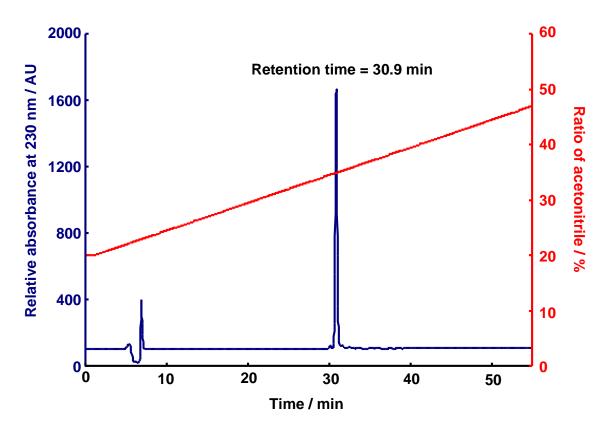
## **Supporting Information**

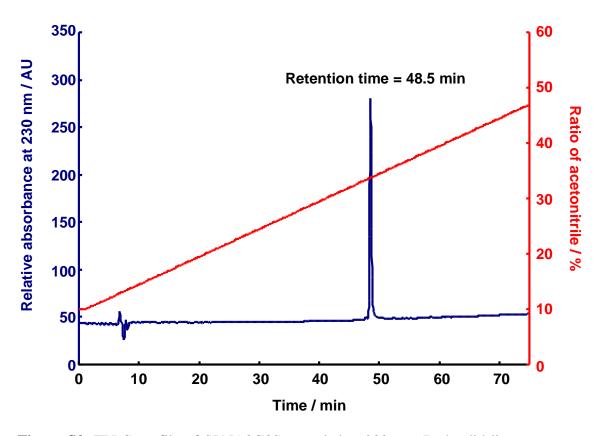
## Time-Dependent Enzyme Activity Dominated by Dissociation of J-Aggregates Bound to Protein Surface

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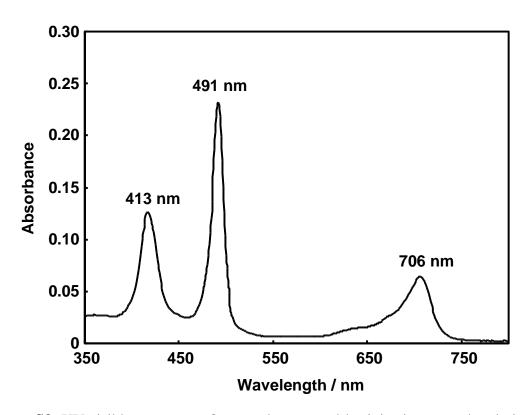
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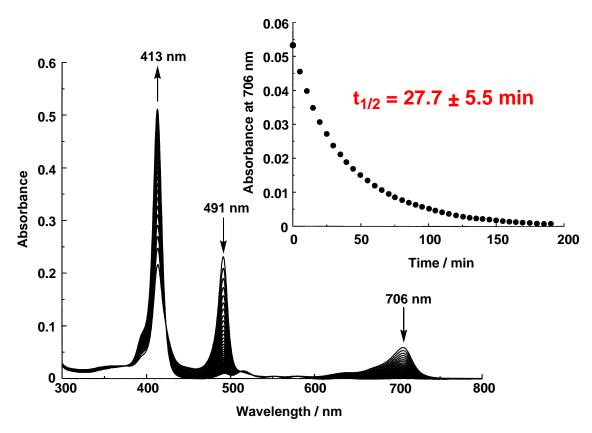
**Figure S1.** FPLC profile of SPNA3G3E recorded at 230 nm. Red solid line represents the ratio of acetonitrile in the eluent (acetonitrile/water containing 0.1 v/v% TFA). A COSMOSIL 5C18-AR-II (4.6 mm x 250 mm) column (Nacalai Tesque, Kyoto Japan) was used for the analysis. Flow rate of the eluent was 0.5 mL/min.



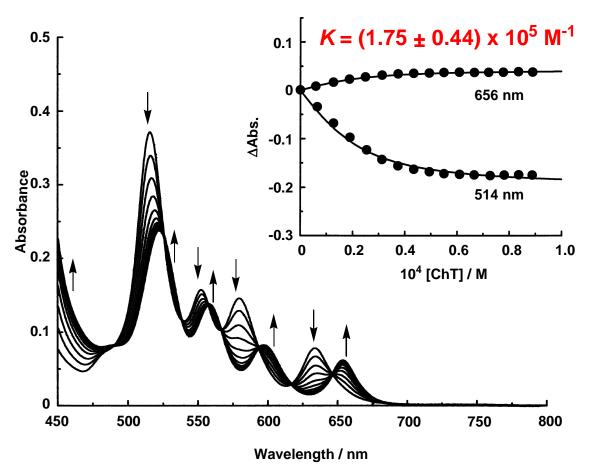
**Figure S2.** FPLC profile of SPNA3G3S recorded at 230 nm. Red solid line represents the ratio of acetonitrile in the eluent (acetonitrile/water containing 0.1 v/v% TFA). A COSMOSIL 5C18-AR-II (4.6 mm x 250 mm) column (Nacalai Tesque, Kyoto Japan) was used for the analysis. Flow rate of the eluent was 0.5 mL/min.



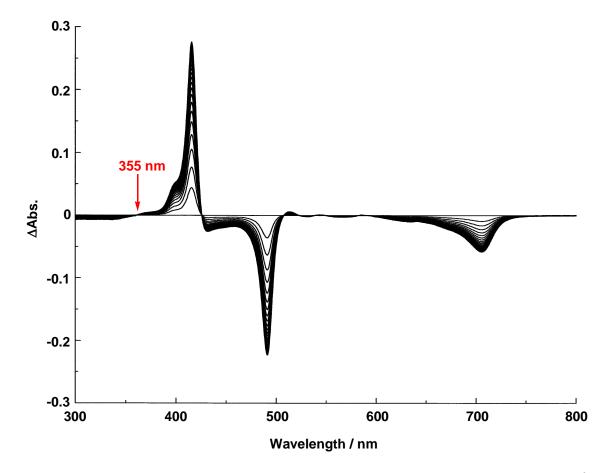
**Figure S3.** UV-visible spectrum of a sample prepared by injecting a stock solution of J-aggregates of  $H_4TPPS^{2-}$  (2.0 µL, 2.0 x 10<sup>-3</sup> M) into 4 mL of phosphate buffer (pH 7.0, 1.0 x 10<sup>-2</sup> M) containing ChT (1.0 x 10<sup>-4</sup> M) at 25 °C. The total concentration of the porphyrin in the system was 1.0 x 10<sup>-6</sup> M.



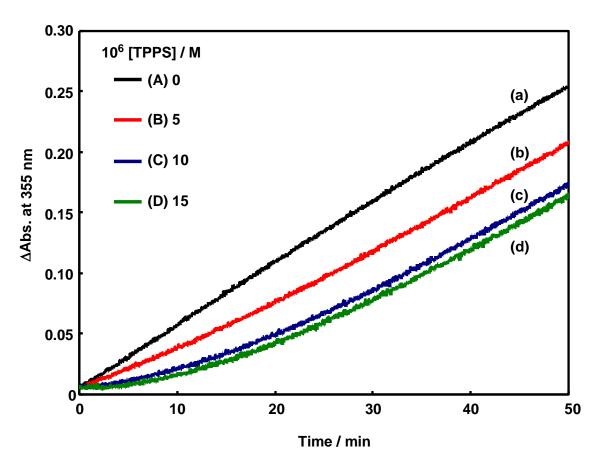
**Figure S4.** Progressive absorption spectral change for decomposition of J-aggregates of  $H_4TPPS^{2-}$  bound to ChT (1.0 x 10<sup>-6</sup> M) into the monomers of  $H_2TPPS^{4-}$  in phosphate buffer (pH 7.0, 1.0 x 10<sup>-2</sup> M) at 25 °C. Inset: Time-course of the absorbance of the J-aggregates at 706 nm. The total concentration of the porphyrin in the system was 1.0 x  $10^{-6}$  M.



**Figure S5.** UV-visible spectral change of  $H_2TPPS^{4-}$  (2.0 x 10<sup>-5</sup> M) upon addition of ChT in phosphate buffer (pH 7.0, 1.0 x 10<sup>-2</sup> M) at 25 °C. Inset: Plots of absorbance changes as a function of [ChT] at 514 nm and 656 nm. The solid lines represent the best fits of the data to an equation for 1:1 complexation for determining *K* value.



**Figure S6.** Differential spectral change for decomposition of J-aggregates of  $H_4TPPS^{2-}$  bound to ChT (1.0 x 10<sup>-6</sup> M) into the  $H_2TPPS^{4-}/TMe-\beta$ -CD inclusion complex in phosphate buffer (pH 7.0 and 1.0 x 10<sup>-2</sup> M) containing TMe- $\beta$ -CD (8.0 x 10<sup>-6</sup> M) at 25 °C. The total concentration of the porphyrin in the system was 1.0 x 10<sup>-6</sup> M.



**Figure S7.** Effects of the amount of J-aggregates on hydrolysis of SPNA3G3E (3.0 x  $10^{-4}$  M) catalyzed by ChT (1.0 x  $10^{-6}$  M) in phosphate buffer (pH 7.0, 1.0 x  $10^{-2}$  M) containing TMe- $\beta$ -CD (1.2 x  $10^{-4}$  M) at 25 °C. [TPPS] = 0 M (A); 5.0 x  $10^{-6}$  M (B); 1.0 x  $10^{-5}$  M (C); 1.5 x  $10^{-5}$  M (D.