Supporting Information for: "**The Catalytic Cycle of Water Oxidation in Crystallized Photosystem II Complexes: Performance and Requirements for Formation of Intermediates"**

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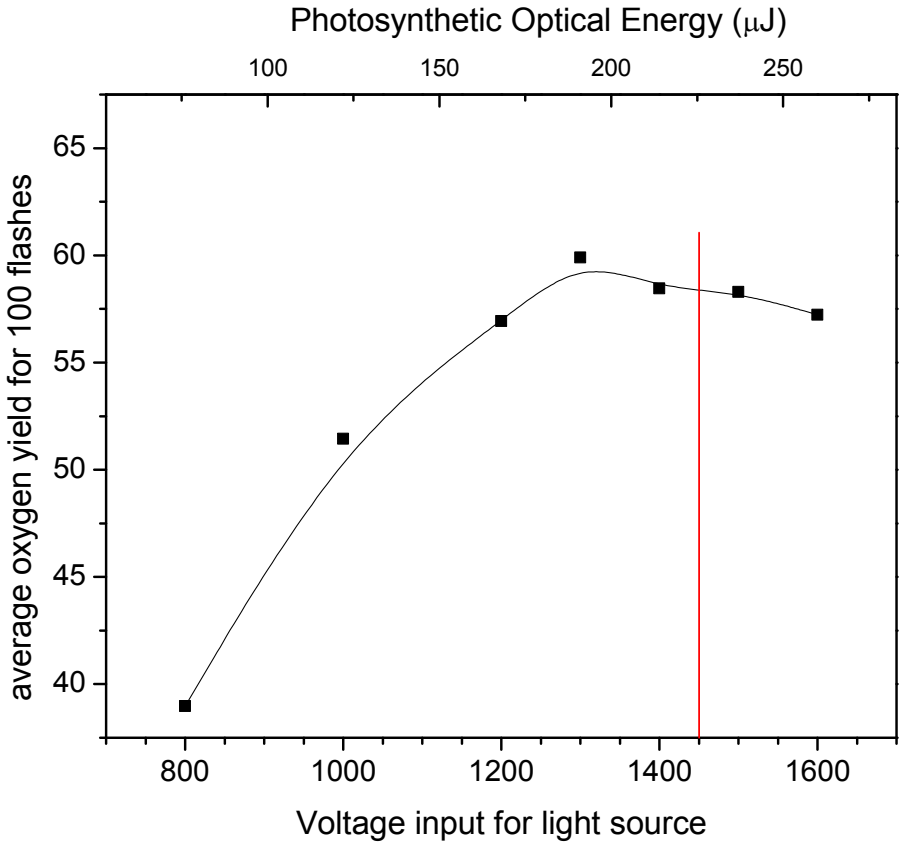

Figure S1. Flash oxygen evolution by dissolved microcrystals (core PSII) as a function of voltage input to the xenon flasher (bottom x axis) and the photosynthetic energy (top x axis).

The oxygen yield does not increase with voltage above 1300 V (174 μJ). This shows light saturation at around 1300 V, and low photoinhibition for higher voltage.

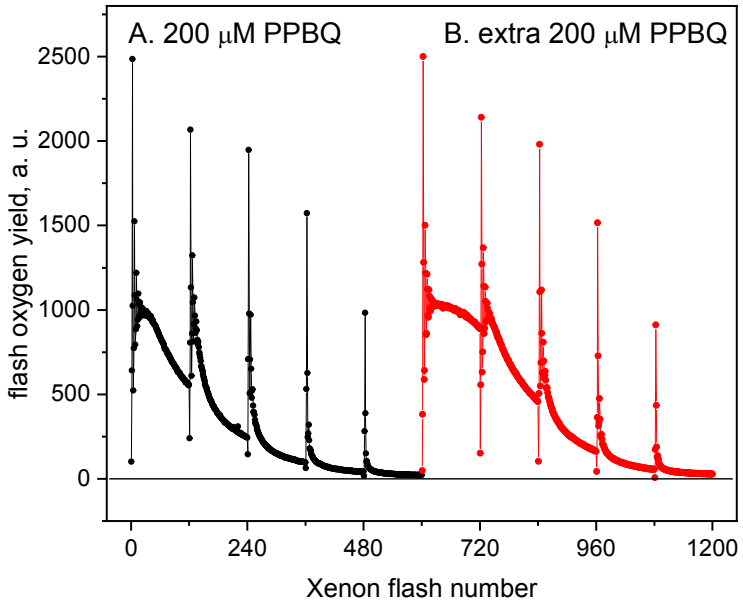

Figure S2. Reactivation of PSII microcrystals after consumption of electron acceptor. Microcrystal sample was supplemented with 200 μM PPBQ and subjected to 5 trains of 120 flashes, resulting in >90% loss of oxygen yield from peak by the last train. Sample was then supplemented with another 200 μM PPBQ, which fully restored oxygen-evolving activity.

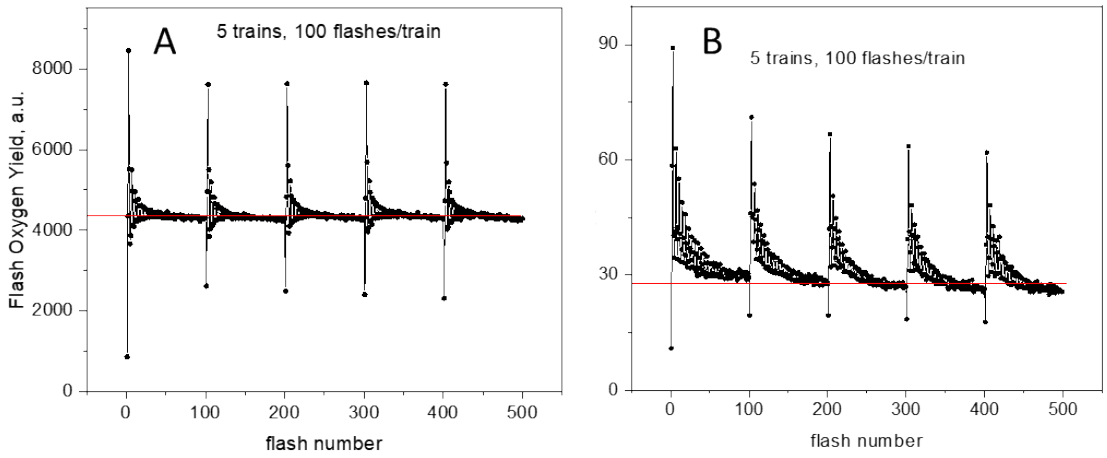

Figure S3. Flash oxygen yield of PSII (A) microcrystals and (B) dissolved PSII core complexes (microcrystals dissolved in 0.015% β-DDM) supplemented with 2 mM FeCN. Measurements were taken in five sequential flash trains of 100 flashes per train with 10 minutes dark pre-incubation time before each train. Both samples were conditioned for 24 hours after dilution in room temperature to ensure dissolution with β-DDM.

In both the microcrystals and the core complexes, the steady state value remains almost constant for each train. Additionally, oxygen yield is recovered for each train in both cases, which implies that there is no long-term electron acceptor limitation. The decrease within each train when using core complexes obtained by treating microcrystals with 0.015% detergent could be due to the generation of ROS during the flashes or to increased availability of reduced electron acceptor.

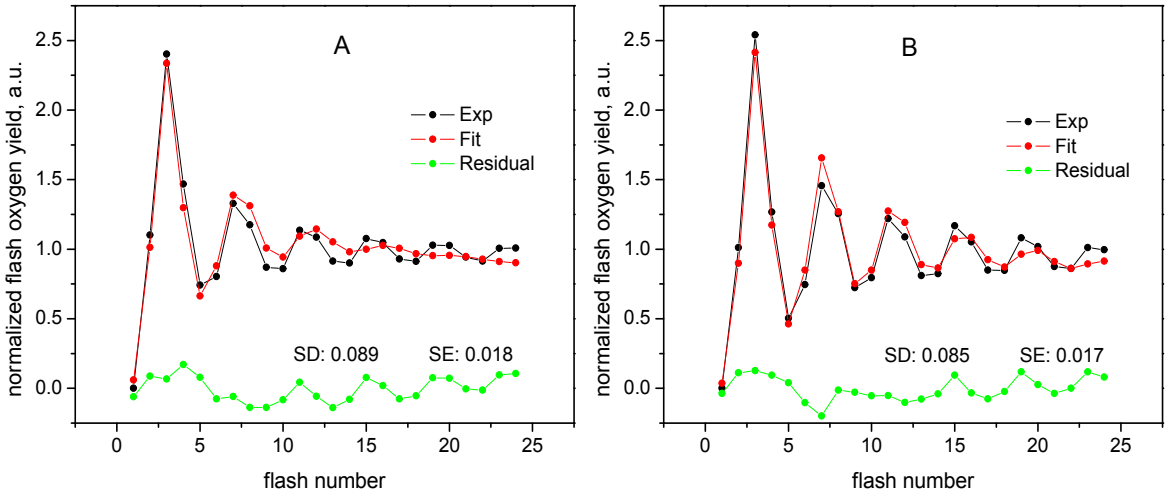

Figure S4. Normalized flash oxygen yield for PSII microcrystals (A) precipitated on the membrane of the oximetric cell, and (B) suspended uniformly in solution in the oxygen cell using bio-gel P-10. The data was fitted with the VZAD model, and the VZAD parameters are shown in Table S1. Raw data are shown in figure 8B.

Table S1. PSII S state populations and inefficiency parameters obtained from the WOC cycle model fits shown in Figure S4. All samples were supplemented with 2 mM ferricyanide and subjected to 10 min dark pre-adaptation time.

VZAD Parameter	Cells *T. elongatus*	PSII microcrystals	
		Precipitated	Suspended in Bio-gel P-10
α, miss	0.100	0.180	0.104
β, double hit	0.051	0.058	0.041
δ, backward transition	0.000	0.000	0.000
ε, inactivation	0.010	0.046	0.047
S0 dark population	0.389	0.065	0.161
S1 dark population	0.444	0.751	0.666
S2 dark population	0.166	0.183	0.173
S3 dark population	0.009	0.000	0.000
Experimental FT Period	4.160	4.074	4.044
Theory FT Period	4.206	4.457	4.201

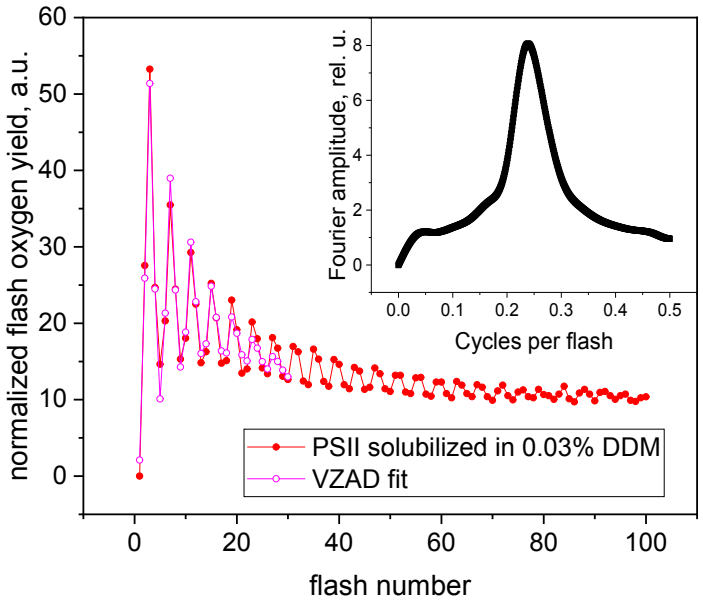

Figure S5. Flash oxygen yield for PSII core complexes acquired from dissolution of PSII microcrystals in 0.03% DDM. The data was fitted with the VZAD model and a Fourier Transform analysis was performed (shown in insert).

The flash oxygen yield from PSII core complexes shows a rapid initial decay, as in microcrystals, but even faster, which is consistent with increased access to the core complexes by quinols which cannot penetrate the crystal matrix on the timescale between flashes.