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PROPERTIES OF PHOTOREDUCTIONS BY PHOTOSYSTEM II IN ISOLATED CHLOROPLASTS

AN ENERGY-CONSERVING STEP IN THE PHOTOREDUCTION OF BENZOQUINONES BY PHOTOSYSTEM II IN THE PRESENCE OF DIBROMOTHYMOQUINONE

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SUMMARY

Dibromothymoquinone has been used to block photosynthetic electron flow in chloroplasts between Photosystem I and II. The inhibition by dibromothymoquinone of electron flow and of coupled ATP formation in photoreductions by Photosystem I is reversed by the addition of plastoquinone, but not of other substituted benzoquinones.

Because dibromothymoquinone does not permit electron flow from water through Photosystem I, Hill reactions in the presence of dibromothymoquinone are attributed to photoreductions by Photosystem II. The photoreduction of *p*-benzoquinone and 2,6-dimethylbenzoquinone in the presence of dibromothymoquinone is coupled to ATP formation with an ATP/e_2 ratio of 0.3. Both photoreductions are stimulated by the addition of uncouplers. Ferricyanide photoreduction in the presence of dibromothymoquinone has a P/e_2 ratio of only 0.16 and is not stimulated by an uncoupler. It is concluded that photoreductions by Photosystem II include an energy-conserving step and that lipophilic and hydrophilic acceptors are reduced on different sides of the membrane.

INTRODUCTION

Recently we introduced dibromothymoquinone (2,5-dibromo-3-methyl-6-isopropyl-*p*-benzoquinone) as an effective inhibitor of photosynthetic electron transport in chloroplasts between Photosystem II and I[1,2]. Its side of inhibition was localized on the reduced side of plastoquinone. Further experiments by Cramer and coworkers[3,4] supported this view and showed that the inhibition of electron flow by dibromothymoquinone occurs between cytochrome b_{559} and cytochrome *f*. Recent results by Lozier and Butler[5] as well as by W. Haehnel (personal communication) gave

Abbreviations: DCIP, 2,6-dichlorophenolindophenol; DCMU, 3-(3,4-dichlorophenyl)-1,1-dimethylurea.

more direct evidence that dibromothymoquinone inhibits the oxidation of plastohydroquinone by Photosystem I but not the reduction of plastoquinone by Photosystem II.

Hill reactions involving both Photosystems I and II like the photoreduction of $NADP^+$ and of low potential quinones at the expense of water as electron donor are completely inhibited by $5 \cdot 10^{-7}$ M dibromothymoquinone. The photoreduction of ferricyanide and DCIP in intact chloroplasts is partly inhibited by dibromothymoquinone, but not at all in fragmented chloroplasts[2]. We concluded from this, that photoreductions by Photosystem II are not affected by dibromothymoquinone provided the structural state of the chloroplasts permits access of polar Hill reagents to Photosystem II. The inhibition of electron flow from Photosystem II to Photosystem I by dibromothymoquinone may be reversed by the addition of plastoquinone[2]. Gimmler and Avron[6] reported that also *p*-benzoquinone seems to reverse the inhibition of photosynthesis by dibromothymoquinone in whole cells. Since this effect could be interpreted as either a reversal of the inhibition or as non-inhibition of benzoquinone photoreduction, we have reinvestigated in more detail the specifity of the reversal of dibromothymoquinone inhibition by plastoquinone and other substituted benzoquinones. We conclude that the photoreduction of certain quinones in the presence of dibromothymoquinone is a photoreduction by Photosystem II. Thus the properties of photoreductions by Photosystem II may be investigated under conditions, in which dibromothymoquinone prevents electron flow through Photosystem I. The results indicate that certain *p*-benzoquinones are advantageous electron acceptors of Photosystem II even in intact chloroplasts and that there is an energy-conserving step in photoreductions by Photosystem II.

METHODS

Spinach chloroplasts have been prepared according to Nelson *et al.*[7]. The particles were washed once in 5 mM Tricine buffer, pH 8.5.

For the assay of photosynthetic activity chloroplasts with 0.2 mg chlorophyll were illuminated at 15 °C with white light of 30000 lux in 3 ml Tricine buffer, pH 8.0. Electron flow was measured by the evolution of oxygen, followed manometrically and ATP formation by the incorporation of ^{32}P into organic phosphate, according to the method of Sugino and Miyoshi[8].

2,6-Dimethylbenzoquinone, obtained from Schuchardt, Munich, was dissolved in a trace of methanol and brought up to the concentration needed by adding water.

Dibromothymoquinone has been kindly synthetised by Dr Draber.

RESULTS

As already reported coupled pseudocyclic electron transport (or pseudocyclic photophosphorylation) with an acceptor of Photosystem I like anthraquinone sulfonate is completely inhibited by low concentrations ($5 \cdot 10^{-7}$ M) of dibromothymoquinone[1,2]. The inhibition of the photoreduction of anthraquinone sulfonate and of coupled ATP formation by $5 \cdot 10^{-7}$ M dibromothymoquinone is partly reversed in a competitive way by the addition of plastoquinone[2]. As Table I indicates other substituted benzoquinones including thymoquinone are ineffective in reversing

TABLE I

REACTIVATION BY QUINONES OF PSEUDOCYCLIC ELECTRON TRANSPORT INVOLVING PHOTOSYSTEM I AND II INHIBITED BY DIBROMOTHYMOQUINONE (ANTHRAQUINONE SULFONATE AS AUTOXIDIZABLE ACCEPTOR)

Conditions: 10 min light in air, the reaction medium contained 10 μmoles ADP and P_i, 5 μmoles $MgCl_2$ and 0.2 μmole anthraquinone sulfonate.

Additions to $5 \cdot 10^{-7}$ M dibromothymoquinone	*μequiv O_2 taken up*	*μmoles ATP formed*
No addition	0.5	0.5
Plastoquinone-45 10^{-4} M	2.3	2.6
Ubiquinone-30 10^{-5} M	0.7	0.6
Ubiquinone-30 10^{-4} M	1.6	0.5
p-Benzoquinone 10^{-4} M	0.9	0.3
2,6-Dimethylbenzoquinone 10^{-4} M	0.8	0.5
2,6-Dimethoxybenzoquinone 10^{-4} M	0.3	0.4
2,6-Dimethoxymethylbenzoquinone 10^{-4} M	0.2	0.5
Trimethylbenzoquinone 10^{-4} M	0.1	0.2
Thymoquinone 10^{-4} M	0.5	0.5
Duroquinone 10^{-4} M	1.3	0.9
Control without dibromothymoquinone	3.4	3.5

dibromothymoquinone inhibition. Only plastoquinone is able to restore electron flow as well as coupled ATP formation in pseudocyclic electron flow inhibited by dibromothymoquinone. Ubiquinone also shows a perceptible effect in reversing the inhibition of electron transport, but not of coupled ATP formation (ubiquinone itself is, however, already an inhibitor of the control reaction, 10^{-4} M ubiquinone-30 inhibiting about 30%). We interpret the slight stimulation of electron flow by duroquinone (see Table I) not as a reversal of inhibition but rather as a bypassing of the inhibitor site because durohydroquinone is an electron donor for Photosystem I (ref. 9). The slight stimulation of ATP formation in the duroquinone experiment is due to an ATP formation at the Photosystem II site, as will be discussed later. We have already observed that also DCIP shows a similar effect[2], because DCIP is an acceptor of electrons at Photosystem II and reduced DCIP is an electron donor for Photosystem I after the inhibition site of dibromothymoquinone. We define as a reversal of electron flow to Photosystem I only those conditions in which electron flow as well as full stoichiometry of ATP formation is restored. Therefore, we conclude from the results of Table I that only plastoquinone is able to reverse the inhibition of dibromothymoquinone of electron flow from Photosystem II to Photosystem I restoring ATP formation as well.

Dibromothymoquinone does not inhibit photoreductions by Photosystem II (ferricyanide or DCIP as acceptor) in sonicated chloroplasts[2] or chloroplasts obtained from *Euglena* cells[10]. In intact chloroplasts dibromothymoquinone does inhibit the photoreduction of ferricyanide and DCIP to some extent[2]. Because of the high polarity and hydrophilicity ferricyanide does not get easy access to Photosystem II buried in the membrane of intact chloroplasts. It is, therefore, reduced preferentially

by Photosystem I in such chloroplasts. This in turn makes these photoreductions sensitive to dibromothymoquinone as already discussed in detail[2,10].

Because of the high lipophilicity of uncharged benzoquinones we expected that these compounds would be able to penetrate to Photosystem II even in intact chloroplasts. This is indeed the case, because benzoquinones in catalytic amounts stimulate a dibromothymoquinone insensitive ferricyanide reduction (Fig. 1 and Table II). The photoreduction of benzoquinones in substrate amounts is only slightly diminished in the presence of dibromothymoquinone as against the part inhibition of ferricyanide and full inhibition of anthraquinone photoreduction (Table III, Figs 2 and 3).

Fig. 1 indicates that a *p*-benzoquinone stimulates the reduction of ferricyanide in the presence of dibromothymoquinone. This can best be explained by a shuttle of electrons between the acceptor site of Photosystem II in the membrane and the

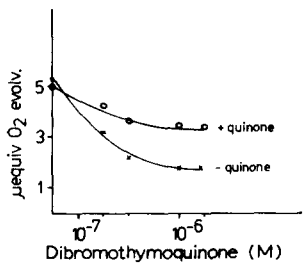

Fig. 1. Reversal by 2,6-dimethylbenzoquinone of the inhibition of ferricyanide photoreduction by dibromothymoquinone. Conditions as in Table II; 10^{-4} M 2,6-dimethylbenzoquinone added.

TABLE II

REACTIVATION BY QUINONES OF THE PHOTOREDUCTION OF FERRICYANIDE INHIBITED BY DIBROMOTHYMOQUINONE

Conditions: 10 min light in N_2, the reaction medium contained 10 µmoles ADP and P_i, 5 µmoles $MgCl_2$ and 20 µmoles potassium ferricyanide.

Additions to dibromothymoquinone	*µequiv O_2 evolved*	*µmoles ATP formed*
$5 \cdot 10^{-7}$ M dibromothymoquinone		
No addition	2.5	1.4
Plastoquinone 10^{-4} M	4.5	4.1
p-Benzoquinone 10^{-5} M	2.9	1.5
p-Benzoquinone 10^{-4} M	3.6	1.7
2,6-Dimethylbenzoquinone 10^{-5} M	3.4	2.0
2,6-Dimethylbenzoquinone 10^{-4} M	3.9	2.3
Control without dibromothymoquinone	5.8	5.6
$2 \cdot 10^{-6}$ M dibromothymoquinone		
No addition	2.1	0.3
2,6-Dimethylbenzoquinone 10^{-4} M	3.2	0.5
2,6-Dimethylbenzoquinone 10^{-4} M + DCMU $2 \cdot 10^{-6}$ M	< 0.1	< 0.1

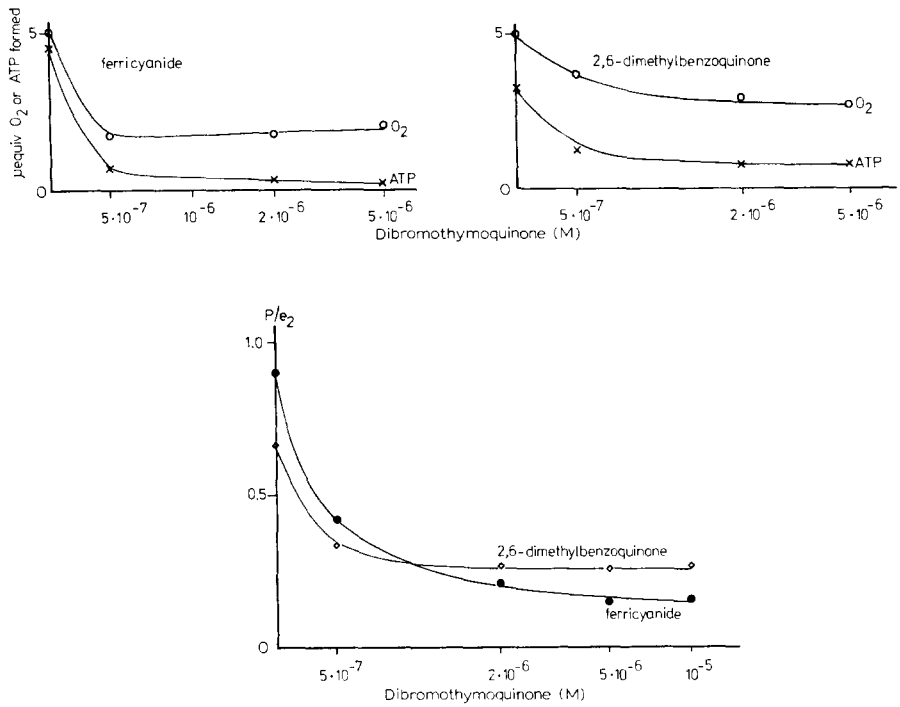

Fig. 2. ATP formation and P/e_2 ratio in photoreductions by Photosystem II (*i.e.* in the presence of dibromothymoquinone). Conditions as in Table III, except at pH 8.0.

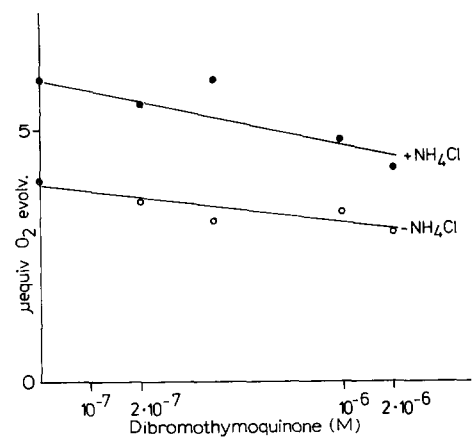

Fig. 3. Stimulation of the photoreduction of *p*-benzoquinone by an uncoupler (NH_4Cl) in the presence of dibromothymoquinone. Conditions as in Table III.

polar ferricyanide outside the membrane. This stimulation of ferricyanide reduction by benzoquinone in the presence of dibromothymoquinone is a draining off of electrons before the inhibition site and not a reversal of inhibition or a bypassing of the inhibition site, which would allow again electron flow through Photosystem I. This interpretation may be deduced from the experiments of Table II, which show

that only plastoquinone restores electron flow as well as ATP formation in the ferricyanide Hill reaction whereas *p*-benzoquinone and dimethylbenzoquinone stimulate electron flow but hardly ATP formation.

The results of Fig. 1 and Table II indicate that benzoquinones are better electron acceptors for Photosystem II in intact chloroplasts than ferricyanide. The photoreduction of benzoquinones in substrate amounts in the presence of dibromothymoquinone should therefore be a testsystem to study the properties of Hill reactions involving only Photosystem II.

Fig. 2 compares the sensitivity towards dibromothymoquinone of the Hill reaction with ferricyanide with the reduction of substrate amounts of 2,6-dimethylbenzoquinone. The quinone photoreduction as measured by oxygen evolution is less sensitive to dibromothymoquinone and a rate of 150 μequiv oxygen/h per mg chlorophyll remains even in the presence of 10^{-5} M inhibitor. More significantly an appreciable rate of ATP formation remains in the Hill reaction particularly with the quinone even at the high concentration of 10^{-5} M dibromothymoquinone. By computing the ratio of electron transport to ATP formation form these figures (Fig. 2b) the ratio of electron flow to ATP formation in the quinone Hill reaction is about 0.3 ATP to 2 *e* transferred. This ratio of P/e_2 is about twice the ratio in ferricyanide reduction by Photosystem II (*i.e.* in the presence of dibromothymoquinone). The ratio of ATP formation to electrons transferred in photoreductions by Photosystem I (*i.e.* in the absence of dibromothymoquinone) is about one, for example in anthraquinone photoreduction or ferricyanide reduction (Fig. 2). The ratio P/e_2 is lower than one in the quinone Hill reaction (in the absence of the inhibitor!). This has been observed already 10 years ago[11,12] and has been reported

TABLE III

STIMULATION OF PHOTOREDUCTIONS BY PHOTOSYSTEM II (*i.e.* IN THE PRESENCE OF DIBROMOTHYMOQUINONE) BY UNCOUPLING AT pH 7.2

General conditions: 10 min light in N_2; coupled = addition of ADP/P_i/Mg^{2+}; uncoupled = addition of $2 \cdot 10^{-3}$ M NH_4Cl or 15 μg gramicidin; 20 μmoles ferricyanide or 6 μmoles benzoquinone.

Electron acceptor	*Conditions*	*μequiv O_2 evolved*		
		No addition	*Dibromothymoquinone added*	
			$2 \cdot 10^{-7}$ M	*$2 \cdot 10^{-6}$ M*
Ferricyanide	Coupled	2.4	2.8	3.0
	Uncoupled	4.4	2.5	2.7
p-Benzoquinone	Coupled	2.8	2.8	2.6
	Uncoupled	5.5	4.6	4.2
2,6-Dimethylbenzoquinone	Not coupled	1.5		2.1
	Coupled	2.8	2.7	3.0
	Uncoupled (NH_4Cl)	6.6	5.3	5.9
	Uncoupled (gramicidin)	5.4	4.9	4.7
	Uncoupled + $2 \cdot 10^{-6}$ M DCMU	< 0.1	< 0.1	< 0.1

recently also by Saha *et al.*[13]. It suggests that already in intact chloroplasts benzoquinones are reduced by Photosystem II to a certain extent.

ATP formation in a Hill reaction involving only Photosystem II suggests an energy conserving step connected with Photosystem II. This is supported by the observation that such Hill reactions show a response of the electron flow rate to uncoupling (Table III and Fig. 3). At pH 7.2 the Hill reaction with *p*-benzoquinone and with 2,6-dimethylbenzoquinone is stimulated by adding an uncoupler (NH_4Cl or gramicidin). Significantly the stimulation of electron flow is independent of whether dibromothymoquinone is present or not, *i.e.* it is independent whether the quinone may be reduced by either Photosystem I and II together (*i.e.* absence of dibromothymoquinone) or can be reduced by Photosystem II only (*i.e.* in the presence of dibromothymoquinone). As against the quinone reaction this stimulation by an uncoupler does not occur in the ferricyanide Hill reaction in the presence of the inhibitor (Table III).

DISCUSSION

Quinones have been used extensively as artificial electron acceptors and cofactors for photosynthetic electron transport in chloroplasts[14]. Warburg[15] already in 1944 used *p*-benzoquinone as acceptor in a Hill reaction. Menadione proved to be a good cofactor of cyclic photophosphorylation[16]. A thorough comparison of quinones as Hill acceptors in 1961 showed that the photoreduction of all quinones is coupled to ATP formation[11,12,17]. Some quinoid compounds with a redox potential lower than about zero volt in addition are also cofactors of cyclic photophosphorylation[11,12]. *p*-Benzoquinones, which have redox potentials more positive than 0 V were used to titrate the redox potential of the acceptor side of Photosystem II in mutant strains of algae which have no Photosystem I activity[18,19]. These experiments[18,19] with whole cells were not intended to show details of the mechanism and energy coupling in the photoreductions by Photosystem II.

Ferricyanide and DCIP photoreduction were also shown to be Photosystem II photoreductions, depending on conditions (lit. cit. [2,10]). With the introduction of the potent plastoquinone antagonist dibromothymoquinone[1,2] it was possible to settle some of the discussion as to whether ferricyanide and DCIP photoreductions are mainly Photosystem I reactions in isolated chloroplasts but Photosystem II reactions in fragmented chloroplasts[2,10]. Unfortunately in the later chloroplasts energy control of electron flow is almost lost. Therefore the possibility of an energy conserving step in Photosystem II can not be studied in such chloroplasts by our methods.

As we have already observed in the early paper[11,12] on quinone–Hill reactions the stoichiometry of ATP formation to electron transport in a Hill reaction with *p*-benzoquinones is less than one as generally measured in the ferricyanide Hill reaction or in the Mehler type pseudocyclic electron transport with anthraquinone as acceptor. In retrospect this lower stoichiometry (*i.e.* low P/e_2 ratio) can now be explained by a simultaneous photoreduction of the quinone by Photosystem II and by Photosystem I, only the later part giving high ATP values. Saha *et al.*[13] recently reported again on the lower ratio of P/e_2 in quinone Hill reactions. Their explanation will be discussed below.

By using dibromothymoquinone as an effective inhibitor of electron flow connecting Photosystem II and I, the properties of photoreductions by Photosystem II alone may be studied in intact chloroplasts with undisturbed energy control. The results of this paper suggest an energy-conserving step in electron flow between water oxidation and the acceptor site after Photosystem II. This is based on the observation that (a) Hill reactions, with certain *p*-benzoquinones in the presence of dibromothymoquinone are still controlled by coupling conditions and are stimulated by uncouplers, (b) these Hill reactions by Photosystem II are coupled to ATP formation. The ratio of ATP formed to electron transferred (P/e_2) is about 0.3. The results on an energy-conserving step in the electron flow system close to Photosystem II are a support of the idea that there are two coupling sites in non cyclic photophosphorylation, as argued since many years by Izawa and Good[20].

The assumption of an energy-conserving site close to Photosystem II and possibly between water and Photosystem II is suggested by the results reported here is in accordance with earlier results by ourselves[21]. They furthermore are supported by experiments of Gimmler[22,23] with whole cells. By reversing the dibromothymoquinone inhibition of light induced energy depended conformational changes in the algae *Dunaliella* by benzoquinone Gimmler[22] noted that this reaction is still under control by the phosphorylation condition. In addition he[23] observed that fluorescence of Photosystem II is under energy control, thus providing an additional methodical approach to the problem. Also the experiments of Schliephake *et al.*[24] indicated coupling sites at both photosystems.

One explanation of the control of Photosystem II by the phosphorylating system would be to visualize the energy-conserving step in the Photosystem II system connected with the protons released in the water-splitting reaction. The assumption is based on experiments particularly of Junge and co-workers[25,27] and Schröder *et al.*[26], who suggested that the water-splitting reaction occurs inside the thylakoid membrane. Protons from this reaction lower the pH inside the membrane. These protons left behind in the water-splitting reaction would contribute to the pH gradient generated by the additional protons translocated by electron and proton carriers between the two light reactions (possibly plastohydroquinone). This total pH gradient would be responsible for the overall stoichiometry of about 1.0 to 1.3 ATP formed per 2 *e* transferred in the Hill reaction with electron flow from water to Photosystem I. The theoretical ratio of $P/e_2 = 1.3$ results because according to Rumberg *et al.*[25,26], the experiments at present suggest that 4 protons per 2 *e* are generated inside the membrane and $3H^+$ are needed to drive the formation of one ATP. Reeves *et al.*[28] have summarized the conditions under which these ratios of ATP/2 *e* above 1 are also experimentally obtained in Hill reactions involving both photosystems.

In photoreductions by Photosystem II as presented in this paper only the protons of the water splitting reactions would be available and responsible for energy control of electron flow and for ATP formation. From Rumberg's computation above it is obvious that P/e_2 values are not necessary whole numbers. The experimental value of P/e_2 obtained is 0.3 in photoreductions by Photosystem II, when benzoquinones are used as acceptors. This ratio is less than the 0.65 one might expect (half of the theoretical value of 1.3 for photoreductions including both photosystems). A constant leakage rate of protons (or membrane potential decay) would

effect the ratio in photoreductions by Photosystem II alone more than in the complete electron flow system. Possibly also the effectiveness of the energy-conserving site at Photosystem II is lower than the one between the two light reactions. Certainly in the Hill reaction with Photosystem I the energy conserving site at plastoquinone is controlling the overall rate.

A second explanation for the control of electron flow through Photosystem II may be based on the result that the control is observed only in the reduction of quinones. The photoreduction of ferricyanide by Photosystem II, *i.e.* its reduction in the presence of dibromothymoquinone is not stimulated by an uncoupler. Also the ratio of ATP/2 *e* is only about 0.16. This difference to the photoreduction of a quinone by Photosystem II is probably due to the different polarity and lipophilicity. Because the rate of access of the polar ferricyanide to Photosystem II is limiting (otherwise quinones would not stimulate electron flow, Table II) stimulation by uncouplers can not be expected. We suggest that under these conditions ferricyanide and quinones are reduced on different sides of the membrane. One would assume that the polar ferricyanide reacts on the outer, the lipophilic quinone on the inner side of the membrane. Both presumably oxidize plastohydroquinone because dibromothymoquinone inhibits the oxidation of plastohydroquinone by Photosystem I but not the reduction of plastoquinone by Photosystem II, as already discussed above (refs 1, 2 and 5; Haehnel, W., personal communication). If one follows these assumptions then also in the presence of dibromothymoquinone plastohydroquinone would be able to transport a proton across the membrane, when oxidized on the inside of the membrane by a quinone but not when oxidized by ferricyanide on the outside. This would account for the observed result of coupling and control in the first and no control in the latter case.

Saha *et al.*[13] recently have divided electron acceptors in Hill reactions into different classes according to their polarity; the reduction of ferricyanide and of charged quinones possibly involving two ATP forming sites and of quinodiimines only one. They suggest that they are reduced on different sites of the membrane. They[13] noted that lipophilic quinones seemed to behave in between, *i.e.* their ratio of P/e_2 is lower than in the ferricyanide reaction. They offer the explanation that one class may be reduced at the hydrophilic Photosystem I, the other class at the lipophilic Photosystem II. Our data here give evidence for their hypothesis. We agree with them on their principal emphasis on polarity of the acceptor used and would like to extend their hypothesis. Our data suggest that there is not just one hydrophilic and one lipophilic site in the electron transport chain for the possible reduction of polar or lipophilic acceptors, respectively, but at least two. Both photosystems have lipophilic and hydrophilic regions, though indeed Photosystem I is definitely more hydrophilic. At the lipophilic region of Photosystem II quinones are reduced, at the hydrophilic site ferricyanide is reduced. According to the theory of Mitchell loops in the electron transport chain from inside the membrane towards the outside and back are leading to a proton gradient across the membrane. One might expect as many hydrophilic areas as loops emerge on the outer membrane surface. Our results support the notion that there are two such loops and that polar acceptors may react, though with different rates, with both ends, *i.e.* the acceptor site of Photosystem I but also of Photosystem II. The same argumentation of course yields several reducing sites for lipophilic compounds also.

In addition to considerations of chemical parameters of the acceptors the structural state of the chloroplast is of importance. In fragmented chloroplasts lipophilic region of the electron transport chain are opened up and polar acceptors get faster access to sites buried in intact chloroplasts[10].

The site of reduction of acceptors has marked influence on the stoichiometry of phosphorylation to electron flow. In intact chloroplasts the polar ferricyanide is reduced by Photosystem I with a high P/e_2 ratio, whereas the lipophilic quinones are reduced already to a certain extent by Photosystem II, thus excluding an energy-conserving site. The P/e_2 ratio of 0.7 Saha *et al.*[13] observed in one class *i.e.* half the maximal ratio of the other class is purely accidental in our view. If electron flow through Photosystem I is prevented, either by dibromothymoquinone or fragmentation, only Photosystem II can reduce electron acceptors of suitable redoxpotential. Again they may be reduced in- or outside the membrane. Under these conditions the polar ferricyanide becomes a very poor acceptor, its P/e_2 ratio is low and the reaction shows no response to uncoupling. Lipophilic quinones (in substrate amounts) are now good acceptors with a (relatively) high P/e_2 ratio, but not exactly half the ratio in undisturbed chloroplasts. The reduction is stimulated by uncoupling.

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REFERENCES

1 Trebst, A., Harth, E. and Draber, W. (1970) *Z. Naturforsch.* 25b, 1157
2 Böhme, H., Reimer, S. and Trebst, A. (1971) *Z. Naturforsch.* 26b, 341
3 Böhme, H. and Cramer, W. A. (1971) *FEBS Lett.* 10, 866
4 Cramer, W. A., Fan, H. N. and Böhme, H. (1971) *J. Bioenerg.* 2, 289
5 Lozier, R. H. and Butler, W. L. (1972) *FEBS Lett.* 26, 161
6 Gimmler, H. and Avron, M. (1972) *Proc. 2nd Int. Congr. Photosynth. Res., Stresa, 1971,* Vol. 1, p. 789, Dr W. Junk N.V. Publishers, The Hague
7 Nelson, N., Drechsler, Z. and Neumann, J. (1972) *J. Biol. Chem.* 246, 143
8 Sugino, Y. and Miyoshi, Y. (1964) *J. Biol. Chem.* 239, 2360
9 Vernon, L. P. and Shaw, E. R. (1965) *Biochemistry* 4, 132
10 Trebst, A. (1972) *Proc. 2nd Int. Congr. Photosynth. Res., Stresa, 1971,* Vol. 1, p. 399, Dr. W. Junk N.V. Publishers, The Hague
11 Trebst, A. and Eck, H. (1961) *Z. Naturforsch.* 16b, 44
12 Trebst, A., Eck, H. and Wagner, S. (1963) in *Photosynthetic Mechanisms of Green Plants* (Kok, B. and Jagendorf, A. T., eds), p. 174, Natl. Acad. Sci. Natl. Res. Counc., Washington, D.C.
13 Saha, S., Ouitrakul, R., Izawa, S. and Good, N. E. (1971) *J. Biol. Chem.* 246, 3204
14 Arnon, D. I. (1961) *Fed. Proc.* 20, 1012
15 Warburg, O. and Lüttgens, W. (1944) *Naturwissenschaften* 32, 301
16 Arnon, D. I., Allen, M. B. and Whatley, F. R. (1954) *Nature* 174, 394
17 Hill, R. and Walker, D. A. (1959) *Plant Physiol.* 34, 240
18 Kok, B. and Datko, E. A. (1965) *Plant Physiol.* 40, 1171
19 Kok, B., Malkin, S., Owens, O. and Forbush, B. (1966) *Brookhaven Symp. Biol.* 19, 446
20 Izawa, S. and Good, N. E. (1968) *Biochim. Biophys. Acta* 162, 380
21 Böhme, H. and Trebst, A. (1969) *Biochim. Biophys. Acta* 180, 137
22 Gimmler, H. (1973) *Z. Pflanzenphysiol.* 68, 289
23 Gimmler, H. (1973) *Z. Pflanzenphysiol.* 68, 385
24 Schliephake, W., Junge, W. and Witt, H. T. (1968) *Z. Naturforsch.* 23b, 1571
25 Junge, W., Rumberg, B. and Schröder, H. (1970) *Eur. J. Biochem.* 575, 14

26 Schröder, H., Muhle, H. and Rumberg, B. (1972) in *Proc. 2nd Int. Congr. Photosynth. Res., Stresa, 1971*, Vol. 2, p. 919, Dr W. Junk N.V. Publishers, The Hague
27 Junge, W. (1972) *Abstr. Int. Photobiol. Congr., Bochum*, p. 34
28 Reeves, S. G., Heathcoke, P. and Hall, D. O. (1972) *Abstr. Int. Photobiol. Congr., Bochum*, p. 284