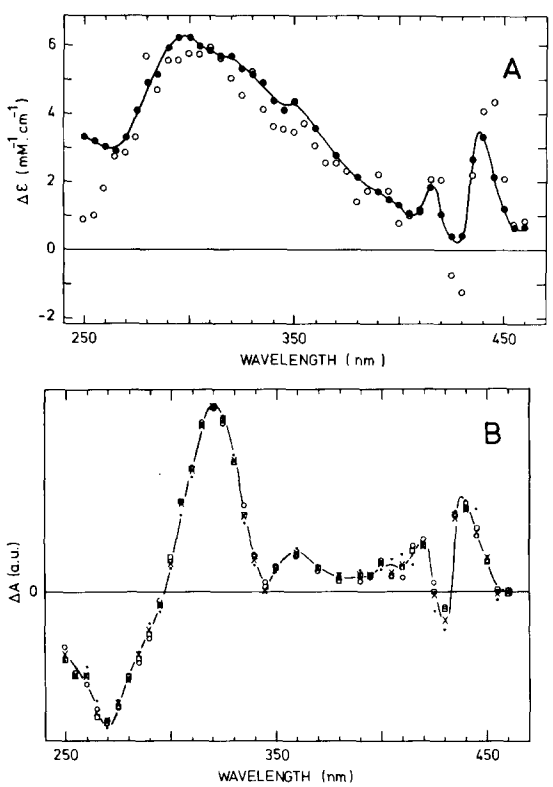

Fig. 7. (A) Spectrum attributed to each of the $S_0 \rightarrow S_1$, $S_1 \rightarrow S_2$ and $S_2 \rightarrow S_3$ transitions (solid circles), determined from Fig. 6 as described in the text. The open circles indicate the absorbance change due to the first flash in preparations with DCMU, from which the change due to Q reduction was subtracted (from Ref. 10). (B) Spectrum of the additional absorbance changes contained in the spectra of the $S_0 \rightarrow S_1$ (points), $S_1 \rightarrow S_2$ (crosses), $S_2 \rightarrow S_3$ (circles) and $S_3 \rightarrow S_0$ (squares) transitions of Fig. 6, normalized at 320 nm.

The most important conclusion from this analysis is that all S-state transitions are characterized by the same difference spectrum, with an amplitude sequence of +1, +1, +1, −3. Thus, if the earlier assignment of this difference spectrum to an Mn(III) to Mn(IV) transition is correct [10], each step in the advancement of the S-states involves the oxidation of one manganese from a valency of 3 to 4.

It should be kept in mind, however, that any change which appears and disappears alternatingly in the difference spectra of the successive S-states is included in the spectrum of Fig. 7B. Especially above 370 nm a contribution to this spectrum by differences between even- and odd-numbered S-states cannot yet be excluded. Such a contribution might be distinguished from the acceptor side changes by a slightly different oscillation pattern.

Fig. 8A shows that the observed oscillation

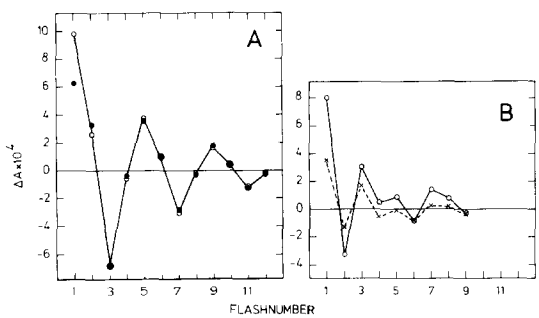

Fig. 8. (A) Comparison of the measured absorbance changes at 300 nm as shown in Fig. 5A (open circles) and the values calculated using the same parameters as in Fig. 3 and assuming that the absorbance change of Fig. 7A occurs in an amplitude sequence +1, +1, +1, −3 during the $S_0 \rightarrow S_1$, $S_1 \rightarrow S_2$, $S_2 \rightarrow S_3$ and $S_3 \rightarrow S_0$ transitions, respectively (solid circles). (B) Differences of the measured and simulated values at 320 nm (circles) and at 440 minus 430 nm (crosses).

pattern at 300 nm, near the isosbestic point of the acceptor side changes, is reproduced closely by the fit when a +1, +1, +1, −3 sequence for the donor side change is assumed. Again, only the first flash deviates considerably. Fig. 8B shows the deviation from this pattern at 320 nm (circles) and for the band shift (440 minus 430 nm, crosses). The somewhat irregular pattern at 320 nm, which was reflected also in the calculated contribution to each S-state transition, is followed by the oscillation of the band shift as well. Apparently, the band shift is caused by the secondary acceptor. The oscillating acceptor side contribution persisted between flashes; the pattern was the same at times from 10 to 150 ms after the flash (not shown).

Finally, the outcome of the analysis may be checked by the shape and amplitude it predicts for the difference spectrum of the 1–1.5 ms transient. The dashed line in Fig. 4A is the sum of 3-times the donor spectrum of Fig. 7A, plus the difference spectrum of Z (at pH 6.0) in a Tris-washed PS II preparation [10]. The similarity of these spectra confirms that the change calculated for the $S_3 \rightarrow S_0$ transition occurs during this kinetic phase, and also suggests that Z^+ is involved in the transient, as has been concluded on the basis of EPR measurements [14].

Discussion

In a previous study [10] we determined the difference spectrum of the ultimate electron donor in dark-adapted PS II preparations in the presence of DCMU, and interpreted this as an Mn(III) → Mn(IV) transition. The change was expected to reflect largely the $S_1 \rightarrow S_2$ transition in Kok's terminology [2], and its oscillation pattern in a series of flashes in the absence of DCMU [11,22] suggested qualitatively a sequence of 0, +1, 0, −1 for the occurrence of the absorbance change during the $S_0 \rightarrow S_1$, $S_1 \rightarrow S_2$, $S_2 \rightarrow S_3$ and $S_3 \rightarrow S_0$ transitions, respectively [11].

The quantitative analysis presented here shows that this sequence actually is +1, +1, +1, −3. The spectrum of the absorbance change was found to be the same for each S-state transition and corresponds, both in shape and in amplitude, to that reported previously [10], except below 260 nm. The oscillation pattern, difference spectrum and amplitude of the 1–1.5 ms transient, ascribed to the $S_3 \rightarrow S_0$ transition plus the reduction of the semiquinone cation Z^+, were consistent with these conclusions.

It has since long been assumed that manganese is in some way involved in photosynthetic oxygen evolution, and various figures have been reported for the amount of Mn per PS II reaction center [23]. Recent data tend to converge to a number of 4, in chloroplasts [24] as well as in oxygen-evolving PS II preparations from spinach [25,26] and from a cyanobacterium [27,28]. Yamamoto and Nishimura [29] reported a lower content, but may have overestimated the PS II reaction center concentration and do not present evidence that all centers were active in oxygen evolution. Klimov et al. [30] have claimed that two of the four manganese ions could be replaced by Mg^{2+}, and thus are not involved in the mechanism of oxygen evolution, but Velthuys [31] has offered a different explanation for those experiments.

Our results show that at least three Mn undergo redox changes in the mechanism of oxygen evolution, but a change of the fourth Mn was not detected. Nevertheless, one might speculate that its oxidation, like that of the other Mn ions, is required for oxygen evolution, and that this reaction determines the 1–1.5 ms halftime of the relaxation to S_0.

Our data do not seem to support any of the models proposed earlier, often based on less direct evidence (see for a review, Ref. 23). Further information on the manganese cluster may be provided by the multiline EPR signal, which is apparently specific for the S_2-state [5–7]. If it is ascribed to an Mn(III)/Mn(IV) dimer [9,5], the manganese atoms involved are those oxidized in the $S_1 \rightarrow S_2$ and $S_2 \rightarrow S_3$ transitions. It cannot be ascribed to an $Mn(III)_3/Mn(IV)$ cluster [8] if at least two Mn(IV) are present in the S_2-state.

Another possible source of information might be the Chl *a* band shift which accompanies the oxidation of manganese. Its amplitude may depend on the net charge of, or the charge distribution in the oxygen-evolving complex. Unfortunately, absorbance changes with a very similar shape are caused by the period-2 oscillation at the acceptor side, and an unambiguous quantitative interpretation of these band shifts is not yet possi-

ble. The larger amplitude of the shift attributed to the $S_1 \rightarrow S_2$ transition in Ref. 10 might indicate that the oscillation pattern of the shift is approx. 0, +1, 0, −1, due to neutralization by proton release on the $S_0 \rightarrow S_1$ and $S_2 \rightarrow S_3$ transitions [cf. 32]. This pattern is approached if about half of the amplitude of the shift now attributed to the acceptor side is assigned to the donor side, without changing the somewhat irregular period-2 oscillation.

This irregular period-2 oscillation is noteworthy in itself. No oscillation was observed after addition of the electron donor tetraphenylboron, while this compound in the absence of DCBQ was able to support a quite normal period-2 oscillation (Dekker, J.P., unpublished data). An oscillation pattern like that shown in Fig. 8B is obtained if the reduced secondary acceptor is partially oxidized by DCBQ between flashes in the state S_0 only. The influence of the S-states on electron transfer at the acceptor side, and the additional changes observed on the first flash, may be responsible for the fact that difference spectra previously attributed to the donor side of PS II [17,11] contain obvious contributions from the acceptor side. The complicated kinetics we observed at the acceptor side may in part be due to different accessibilities to DCBQ or other, perhaps artefactual, heterogeneities.

At the donor side, however, no complications were observed and the simple Kok scheme explains our data, if the S-state transitions $S_0 \rightarrow S_1$, $S_1 \rightarrow S_2$ and $S_2 \rightarrow S_3$ are all accompanied by the same absorbance changes in the ultraviolet, which probably reflect the oxidation of Mn(III) to Mn(IV). The fourth oxidant required for water oxidation is either too short-lived to detect, or is the oxidized secondary donor Z^+ itself, since the semiquinone cation and the three Mn(IV) ions are rereduced together in about a millisecond.

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