## Supporting Information

# Visible-Light-Promoted Organic-Dye-Catalyzed Sulfonylation/Cyclization to Access Indolo[2,1- $\alpha$ ]isoquinoline Derivatives 

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## 1. General Information

All reagents and solvents were purchased from commercial suppliers and used without purifications. TLC was performed on silica gel plates (200-300mesh) using UV light ( $254 / 365 \mathrm{~nm}$ ) for detection and column chromatography was performed on silicagel (200-300 mesh).The ${ }^{1} \mathrm{H}$ NMR and ${ }^{13} \mathrm{C}$ NMR spectra were recorded at $25{ }^{\circ} \mathrm{C}$ in $\mathrm{CDCl}_{3}$ at 400 and 100 MHz , respectively, with TMS as the internal standard. Chemical shifts $(\delta)$ are expressed in ppm and coupling constants $J$ are given in Hz. All reactions were performed on the photoreaction instrument (WP-TEC-1020SL), which are purchased from WATTCAS, China (Figure S1).


Figure S1.Photoreactor for photoreaction

## 2. Experimental Section

General procedures for the synthesis of ester substituted indolo[2,1- $\alpha$ ]isoquinolines and benzimidazo[2,1- $\alpha$ ]isoquinolin- $6(5 H)$-ones.


To a suspension of 2-aryl- $N$-acryloyl indoles ( 0.2 mmol ) in DMSO ( 2 mL ) was added sulfonyl hydrazine( $0.6 \mathrm{mmol}, 3.0 \mathrm{eq}$.), Eosin B ( $5 \mathrm{~mol} \%$ ) and $\left(\mathrm{NH}_{4}\right)_{2} \mathrm{~S}_{2} \mathrm{O}_{8}(0.6$ mmol, 3.0 eq.) at room temperature, and the mixture was stirred in air under a 10 W blue LEDs and irradiated for 15 hours. The temperature was maintained at $20 \sim 25^{\circ} \mathrm{C}$ when the LED light was on. After the reaction was complete, the reaction mixture was diluted with a brine solution ( 25 mL ) and extracted with EtOAc $(3 \times 50 \mathrm{~mL})$. The combined organic phase was dried over anhydrous $\mathrm{Na}_{2} \mathrm{SO}_{4}$, filtered and concentrated in vacuo. The residue was purified by flashcolumn chromatography to afford the desired products.

## 3.Fluorescence quenching experiments

The fluorescence emission intensities were carried out on an F-7000 FL spectrophotometer (Hitachi Ltd, Japan) with excitation slit set at 5 nm and emission at 5 nm . The excitation wavelength was fixed at 323 nm , and the emission wavelength was measured at $350 \sim 550 \mathrm{~nm}$. The samples were prepared by mixing Eosin B $\left(1.0 \times 10^{-8} \mathrm{~mol} / \mathrm{L}\right)$ and different amount of $\left(\mathrm{NH}_{4}\right)_{2} \mathrm{~S}_{2} \mathrm{O}_{8}$ in DMSO in a light path quartz fluorescence cuvette. The concentration of $\left(\mathrm{NH}_{4}\right)_{2} \mathrm{~S}_{2} \mathrm{O}_{8}$ stock solution is $1.3 \times 10^{-6} \mathrm{~mol} / \mathrm{L}$ in DMSO. For each quenching experiment, 0.1 mL of $\left(\mathrm{NH}_{4}\right)_{2} \mathrm{~S}_{2} \mathrm{O}_{8}$ stock solution was titrated to a mixed solution of Eosin B $(0.1 \mathrm{~mL}$, in a total volume $=$ 1.0 mL ). Then the emission intensity was collected and the results were presented in Figure S1.


Figure S1 Quenching of Eosin B fluorescence emission in the presence of $\left(\mathrm{NH}_{4}\right)_{2} \mathrm{~S}_{2} \mathrm{O}_{8}$.

## 4. ${ }^{1} \mathrm{H}$ and ${ }^{13} \mathrm{C}$ NMR spectra of products



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