

Figure S1 (a) TEM image, line scan elemental analysis spectrum of (b) Ti, (c) Fe, (d) O elements in MIL-100(Fe)/TiO<sub>2</sub>.



Figure S2 TEM images of MIL-100(Fe)/TiO<sub>2</sub> synthesized at (a) 140  $^{\circ}$ C and (b and c)280  $^{\circ}$ C.



Figure S3 SEM images of MIL-100(Fe)/TiO<sub>2</sub>/CoO<sub>x</sub> after (a) 4 and (b) 18 hours of photocatalytic reaction.



**Figure S4.** N<sub>2</sub> adsorption-desorption isotherms of (a) MIL-100(Fe), (b) MIL-100(Fe)/TiO<sub>2</sub> (c) MIL-100(Fe)/TiO<sub>2</sub>/CoO<sub>x</sub> samples.



Figure S5 Reusability results of the  $O_2$  production for the MIL-100(Fe)/TiO<sub>2</sub>/CoO<sub>x</sub>20 sample.

| Photocatalysts   | Condition   | OER (µmol<br>h <sup>-1</sup> g <sup>-1</sup> ) | Refs         |
|--|---|--|--------------|
| MIL100(Fe)/TiO <sub>2</sub> /CoO<br>x                              | 10 mg, 0.1 M FeCl <sub>3</sub> , 300 W<br>Xe lamp as the light source   | 558.3  | This<br>work |
| Ov-WO <sub>3</sub> -Pt   | 50 mg, 0.1 M Na <sub>2</sub> S <sub>2</sub> O <sub>8</sub> , pH 10.5 adjusted by NaOH                                   | 683  | [1]          |
| CN/a-ZnO   | under simulated sunlight irradiation  | 532.4  | [2]          |
| Ag <sub>3</sub> PO <sub>4</sub> /TiO <sub>2</sub>                  | 50 mg catalyst in 50 mL solution under illumination by a 300W Xe-lamp   | 537  | [3]          |
| FeOOH/BiOBr  | 300 W simulated sunlight<br>using AgNO <sub>3</sub> as the sacrificial<br>agent   | 284.34   | [4]          |
| TiO <sub>2</sub> /C HNTs   | under simulated solar light irradiation   | 539  | [5]          |
| 5% $CoO_x/\alpha$ -Fe <sub>2</sub> O <sub>3</sub>                  | visible light ( $\lambda > 400$ nm)<br>using AgNO <sub>3</sub> as the electron<br>scavenger                             | 195.19   | [6]          |
| Cu/ZIS/TiO <sub>2</sub>  | 300 W Xe lamp as the light source   | 292.75   | [7]          |
| r-TiO <sub>2</sub>   | 5 mg of catalyst in 30 mL of<br>0.02 M AgNO <sub>3</sub> solution under<br>a 300 W Xe lamp                              | 71.9   | [8]          |
| Ni SA-<br>NG/SrTiO <sub>3</sub> (Al)/CoO <sub>x</sub>              | Full arc irradiation  | 230  | [9]          |
| RuO <sub>x</sub> /CoO <sub>x</sub> /Ta <sub>3</sub> N <sub>5</sub> | 50 mg, 0.1 M Na <sub>2</sub> S <sub>2</sub> O <sub>8</sub> , under visible light irradiation (420 $< \lambda < 800$ nm) | 159  | [10]         |

**Table S1** Comparison of photocatalytic oxygen evolution performance of MIL- $100(Fe)/TiO_2/CoO_x$  with other reported photocatalysts.

## Reference

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