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# PICOSECOND FLUORESCENCE KINETIC STUDIES OF ELECTRON ACCEPTOR Q REDOX HETEROGENEITY

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**We have investigated the influence of chloroplast organization on the nature of chemical reductive titrations of Photosystem II fluorescence decay kinetics in spinach chloroplasts. Structural changes of the chloroplast membrane system were induced by varying the ionic environment of the thylakoids. A single-photon timing system with picosecond resolution monitored the kinetics of the chlorophyll *a* fluorescence emission. At all ionic concentrations studied, we have observed biphasic potentiometric titration curves of fluorescence yield; these have been interpreted to be suggestive of electron acceptor Q heterogeneity (Karukstis, K.K. and Sauer, K. (1983) Biochim. Biophys. Acta 722, 364–371; Cramer, W.A. and Butler, W.L. (1969) Biochim. Biophys. Acta 172, 503–510). A direct relation is observed between the $E_m$ value of the low-potential component of Q and the $Mg^{2+}$ concentration of the chloroplast suspending medium. We have attributed these midpoint potential variations to the thylakoid structural rearrangements involved in cation-regulated grana stacking. Ionic effects on the fluorescence decay kinetics at the redox transitions are discussed in terms of the heterogeneity of Photosystem II units ($\alpha$- and $\beta$-centers) and the mechanism of deexcitation at a closed reaction center (fluorescence or nonradiative decay).**

## Introduction

The chlorophyll fluorescence yield in higher plants varies with the state of the photochemical reaction centers in PS II. Changes in the fluorescence yield are correlated with the redox state of the PS II electron acceptor Q [1]. The transition of a PS II reaction center from a photochemically active open state to a photoinactive closed state upon chemical reduction or photoreduction of Q increases the fluorescence yield from a minimal level, $F_0$, to a maximum, $F_{max}$. Chemically induced titrations of fluorescence yield permit the degree of Q reduction to be controlled by the amount of reductant added to poise the redox potential of the chloroplast sample. Chemical reductive titrations of fluorescence yield in chloroplasts at room temperature reveal two distinct redox components designated as the high- and low-potential forms of Q, $Q_H$ and $Q_L$, respectively. This biphasic character of the potentiometric titration of chlorophyll fluorescence yield has been interpreted to be indicative of PS II electron acceptor Q heterogeneity [2–10].

Various hypotheses have been proposed to account for the origin of this electron acceptor heterogeneity [4,11–14]. One model [4,11] assigns $Q_H$ and $Q_L$ as the electron acceptors of PS II reaction centers in separate units ($\beta$-centers) and in statistical pigment beds ($\alpha$-centers), respec-

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Abbreviations: DCMU, 3-(3′,4′-dichlorophenyl)-1,1-dimethylurea; Hepes, *N*-2-hydroxyethylpiperazine-*N*′-2-ethanesulfonic acid; $E_m$, reduction potential midpoint; PS, photosystem; Chl, chlorophyll.

tively. A second model [4,12–14] attributes the $Q_H$ and $Q_L$ transitions to closure of physically distinct PS II reaction centers, with $Q_H$ in the unstacked thylakoids exposed to the aqueous environment of the stroma and $Q_L$ in the partition regions of the grana stacks. These two models may be different descriptions of the same phenomena as a consequence of the recent proposal that $\beta$-centers are located in thylakoid membranes directly exposed to the stroma while $\alpha$-centers are in the partitions of the grana [15,16]. A third model [5] equating $Q_H$ and $Q_L$ to the singly ($Q^-$) and doubly reduced ($Q^{2-}$) forms of the quinone acceptor is not widely held [3–5]. Finally, the redox interaction of two closely associated PS II reaction centers in the grana to 'create' $Q_L$ has been invoked to explain the existence of an additional acceptor [14]. In such a model the increased interaction associated with membrane stacking leads to delocalization of electronic charge; and, as a consequence, the $Q_H$ transition in grana corresponds to $[QQ] \xrightarrow{e^-} [QQ]^-$ and the associated $Q_H$ transition to $[QQ]^- \xrightarrow{e^-} [QQ]^{2-}$.

Correlations of the existence of $Q_H$ and $Q_L$ to structural and compositional differences in chloroplasts have been used to evaluate the nature of the electron acceptor heterogeneity. The distinct membrane organizations of $MG^{2+}$-depleted chloroplasts, deficient in thylakoid stacks, and $Mg^{2+}$-enriched chloroplasts, with both granal and stromal membranes, are ideal systems to investigate the effect of PS II organization and structure on the redox properties of the electron acceptor Q. An earlier study [18] of the organizational differences of $Mg^{2+}$-depleted and -enriched chloroplasts has been successfully performed with measurements of chlorophyll fluorescence decay kinetics as a function of illumination conditions, revealing organizational effects on fluorescence which occur at different levels of added cation. In the absence of added $Mg^{2+}$, communication between PS II and PS I in the form of excitation energy transfer is at its highest level and decreases with the addition of $Mg^{2+}$, saturating at $[Mg^{2+}] = 0.75$ mM. These changes are reflected in an increase in the lifetime of the slow decay component of fluorescence at $F_0$ and $F_{max}$. At higher levels of $Mg^{2+}$ (greater than 2 mM), communication between PS II units develops and the effective absorption cross-section of PS II increases, as exemplified by increases in the $F_{max}$ level slow phase yield and decreases in the $F_0$ level slow phase lifetime with increasing $Mg^{2+}$ concentration.

Thus, an analogous study of the fluorescence decay kinetics during potentiometric titrations of Q at variable levels of $Mg^{2+}$ offers a possibility for distinguishing the effect of chloroplast organization on Q heterogeneity. We have recently reported the results of a potentiometric titration of the fluorescence decay kinetics in $Mg^{2+}$-enriched (5 mM) chloroplasts [10]. In the present paper we present an analysis of the effect of reduction potential on the fluorescence decay components of chloroplasts isolated in the absence of added $Mg^{2+}$ and in the presence of 0.50 mM $Mg^{2+}$. We use a comparison of results at all levels of cation concentration to discuss the possible origin of the PS II electron acceptor heterogeneity.

## Materials and Methods

Broken chloroplasts were isolated from growth-chamber spinach by grinding depetioled leaves in a blender for 10 s in a medium of 0.4 M sucrose, 50 mM Hepes-NaOH, pH 7.5, and 10 mM NaCl, followed by centrifugation at 6000 × *g* for 10 min. A wash with fresh grinding medium was followed by centrifugation under the same conditions. The pellet was resuspended in a medium of 0.1 M sucrose, 10 mM Hepes-NaOH, pH 7.5, and 10 mM NaCl, and then centrifuged at 6000 × *g* for 10 min. The isolated chloroplasts were resuspended in a medium of 0.1 M sucrose, 50 mM Hepes-NaOH, pH 7.5, 5 mM NaCl, and the desired $MgCl_2$ concentration (either 0 or 0.50 mM) to give approx. 1 mg Chl/ml. The chloroplasts were allowed to equilibrate in these buffers for at least 1 h. For fluorescence measurements the chloroplast suspension was diluted with this final buffer, deaerated with nitrogen gas, to a concentration of 17 μg Chl/ml.

The reduction potential of the medium was measured using a PAR model 173 potentiostat and a platinum electrode with an Ag/AgCl electrode (saturated KCl solution) as reference, calibrated against a quinhydrone electrode. All reduction potentials are reported with respect to the standard hydrogen electrode (pH 0). The reduction potential of the suspension was adjusted under anaerobic conditions by additions of solid dithionite or small aliquots of 250 mM potassium ferricyanide (equilibration time 15 min). Redox mediators included indophenol ($E_{m,7} = +228$ mV), 1,4-naphthoquinone (+60 mV), duroquinone (0 mV), indigotetrasulfonate (−46 mV), 2,5-dihydroxybenzoquinone (−60 mV), indigotrisulfonate (−81 mV), indigodisulfonate (−125 mV), anthraquinone-2,6-disulfonate (−184 mV), and anthraquinone-2-sulfonate (−225 mV).

The fluorescence excitation source was a Spectra-Physics synchronously pumped, mode-locked dye laser (SP 171 argon ion laser, SP 362 mode locker, and modified SP 375 dye laser). Chloroplast samples were excited with output pulses at 620 nm with a full-width half-maximum duration of 8 ps. We used a low excitation intensity to measure the initial level of fluorescence and to prevent appreciable steady-state reduction of Q. (A fluorescence photon count ratemeter monitored the fluorescence intensity during an experiment.) Fluorescence at 680 nm was detected at right angles. The single-photon timing system and numerical analysis methods have been discussed previously [18–20]. All fluorescence decay data were resolved into a sum of three exponential decays with a lifetime resolution limit of 25 ps. An average lifetime of a fluorescence decay is calculated using the equation

$$\tau_{\mathrm{mean}} = \sum_{i=1}^{3} \alpha_i \tau_i$$

where $\alpha_i$ and $\tau_i$ are the amplitude and lifetime of the $i$-th component, respectively.

## Results

### *Titration in the Absence of Added $Mg^{2+}$*

Fig. 1 presents the potentiometric titration of the total fluorescence yield and the yield of the three fluorescence decay components for spinach chloroplasts at pH 7.5 in the absence of $Mg^{2+}$. Both the oxidative and reductive titrations are reversible processes, verifying that stable reduction potentials had been attained. The total fluorescence yield increases 2.4-fold as the reaction centers are closed upon chemical reduction. The solid curve through the experimental total yield data represents Nernst behavior for two successive one-electron redox transitions previously attributed to $Q_H$ and $Q_L$, the high- and low-potential components of Q, respectively. The midpoint reduction potentials, $E_{m,7.5}$ values, are +99 and −295 mV for $Q_H$ and $Q_L$, respectively, with $Q_H$ accounting for 57% of the variable fluorescence and $Q_L$ for 43%. Changes in the slow phase yield are also fitted to a composite of two one-electron transitions with $E_{m,7.5} = +113$ and −287 mV and with corresponding relative contributions to the fluorescence yield of 55 and 45%. There is a 6-fold increase in the slow phase yield as the reaction centers are closed. The yield of the fast decay component is essentially insensitive to the state of the PS II reaction center, while the middle phase yield increases by 30% as the potential is lowered.

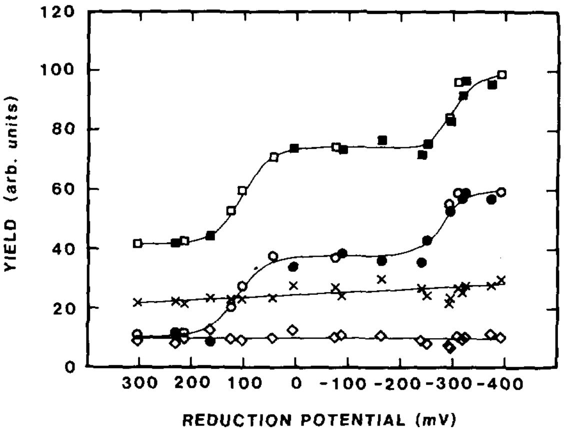

Fig. 1. Potentiometric titration of the total fluorescence yield and the yields of the components of the fluorescence decay in spinach chloroplasts at pH 7.5 in the absence of added $Mg^{2+}$. (□) Total fluorescence yield; (○) yield of the slow phase; (×) yield of the middle phase; and (◇) yield of the fast phase. For the total and slow phase yields, open symbols, oxidative titration; closed symbols, reductive titration.

The lifetimes of the three components of the fluorescence decay as functions of reduction potential are plotted in Fig. 2. The slow phase has a constant lifetime of approx. 1500 ps during $Q_H$ reduction and increases to about 1700 ps during $Q_L$ reduction. The middle phase lifetime increases during $Q_H$ reduction from approx. 350 to 500 ps. The lifetime of the fast phase remains constant at

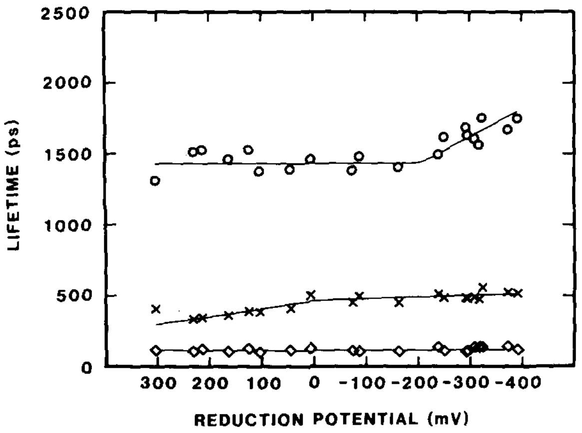

Fig. 2. Lifetimes of the components of the fluorescence decay in spinach chloroplasts at pH 7.5 in the absence of $Mg^{2+}$ as a function of the reduction potential of the medium. (○) Lifetime of the slow phase; (×) lifetime of the middle phase; (◇) lifetime of the fast phase.

approx. 120 ps. The mean lifetime doubles during the course of Q reduction from 600 to 1200 ps. These effects are essentially the same as those observed in a recent investigation [18] of the state of the PS II reaction center of $Mg^{2+}$-depleted chloroplasts as determined by illumination conditions.

*Titration in the Presence of 0.50 mM* $Mg^{2+}$

Fig. 3 presents the potentiometric titration of the total fluorescence yield and the yields of the three fluorescence decay components for spinach chloroplasts isolated at pH 7.5 in the presence of 0.50 mM $Mg^{2+}$. The equivalent titrations in the presence and absence of redox mediators verify that stable reduction potentials were attained. The redox transitions of the total fluorescence yield are best described by the sum of two one-electron transitions with $E_{m,7.5} = +70$ and $-345$ mV. $Q_H$ contributes 55% to the total variable fluorescence and $Q_L$, 45%. Two successive increases in the slow phase yield exactly parallel those of the total yield, with $E_{m,7.5} = +70$ and $-346$ mV and relative amplitudes of 54 and 46%, respectively. The fluorescence yield of the fast component is independent of the redox state of the reaction center, although the yield of the middle component increases by 30% as the reaction centers are closed. The dependence on reduction potential of the fluorescence lifetimes of the decay components is presented in Fig. 4. The lifetime of the fast component is constant at approx. 100 ps and that of the middle phase is also relatively constant at 400–500 ps. An increase in the lifetime of the slow component from 1100 to 1500 ps occurs during the $Q_H$ transition, while a lifetime in the range of 1500–1700 ps is characteristic of the $Q_L$ transition. The mean lifetime increases from 600 to 1200 ps

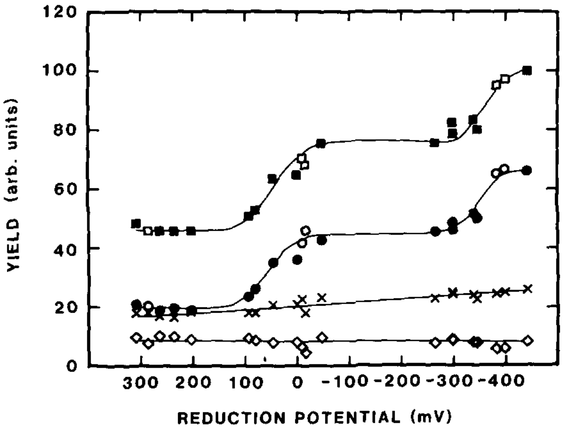

Fig. 3. Potentiometric titration of the total fluorescence yield and the yields of the components of the fluorescence decay in spinach chloroplasts at pH 7.5 in the presence of 0.50 mM $Mg^{2+}$. (□) Total fluorescence yield; (○) yield of the slow phase; (×) yield of the middle phase; and (◇) yield of the fast phase. For the total and slow phase yields, open symbols, no redox mediators; closed symbols, redox mediators present.

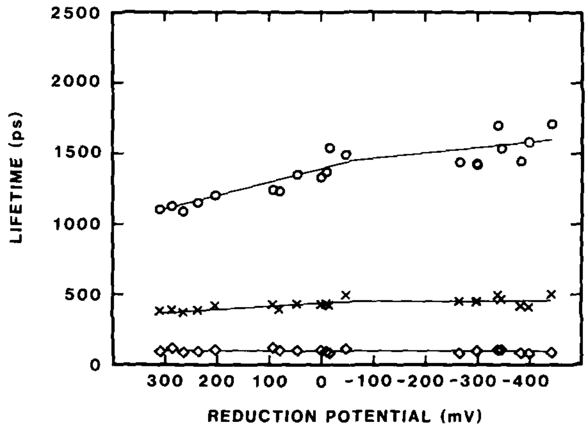

Fig. 4. Lifetimes of the components of the fluorescence decay in spinach chloroplasts at pH 7.5 in the presence of 0.50 mM $Mg^{2+}$ as a function of the reduction potential of the medium. (○) Lifetime of the slow phase; (×) lifetime of the middle phase; and (◇) lifetime of the fast phase.

TABLE I

CHARACTERISTICS OF THE POTENTIOMETRIC TITRATION OF ELECTRON ACCEPTOR Q AS A FUNCTION OF $Mg^{2+}$ CONCENTRATION

$E_{m,7.5}$ values relative to the standard hydrogen electrode.

| $[Mg^{2+}]$ (mM) | $Q_H$ | | | | $Q_L$ | | | |
|---|---|---|---|---|---|---|---|---|
| | $\phi_{total}$ | | $\phi_{slow}$ | | $\phi_{total}$ | | $\phi_{slow}$ | |
| | $E_{m,7.5}$ (mV) | % | $E_{m,7.5}$ (mV) | % | $E_{m,7.5}$ (mV) | % | $E_{m,7.5}$ (mV) | % |
| 0 | +99 | 57 | +113 | 55 | −295 | 43 | −287 | 45 |
| 0.50 | +70 | 55 | +70 | 54 | −345 | 45 | −346 | 46 |
| 5.0 | +106 | 50 | +119 | 54 | −384 | 50 | −395 | 46 |

as the reaction centers are closed.

Table I summarizes the results of this study for the potentiometric titration of fluorescence decay kinetics for $Mg^{2+}$-depleted chloroplasts and chloroplasts isolated in the presence of 0.50 mM $Mg^{2+}$. Included for comparison are the results of the earlier [10] potentiometric titration of the fluorescence decay kinetics of $Mg^{2+}$-enriched chloroplasts isolated in the presence of 5.0 mM $Mg^{2+}$.

## Discussion

### *Cationic Changes in Chloroplast Structure and Function*

The stacking of thylakoid membranes to form grana is induced in higher plants by low concentrations of divalent cations (2–5 mM) or high concentrations of monovalent cations (100 mM) [21–26]. In addition to the formation of appressed thylakoids in grana, it has been generally held that a conversion of isolated PS II β-centers into communicating PS II α-centers occurs upon addition of $Mg^{2+}$ via enhanced coupling of the light-harvesting Chl *a/b* antenna to the reaction centers [27]. Melis and Ow [16] have recently demonstrated, however, using absorbance difference spectrophotometry in the ultraviolet region and measurements of fluorescence induction, that the absolute number of α- and β-centers remains unchanged by $Mg^{2+}$ treatment and that the relative number of α- and β-centers is approx. 65 and 35% of the total, respectively. Furthermore, recent reports [15,16] indicate that, in grana-containing chloroplasts, α-centers are located in appressed membranes only and β-centers are found in the stroma-exposed thylakoids.

In addition to effecting conformational changes, the cationic composition of the chloroplast suspending medium induces fluorescence yield variations. One such chloroplast functional change is the well documented [24,28–30] 2-fold increase in the room-temperature fluorescence yield of DCMU-poisoned chloroplasts upon the addition of 2–5 mM $Mg^{2+}$. This observed increase in PS II quantum efficiency has been hypothesized to result from either a decrease in the spillover rate between PS II and PS I [29–32], an increase in the absorption cross-section of PS II [33,34], or a combination of the two effects [18]. However, Melis and Ow [16] recently concluded that the cation-dependent yield of chlorophyll fluorescence in chloroplasts at room temperature is entirely accounted for by an $Mg^{2+}$-dependent nonradiative decay rate constant for PS II α-centers. They observed a 7-fold increase in the variable fluorescence yield of PS II α-centers upon the addition of 5 mM $Mg^{2+}$. The variable fluorescence yield of β-centers and the nonvariable fluorescence yield, $F_0$, are essentially independent of cation concentration [16].

### *Cationic Variations in Potentiometric Titrations of Fluorescence*

This study measured significantly different $E_m$ values for the $Q_L$ transition in spinach chloroplasts isolated in the presence of different ion concentrations. The midpoint potential of $Q_L$ is lowered with increased levels of $Mg^{2+}$; at $Mg^{2+}$ concentrations of 0, 0.50 and 5.0 mM, the $E_{m,7.5}$

values for the transition in the total fluorescence yield are $-295$, $-345$ and $-384$ mV, respectively, with similar $E_{\mathrm{m},7.5}$ values for the slow phase yield, $-287$, $-346$ and $-395$ mV, respectively. Examination of the steady-state titrations of fluorescence yield of Horton [8] in the presence and absence of added $Mg^{2+}$ reveals a similar, but less pronounced trend in $E_{\mathrm{m},8}$ value for $Q_L$, decreasing from $-265$ to $-300$ mV, upon the addition of 5 mM $Mg^{2+}$. In the present study, $Mg^{2+}$ concentration did not significantly alter the relative contribution of $Q_L$ to the total variable fluorescence, although Horton [8] observed a decrease in the relative contribution of $Q_L$ from 30 to 16% upon depletion of $Mg^{2+}$ in steady-state measurements of total fluorescence yield. Our study further indicates that the contribution of $Q_L$ to the slow decay component yield is constant with increasing levels of $Mg^{2+}$. In addition, the presence of both $Q_H$ and $Q_L$ in unstacked $Mg^{2+}$-depleted membranes eliminates the simplifying assignment [4,11] of these acceptors to physically distinct reaction centers.

The present results are suggestive that chloroplast membrane organization changes affect the redox characteristics of the $Q_L$ transition. Variations in the proportion of $Q_L$ have previously been observed [8,17] or predicted [8,17] when the organization of PS II complexes has been altered by such treatments as growth under an intermittent-light regime, isolation of subchloroplast particles with detergents, or the phosphorylation of thylakoids. However, the assumption that fluorescence yield is linearly related to Q concentration is not generally valid [7,13,35]. The present study presents the first correlation of the $E_m$ value of $Q_L$ to membrane organization.

According to recent reports [8,14], the existence of $Q_L$ is associated with the increased complexity of chloroplast membranes upon aggregation of PS II reaction centers. Such a theory is modeled after the demonstration of Wikstrom et al. [36] of the effect of heme-heme interaction on the redox properties of the *a*-type cytochromes in cytochrome oxidase. Differences in the organization of the pigment-protein complexes in the thylakoid membranes occur as a consequence of changes in the ionic environment surrounding the membrane [37]. These organizational changes affect the redox characteristics of the $Q_L$ transition such that the stacking of thylakoids into grana is reflected in a more negative $E_m$ value for $Q_L$. However, the present results do not rule out the possibility for the creation of $Q_L$ by a redox interaction between two closely associated PS II reaction centers [8,14]. In such a case, $Q_H$ corresponds to the transition $[QQ]^- \xrightarrow{e^-} [QQ]^-$ and $Q_L$ to the transition $[QQ]^- \xrightarrow{e^-} [QQ]^=$.

Differences in the $E_{\mathrm{m},7.5}$ value for the $Q_H$ transition in total fluorescence yield are also noted at different levels of $Mg^{2+}$. At a level of added cation of 0.50 mM $Mg^{2+}$, the $E_{\mathrm{m},7.5}$ value, $+70$ mV, is lower than the corresponding value in either the absence or the presence of 5.0 mM $Mg^{2+}$, $E_{\mathrm{m},7.5} \simeq +100$ mV. Complex behavior of both the steady-state fluorescence [34] and the fluorescence decay kinetics [18] at this intermediate level of $Mg^{2+}$ has previously been noted. Although changes in the effective absorption cross-section of PS II and in the rate constant for nonradiative decay have been postulated as the origin of this complex fluorescence behavior, Marsho and Kok [38] also found an increase in the extent of reduction of the secondary electron acceptor plastoquinone pool with the addition of low levels of $Mg^{2+}$. Small variations in the redox state of these intermediate electron carriers, which titrate in the vicinity of $Q_H$ [6,9,39], are a likely source of the observed changes in the $E_m$ value of $Q_H$.

### *Effect of Cation Level on Fluorescence Decay Kinetics*

We have recently proposed [10,18,19] that an increase in the lifetime of the slow component of fluorescence decay, $\tau_{\mathrm{slow}}$, as reaction centers are closed is indicative of energy transfer between PS II units. When communication between reaction centers exists, increases in the lifetime of an excited state in the chlorophyll antenna of a closed reaction center are anticipated. PS II $\alpha$-centers would be expected to exhibit such communication via excitation transfer, the extent of the increase depending on the fraction of open reaction centers. Thus, the state of the reaction center as determined by reduction potential or illumination conditions affects $\tau_{\mathrm{slow}}$ of $\alpha$-centers only. Although the relative number of $\alpha$-centers is constant over all levels of added $Mg^{2+}$, high concentrations of $Mg^{2+}$ (2–5 mM) are required for excitation transfer to be observed [18,19]. Presumably [16], nonradiative decay competes efficiently with excitation transfer as a possible deexcitation mechanism at low $Mg^{2+}$ levels and $\tau_{slow}$ remains constant.

Previous titrations [10] in the presence of 5 mM $Mg^{2+}$ revealed an increase in $\tau_{slow}$ during both $Q_H$ and $Q_L$ reduction, suggesting that both electron acceptors may be present in $\alpha$-centers. For spinach chloroplasts in the presence of 0.50 mM $Mg^{2+}$ the increase in $\tau_{slow}$ during $Q_H$ reduction verifies that $Q_H$ is a possible electron acceptor in $\alpha$-centers. In $Mg^{2+}$-depleted chloroplasts the constant $\tau_{slow}$ during Q reduction is attributed to the absence of communication via excitation transfer between $\alpha$-centers as a consequence of a competitive rate of nonradiative decay of excitation. We are still unable to determine the identity of the electron acceptor in $\beta$-centers.

## Conclusion

We have used measurements of the potentiometric titration of the fluorescence decay kinetics in spinach chloroplasts in suspension media of various ionic concentrations to characterize the observed electron acceptor Q heterogeneity. Our data reflect a direct correlation between the complexity of chloroplast membrane organization and the reduction midpoint potential of the low-potential component of Q in PS II. We have further verified that the yield of the slow phase of fluorescence decay is sensitive to the state of the reaction center and that the redox dependence of the lifetime of the slow phase reflects differences in reaction center organization and deexcitation mechanisms.

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