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# Photocatalytic carbon dioxide reduction coupled with benzylamine oxidation over Zn-Bi<sub>2</sub>WO<sub>6</sub> microflowers<sup>†</sup>

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Using solar energy to realize  $CO_2$  reduction and benzylamine oxidation is of great importance. Here, we design  $Zn-Bi_2WO_6$  microflowers catalyst, which can simultaneously reduce  $CO_2$  to CO and oxidize benzylamine to *N*-benzylbenzaldimine with high activities and selectivities in a simple photocatalytic system.

## Introduction

These days, photocatalytic carbon dioxide reduction reaction (CO<sub>2</sub>RR) has been attracting tremendous attention because it can mitigate the greenhouse effect, achieve the carbon cycle and remit the energy crisis.<sup>1–5</sup> To effectively convert  $CO_2$  into carbon compounds (e.g., carbon monoxide, methane and formic acid), a sacrificial agent like triethanolamine, sodium sulfite or triethylamine is usually a requisite to capture photoexcited holes, which is economically unfeasible and wasteful of holes.4,6-9 The photocatalyst with special micro- and nano-structure can effectively promote the separation of the photocharge and hole, so it can also have a certain photocatalytic efficiency without the sacrificial agent.<sup>2</sup> To date, great efforts have been devoted to the development of photocatalytic CO2RR systems without a sacrificial agent by constructing a catalyst with a special structure, but are restricted mainly in terms of two main issues. First, oxygen evolution reaction is inevitable in many aqueous solution catalytic systems, causing extra steps to separate the reduction products of  $CO_2$  and oxygen.<sup>10,11</sup> Second, the efficiency of  $CO_2RR$  is usually low.<sup>12–16</sup> As a result, it is still difficult to achieve the expected photocatalytic efficiency simply by constructing the catalyst with a special structure. In light of the role of sacrificial reagents (hole consumption) in photocatalytic  $CO_2RR$ , a hole-consuming reaction can provide a sacrificial reagent-free way, which can effectively capture the holes generated in the photocatalytic process and make full use of the photogenerated charges.<sup>17</sup> Therefore, coupling photocatalytic  $CO_2RR$  with a hole-consuming reaction could overcome the limitations of photocatalytic  $CO_2RR$  and maximize photocatalytic efficiency and energy utilization.

Here, we demonstrate for the first time the combination of photocatalytic CO2RR with benzylamine oxidation reaction, which is a hole-consuming reaction and an important approach to synthesize N-benzylbenzaldimine.<sup>18-20</sup> For this strategy, CO<sub>2</sub> consumes electrons to generate CO, while benzylamine consumes holes to get N-benzylbenzaldimine. Therefore, neither sacrificial agent nor additional oxidant (like oxygen) is needed. Zn-Bi<sub>2</sub>WO<sub>6</sub> microflowers assembled by ultra-thin The nanosheets were synthesized as a photocatalyst, which can improve the rapid separation and transfer of photoexcited electrons and holes, so as to ensure that they can participate in the simultaneous CO2RR and benzylamine oxidation reaction. The maximum production rates of CO and N-benzylbenzaldimine can reach 127.5  $\mu$ mol g<sup>-1</sup> h<sup>-1</sup> and 13.1 mmol g<sup>-1</sup> h<sup>-1</sup>, respectively, which are both the highest among the reported values of the separate CO2RR and benzylamine oxidation reaction under comparable conditions. A scheme for the simultaneous photocatalytic CO2RR and benzylamine oxidation reaction by Zn-Bi<sub>2</sub>WO<sub>6</sub> microflowers is illustrated in Scheme 1.

## **Results and discussion**

# Synthesis and structural characterizations of Zn-Bi<sub>2</sub>WO<sub>6</sub> microflowers

The  $Zn-Bi_2WO_6$  photocatalyst was synthesized by a solvothermal strategy. As an example, Fig. 1 shows the characteriz-

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<sup>†</sup>Electronic supplementary information (ESI) available: SEM and TEM images, XRD patterns, EXAFS spectra, XPS spectra and band structures. See DOI: 10.1039/d1gc00676b





**Fig. 1** (a–c) SEM images of Zn-Bi<sub>2</sub>WO<sub>6</sub>. (d) TEM image of Zn-Bi<sub>2</sub>WO<sub>6</sub>. (e) HRTEM image and SAED pattern (inset) of Zn-Bi<sub>2</sub>WO<sub>6</sub>. (f) AFM image of Zn-Bi<sub>2</sub>WO<sub>6</sub>. (g) EDX mappings of Zn-Bi<sub>2</sub>WO<sub>6</sub>. Scale bars: 3  $\mu$ m (a), 500 nm (b), 200 nm (c), 50 nm (d), 3 nm (e), 50 nm (f) and 500 nm (g).

ations of the Zn-Bi<sub>2</sub>WO<sub>6</sub> with Zn content of 1.0 wt%, determined by inductively coupled plasma-mass spectrometry. From scanning electron microscopy (SEM) and transmission electron microscopy (TEM) images, the product has a flowerlike microstructure, which is assembled by nanosheets in a lateral size of ~50 nm (Fig. 1a-d). The high-resolution TEM (HRTEM) image shows the lattice fringes with a distance of 0.32 nm corresponding to the (131) crystal plane of russellite Bi<sub>2</sub>WO<sub>6</sub> (Fig. 1e).<sup>21</sup> Powder X-ray diffraction (XRD) analysis further confirms the formation of russellite Bi<sub>2</sub>WO<sub>6</sub> (JCPDS card no. 39-0256), which is consistent with the selected area electron diffraction (SAED) pattern (inset in Fig. 1e). It is obvious that Zn doping does not change the crystal conformation of Bi<sub>2</sub>WO<sub>6</sub> (Fig. S1<sup>†</sup>). The atomic force microscopy (AFM) image reveals that the average thickness of nanosheets is 4.9 nm (Fig. 1f). Elemental mappings reveal the even distribution of Bi, W, O, and Zn (Fig. 1g). For comparison, Bi<sub>2</sub>WO<sub>6</sub> was synthesized in the absence of zinc nitrate hexahydrate, while the other experimental conditions were the same as those for Zn-Bi<sub>2</sub>WO<sub>6</sub>. Its XRD pattern and morphology (Fig. S1 and S2<sup> $\dagger$ </sup>) are similar to those of Zn-Bi<sub>2</sub>WO<sub>6</sub>.

#### Fine structure of Zn-Bi<sub>2</sub>WO<sub>6</sub> microflowers

The fine structure of Zn-Bi<sub>2</sub>WO<sub>6</sub> was investigated by synchrotron extended X-ray absorption fine structure (EXAFS) spectroscopy. For X-ray absorption near-edge structure (XANES), the pre-edge position of Zn-Bi<sub>2</sub>WO<sub>6</sub> is located closer to that of ZnO (Fig. 2a), suggesting that Zn exists in the form of (+2) oxidation state in Zn-Bi<sub>2</sub>WO<sub>6</sub>. Fourier transform (FT)  $k^3$ -weighted  $\chi(k)$  function of the EXAFS spectrum shows a dominant peak at 1.49 Å, belonging to the coordination bond between Zn and O in Bi<sub>2</sub>WO<sub>6</sub> (Fig. 2b). Compared with ZnO and Zn foil, there is no Zn-Zn coordination peak in Zn-Bi<sub>2</sub>WO<sub>6</sub>. These results reveal that Zn exists in Zn-Bi<sub>2</sub>WO<sub>6</sub> as isolated ions rather than as Zn or ZnO particles. In Bi L<sub>3</sub>-edge XANES, the white line intensity of Zn-Bi2WO6 is stronger compared with that of Bi<sub>2</sub>WO<sub>6</sub> (Fig. S3<sup>†</sup>). Meanwhile, in the FT  $k^3$ -weighted  $\gamma(k)$  functions of the EXAFS spectra, the intensity of Bi-O of Zn-Bi<sub>2</sub>WO<sub>6</sub> is weaker than that of Bi<sub>2</sub>WO<sub>6</sub>. In W L<sub>3</sub>-edge XANES, the white line intensity of Zn-Bi<sub>2</sub>WO<sub>6</sub> is weakened compared with that of Bi<sub>2</sub>WO<sub>6</sub>, and the intensity of W–O of Zn-Bi<sub>2</sub>WO<sub>6</sub> is weaker than that of Bi<sub>2</sub>WO<sub>6</sub> in FT  $k^3$ -weighted  $\chi(k)$  function of the EXAFS spectra. This indicates that Zn doping changes the local atomic structure of Bi2WO6.11 X-ray photoelectron spectra further prove that Zn exists as Zn<sup>2+</sup> and Zn doping changes the surface structure of Bi<sub>2</sub>WO<sub>6</sub> (Fig. S4<sup>†</sup>), which is consistent with the EXAFS results.<sup>22</sup> Since the radius of Zn<sup>2+</sup> (74 pm) is closer to that of  $W^{6+}$  (62 pm) than of  $Bi^{3+}$  (103 pm), it is expected that  $Zn^{2+}$  ions could replace partially the  $W^{6+}$  ions, forming  $Zn^{2+}$ substitutionally doped Bi<sub>2</sub>WO<sub>6</sub>.<sup>23-25</sup> The Zn<sup>2+</sup> ions can act as electron traps to promote the electron-hole separation and enhance the photocatalytic efficiency.

#### Photochemical performance of Zn-Bi<sub>2</sub>WO<sub>6</sub> microflowers

The performance of Zn-Bi<sub>2</sub>WO<sub>6</sub> for the simultaneous photocatalytic CO<sub>2</sub>RR and 4-methylbenzylamine oxidation reaction was studied. The reaction was irradiated by simulated solar light irradiation at atmospheric pressure and room temperature. No sacrificial agent or additional oxidant (like oxygen) was added. After the reaction, the gas and liquid products were analyzed by gas chromatograph and <sup>1</sup>H nuclear magnetic resonance spectroscopy, respectively. The results show the formation of CO and (*E*)-*N*-(4-methylbenzyl)-1-(*p*-tolyl)methanimine (Fig. S5†), and no other products were detected. Fig. 3a



**Fig. 2** (a) Zn K-edge XANES spectra of Zn-Bi<sub>2</sub>WO<sub>6</sub>. (b) Fourier transform (FT)  $k^3$ -weighted  $\chi(k)$  function of the EXAFS spectra of Zn-Bi<sub>2</sub>WO<sub>6</sub>. ZnO and Zn foil were used as contrast samples.



**Fig. 3** (a) Time-dependent CO and (*E*)-*N*-(4-methylbenzyl)-1-(*p*-tolyl) methanimine production amounts by Zn-Bi<sub>2</sub>WO<sub>6</sub>. (b) Photocatalytic CO<sub>2</sub>RR and 4-methylbenzylamine oxidation reaction under various conditions. (c) Stability of Zn-Bi<sub>2</sub>WO<sub>6</sub> for five cycles. (d) Photocatalytic CO<sub>2</sub>RR and diverse *para*-substituted benzylamine oxidation activities of Zn-Bi<sub>2</sub>WO<sub>6</sub>.

shows that CO and (E)-N-(4-methylbenzyl)-1-(p-tolyl)methanimine production amounts increase linearly with reaction time during a 10 h test. It indicates the good stability of the Zn-Bi<sub>2</sub>WO<sub>6</sub> photocatalyst for the duration of the photocatalytic reaction process. The production rates of CO and (E)-N-(4methylbenzyl)-1-(*p*-tolyl)methanimine are 127.5  $\mu$ mol g<sup>-1</sup> h<sup>-1</sup> and 13.1 mmol  $g^{-1}$  h<sup>-1</sup> (with >99% selectivity), respectively. CO<sub>2</sub> and water could act as amine activating agents to favor and accelerate the photocatalytic oxidative coupling of benzylamines,<sup>26</sup> thus leading to a difference in oxidation efficiency and reduction efficiency. For comparison, the simultaneous photocatalytic CO<sub>2</sub>RR and 4-methylbenzylamine oxidation reaction was also studied using Bi<sub>2</sub>WO<sub>6</sub> as a catalyst. The production rates of CO and (E)-N-(4-methylbenzyl)-1-(p-tolyl) methanimine are 42.4  $\mu$ mol g<sup>-1</sup> h<sup>-1</sup> and 5.1 mmol g<sup>-1</sup> h<sup>-1</sup>, respectively, which are much lower than those catalyzed by Zn- $Bi_2WO_6$ . It indicates that the introduction of  $Zn^{2+}$  into  $Bi_2WO_6$ can effectively improve the photocatalytic efficiency. Moreover, the production rates of CO and (E)-N-(4-methylbenzyl)-1-(ptolyl)methanimine from the coupled reaction catalyzed by Zn-Bi<sub>2</sub>WO<sub>6</sub> are higher than those of the reported single reactions under comparable conditions (ESI Table S1,†<sup>11,27-30</sup> Table S2<sup>† 20,31-34</sup>).

Some control experiments for the simultaneous photocatalytic CO<sub>2</sub>RR and 4-methylbenzylamine oxidation reaction catalyzed by Zn-Bi<sub>2</sub>WO<sub>6</sub> were tested under different photocatalytic conditions (Fig. 3b). For all these control experiments, no sacrificial agent or additional oxidant (like oxygen) was added. First, when CO<sub>2</sub> was replaced by air, nearly no CO was detected and the production rate of (*E*)-*N*-(4-methylbenzyl)-1-(*p*-tolyl)methanimine was 11.1 mmol g<sup>-1</sup> h<sup>-1</sup> (B in Fig. 3b and Fig. S6†). Second, the single photocatalytic CO<sub>2</sub>RR catalyzed by Zn-Bi<sub>2</sub>WO<sub>6</sub> was studied. The CO production rate was as low as 6.7 µmol g<sup>-1</sup> h<sup>-1</sup> (C in Fig. 3b). It can be attributed to the fact

that the photogenerated holes cannot be effectively consumed during a single photocatalytic CO<sub>2</sub>RR, so the electrons and holes may recombine in the process of migration to the Zn-Bi<sub>2</sub>WO<sub>6</sub> surface to reduce the number of electrons involved in CO<sub>2</sub>RR. However, when the photocatalytic CO<sub>2</sub>RR and 4-methylbenzylamine oxidation reaction simultaneously proceed, the photogenerated holes can be consumed by the benzylamine oxidation reaction, thus improving the reduction efficiency of CO<sub>2</sub>. Third, the single 4-methylbenzylamine oxidation reaction catalyzed by Zn-Bi<sub>2</sub>WO<sub>6</sub> was tested under a nitrogen  $(N_2)$  atmosphere, which shows a low production rate of (E)-N-(4-methylbenzyl)-1-(p-tolyl)methanimine of 0.7 mmol  $g^{-1}$  h<sup>-1</sup> (D in Fig. 3b). Fourth, the photocatalytic CO<sub>2</sub>RR and 4-methylbenzylamine oxidation reaction catalyzed by Zn-Bi<sub>2</sub>WO<sub>6</sub> were performed without light irradiation. Neither CO nor (E)-N-(4-methylbenzyl)-1-(p-tolyl)methanimine was detected (E in Fig. 3b). Obviously, the efficiency of the simultaneous photocatalytic CO2RR and 4-methylbenzylamine oxidation reaction is much higher than of the two single reactions. In a photocatalytic reaction, CO<sub>2</sub>RR consumes electrons, while benzylamine oxidation reaction consumes holes. Consequently, the recombination of photogenerated electrons and holes is effectively reduced, making them more likely to migrate to the surface of the Zn-Bi<sub>2</sub>WO<sub>6</sub> to participate in reduction and oxidation reactions. From the above results, it can be concluded that there is a synergistic effect for CO<sub>2</sub>RR and 4-methylbenzylamine oxidation reaction during the coupling process, which can promote each other efficiently.

To evaluate the stability of Zn-Bi<sub>2</sub>WO<sub>6</sub>, the performances of the photocatalytic coupling reaction for five cycles were measured. The production rates of CO and (*E*)-N-(4-methylbenzyl)-1-(*p*-tolyl)methanimine are basically stable and no drop was detected during the five cycles (Fig. 3c). The morphology of the used Zn-Bi<sub>2</sub>WO<sub>6</sub> is the same as that of the pristine Zn-Bi<sub>2</sub>WO<sub>6</sub> (Fig. S7†). Besides, the crystallinity of Zn-Bi<sub>2</sub>WO<sub>6</sub> is well maintained after five photocatalytic runs (Fig. S8†).

The photocatalytic CO<sub>2</sub>RR integrated with diverse para-substituted benzylamine oxidation reaction (R = -OCH<sub>3</sub>, -H, -Cl,  $-OCF_3$ ) by Zn-Bi<sub>2</sub>WO<sub>6</sub> was further investigated. The results show that CO<sub>2</sub> and benzylamines can be transformed simultaneously to CO and the corresponding N-benzylbenzaldimines (with >99% selectivity) effectively. For the four different para-substituents, the production rates of CO and N-benzylbenzaldimine determined at 4 h are above 90  $\mu$ mol g<sup>-1</sup> h<sup>-1</sup> and 9 mmol g<sup>-1</sup> h<sup>-1</sup>, respectively (Fig. 3d). The difference in performances of the coupling reactions can be attributed to the electron supplying ability of the substituent. The electron donating groups ( $R = -CH_3$ ,  $-OCH_3$ ) could increase the electron density than the electron withdrawing groups (R = -Cl,  $-OCF_3$ ), thus promoting the efficiency of the coupling reaction.<sup>19</sup>

Moreover, a series of  $\text{Zn-Bi}_2\text{WO}_6$  catalysts with different Zn contents were synthesized. All of them present the russellite  $\text{Bi}_2\text{WO}_6$  crystal structure and flower-like morphologies (Fig. S9–S12†), similar to the above Zn-Bi}\_2\text{WO}\_6 with Zn content

of 1.0 wt%. Their performances for the simultaneous photocatalytic CO<sub>2</sub>RR and 4-methylbenzylamine oxidation reaction were studied. Using Zn-Bi<sub>2</sub>WO<sub>6</sub> catalysts with Zn contents of 0.4, 1.0, 1.8 and 2.3 wt%, the CO production rates determined at 4 h are 80.6, 127.5, 110.2 and 94.2  $\mu$ mol g<sup>-1</sup> h<sup>-1</sup>, while the *N*-benzylbenzaldimine production rates are 10.6, 13.1, 11.7 and 10.1 mmol g<sup>-1</sup> h<sup>-1</sup>, respectively. It is obvious that with the increase of Zn doping in Bi<sub>2</sub>WO<sub>6</sub>, the catalytic performance increases first and then decreases. The optimum Zn content in Bi<sub>2</sub>WO<sub>6</sub> is 1.0 wt% to get the maximum efficiency for the coupling reaction.

#### Photoelectric properties of Zn-Bi<sub>2</sub>WO<sub>6</sub> microflowers

The above results reveal the high efficiency of coupled photocatalytic CO<sub>2</sub>RR and benzylamine oxidation reaction catalyzed by Zn-Bi<sub>2</sub>WO<sub>6</sub>. To get the information on the underlying mechanism, the photoelectric properties of the photocatalyst were investigated. For UV-vis absorption spectra, Zn-Bi<sub>2</sub>WO<sub>6</sub> exhibits a slight red shift of the absorption edge as compared with Bi<sub>2</sub>WO<sub>6</sub>, indicating the enhanced light absorption and reduced band gap (Fig. 4a). The calculated optical band gaps of Zn-Bi<sub>2</sub>WO<sub>6</sub> and Bi<sub>2</sub>WO<sub>6</sub> are 3.19 and 3.24 eV, respectively.<sup>35</sup> The conduction band (CB) positions of Zn-Bi<sub>2</sub>WO<sub>6</sub> and Bi<sub>2</sub>WO<sub>6</sub> were determined by Mott–Schottky plots to be -0.85 V and -0.78 V (V vs. Ag/AgCl, pH = 6.8), respectively (Fig. 4b). The proper band structure manifests that Zn-Bi<sub>2</sub>WO<sub>6</sub> possesses



Fig. 4 (a) UV-vis absorption spectra and (inset) corresponding band gap energies of Zn-Bi<sub>2</sub>WO<sub>6</sub> and Bi<sub>2</sub>WO<sub>6</sub>. (b) Mott–Schottky plots, (c) transient state, (d) steady state photoluminescence spectra, (e) transient photocurrent spectra and (f) electrochemical impedance spectra of Zn-Bi<sub>2</sub>WO<sub>6</sub> and Bi<sub>2</sub>WO<sub>6</sub>.

sufficiently enough reductive and oxidative potentials to  $CO_2RR$  and benzylamine oxidation (Fig. S13†). The transient state photoluminescence (PL) spectra of Zn-Bi<sub>2</sub>WO<sub>6</sub> were measured to evaluate the separation efficiency of photoexcited electrons and holes (Fig. 4c). The average lifetime of Zn-Bi<sub>2</sub>WO<sub>6</sub> is 3.93 ns, which is longer than that of Bi<sub>2</sub>WO<sub>6</sub> (2.50 ns). The prolonged charge lifetime in Zn-Bi<sub>2</sub>WO<sub>6</sub> indicates that the photocarrier can migrate to the surface of the catalyst to participate in the subsequent  $CO_2RR$  and benzylamine oxidation reaction, thus improving photocatalytic performance. In addition, the PL intensity of Zn-Bi<sub>2</sub>WO<sub>6</sub> in steady state PL spectra is lower than that of Bi<sub>2</sub>WO<sub>6</sub> (Fig. 4d), indicating the effectively restrained recombination of photoexcited electronhole pairs by Zn-Bi<sub>2</sub>WO<sub>6</sub>.<sup>36,37</sup>

The transient photocurrent spectra and electrochemical impedance spectra were examined to further reveal the charge separation and transport efficiency of  $\text{Zn-Bi}_2\text{WO}_6$ .  $\text{Zn-Bi}_2\text{WO}_6$  exhibits rapid and uniform photocurrent responses under cycling light on and off, which is significantly higher than that of  $\text{Bi}_2\text{WO}_6$  (Fig. 4e). It indicates that the separation of photo-excited electrons and holes is greatly enhanced in  $\text{Zn-Bi}_2\text{WO}_6$ .<sup>38,39</sup> Moreover,  $\text{Zn-Bi}_2\text{WO}_6$  shows a much smaller arc radius in Nyquist plots than  $\text{Bi}_2\text{WO}_6$  (Fig. 4f), suggesting its lower charge transfer resistance.<sup>4,40</sup> The above analysis shows that the introduction of  $\text{Zn}^{2+}$  into  $\text{Bi}_2\text{WO}_6$  can promote the separation and transfer of photogenerated electrons and holes, and effectively improve the photocatalytic redox efficiency.

Furthermore, the CO<sub>2</sub> adsorption properties of Zn-Bi<sub>2</sub>WO<sub>6</sub> and Bi<sub>2</sub>WO<sub>6</sub> catalysts were measured (Fig. S14<sup>†</sup>). Obviously, the Zn-Bi<sub>2</sub>WO<sub>6</sub> catalyst presents improved CO<sub>2</sub> adsorption capacity than Bi<sub>2</sub>WO<sub>6</sub>. For example, the CO<sub>2</sub> adsorption capacity of Zn-Bi<sub>2</sub>WO<sub>6</sub> reaches 4.6 mg g<sup>-1</sup> at 298 K and 1 atm. Also, the CO<sub>2</sub> adsorption properties of Zn-Bi<sub>2</sub>WO<sub>6</sub> and Bi<sub>2</sub>WO<sub>6</sub> catalysts were further explored *via* CO<sub>2</sub> temperature-programmed desorption (CO<sub>2</sub>-TPD) profiles. Peaks at the temperature of 90 °C were observed in the CO<sub>2</sub>-TPD profile, indicating the weak CO<sub>2</sub> adsorption of the catalysts. The peaks beyond 250 °C correspond to strong CO<sub>2</sub> adsorption. It is obvious that the peak of CO<sub>2</sub> desorption for Zn-Bi<sub>2</sub>WO<sub>6</sub> moves to a higher temperature than that of Bi<sub>2</sub>WO<sub>6</sub>, which indicates that Zn<sup>2+</sup> doping enhances the chemical adsorption of CO<sub>2</sub> for the catalyst.<sup>41</sup>

### Conclusions

In conclusion, we prepared  $\text{Zn-Bi}_2\text{WO}_6$  microflowers as a photocatalyst for coupled  $\text{CO}_2\text{RR}$  and benzylamine oxidation reaction in the absence of a sacrificial agent or oxidant. The  $\text{Zn}^{2+}$  ions embedded in  $\text{Bi}_2\text{WO}_6$  can be used as electron traps to improve the separation efficiency of the photoexcited electrons and holes, thus prolonging charge lifespan. The activities and selectivities of both  $\text{CO}_2$  reduction to CO and benzylamine oxidation to *N*-benzylbenzaldimine are improved in such a simple photocatalytic system. It is anticipated that the photocatalytic  $\text{CO}_2\text{RR}$  will be coupled with other different types of oxidation reactions for improving efficiencies in future.

## Conflicts of interest

There are no conflicts to declare.

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