LIGHT-INDUCED CHANGES IN THE FLUORESCENCE YIELD OF CHLOROPHYLL *A* IN VIVO

IV. The Effect of Preillumination on the Fluorescence Transient of *Chlorella pyrenoidosa*

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ABSTRACT The fluorescence transient of *Chlorella pyrenoidosa*, excited by saturating blue light, has a base level *O*, hump *I*, dip *D*, peak *P*, and at 1.5 sec a quasi-steady level *S* (12). With 2 sec exciting exposures and 4 min dark periods, preillumination-1 ($\lambda \geqq 690$ nm, intensities 1–750 ergs/sec-cm^2 incident), replacing the dark periods, lowers *I* more effectively than preillumination-2 (650 nm $\leqq \lambda \leqq$ 680 nm) in both aerobic and anaerobic cells. Results indicate that the intersystem electron transport pool *A* as well as the primary electron acceptor of pigment system II *Q* (fluorescence quencher) is normally being reduced at *I*. Preillumination-1 lowers and delays *P*. Preillumination-2 (absorbed by both pigment systems) also lowers *P*, but delays *P* only at low intensity; at high intensity it hastens *P*. Preillumination-1 raises *S* while preillumination-2 lowers *S*. With 30 instead of 2 sec exciting light exposures, preillumination-1 causes a large *S* increase, and at low intensity a *P* increase. The *S* effects seem to be of a long-term nature (26–29) rather than rapid changes in the redox state of *Q*. As exciting light intensity increases, fluorescence yield at *P* increases three-fold maximally. The ratio of *P* (anaerobic) to *O* (aerobic) is 5.5. These high ratios restrict the Franck-Rosenberg model of photosynthesis (13), which is based on fluorescence yield doubling.

INTRODUCTION

Fluorescence kinetics of algae and chloroplasts during the first seconds of illumination, which we call the fluorescence transient, have long been analyzed for information about the mechanism of photosynthesis (1). A consensus of recent studies is that *Q*, the fluorescence quencher and electron acceptor for system II, has a ratio of 1 to 150–400 chlorophyll molecules, and that the pool of intersystem intermediates called *A* has a ratio of 1 to 35–100 chlorophylls (2–6).

In *Chlorella pyrenoidosa* the fluorescence transient exhibits a base level *O*, a hump *I* at 50 msec. (at 1.5×10^4 ergs/sec-cm^2), a dip *D*, a peak *P* at 400 msec., and at 1.5 sec a quasi-steady level *S* (7–12). Currently, the *O-I* rise is believed to follow reduction of *Q* and *A* (8–12). The dip *D* after *I* indicates an oxidation of *QH* and *AH* caused by the action of system *I*; the *D-P* rise indicates a reduction of *Q* and *A* caused by development of a block in oxidation of *XH*, the reduced system *I* electron acceptor (12).

Morin (8) showed that under extremely intense light the *O-I* rise in *Chlorella* is purely photochemical. Short and intense preillumination left a partially completed *O-I* rise, indicating that preillumination causes a partial reduction of *Q*. Joliot (11) found that weak preillumination modified the fluorescence transient only slightly, whereas the oxygen rate transient lacked an activation phase. This result suggests that the inactive form of *E* (see reference 4) does not participate in the main features of the fluorescence transient. Delosme (10) reported wavelength independence of the effects of intense preillumination.

Because intersystem intermediates are reduced by system II and oxidized by system I, we expected a clear wavelength dependence of preillumination. Our studies reveal a wavelength dependence of weak preillumination, and show that most weak preillumination effects are sensitized by absorption in system I. Some effects suggest redox changes in intersystem intermediates, and other suggest long-term changes, perhaps involving conformational changes. During development of standard procedures, we found fluorescence yield changes connected with the transient which are too large to be accommodated easily by the Franck and Rosenberg (13) model of photosynthesis.

METHODS

Materials and methods were the same as described in the accompanying paper (12) with some modifications as set forth below.

Preillumination intensity ranged up to 750 ergs/sec-cm^2; a typical value of absorbed preillumination intensity was 10^{11} quanta/sec-cm^2, 1000 times smaller than absorbed exciting light intensity.

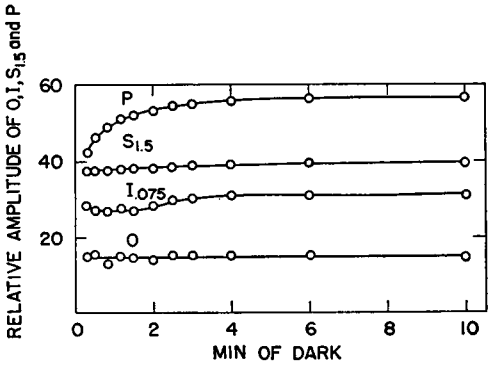

FIGURE 1 Amplitude of points of the transient vs. length of the preceding dark period. *S* was measured at 1.5 sec, and *I* at 75 msec.

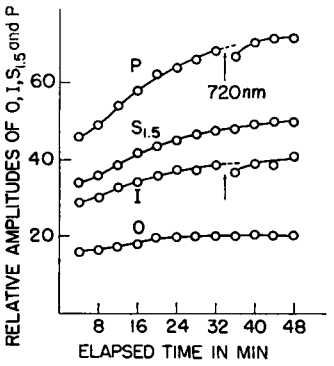

FIGURE 2 Amplitude of points of the transient during an adaptation period at the beginning of one experiment. 720 nm preillumination depressed *P* and *I* but did not affect *O*. *S* was usually increased, but in this culture an *S* effect appeared only at higher 720 nm intensity.

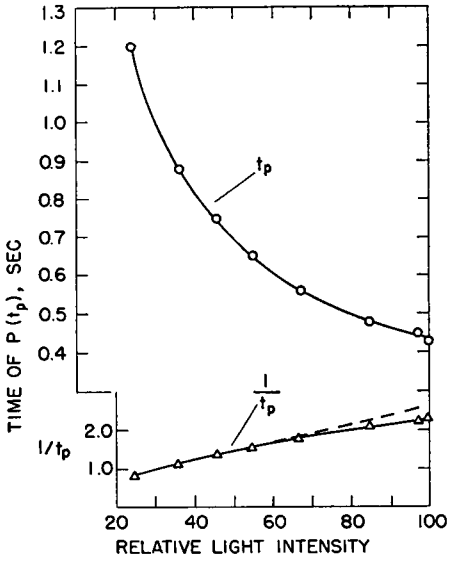

FIGURE 3 Time of the peak, t_p, vs. exciting light intensity. Maximum intensity (100) is 1.5×10^4 ergs/sec-cm^2 incident. The plot of $1/t_p$ is linear at low intensity, indicating that the product of intensity and t_p is a constant.

The standard procedure was a sequence of 2 sec light exposures separated by 4 min dark intervals. Fig. 1 in reference 12 shows a sequence of transients obtained in this way from a single sample. Fig. 1 (in the present paper) shows that 4 min dark intervals allowed *P* to rise to more than 95% of its fully dark-adapted height. After an adaptation period consisting of the initial eight or more cycles, the transient was repeatable, and preillumination tests were then begun. Fig. 2 shows the changes which took place during one adaptation period for cells which had been under continuous illumination just previously. In most experiments, light intensity was adjusted slightly to produce a t_p (time of the peak, *P*) of 400–500 msec. Fig. 3 shows the intensity dependence of t_p. In some early experiments (see reference 14) t_p was less than 300 msec, the light exposures were for 30 rather than 2 sec, and the dark intervals were 4½ rather than 4 min.

In preillumination experiments, preillumination replaced the dark period. It was previously noticed that preillumination effects persisted for several minutes (see reference 14); thus a wait of three light-dark cycles was required between preillumination trials to allow the sample

condition to return to normal. When studying intensity effects, we employed a monotonic sequence of increasing preillumination intensities, or a sequence of first decreasing and then increasing exciting light intensities.

RESULTS

I. Light Intensity and the Transient

During standardization of procedures, we measured several times the relation between exciting light intensity and fluorescence intensity at O, I at 50 msec, P, S at 1.5 sec. ($S_{1.5}$), and S between 5 and 30 sec (S_5). Fig. 4 shows results from one sample. Fig. 3 was obtained from the same experiments. (We previously published a similar figure in reference 14.)

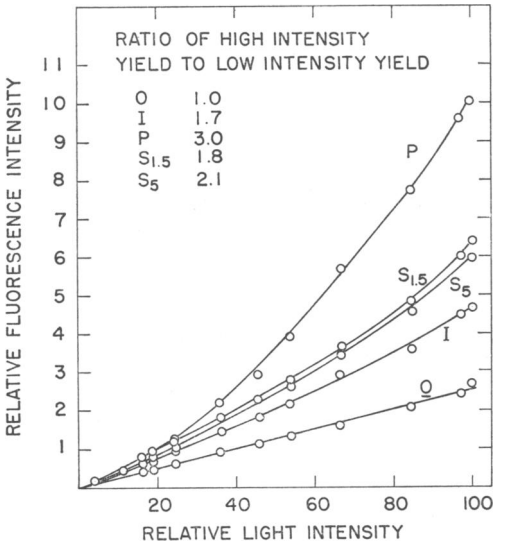

FIGURE 4 Fluorescence intensity at different points of the transient vs. exciting light intensity. O, I, P, and $S_{1.5}$ were determined in successive 2 sec light exposures (with 4 min dark intervals) at increasing then decreasing light intensity. S_5 was determined by rapid intensity changes during a subsequent 30 sec light exposure. The yield ratios were determined from the slopes at high and low light intensities.

The O curve has a constant slope. This feature of O was first observed by Lavorel (15). P becomes distinguishable from S_5 and $S_{1.5}$ as their yields begin to change. The yield increase is greatest for P. The ratios of high intensity yields to low intensity yields are, for O—1.0, I—1.7, $S_{1.5}$—1.8, S_5—2.1, and for P—3.0. Relatively similar values were obtained in reference 14.

II. The Effect of Preillumination on the Transient

A. The Aerobic Case. The effects were studied at 8 wavelengths between 650 nm and 725 nm. System II effects are expected to predominate at $\lambda \leqq 680$ nm, although both systems absorb light of these wavelengths, and system I effects are expected at $\lambda \geqq 690$ nm. Beyond 720 nm clear system I effects can be observed (6, 16, 17).

1. Intensity dependences for several wavelengths. An intensity study at one wavelength required more than 6 hr. We were thereby forced to study different cultures in order to compare effects at different wavelengths. But consistent results were obtained in 25 preliminary experiments and in three careful experiments at each of 7 wavelengths with only one complete experiment at 725 nm. Percentage changes in I at 50 msec, P, $S_{1.5}$, and in t_p are plotted vs. absorbed intensity of preillumination. Results are shown for single experiments at 710 and 650 nm in Fig. 5 and 6. 725 and 700 nm results, not shown here, were similar to results at 710 nm; 670 and 660 nm results were similar to those at 650 nm, and, 690 and 680 nm results represented a transition between the longer and the shorter wavelengths.

The point O is not affected by preillumination at the intensities used in Figs. 5 and 6. Fig. 2 shows that 720 nm preillumination which affected P and I did not affect O. I is decreased for all wavelengths by 10%, and the effect saturates at very weak intensities. P is decreased for all wavelengths, when the exciting light exposure is 2 sec. Results with 30 sec exposures are different (see the next paragraph). The P curves in Figs. 5 and 6 show two segments, a steep segment which saturates at the same intensity as the effect on I, and a mildly sloping segment which continues until P is indistinguishable from I and S. t_p increases for all wavelengths at low intensities, and the increase saturates near the same intensities as effects on P and I. High intensity preillumination-2 causes t_p to decrease. This decrease also saturates. S is increased by preillumination-1 and decreased by preillumination-2, and the effects saturate. The saturation intensities for all variables increase as the wavelength changes from 710 to 650 nm (the 725 nm result was an exception, but absorption values at 725 nm are small and subject to relatively greater error).

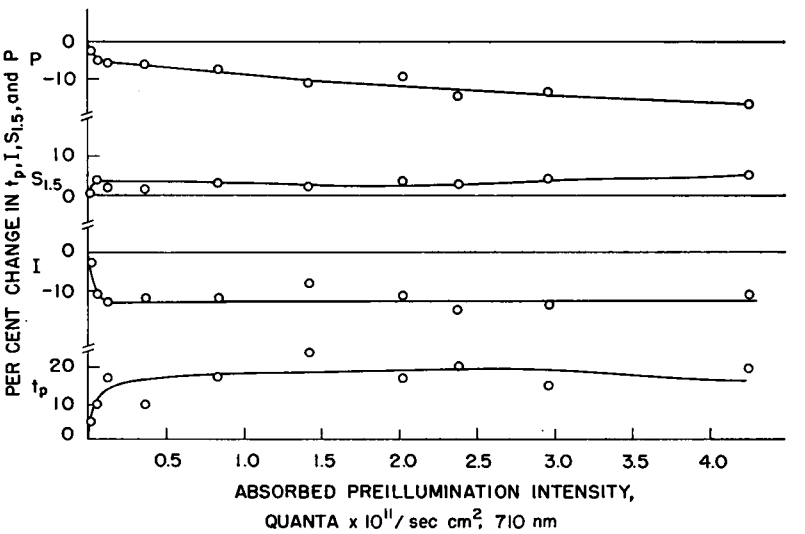

FIGURE 5 The effects on points of the transient caused by different intensities of 710 nm preillumination replacing the usual 4 min dark period. Exciting light exposures: 2 sec.

In earlier experiments, the exciting light exposure was for 30 rather than 2 sec, and the dark periods were 4½ rather than 4 min. During the 30 sec light exposure, at our exciting light intensity, S declines about 6%. For 690 nm preillumination we observed a P increase at low intensity and a large S increase at all intensities, while at 640 nm, we found small P and S increases at low intensity and decreases at higher intensity (Figs. 7, 8). In other experiments with 30 sec exciting light exposures, we

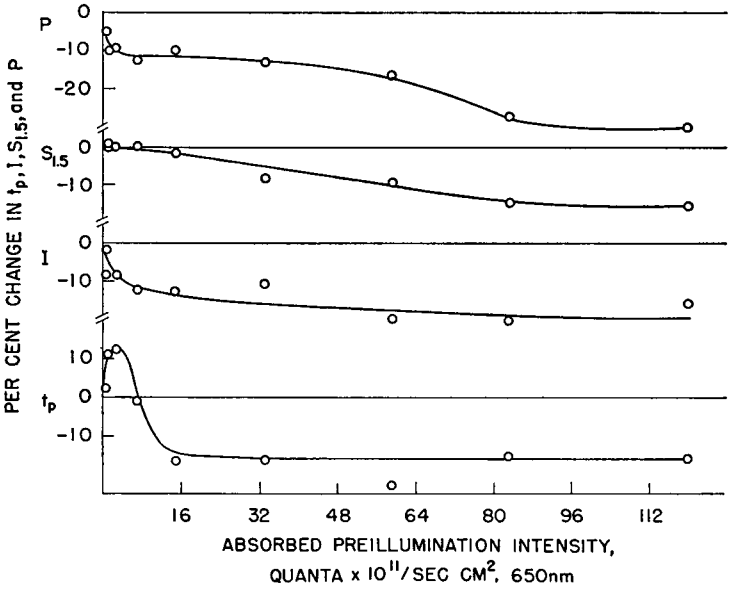

FIGURE 6 The effects of 650 nm preillumination (see legend of Fig. 5).

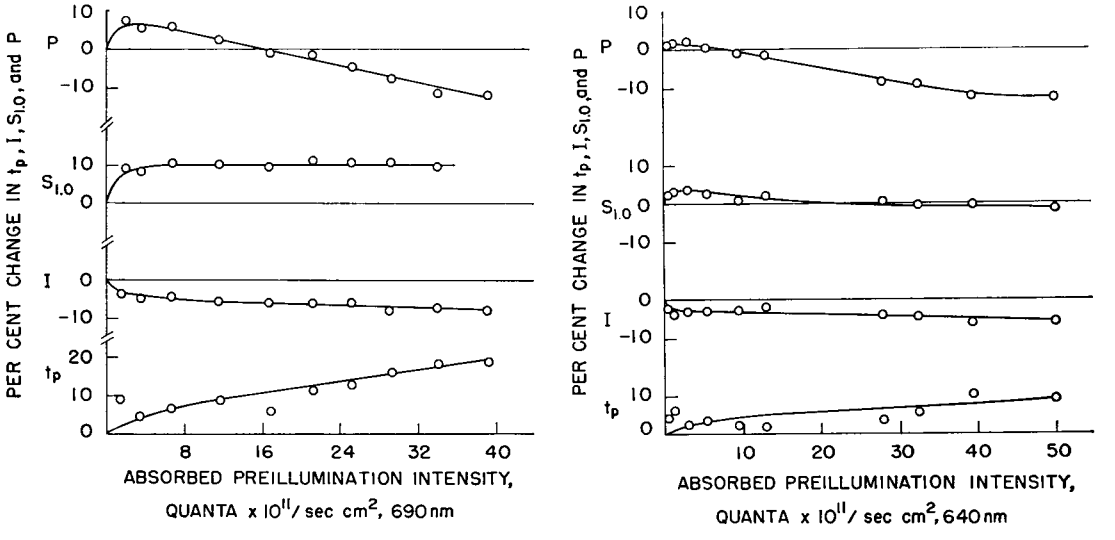

FIGURE 7 The effects of 690 nm and 640 nm preillumination under 30 sec exciting light exposures separated by 4½ min.

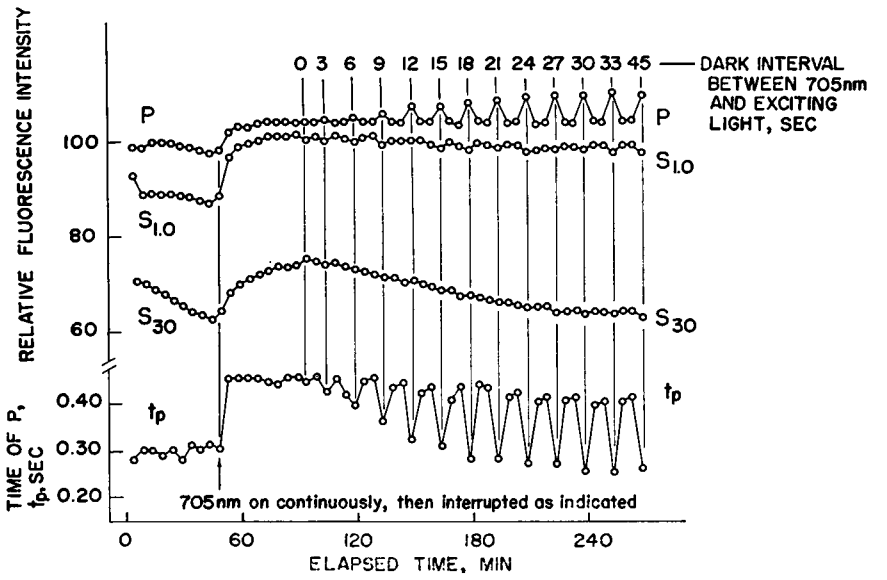

FIGURE 8 The effects of continuous, then interrupted 705 nm background light. 30 sec exciting light exposures separated by 4½ min.

used 705 nm background light (equivalent to preillumination plus supplementary light during the transient). P, S_1, and S_{30} then increased over a 30 min period. Fig. 8, from one of these experiments, shows the long-term increases in P and S. It also shows that interruption of the 705 nm light several seconds before the exciting light exposure allowed P to rise further.

2. Quantum yield spectra from single samples. The increase, at shorter wavelengths, of saturation intensities for the preillumination effects, suggested that the effects are sensitized by long wavelengths. However, the foregoing results were obtained from different cultures. To test the hypothesis on single samples, we performed experiments in which each wavelength of preillumination was used only twice. The intensities were chosen to fall below the saturation intensities. From two of the experiments, a relative quantum yield spectrum has been constructed which shows which preillumination wavelengths were maximally effective in changing I, P, and t_p (Fig. 9). Long wavelengths, especially 700 and 720 nm, were the most effective. A hump at 660 nm (in P and I curves) may be of significance since a dip at this wavelength has been often seen in the action spectrum ($\Phi_{o_2} = f(\lambda)$) of photosynthesis and fluorescence in *Chlorella*.[1]

B. The Anaerobic Case. In anaerobic cells, I is a high spike, D is a deep trough (but still higher than in aerobic cells), and P is high and persistent for several

[1] The dip at 660 nm in the $\Phi_{o_2} = f(\lambda)$ curve of both fluorescence and oxygen evolution in *Chlorella* may be caused by a shortwave component of Chlorophyll *a* in pigment system I (see R. Govindjee, E. Rabinowitch, and Govindjee [31]).

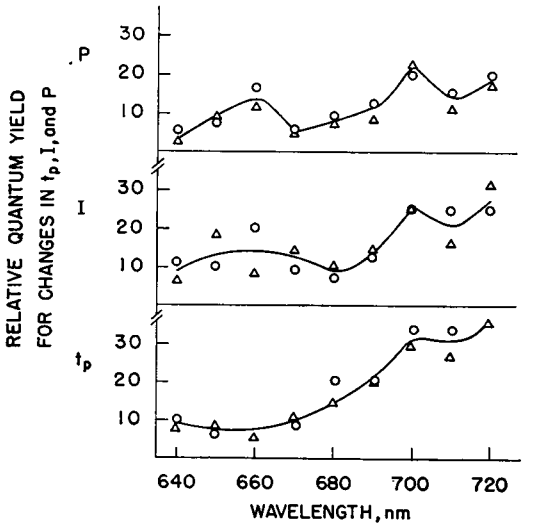

FIGURE 9 The relative quantum yield for the increase of t_p and the decreases of I and P, vs. preillumination wave length. Preillumination intensities were below the saturation regions of Figs. 5–7. 2 sec exciting light exposures, 4 min intervals.

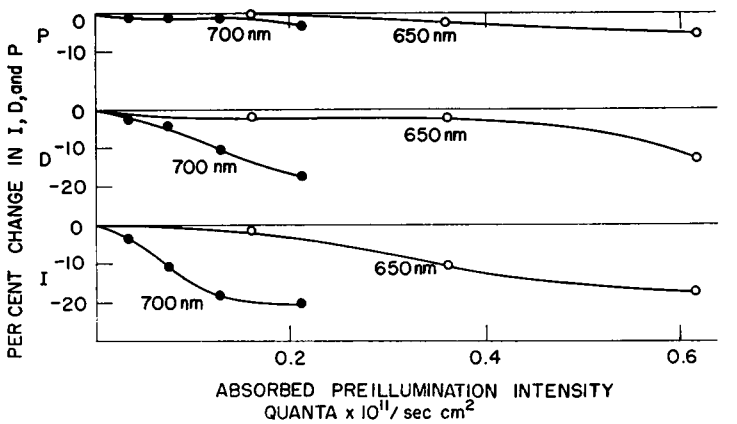

FIGURE 10 The effects of 700 and 650 nm preillumination in anaerobic cells. Plots for t_p and $S_{1.5}$ are omitted; t_p and $S_{1.5}$ are unmeasurable because P does not decline for several seconds in anaerobic cells. 2 sec exciting light exposure, 4 min intervals.

seconds (see Fig. 3 in reference 12). The clear separation of I from P allows an unambiguous determination of the effect and wavelength dependency of preillumination on I. Single samples were tested for effects of 700 and 650 nm preillumination, and the changes in I, D, and P observed in one of several experiments are shown in Fig. 10. No t_p and S changes can be identified because there is no P to S decline in anaerobic cells; therefore, t_p and S plots are omitted. The results—700 nm was more effective than 650 nm in decreasing I, D, and P.

C. Dark Decay of the Preillumination Effect. In study of the dark decay, we utilized a 3 sec exposure of 700 nm preillumination followed by different dark

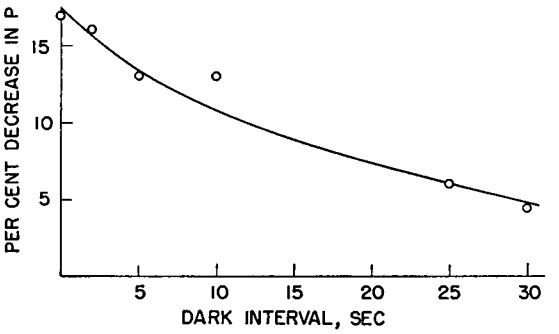

FIGURE 11 Dark decay of the effect on P of 700 nm preillumination. 2 sec exciting light exposures, 4 min intervals.

intervals before the exciting light exposure. The preillumination intensity was sufficient to decrease P by 15%. (This intensity was much higher than that producing the same effect when the duration was 4 min.) Results from several experiments were similar. In one experiment (Fig. 11), the decrease of P decayed to one-half maximum value in a dark interval of 15 sec. In the experiment of Fig. 8 the half-time was 14 sec.

DISCUSSION

I. Transient Fluorescence Yield and the Franck and Rosenberg Model

Franck and Rosenberg (13) proposed in 1964 a model for photosynthesis which has been widely discussed as an alternative to the series scheme. Its main details are that the two photochemical reactions of photosynthesis alternate at a single reaction center rather than taking place at separate centers arranged in series. One reaction utilizes triplet-state energy, which requires triplet-state formation. Fluorescence during optimal photosynthesis is emitted in competition with triplet-state formation. If photosynthesis stops no energy is used for the singlet reaction; all energy, rather than half, becomes subject to triplet formation and fluorescence, and fluorescence yield must double.

In several instances, we observe fluorescence increases which are significantly greater than the predicted doubling. During the transient the level corresponding to optimal use of excitation energy is the O level, and the ratio of P to O is 4.0 at our highest excitation intensity (see Fig. 4). The yield increase for P as light intensity increases is by a factor of 3.0. In addition, the ratio P (anaerobic) to O (aerobic) is 5.5, where P (anaerobic) is considered the fluorescence level when no photosynthesis is occurring (see Fig. 3 in the accompanying paper (12)).

It may be argued that fluorescence at O is not associated with either photoreaction and thus is not in competition with photosynthesis, and that another fluorescence level should represent the fluorescence in competition with optimal photosynthesis.

Franck and Rosenberg suggested the steady-state level, *S*. Even then, with the level *O* (aerobic) subtracted away because it is to be considered nonimportant, the ratio of *P* (anaerobic) to *S* (aerobic) equals 3.0 (see Fig. 3 in the accompanying paper (12)).

Other workers have noticed similarly large fluorescence yield increases. Wassink and Katz (18), Shiau and Franck (19), and Butler and Bishop (20) have observed increases of the order of three with inhibition of photosynthesis.

Franck and Rosenberg recognized that fluorescence might more than double at higher intensity if photooxidation of chlorophyll occurred. If the model is correct, our results require that such photooxidation become important even before light intensity becomes high enough to saturate photosynthesis. However, Krey and Govindjee (21) found in *Porphyridium* that the fluorescence yield was constant for light intensities twice the saturation intensity. If photooxidation were involved at these intensities, the yield should be constantly increasing with light intensity.

These observations require that the Franck and Rosenberg model be modified with appropriate assumptions in order to remain viable.

II. Significance of the Preillumination Effects

A. The O Level. After Kok et al. (22) we believe all *Q* is oxidized at *O*. *O* is not affected by either preillumination-1 or preillumination-2 (at our weak intensities). Therefore, weak preillumination-2 does not reduce *Q* faster than it is oxidized. Also, dark-period oxidation of *QH* is complete, even without the aid of preillumination-1 (if otherwise, then preillumination-1 would lower *O*).

B. The I level. *I* is decreased by preillumination-1. This and the fact that dark-oxidation of *QH* is complete signify that *I* is affected by the concentrations of intersystem intermediates other than *Q* (we call these other intermediates *A*). It follows that the *O-I* rise, under our exciting light intensity, involves reduction of *A* as well as *Q*.

Since preillumination-1 decreases *I*, it must be that a dark period does not completely oxidize *AH*. Furthermore, from Fig. 1, 10 min dark lowers the *I* level no more than 15 sec dark. Thus, a partial oxidation of *AH* occurs within 15 sec dark, and subsequently the *AH* level remains constant. We can conceive of two possible reasons why the dark oxidation of *AH* is partial rather than complete: (1) the concentration of oxidants which oxidize *AH* in the dark is smaller than the concentration of *AH*, or (2) *A* is reduced in the dark at a moderate rate.

The decrease in *I* was generally about 10%. Saturation of the decrease indicates when oxidation of *AH* is complete.

Saturation intensities for the *I*-decrease are higher for preillumination-2 than for preillumination-1, indicating that long wavelengths preferentially decrease *I*. The quantum yield spectrum (aerobic conditions) also indicates that long wavelengths preferentially decrease *I*. In anaerobic conditions, long wavelengths again preferen-

tially decrease *I*. These results are consistent; *I* is preferentially decreased by wavelengths beyond 690 nm. The results suggest that the decrease in *I* is due to quanta which reach system I.

It is puzzling, however, that preillumination-2 also decreases *I*. One expects, since most quanta of preillumination-2 are absorbed by system II, that the net effect of preillumination-2 would be to reduce *A*, and thereby increase *I*. Because of the decrease in *I*, we speculate that low intensity preillumination absorbed by system II does not effect transfer of electrons to *A*. Perhaps the quanta are wasted in some manner. Other workers have decided that system II may waste dim light. Murata et al. (5) studied the steady-state fluorescence yield of isolated chloroplasts and decided that the change in yield at very low exciting light intensities (far below the intensity which saturates photosynthesis) can be explained in terms of a back reaction between QH and an intermediate on the water side of system II. Joliot (11) believes the change in yield involves deactivation of EH to E_iH (an inactive form of the system II oxygen intermediate). Both interpretations suggest that *A* would not receive electrons in weak system II light. Malkin (23) believes the change in yield involves photoinduced electron transport with oxygen as the terminal electron acceptor. Some of his evidence suggests that system I is involved as well as system II. The Malkin explanation, in contrast to the others, seems to imply that *A* would receive electrons in weak system II light.

C. The D Level. In the accompanying paper (12), we have analyzed the dip *D*, following *I*, as an oxidation of QH via the action of system I. We suggest that the peak *P* is due to reduction of *Q*, caused by development of a block in oxidation of XH. At *P*, then, *X*, *A*, and *Q* are largely reduced.

D. The P level. Preillumination-1 causes *P* to decrease, the curves having two segments. We think the first segment is due to an increase in *A* caused by the action of system I: the preillumination reduces *X* and oxidizes AH. Because the preillumination is weak, the carbon cycle easily drains electrons from XH; meanwhile, AH is oxidized. During the next exciting light exposure, the higher concentration of *A* delays completion of the reduction of *X*, *A*, and *Q*, resulting in a prolongation and depression of *P*.

The second segment of the *P* decrease does not involve further increase in *A*, because the *I* decrease, which is responsive to changes in *A*, is already saturated. We can only speculate that the second segment has the following explanation: From Fig. 1, dark restoration of the *P* level after a light exposure in which the level *S* has been reached requires several minutes of dark. Perhaps preillumination simply prevents dark restoration.

We comment in passing that the dark restoration of *P* is not yet adequately explained. If *P* were due, as many workers believe, to reduction only of *A* and *Q*, then *P* and *I* should be restored to the same degree by the same length of dark period. From Fig. 1, *I* is restored within 15 sec of dark; thus, most AH is oxidized within that time. However, 95% dark restoration of *P* requires 4 min. This long time

suggests that P has a more complex origin than reduction only of A and Q. In the accompanying paper (12) we propose that P involves development of a block in oxidation of XH which prevents oxidation of A and Q. We do not have any indication of the location or nature of the block except that it is beyond XH toward the carbon cycle.

Preillumination-2 decreases P. The curves have two segments. Saturation intensities for the first segment are higher for preillumination-2 than for -1; the quantum yield spectrum shows long wavelength peaks; and in anaerobic conditions long wavelengths are more effective in decreasing P. All these results are consistent in indicating long wavelength and therefore system I sensitization of the decrease. We speculate that low intensity preillumination absorbed by system II may be wasted (see discussion in part B).

E. The Variable t_p. An increase in t_p representing a delay in P occurs for all low intensity preillumination. We hypothesize that this increase results from an increase in A caused by the action of quanta reaching system I (above, in part D, we explained how this increase might delay P).

Preillumination-2 at high intensity causes a t_p decrease. At first thought, this decrease might be explained conversely to a t_p increase. Thus we might expect that the decrease involves reduction of A to AH. However, this explanation cannot be allowed because I is still decreased at the high intensity indicating that AH has been oxidized. Perhaps the t_p decrease is simply an obligatory coincidence with elimination of P, due to details of the kinetic relationships involved. However, high intensity preillumination-1 nearly eliminates P, but does not decrease t_p.

F. The S Level. Preillumination-1 raises S while preillumination-2 decreases S. Such changes are opposite to those reported from our laboratory (24) and elsewhere (3, 20, 25) in which simultaneous absorption of light in system I decreases fluorescence from system II, and which are now understood as the effects of system II and I on the redox levels of Q and A. Our results for S cannot be explained in this way. We believe instead that our results involve long-term changes in bulk fluorescence yield. The relevant facts: when we employed 30 sec rather than 2 sec light exposures (Fig. 7 and Fig. 3 in reference 14), the S level declined about 6% between 1 and 30 sec. Then, preillumination-1 caused a large S increase and preillumination-2 at low intensity also caused an increase. From Fig. 8, also involving 30 sec light exposures, system I illumination caused long-term S and P increases. When we employed 2 sec light exposures the S increase caused by preillumination-1 was small, and preillumination-2 caused only a decrease. Furthermore, when in measuring the dark decay of the preillumination effects we used 3 sec of preillumination rather than 4 min, the S changes were negligibly small. Thus, short exposure and preillumination durations produce small S changes, and long durations produce large S changes.

Perhaps our S changes are related to long-term fluorescence induction. Papa-

georgiou (25) and Papageorgiou and Govindjee (27–29) have studied fluorescence induction in *Chlorella* and *Anacystis* during the first hour of illumination, and they have suggested that long-term induction involves lamellar conformational changes associated with ATP-coupled electron transport.

G. The Dark Decay. The persistence of the effect of preillumination-1 for about 15 sec is similar to results reported by French (30). Enhancement of oxygen production in *Porphyridium* occurred when 5 sec of red light (system I) preceded 5 sec of green light (system II). The enhancement had a half-life of 18 sec. When the green light came first, however, there was no enhancement. The dark decays of enhancement and the preillumination effect are explainable by the view that *A* remains oxidized after system I activity for 15 sec. Obviously, *A* is partially but not completely protected from strong reductants.

This work is part of a thesis submitted by John C. Munday to the University of Illinois (1968) in partial fulfillment of the requirements of a Ph.D. in Biophysics.

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