1	Microfluidic chip-based one-step fabrication of artificial photosystem I
2	for photocatalytic cofactor regeneration
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## 16 Supporting information

The thickness of the few-layer  $g-C_3N_4$  is a necessary factor in identifying the layer number of the material. Here, we characterized the thickness of the few-layer  $g-C_3N_4$  by AFM and the diluted dispersion of the products obtained after ultrasonication treatment for 1 h. The typical result of AFM image was shown in Figure S1A and a typical height image of a few-layer g- $C_3N_4$  nanosheet was 1.6 nm (Fig. S1B). Since the reported height is 5 Å for the single layer g- $C_3N_4$  film<sup>[1]</sup>, the nanosheet is estimated to have ~3 layers on average.

In the traditional method of synthesis M, more than four reagents were used, increasing the opportunities to get impurities, for example, the reagents themselves and unwanted elements inside. The NMR peaks of M in the traditional method (as shown in Fig. S2A), is consistent with the reported structure<sup>[2]</sup> of M: Cp\*[Rh(2,2'-bpy)]  $\delta$ (ppm) =9.02 (d, 2H, H-3,

1 3'), 8.84 (d, 2H, H-6, 6'), 8.24 (t, 2H, H-5, 5'), 7.81 (t, 2H, H-4, 4'), 1.74 (s, 15H, Cp\*). However, obvious residuals of diethyl ether and other impurities were found, as shown in Fig. 2 S2A, by the peak at 3.45 ppm and other miscellaneous peaks. The NMR spectrum of M in the 3 one-step method (without g-C<sub>3</sub>N<sub>4</sub>) is much more clear (Fig. S2B). In the IAPSI microreactor, 4 the formation of M was with few-layer g-C<sub>3</sub>N<sub>4</sub> in the ethanol, therefore the structure of M in 5 this condition was measured. From Fig. S2C, there is no difference between this NMR 6 spectrum and the pure M in Fig. S2B. Thus, we confirmed that the electron mediator formed 7 from the one-step method possesses the correct structure. We also found that the one-step 8 synthesis of M could be also applied in other reagents, such as methanol and chloroform. 9

In the regeneration part, we choose the static flow (i.e., the flow rate is 0) during 10 coenzyme regeneration. Although the flow shear stress is not an influential factor, for the 11 completeness of this research, we did the shear stress experiment. Here, we chose the bulk g-12  $C_3N_4$  as a typical sample since it is much easier to count the density under the optical 13 microscope. Thus, we formed the bulk g-C<sub>3</sub>N<sub>4</sub>-IAPSI microreactor and the initial state is 14 shown in Fig. S3A. We then increase the flow rate to flush the channel. Here, the 15 corresponding shear stress  $\tau$  could be calculated from the flow rate Q and the microchannel 16 geometry based on the equation:  $\tau = 6\eta Q/h^2 w$ , where h is the channel height, w the 17 channel width and  $\eta$  the viscosity of fluid. In this part, h is 40  $\mu$ m, w is 2000  $\mu$ m and  $\eta$  is 18 0.001 Pa • s (20 °C). Thus, the shear stress for 700  $\mu$ L/min is 876 Pa, which is large enough 19 for most experiments. Here, the flow rate was fast, and we found an obvious drop of particle 20 density from 0 to 20 s (Fig. S3B), caused by the loss of nanoparticles that were bound loosely, 21 occupying only 5 percent. Then, there was little change from 1 min to 3 min in Fig. S3C and 22 D. The corresponding calculation data were shown in Fig. S3E. 23

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## 26 **References**

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- 1 Figure S1.
- 2



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4 Fig. S1. (A) AFM images of the representative few-layer  $g-C_3N_4$ , (B) a typical 5 nanosheet (marked with red line in Fig. S1A) with a height of 1.6 nm.

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5 Fig. S2. Comparison of the nuclear magnetic resonance (NMR) spectra of the electron 6 mediator M. (A) The NMR spectrum of M in the traditional method, containing several 7 unwanted impurities. (B) The NMR spectrum of M in our one-step method without  $g-C_3N_4$ , 8 which is much purer with less impurities. (C) The NMR spectrum of M in our one-step 9 method with the few-layer  $g-C_3N_4$ , which is the same to the spectrum in (B), proving that the 10 existence of few-layer  $g-C_3N_4$  has a negligible effect on the M formation, thus the feasibility 11 of our one-step method.

- 1
- 2 Figure S3.



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5 Fig. S3. Shear stress tests of the bulk  $g-C_3N_4$  by applying a flow rate of 700  $\mu$ L/min. Particle 6 densities on the glass slide decrease in the first 20 s, but show little drop afterwards.