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## Time-Resolved FTIR Study of Light-Driven Sodium Pump Rhodopsins

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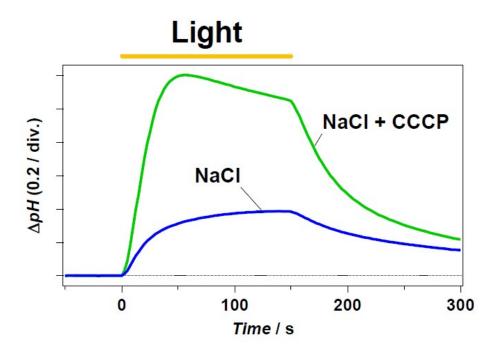
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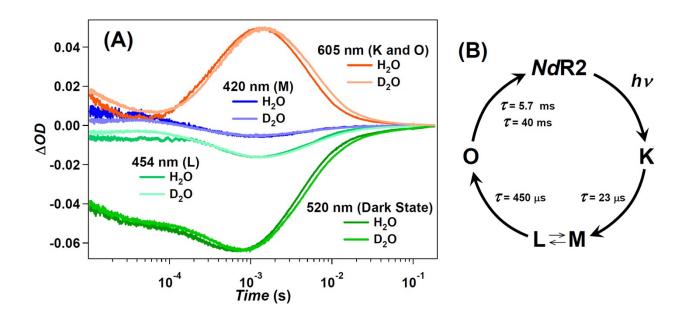
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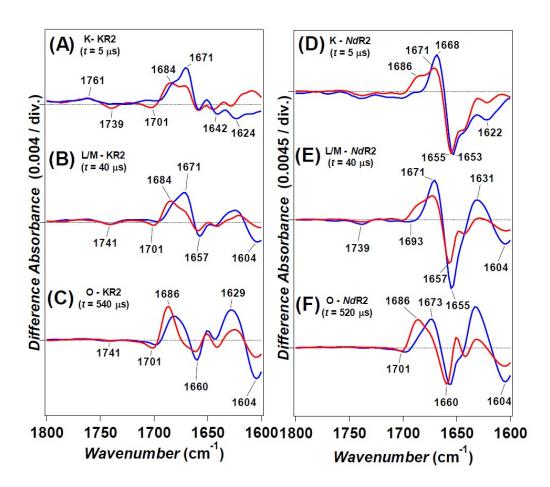
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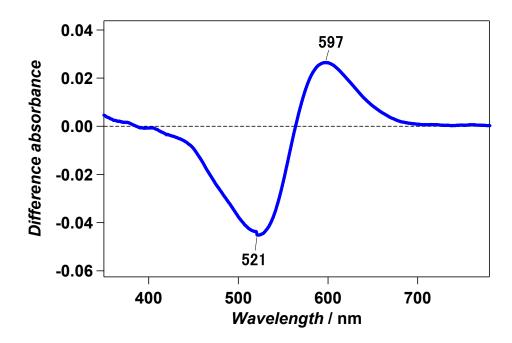
**Figure S1.** Sodium-pump activity of NdR2.



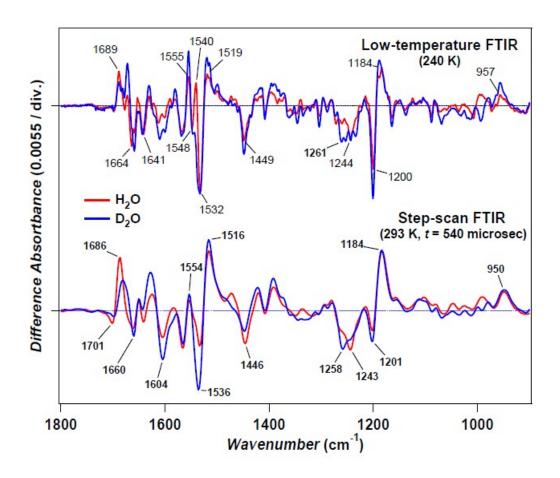
**Figure S2.** Flash photolysis data of NdR2. (A) Time traces of absorption changes of NdR2 at 420 (blue), 454 (light green), 520 (green) and 605 nm (orange) probe wavelengths. The samples were hydrated with  $H_2O$ . (B) Photocycle of NdR2.



**Figure S3.** Time-resolved difference spectra for KR2 (A-C) and NdR2 (D-F) at 1800-1600 cm<sup>-1</sup>. The spectra are obtained at 5 μs (A, D), 40 μs (B, E), 540 μs (C), and 520 μs (F), which mainly monitor the K intermediate of KR2 and NdR2, the L/M intermediate of KR2 and NdR2, the O intermediate of KR2, and the O intermediate of NdR2, respectively. The sample was hydrated with  $H_2O$  (red lines) or  $H_2O$  (blue lines).



**Figure S4.** Low-temperature light-induced UV-visible absorption spectrum of KR2 upon illumination at  $\lambda > 500$  nm at 240 K.



**Figure S5.** Comparison of the O-minus-KR2 spectra between low (upper trace) and room (lower trace) temperatures. The upper spectra are obtained by stationary measurements at 240 K, where the sample is illuminated at 500 nm, while the lower spectra are obtained at 540  $\mu$ s after laser excitation at 532 nm in the step-scan time-resolved measurements. The sample was hydrated with  $H_2O$  (red lines) or  $H_2O$  (blue lines).