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Charge equilibrium between the water-oxidizing complex and the electron donor tyrosine-D in Photosystem II

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The decay kinetics for the S_2 and S_3 states of the water-oxidizing complex have been measured with an unmodulated Joliot-type oxygen electrode in isolated spinach thylakoids. The S_2 and S_3 states decay biphasically (Vermaas, W.F.J., Renger, G. and Dohnt, G. (1985) Biochim. Biophys. Acta 764, 194–202) with half-decay times of 1–1.5 s and 30–35 s at room temperature. The proportion of the fast phase is negligible in preilluminated thylakoids but increases during dark adaptation to 22–24% for both S_2 and S_3. This process, $t_{1/2} \approx 10$ min, is accompanied with the conversion of the S_0 state to S_1 in about 25% of the centers. Chemical reduction of tyrosine-D$^+$, which gives rise to the EPR Signals II_{slow}, by dichlorphenolindophenol/ascorbate increases the proportion of the fast decaying phase of S_2 and S_3 to about 70–80%. The decay of S_2 is accompanied by the accumulation of S_1 and the decay of S_3 results in a transient increase of S_2. These data led us to conclude that the fast phase in the S_2 and S_3 decay is correlated with one-electron donation from tyrosine-D to the water-oxidizing complex located within the same center. This process results in the $S_3D \rightarrow S_2D^+$ ($\rightarrow S_1D^+$) and $S_2D \rightarrow S_1D^+$ univalent sequences of deactivating reactions. The electron transfer from tyrosine-D to the S_2 and S_3 states is strongly temperature-dependent and shows 0.46 and 0.49 eV activation energy, respectively, over the +8 to +37°C temperature range. The deactivation process which is reflected by the slower phase of S_2 and S_3 decay has an activation energy of 0.65 and 0.76 eV, respectively. An extension of the Kok model of oxygen evolution is also presented taking into account the effect of fast electron donation from tyrosine-D to the water-oxidizing complex.

Introduction

Photosynthetic oxidation of water to molecular oxygen occurs via a four-step univalent reaction sequence that is catalysed by the Mn-containing water-oxidizing complex of Photosystem II (PS II). Recent experimental findings suggest that the D1/D2 protein complex of the PS II reaction center, which carries the reaction center chlorophyll P680 [1], provides binding sites for the catalytic Mn cluster required for water-oxidation [2–4]. The D1/D2 heterodimer contains also two redox active tyrosines. Z, tyrosine-161 in the D1 protein [5], mediates electron transfer between P680 and the water-oxidizing complex. D, tyrosine-160 in the D2 protein [6,7], is also involved in charge exchange with the water-oxidizing complex. Illumination of PS II oxidises these electron donors and induces characteristic EPR signals, called Signal $II_{very\text{-}fast}$ from Z^+ [8] and Signal II_{slow} from D^+ [9] having identical lineshape but different kinetics.

Flash-induced oxygen evolution reveals a characteristic period-four oscillation having the first maximum at the third flash [10]. This phenomenon reflects the cycling of water-oxidizing complex through five redox states, denoted S_0–S_4 [11]. The oxygen molecule is released in the S_3 to S_0 transition in which S_4 is a transient state. The stability and distribution of S states has been a central question in studies of the process of water-oxidation. In the light, S_0 to S_3 are equally populated, but S_2 and S_3 decays back to S_1 when the light is switched off [12]. This deactivation process results in an approx. 75% S_1 and 25% S_0 distribution observed normally after 3–5 min dark adaptation. In the original S-state model, both S_0 and S_1 were assumed to be stable in the dark [11]. The reality of dark stable S_0 state was questioned by Velthuys and Visser who proposed that the reduction of S_2 and S_3 by donor D might be

Abbreviations: Chl, chlorophyll; D1, *psbA* gene product; D2, *psbD* gene product; P680, primary electron donor of PS II; PS II, Photosystem II; Q_A, primary quinone acceptor of PS II; Q_B, secondary quinone acceptor of PS II; RC, reaction center of PS II.

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responsible for the apparent S_0 fraction present after dark adaptation [13]. Vermaas et al. found that indeed an almost 100% S_1 oxygen flash-pattern was observed in thoroughly dark adapted thylakoids if flashes were given 0.25 s apart while the usual 25% S_0 and 75% S_1 pattern was observed when flashes were separated by 2 s intervals [14]. This effect was correlated with donation of an electron from D in 25% of centers. It was also shown that S_0 was slowly converted to S_1 in the dark with a 10–15 min half-time [14]. Subsequently, Styring and Rutherford demonstrated by elegant EPR measurements that D^+ was the electron acceptor in this reaction, i.e., centers in the D^+S_0 state were converted to DS_1 during dark adaptation [15].

The dark decay of the S_2 and S_3 redox states to S_1 occur mainly in the range of tens of seconds, partially by recombination with electrons from the reduced electron acceptors [16]. In dark-adapted thylakoids a fast decaying phase of S_2 and S_3 was also observed ($t_{1/2} \approx$ 1–1.5 s), and was tentatively assigned to electron donation from D [14].

The characteristics and redox state of D have been studied mainly by measurements of EPR Signal II_{slow}. The above literature data, however, indicate a more or less direct effect of D on the distribution and stability of S_2 and S_3 as revealed by oxygen flash pattern. The quantitative establishment of such correlation is expected to provide an additional tool to monitor the redox state of D by simple flash-oxygen yield measurements.

In this paper we demonstrate that fast electron donation from D to the water-oxidizing complex is clearly related to the fast decaying phase of S_2 and S_3. We also present an extension of the conventional Kok-model of flash-induced oxygen evolution to interpret quantitatively the effect of electron donation from D on flash-oxygen sequences.

Materials and Methods

Thylakoids were isolated from market spinach as described earlier [17], resuspended in 0.4 M sorbitol, 5 mM $MgCl_2$, 10 mM NaCl, 1 mM $MnCl_2$ 40 mM Hepes (pH 7.5) at 2–3 mg Chl/ml and stored at $-70\,^\circ$C until use. For chemical reduction of D^+ 75 μM DCPIP and 0.75 mM ascorbate was added to the thylakoid suspension just before the oxygen measurement.

Flash-induced oxygen yield was measured with an unmodulated Joliot-type O_2 electrode [18], constructed similarly as in Ref. 19. Thylakoids were diluted to 0.75 mg/ml with the above medium and transferred to the Pt electrode in very dim green light. After 3–5 min dark period, required for the equilibrium of the O_2 electrode, thylakoids were illuminated by a train of saturating Xe flashes (General Radio Stroboslave, 3 μs, 0.5 J). In experiments with short dark adaptation, thylakoids were illuminated by 50 preflashes on the electrode before measurement. For longer than 5 min dark adaptation, preillumination was done with continuous white light for 30 s in a flat petri-dish during continuous stirring. Thylakoids were then distributed to 50 μl aliquots in the dark, kept at room temperature for the required period of time. Dark-adapted samples were stored at $-30\,^\circ$C in order to keep them in the same condition until measurements were performed within 4–5 h. The amplified oxygen signals were fed to a multichannel analyzer (ICA70 KFKI, 2.5–10 ms/point). The measurements were controlled by a Commodore 64 computer which was also used for calculation and storage of oxygen yields.

For the measurements of S_2 and S_3 decay thylakoids were preilluminated with one or two flashes (4 Hz), respectively, followed by various intervals of dark relaxation and then by a train of ten measuring flashes (4 Hz). The relative concentration of S_2 was calculated from the normalized oxygen yield in the second flash of the flash train (Y_2) corrected for the Y_2 obtained without preflashes ($Y_{2,c}$) i.e. $S_{2,rel} = Y_2 - Y_{2,c}$. The relative amount of S_3 was obtained as $S_{3,rel} = Y_1$. The oxygen yields were all normalized to the average yield obtained between the third and sixth flashes, including the preflashes. In some experiments the fraction of centers in the S_1 state was also calculated from the oxygen sequences obtained after one preflash taking into account the effect of misses. The S_2 values produced via consecutive reaction from S_3 were calculated as indicated above from oxygen sequences measured after two preflashes. The calculated S_2 and S_3 decay curves were resolved into exponential components by a least-squares fit computer program.

Experimental oxygen yield sequences were analyzed by a simplex least-squares fit method minimizing the squared difference-sum between the experimental and theoretical oxygen-yield sequences up to 20 flashes. Theoretical oxygen-yield sequences were calculated by using the matrix formalism of S-state turnovers [20,21] either with the conventional Kok model or with its extension including the charge equilibrium with D as discussed later.

Results and Discussions

Effect of dark storage on S_2 and S_3 decays

Vermaas et al. reported that in thoroughly dark-adapted pea thylakoids the decay of S_2 and S_3 states showed an about 25% fast phase ($t_{1/2} = 1$–1.5 s) [14]. This phenomenon was suggested to reflect an electron transfer process from a reduced one-electron donor of PS II to the water-oxidizing complex [14]. In order to study the formation of this electron donor we measured the decay course of the S_2 and S_3 states after various dark-adaptation periods. As Fig. 1 shows, both S_2 and

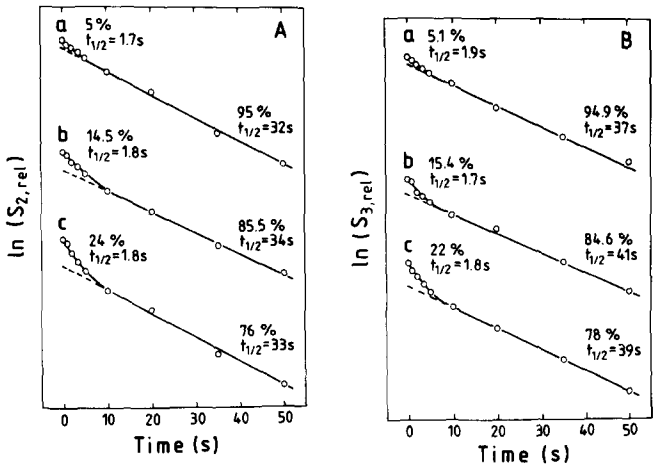


Fig. 1. Decay kinetics of the S_2 (A) and S_3 (B) states in spinach thylakoids dark adapted for 3 (a), 8 (b) and 30 (c) min. Relative amounts of S_2 and S_3 were calculated from oxygen-yield sequences measured at 22°C after one and two preflashes, respectively as described in Materials and Methods.

S_3 decay biphasically as reported earlier [14], having a fast component of 1.7–1.9 s half-time and a slow component of 32–41 s half-time. These half-times vary somewhat between different preparations and also with the temperature (see below). The amplitude of the fast decaying phase of both S_2 and S_3 increases from about 5% to 22–24% during 30 min dark adaptation (Fig. 1A and B). Fig. 2 shows that the real, frequency independent S_0 population, estimated by fitting of oxygen yield sequences obtained with 4 Hz flash frequency, decreases with about 10 min half-time during the dark adaptation. The amplitude of the fast phase in S_2 and S_3 increases in a complementary way. These data demonstrate that the reduced form of the donor species which quickly deactivates S_2 and S_3 by electron donation is formed in the dark concomitantly with the slow conversion of S_0

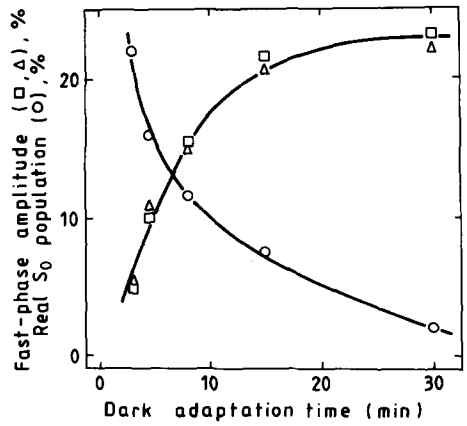


Fig. 2. Dependence of S_0 population and fast phase of S_2 and S_3 decays on dark adaptation time. The fraction of centers in the S_0 state (○) was calculated from the experimental oxygen yield sequences obtained with 4 Hz flash frequency, using the conventional Kok model. The amplitude of the fast decaying phase of S_2 (□) and S_3 (△) was obtained as shown in Fig. 1.

to S_1. Styring and Rutherford have recently shown that D^+ is slowly reduced to D by S_0 during dark storage of thylakoids ($t_{1/2} \approx 20$ min) [15]. The comparison of our data with this finding indicates that fast electron donation from D to S_2 and S_3 is responsible for their fast decaying phase. This conclusion is corroborated by the results of Babcock and Sauer, who showed that D^+ is formed with about 1 s half-time after one flash in chloroplasts prepared from dark-stored spinach leaves [9]. It is of note, however, that the half-time of charge recombination between S_2 and Q_A^- is also about 1–2 s [14,22] and a similar half-time is found for the $S_3Q_A^-$ recombination [23]. In thylakoid preparations only in the minority of the centers (in which the acceptor side is damaged and Q_A is disconnected from Q_B) has S_2 or S_3 reduced Q_A as counterpart, i.e., only a minor fraction of the fast phase can be assigned to charge recombination with reduced Q_A. This effect, however, should not be neglected if the amount of Q_A^- is high, as in the case of triazine-resistant thylakoids [14].

The amplitude of the fast-decaying phase of S_2 and S_3 is saturated at around 22–24% after 30 min dark adaptation as shown above. This agrees with the reduction of D^+ in the S_0 centers. However, when thylakoids were stored at −70°C for a long time a much larger fast phase appeared in the decay of both S_2 and S_3 (70–80% after 6 months). The first maximum of the oxygen flash pattern was also shifted from the third flash to the fourth flash when measured with 0.5 Hz flash frequency but not with 4 Hz (not shown). This indicates a very slow reduction of D^+ at −70°C even in the S_1 centers by an as yet unspecified donor.

Chemical reduction of D^+

Addition of DCPIP/ascorbate is known to reduce D^+ [13]. In these conditions the first maximum of the oxygen flash pattern appears at the 4th flash when measured with 0.5 Hz (Fig. 3, circles), in agreement with data published earlier [13,24]. When flashes were fired with 4 Hz the first oxygen maximum was shifted back to the 3rd flash (Fig. 3, triangles) indicating a large population of reduced D. Illumination with 50 preflashes did not restore the normal flash pattern when it was measured with 0.5 Hz (Fig. 3, squares). This shows that D^+ formed during illumination is rereduced by DCPIP/ascorbate during the 3 min dark period preceding the measuring flash series. DCPIP/ascorbate addition increased also the fast decaying phase to about 80% both in S_2 (Fig. 4A) and S_3 (Fig. 4B), which gives further support for the assignment of the fast decay to the electron donation from D to the water-oxidizing complex. The equal amplitude of the fast phase of S_2 and S_3 decay shows also that electron donation from D occurs only to the water-oxidizing complex located in the same center. If D could donate to other centers as well, e.g., via a mobile carrier, then the S_2 centers just

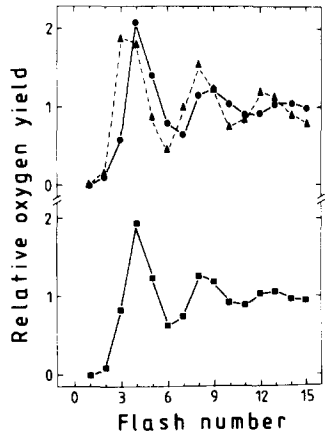


Fig. 3. Effect of chemical reduction of D^+ on oxygen production by spinach thylakoids. Samples were incubated with 0.075 mM DCPIP and 0.75 mM ascorbate. Oxygen evolution was measured with 0.5 (●) and 4 (▲) as well as with 0.5 Hz flash frequency after 50 preflashes (■).

formed from S_3 would also be reduced by D. This process would obviously decrease the D population available for S_3 reduction and would result in an about 50% decrease in the amplitude of fast phase in S_3 decay as compared to that of S_2. It should be noted that half-times of both the fast and slow phases of S_2 and S_3 decay in Fig. 4A and B are shorter than as shown in Fig. 1. This is due mainly to the higher temperature (28° C) during the measurement shown in Fig. 4A and 4B, compared to that of in Fig. 1 (22° C).

With respect to the pathway of S-state deactivations most of the data support the $S_3 \rightarrow S_2 \rightarrow S_1$ univalent deactivation sequence [25–27]. Other results on *Chlorella*, however, favour the more complex $S_3 \rightarrow S_1$ and $S_2 \rightarrow S_0$ bivalent sequence, coupled via an unknown component C [28]. As Fig. 4A shows the fast and slow decaying phase of S_2 (curve A) is accompanied with a fast and slow increase of S_1 (curve b), respectively. The decay of S_3, however, results in a transient increase of S_2 (Fig. 4B curves a and b, respectively). These results imply that the fast reduction of S_3 by D results in the formation of slowly decaying S_2D^+ and thus the transient accumulation of S_2. Similarly, the fast decay of S_2 leads to the accumulation of stable S_1D^+ state, and the slow decay of S_2D^+ increases further the S_1D^+ population. This interpretation demonstrates the validity of the earlier suggested one-electron characteristics of D [14], and confirms also that D donates to the water-oxidizing complex located in the same center as concluded from the equal amplitude of the fast phase in S_2 and S_3 (Figs. 1A, B and 4A, B).

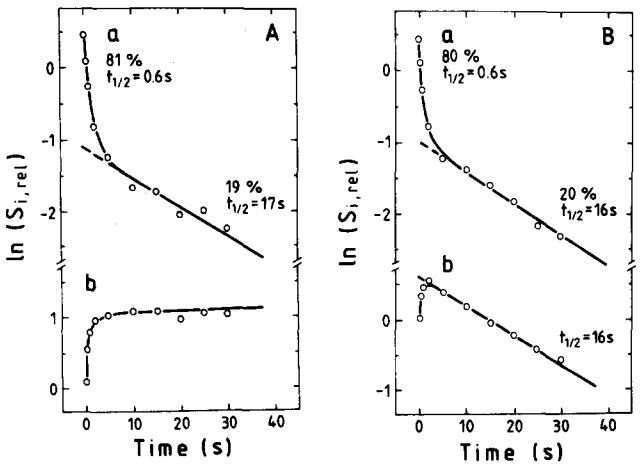


Fig. 4. Effect of chemical reduction of D^+ on the kinetics of the S_2 to S_1 and S_3 to S_2 conversion in spinach thylakoids. Samples were incubated with 0.075 mM DCPIP and 0.75 mM ascorbate. (A) The amount of S_2 (a) and S_1 (b) was calculated from oxygen-yield sequences measured after one preflash followed by various dark periods. (B) The amount of S_3 (a) and S_2 (b) was obtained from oxygen-yield sequences measured after two preflashes as described in Materials and Methods. The measurements were done at 28° C.

TABLE I

Effect of temperature on the half-times of the fast and slow phases of the S_2 and S_3 decays in isolated spinach thylakoids

Temperature (° C)	Half-times (s)			
	$S_{2,fast}$	$S_{2,slow}$	$S_{3,fast}$	$S_{3,slow}$
8.5	5.0	128	3.9	238
15.0	2.2	67	2.1	108
23.5	1.4	29	1.6	47
28.5	0.94	22	1.1	24
32.0	0.89	16	0.87	19
37.0	0.70	15	0.55	12

Temperature-dependence of S_2 and S_3 deactivations

In order to obtain more detailed information about the deactivation reactions of S_2 and S_3 states, the temperature-dependence of these processes was studied between 8 and 37° C. The half-decay times obtained reveal a strong temperature-dependence of both the fast and slow phases of S_2 and S_3 states (Table I). The Arrhenius plot of the half-times is shown in Fig. 5. The calculated activation energies are 0.49 and 0.46 eV for the fast phases, 0.65 and 0.76 eV for the slow phases of S_2 and S_3, respectively. The temperature-dependence for the direct electron transfer between D and the Mn cluster has recently been reported by Inui et al. [30] based on EPR measurements of Signal II_{slow}. The activation energy they obtained is 16.6 kJ/mol (0.172 eV) [30] which is about half our value. This difference probably arises from the different temperature ranges studied. Inui et al. [30] measured electron donation from D below −23° C, i.e., in the frozen state of samples. In our measurements, however, samples were in liquid phase between 8 and 37° C. Another possibility is that the difference in the experimental material and pH, PS II membrane preparations at pH 6.9 in Ref.

30 and thylakoids at pH 7.5 in our study, is responsible for the different activation energy.

The slower phase of S_2 and S_3 decay reflects deactivation which occurs partially by back reactions with electrons from the reduced electron acceptors [16]. The about 30 s half-decay time indicates that this acceptor is most probably Q_B^-, which recombines with S_2 and S_3 with similar half-decay time [22] at room temperature. The activation energies obtained for the $S_{2(3)}Q_B^-$ recombination from thermoluminescence measurements are in the range of 0.7–1.0 eV [31,32] in different preparations. Analysis of thermoluminescence curves measured on the same batch of thylakoids as the oxygen-yield sequences (not shown) resulted 0.62 and 0.65 eV activation energy for the $S_2Q_B^-$ and $S_3Q_B^-$ recombinations, respectively. The temperature-dependence of S_2 decay was also measured by detecting the decay of the so called S_2-multiline EPR signal [33]. The half-time of S_2 decay was 40 s at 22 °C and about 120 s at 6 °C [33], which data are in good correlation with our results. Via recombination with Q_B^-, however, only part of S_2 and S_3 can directly decay. In centers where Q_B is oxidized after the flashes another electron donor deactivates the S_2 and S_3 states. This could be plastoquinol, as suggested by Rutherford et al. [34]. If no electrons are available on Q_B and in the plastoquinone pool, as in the presence of phenylparabenzoquinone, the stability of S_2 and S_3 was shown to increase to 3–4 min in PS II membranes [27].

It is of note that the measured oxygen sequences showed an increased probability of misses with increasing temperature (from 0.11 at 8 °C to 0.15 at 35 °C), in agreement with earlier data [29].

Effect of tyrosine-D on S-state turnovers

The complex charge equilibrium between D and the water-oxidizing complex is summarized in Scheme 1. The stability of D in the S_1 and that of D^+ in the S_1 and S_2 states shows that

$$k_{10} \ll k_{01},\ k_{21} \gg k_{12} \text{ and } k_{32} \gg k_{23}.$$

From the similar decay times of the fast phase of S_2 and S_3 it follows that

$$k_{21} \approx k_{32} = k_D.$$

During illumination, D is oxidized; thus the $S_0D^+, \ldots, S_3D^+$ states are equally populated. After switching off the light S_3D^+ and S_2D^+ decays back to S_1D^+ resulting in an approx. 25% S_0D^+ and 75% S_1D^+ distribution after 3–5 min dark-adaptation. Prolonged dark-adaptation leads to the slow conversion of S_0D^+ to S_1D ($t_{1/2} = 10$–20 min), and eventually to an about 75% S_1D^+ and 25% S_1D distribution. Flash illumination induces S-state turnovers in both populations. The formation of S_2D and S_3D is followed by their fast decay to S_1D^+ and S_2D^+, respectively. The amount of centers converted during the Δt time interval between two flashes is given by

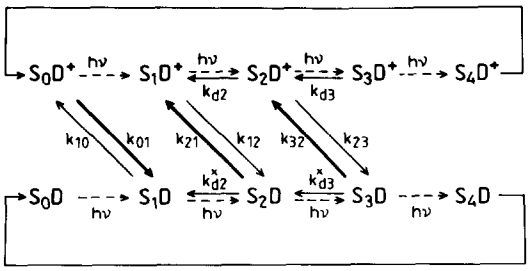
$$\Delta S_{2(3)} = [S_{2(3)}D]_0 \cdot (1 - \exp(-k_D \cdot \Delta t))$$

where the 0 subscript indicates the amount of centers just after firing the flashes. This process decreases the $S_{2(3)}D$ population and increases the $S_{1(2)}D^+$ population to the same extent

$$[S_{2(3)}D] = [S_{2(3)}D]_0 - \Delta S_{2(3)}$$

$$[S_{1(2)}D^+] = [S_{1(2)}D^+]_0 + \Delta S_{2(3)}$$

resulting eventually in the oxidation of all D's. If the repetition time of flashes is long enough relative to the half-time of electron donation, then S_2D is converted to S_1D^+ between the first and second flash, which mimics an apparent S_0 fraction in the oxygen yield sequence. When a short repetition time is used, the decay of S_2D to S_1D^+ is not significant between the first two flashes and several flashes are needed to oxidize D by S_2 and S_3. This process induces a small increase in the calculated miss factor rather than a large increase in S_0.



Scheme 1

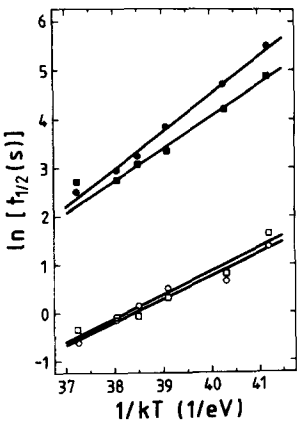


Fig. 5. Arrhenius plots of S_2 and S_3 half-decay times in isolated spinach thylakoids. Decay kinetics of the S_2 (□,■) and S_3 state (○,●) was measured as shown in Fig. 1. The logarithms of half-times obtained for the fast (□,○) and slow phases (■,●) were plotted as a function of $(kT)^{-1}$, where k is the Boltzmann's constant and T is the temperature.

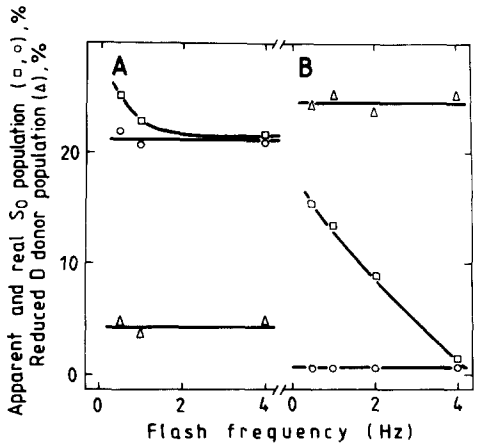


Fig. 6. Flash frequency dependence of the apparent and real S_0 populations as well as the reduced D fraction in spinach thylakoids. Oxygen flash sequences were measured after 3 (A) or 30 min (B) dark-adaptation with flash frequencies varying between 0.5 and 4 Hz. The apparent S_0 fraction (□) was calculated by using the conventional Kok model. The amount of real S_0 (○) and reduced D (△) was calculated by the extension of the Kok model presented in the text.

For the analysis of oxygen flash patterns measured in samples with large population of reduced D the conventional Kok model was extended according to Scheme I. Besides the parameters of the Kok model (miss, single hit, S_0 fraction of centers) two additional parameters were introduced, the amount of centers having reduced D and the rate constant of electron donation from D. Theoretical oxygen-yield sequences were calculated applying the matrix formalism of S-state turnovers [20,21] taking into consideration the conversion of the S_nD centers to $S_{n-1}D^+$ centers during the dark period between flashes. The results of fitting oxygen sequences with the conventional and extended Kok model is summarized in Fig. 6. The real S_0 population (circles) and the amount of reduced D (triangles) is independent of flash frequency either after 3 (Fig. 6A) or 30 min (Fig. 6B) dark adaptation. The apparent S_0 (squares) population obtained with the conventional Kok model, however, shows a weaker or stronger frequency dependence after 3 or 30 min dark relaxation, respectively. It can also be seen that calculation of S_0 concentration by the conventional Kok model from oxygen patterns obtained with 4 Hz flash frequency gives a good estimation of the real, frequency-independent S_0 population. The amount of D and the rate constant of fast electron donation obtained from the fit of oxygen flash patterns are in fair agreement with those calculated from the S_2 and S_3 decay curves.

Concluding remarks

The flash-oxygen data presented here confirm earlier observations concerning the conversion of the S_0 state to S_1 [14] by reducing D^+ during prolonged dark-adaptation [15]. We also demonstrate that the fast phases of the S_2 and S_3 decays appear in parallel with the dark conversion of S_0 to S_1, and reflects a fast one-electron donation from D to the water-oxidizing complex located within the same center. Thus, the amplitude of the fast decaying phase of S_2 and S_3 provides a sensitive indicator of the redox state of D, which probably plays an important role in stabilizing the catalytic Mn cluster of water-oxidation [15].

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