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Light-Dependent Incorporation of Adenine Nucleotide into Noncatalytic Sites of Chloroplast ATP Synthase

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Abstract—The binding of ADP and ATP to noncatalytic sites of dithiothreitol-modified chloroplast ATP synthase was studied. Selective binding of nucleotides to noncatalytic sites was provided by preliminary light incubation of thylakoid membranes with [^{14}C]ADP followed by its dissociation from catalytic sites during dark ATP hydrolysis stimulated by bisulfite ions ("cold chase"). Incorporation of labeled nucleotides increased with increasing light intensity. Concentration-dependent equilibrium between free and bound nucleotides was achieved within 2-10 min with the following characteristic parameters: the maximal value of nucleotide incorporation was 1.5 nmol/mg of chlorophyll, and the dissociation constant was 1.5 μM. The dependence of nucleotide incorporation on Mg^{2+} concentration was slight and changed insignificantly upon substituting Ca^{2+} for Mg^{2+}. Dissociation of nucleotide from noncatalytic sites was illumination-dependent. The dissociation kinetics suggested the existence of at least two nucleotide-binding sites with different dissociation rate constants.

Key words: ATP synthase, CF_0F_1, noncatalytic sites, chloroplasts

ATP synthases (F_1F_0) of chloroplasts, mitochondria, and bacteria catalyze phosphorylation of ADP using the transmembrane gradient of proton electrochemical potential. Proton transfer is performed by a membrane part of the complex (F_0), while enzymatic function is fulfilled by its peripheral part, i.e., by the coupling factor F_1 comprising 3α, 3β, γ, δ, and ε subunits. At the interface between α and β, there are three catalytic sites and three sites termed "noncatalytic" [1]. The latter are capable of nucleotide binding but display no catalytic properties [2]. In solution, coupling factors of various origins catalyze ATP hydrolysis. The F_1-ATPase activity is inhibited in the presence of ADP and Mg^{2+} [3-6]. This inactivation is accompanied by tight binding of MgADP to one catalytic site, by MgATP binding to another catalytic site, and by nucleotide dissociation from the third one [1, 7]. An elevated ATP concentration [8, 9] and the presence of weak acid oxyanions (sulfite, carbonate, borate, etc.) in the reaction mixture [10-12] result in reactivation of ATPase. As reported by Boyer and colleagues and by Allison and colleagues, the reversible reactions of F_1-ATPase inactivation/reactivation involve not only catalytic but also noncatalytic nucleotide binding sites [9, 13]. A prerequisite to F_1-ATPase reactivation was ATP binding to a noncatalytic site [9, 14, 15], which expedited dissociation of ADP tightly bound to a catalytic site [16]. Presumably, a similar mechanism is used for oxyanion binding to noncatalytic sites [17].

Unlike those of F_1, the noncatalytic sites of F_1F_0 have not been adequately investigated. The reported data on their role and properties are comparatively few in number and contradictory. According to Pitard et al. [18], ATP binding to noncatalytic sites of ATP synthase from thermophilic bacteria PS3 that had been reconstituted into liposomes together with bacteriorhodopsin resulted in a higher photophosphorylation rate. As reported by Possmayer and colleagues, noncovalent binding of 2-azido-ATP (an ATP analog) to noncatalytic sites of chloroplast ATP synthase (CF_0F_1) reconstituted into liposomes produced no effect on photophosphorylation [19]. Here, energization of a phospholipid membrane by $\Delta pH/\Delta\varphi$ did not affect the rate of 2-azido-ATP dissociation. Unlike CF_1 catalytic sites that are capable of binding both ATP and ADP [13, 20], those of CF_0F_1 were reported [19] to bind only 2-azido-ATP. The above contradictions could be a result of different properties of ATP synthases of different origin (from thermophilic bacteria and from chloroplasts) and also could be caused by different experimental conditions or by peculiarities of interaction between azido derivatives and noncatalytic sites of the enzyme.

Abbreviations: PMS) phenazine methosulfate; DTT) dithiothreitol.

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The current paper focuses on [^{14}C]ADP and [^{14}C]ATP binding to noncatalytic sites of ATP synthase integrated into a native chloroplast thylakoid membrane. The thylakoid membrane was energized by light. To estimate nucleotide binding to ATP synthase, we modified the "cold chase" technique that had been used to study noncatalytic sites of coupling factors from chloroplasts, mitochondria, and bacteria [13, 14, 20-22].

MATERIALS AND METHODS

Chloroplast thylakoid membranes were isolated from pea leaves as described by Semenova et al. [23] with the following modifications. Washed leaves were incubated in a refrigerator for 24 h and then homogenized for 5 min in medium containing 0.4 M sucrose, 20 mM Tris-HCl (pH 7.8), and 20 mM NaCl. The homogenate was filtered through four layers of gauze and centrifuged at 1000*g* for 5 min. The supernatant was centrifuged once again at 5000*g* for 5 min, and the pellet resuspended in solution containing 0.2 M sucrose, 10 mM Tricine-KOH (pH 7.8), 10 mM NaCl, 5 mM $MgCl_2$. The membranes were collected by centrifugation for 5 min at 5000*g* and washed once again with the same solution. Sedimented membranes were resuspended in 20 mM Tricine-KOH (pH 7.8), 0.2 M sucrose, 20 mM KCl, and 1 mM $MgCl_2$ (TKM buffer) to a chlorophyll concentration of 2.5-4.0 mg/ml and stored in the dark at 4°C. Chlorophyll was determined according to [24]. Using an infrared radiation filter SZS-27, the membranes were illuminated with white light at 560 W/m^2 for 5 min in reaction medium containing TKM buffer, 50 μM phenazine methosulfate (PMS), 10 mM dithiothreitol (DTT), and 0.5-1.0 mg/ml chlorophyll, sedimented by centrifugation, and resuspended in TKM buffer to give a final chlorophyll concentration of 1-2 mg/ml.

Nucleotide was bound to CF_0F_1 of thylakoid membranes as follows: the membranes were illuminated with white light (560 W/m^2) at room temperature in 10 ml reaction medium containing [^{14}C]ADP, TKM buffer, 50 μM PMS, and 40-50 μg/ml chlorophyll. The modified "cold chase" technique was used to stop binding of labeled nucleotides and to start their selective dissociation from catalytic sites. At indicated intervals, 1 ml aliquots were taken and mixed with 100 μl quenching solution containing 22 mM ATP, 2.2 mM $MgCl_2$, 0.66 M $KHSO_3$ (pH 7.8), 50 μM diadenosine pentaphosphate, and 50 μM gramicidin. After 2 min the mixture was centrifuged for 1 min at 10,000*g*, the pellet was resuspended in 0.2 M sucrose, 50 mM Tricine-KOH (pH 7.8), 50 mM KCl, 5 mM $MgCl_2$, and 1 mM ATP (TKMA buffer), and free nucleotides were removed by forced gel filtration using a Sephadex G-50 (fine) column (0.5 × 8 cm) equilibrated with TKMA buffer. Alternatively, to remove free nucleotides the pellet obtained after centrifugation was washed three times and resuspended in 90 μl TKMA buffer. For chlorophyll determination, 20-40 μl of the resultant suspension was utilized, and the rest was used for nucleotide extraction by the 0.5 M $HClO_4$/KOH procedure. To determine radioactivity, 20 μl extract aliquots were placed into vials with scintillation liquid. Kinetic data was processed using Origin 6.0 (Microcal Software).

RESULTS

To estimate nucleotide incorporation into CF_0F_1 noncatalytic sites, a technique providing selective dissociation of radioactive nucleotides from catalytic sites ("cold chase") was used. The technique depends on different nucleotide exchange rates at catalytic and noncatalytic sites during ATP hydrolysis [2, 25]. As major events, this technique involves enzyme incubation with radioactive nucleotides that provides label binding to sites of the both types, introduction of unlabeled ATP into the reaction mixture to displace labeled nucleotides from catalytic sites, separation of free nucleotides from those tightly bound to noncatalytic sites, and finally, estimation of the amount of noncatalytic site-bound nucleotides from their radioactivity. Applicability of this technique to investigation of properties of CF_0F_1 noncatalytic sites depends on two parameters: first, on whether or not ATPase activity of thylakoid membranes can be enhanced, because in the dark this activity, and hence, the nucleotide exchange rate at catalytic sites, is extremely low; and second, on conditions of free nucleotide separation from thylakoid membranes under which nucleotide dissociation from noncatalytic sites is negligible. In 1989, Larson and colleagues discovered that ATPase activity of DTT-modified thylakoid membranes can be many tens of times higher in the presence of ammonium sulfate and at a high sulfite concentration [26].

Our preliminary experiments showed that an optimal reaction mixture must contain 60 mM sulfite, 5 μM gramicidin, and 1-2 mM $MgCl_2$. Unlike spinach chloroplasts [26], those of pea were not stimulated by ammonium sulfate. At 20°C and with 2 mM ATP added, the hydrolysis rate was 200-400 nmol/min per mg chlorophyll, i.e., when converted to CF_0F_1 activity, 200-400 min^{-1} (assuming the ratio to be 1 nmol of CF_0F_1 per mg chlorophyll [27]). Since at the indicated ATP concentrations the reaction consecutively involved all the three catalytic sites [28, 29], an easy calculation gives 70 to 130 catalytic cycles per min happening at each of them. The rate of nucleotide dissociation from DTT-reconstituted CF_1 noncatalytic sites ranged from 10^{-3} (ATP) to $1.5 \cdot 10^{-1}$ (ADP) min^{-1} [20]. Such a great rate difference between catalytic and noncatalytic sites allows, in principle, applying the "cold chase" technique for assessment of nucleotide binding to CF_0F_1 noncatalytic sites.

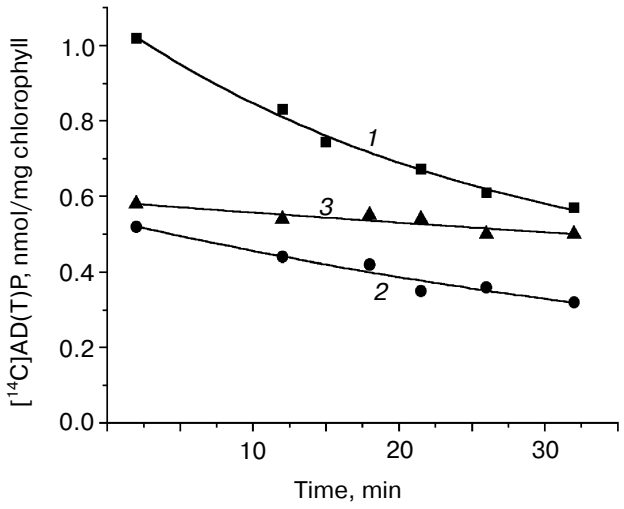

Fig. 1. Effect of conditions of free nucleotide separation on the content of labeled nucleotides tightly bound to thylakoid membranes. Light- and DTT-activated thylakoid membranes (1.4 mg chlorophyll/ml) were incubated with 0.2 M sucrose, 20 mM Tricine (pH 7.8), 20 mM KCl, 1 mM $MgCl_2$, 50 μM PMS, and 4.6 μM [^{14}C]ADP for 4 min at 20°C. To make labeled nucleotides dissociate from catalytic sites, the reaction mixture was supplemented with 2 mM ATP, 60 mM $KHSO_3$, and 5 μM gramicidin D (final concentrations) (curves *2* and *3*). Free nucleotides were separated either by gel filtration (the initial points of the curves) or by 3-7 centrifugation/re-suspension procedures (the remaining points of the curves) in TKM buffer (curves *1* and *2*) or in TKMA buffer (curve *3*). Tightly bound nucleotides were extracted by treating thylakoid membranes with 0.6 M $HClO_4$ and KOH followed by centrifugation.

The initial point on curve *1* of Fig. 1 shows the amount of tightly bound nucleotides after light incubation of thylakoid membranes in the presence of [^{14}C]ADP, and after removal of loosely bound nucleotides using a Sephadex G-50 fine column. Presumably, tightly bound nucleotides include both ADP and ATP, the latter resulting from adenylate kinase reaction that occurs in the reaction mixture [30]. Addition of ATP and sulfite to the reaction mixture prior to application of thylakoid membrane suspension onto a column (cold chase) results in about two-fold decrease in the amount of tightly bound nucleotides (first point of curve *2*). The difference in amount of tightly bound nucleotides can possibly be attributed to replacement of radioactive nucleotides at catalytic sites by unlabeled ones in the course of a catalytic reaction. Radioactive nucleotides retained by thylakoid membranes should, by definition [2], pertain to noncatalytic sites. As seen from curves *1* and *2*, a longer washing procedure causes a slow decrease in the amount of nucleotides tightly bound to thylakoid membranes. Increasing Mg^{2+} concentration from 1 to 5 mM (not shown) and ATP (curve *3*) had a stabilizing effect. It should be noted that dissociation of a portion of tightly bound nucleotides in the presence of ATP and sulfite cannot be caused by membrane energization due to ATP hydrolysis-induced transmembrane proton gradient because light cessation was accompanied by addition of 5 μM gramicidin D concurrently with ATP and sulfite.

The effect of light intensity on nucleotide incorporation into CF_0F_1 noncatalytic sites is shown in Fig. 2. In experiments, light intensity was varied using neutral gray filters of different density. The indicated light intensity values are average of those in front of and behind the reaction mixture (the difference was never more than 30%). These results show that nucleotide incorporation into noncatalytic sites is activated by light. The incorporation rate increases with increasing nucleotide concentration (Fig. 3). In the 0.7-12.6 μM nucleotide range, equilibrium between bound and free nucleotides is reached within 1-10 min. Figure 4 shows nucleotide incorporation into noncatalytic sites versus nucleotide concentration. The Origin 6-computated dissociation constant was 1.5 μM, and the maximal incorporation was 1.5 nmol nucleotides per mg chlorophyll. Of note, different samples of thylakoid membranes showed different maximal incorporations (difference range, 0.5-1.6 nmol/mg chlorophyll) but almost the same dissociation constants and their rates (see below). The mentioned differences may result from different extents of coupling of electron transport with transmembrane proton transfer, which affects energization of thylakoid membrane, as

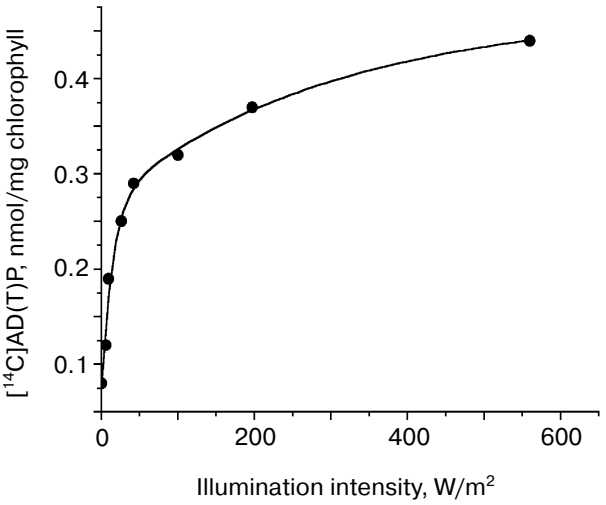

Fig. 2. Effect of light intensity on nucleotide incorporation into noncatalytic sites of chloroplast ATP synthase. Light- and DTT-activated thylakoid membranes (0.15 mg chlorophyll/ml) were incubated with 0.2 M sucrose, 20 mM Tricine-KOH (pH 7.8), 20 mM KCl, 1 mM $MgCl_2$, 50 μM PMS, and 4.8 μM [^{14}C]ADP for 40 sec. After dissociation of labeled nucleotides from catalytic sites, free nucleotides were removed from the thylakoid membranes by washing with TKM buffer (three times); then tightly bound nucleotides were extracted with $HClO_4$/KOH, and their radioactivity was determined using a scintillation counter.

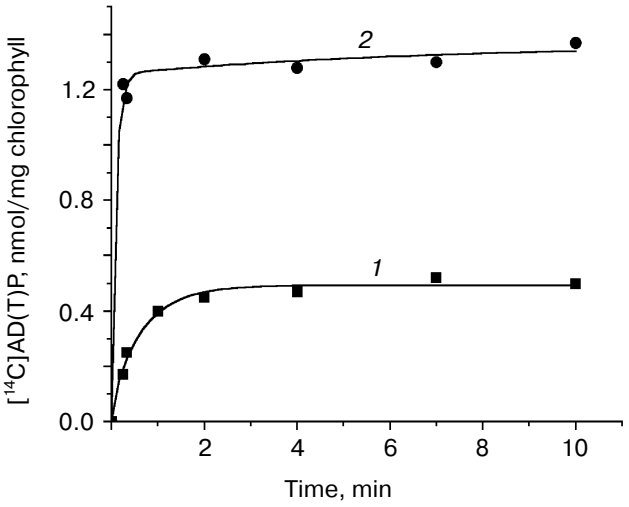

Fig. 3. Kinetics of ^{14}C-labeled nucleotide binding to CF_0F_1 noncatalytic sites. Light- and DTT-activated thylakoid membranes (0.05-0.15 mg chlorophyll/ml) were incubated with 0.7 (*1*) or 12.6 μM (*2*) [^{14}C]ADP in reaction mixture containing TKM buffer (pH 7.8) and 50 μM PMS. After light cessation and removal of ^{14}C-labeled nucleotides from catalytic sites using 2 mM ATP and 60 mM K_2SO_3, the thylakoid membranes with nucleotides tightly bound to their noncatalytic sites were separated as described under "Materials and Methods".

well as from a changing chlorophyll/CF_0F_1 ratio [31]. Over 40 experiments conducted in the course of a year gave the average nucleotide concentration in pea chloroplasts as 3.8 nmol (ADP + ATP) per mg chlorophyll, which agrees with the literature [32]. However, the maximal and minimal values differed by more than twofold (2.1 and 5.7 nmol (ADP + ATP) per mg chlorophyll, respectively). Presumably, these differences must comply with such of the chlorophyll to CF_0F_1 ratio, because the amount of tightly bound nucleotides is determined by properties of the enzyme.

As seen from Fig. 5, increasing concentration of Mg^{2+} in the reaction mixture is accompanied by decreasing rate of nucleotide dissociation from ATP synthase noncatalytic sites. In the presence of 1 mM $MgCl_2$, the dissociation kinetics correlate with nucleotide binding to two sites with dissociation rate constants of 5.0 ± 0.5 and 0.24 ± 0.05 min^{-1}. With $MgCl_2$ concentration as high as 10 mM, the former decreases insignificantly (to 4.0 ± 0.4 min^{-1}), while the latter almost 6-fold (to 0.04 ± 0.006 min^{-1}). The level of nucleotide incorporation depended on Mg^{2+} concentration only slightly (Fig. 6). Nucleotide binding appeared to be unaffected by replacement of Mg^{2+} with calcium ions, which support hydrolysis but, unlike Mg^{2+}, cannot support ATP synthesis.

DISCUSSION

To date, there is convincing evidence for participation in the reaction of all the three catalytic sites, provided ATP concentration is above K_m [29]. This means that after preincubation of thylakoid membrane with ^{14}C-labeled nucleotides followed by incubation with 2 mM ATP and 60 mM sulfite, ATP synthase retains only

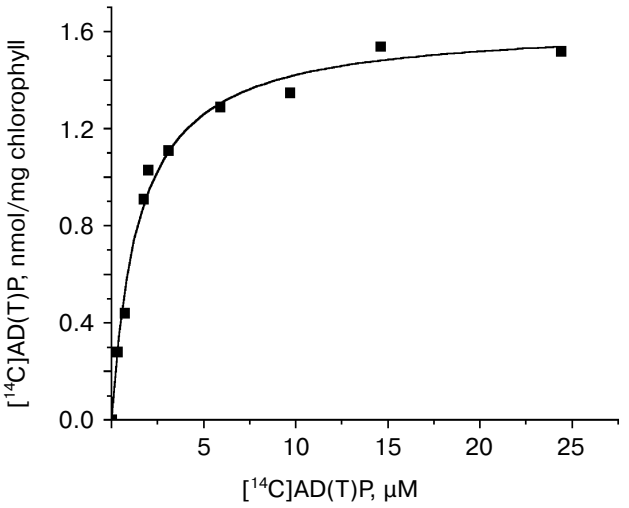

Fig. 4. Concentration dependence of nucleotide incorporation into CF_0F_1 noncatalytic sites. Light- and DTT-activated thylakoid membranes (0.05-0.15 mg of chlorophyll/ml) were incubated with [^{14}C]ADP of various concentrations in reaction mixture containing TKM buffer (pH 7.8) and 50 μM PMS for 10 min. The extent of nucleotide binding to noncatalytic sites was determined as described under "Materials and Methods".

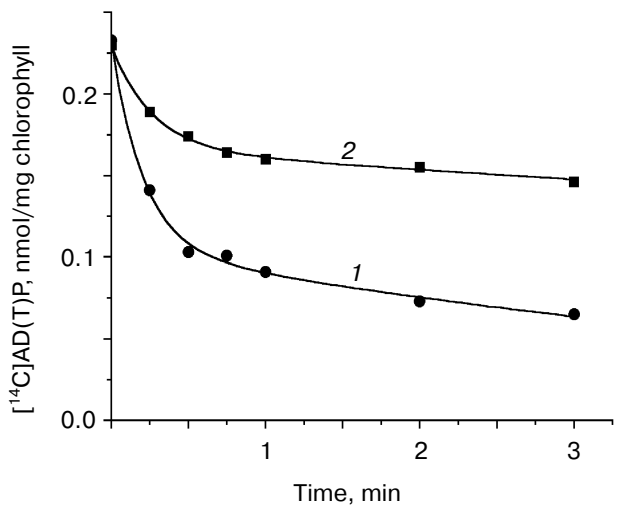

Fig. 5. Kinetics of nucleotide dissociation from CF_0F_1 noncatalytic sites. Light- and DTT-activated thylakoid membranes containing 0.3 nmol of ^{14}C-labeled adenine nucleotides per mg chlorophyll were incubated in the light with 1 mM (*1*) or 10 mM (*2*) $MgCl_2$ and 2 mM unlabeled ATP. Aliquots were taken after indicated time intervals, and the amount of labeled nucleotides was determined.

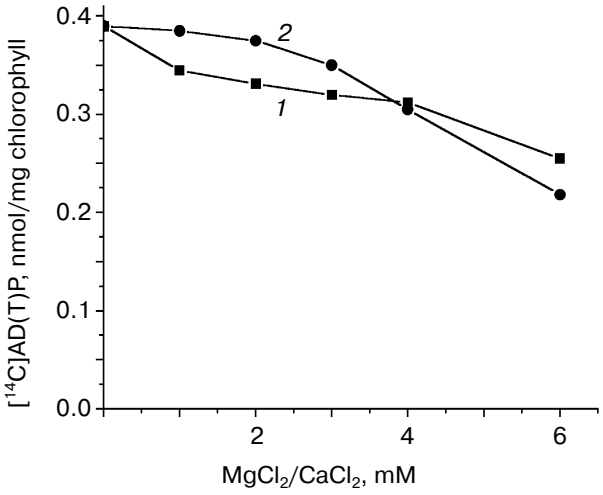

Fig. 6. Dependence of nucleotide incorporation into CF_0F_1 noncatalytic sites on Mg^{2+} (*1*) and Ca^{2+} (*2*) concentration. Light- and DTT-activated thylakoid membranes (0.05-0.15 mg chlorophyll/ml) were incubated in the light for 5 min with 0.9 μM [^{14}C]ADP and with Mg^{2+} (*1*) or Ca^{2+} (*2*) of various concentrations. The initial points on both curves reflect addition to the reaction mixture of 0.05 mM EDTA instead of Mg^{2+} or Ca^{2+}.

labeled nucleotides located at noncatalytic sites. In our experiments, the maximal nucleotide binding to noncatalytic sites amounted to 1.5 nmol/mg chlorophyll (Fig. 4). Making allowance for nucleotide loss that occurs during membrane washing and gel filtration (~10% according to our estimates) and during membrane precipitation with perchloric acid (~12% [32]), the incorporation is about 2 nmol of bound nucleotides per mg chlorophyll. With a value of 1 nmol CF_0F_1 per mg chlorophyll presently accepted in the literature [27], this corresponds to filling two noncatalytic nucleotide binding sites. Mathematical description of light-dependent dissociation requires participation of at least two sites as well, with different dissociation rate constants (Fig. 5). The lack of nucleotide binding at the third noncatalytic site could be a result of its comparatively high dissociation constant or of a high nucleotide dissociation rate, which may be the cause of nucleotide loss at the step of removal of loosely bound nucleotides. The above results are in agreement with previously reported heterogeneity of noncatalytic sites of isolated coupling factor CF_1 [20] and with a paper by Possmayer et al. [19] according to which one noncatalytic site of liposome-integrated CF_0F_1 displays about 25 times lower affinity for 2-azido-ATP as compared with its two other sites. However, it should be noted that due to variability of the CF_0F_1/chlorophyll ratio of different chloroplast preparations (see "Results"), a possibility of nucleotide tight binding to the third noncatalytic site cannot be ruled out.

Light incorporation of labeled nucleotides into thylakoid membranes is probably a result of exchange between nucleotides of the reaction mixture and endogenous nucleotides of ATP synthase noncatalytic sites. The observed light dependence of the incorporation (Fig. 2, also cf. Figs. 1 and 5) indicates that this exchange is energy-dependent. However, unlike the exchange at a catalytic site, this event is much slower (Figs. 2 and 5). The rate constant of nucleotide binding to noncatalytic sites calculated from dissociation constant and from dissociation rate constant is about 3 $min^{-1}\cdot\mu M^{-1}$ (with 1 mM $MgCl_2$). This means that at physiological nucleotide concentrations the exchange at noncatalytic sites must be limited by nucleotide dissociation.

A considerable nucleotide incorporation observed at Mg^{2+} (Ca^{2+}) concentrations below 1 mM (Fig. 6) can be explained on the analogy with catalytic sites. Tight binding of ADP to one of the catalytic sites is known to require no Me^{2+} [34]. In the absence of metal ions it is probably mostly ADP that binds to noncatalytic sites. As shown previously, noncatalytic sites of chloroplast coupling factor can bind both ATP and ADP [14, 20].

Asymmetric effects of Ca^{2+} on ATP synthesis and hydrolysis reported by Racker in his earliest paper on properties of chloroplast coupling factor [35] have not been explained yet. As known, Ca^{2+} catalyzes hydrolysis but does not support ATP synthesis, although it provides the same level of proton transmembrane gradient as Mg^{2+}. The above illustrated similar Mg^{2+} and Ca^{2+} effects of nucleotide binding (Fig. 6) suggest that the particular Ca^{2+} effect on photophosphorylation is unconnected with its effect on interaction between nucleotides and noncatalytic sites of chloroplast ATP synthase.

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