

Solar-driven co-production of C₂H₄ and H₂O₂ from CO₂ and H₂O

Corresponding Author: Professor Weidong Shi

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

This manuscript reports the preparation of the ternary composite, Cu/AgBr/TiO₂, as an efficient photocatalyst by an oil-bath based route. This as-prepared Cu/AgBr/TiO₂ exhibits the photocatalytic performances for C₂H₄ and H₂O₂ evolution from utilizing CO₂ and H₂O. A range of in situ/ex situ characterizations were utilized to explore the reaction mechanism. The results reveal that the Cu nanocluster (NC) can serve as catalytic centres for capturing [•]OH to efficiently inhibiting the overoxidation of [•]OH and also limiting the [•]H-[•]OH recombination to accelerate the [•]H mass transfer. This manuscript exhibits certain novelty and importance. Nevertheless, there are still many things to be resolved before reconsideration of its publication:

1. In Figure S13, only very limited or negligible amount of H₂O₂ evolution is detected in the gas phase, although the authors claim that H₂O₂ generation was obtained in liquid and gas phases. Thus, it is recommended to report the UV-vis results for H₂O₂ quantification and the calibration curves for H₂O₂ generation.
2. What are the H₂O₂ evolution results after 72 h reaction?
3. In Figure 5a and b, there are no peaks related to the generation of H₂O₂. It is recommended that authors provide more evidence on this through in situ IR technique.
4. It is recommended that the quenching test results for generation of H₂O₂ and ethylene can be provided.
5. How about the structure and composition change after the 72-hour reaction for Cu(9)/AgBr(10)/TiO₂? It is recommended that the authors provide some results on the structure/composition change.
6. Authors claim that the Cu²⁺ of the catalyst are reduced under light irradiation. How about the status of Cu element after the reaction without light irradiation?

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I have reviewed the Review 3 comment and the response from the authors.

There are still some issues to be resolved:

- i. In Figure 6e, the deconvolution of Br 3d peak in the dark is not correct. The peak area ratio of Br 3d_{3/2} peak to Br 3d_{5/2} peak ought to be 2 : 3.
- ii. In Figure S22e and f, the line 1 and 2 potential values are ~750 and ~600 mv, respectively, corresponding to the potential values on the surface of Cu-modified AgBr and TiO₂ particles. However, according to the mechanism in Figure 7b and c, the photo-generated electrons remain at Cu-modified AgBr and the photo-generated holes remain at TiO₂ particles, respectively. Thus, the claims and the AFM-KPFM test results are contradictory.
- iii. In Figure S14, the XRD results of the reacted catalyst exhibits the clear reduced peak intensity and the appearance of apparent bump at ~20 degrees, obviously indicating the corrosion and reduced crystallinity of the reacted catalyst and also the appearance of amorphous structure in the reacted catalyst. All these results indicate the stability of catalyst is not that good. Authors ought to revise the claim.
- iv. In Figure 6b, there are sub-peaks in the XPS results of Ag 3d_{3/2} and Ag 3d_{5/2} peaks. However, the authors might try to deconvolute the peaks.

Reviewer #2

(Remarks to the Author)

The authors have thoroughly addressed all major concerns from the previous review, and the manuscript has been significantly improved. I find that its quality now meets the high standards required by Nature Communication. Therefore, I recommend acceptance.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

This research can be considered for publication after this revision.

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REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

This manuscript reports the preparation of the ternary composite, Cu/AgBr/TiO₂, as an efficient photocatalyst by an oil-bath based route. This as-prepared Cu/AgBr/TiO₂ exhibits the photocatalytic performances for C₂H₄ and H₂O₂ evolution from utilizing CO₂ and H₂O. A range of in situ/ex situ characterizations were utilized to explore the reaction mechanism. The results reveal that the Cu nanocluster (NC) can serve as catalytic centres for capturing ^{*}OH to efficiently inhibiting the overoxidation of ^{*}OH and also limiting the ^{*}H-^{*}OH recombination to accelerate the ^{*}H mass transfer. This manuscript exhibits certain novelty and importance. Nevertheless, there are still many things to be resolved before reconsideration of its publication:

Response to comments: Thank you very much for your further constructive comments, which have helped us improve and refine our work. We are also grateful for your previous communication with us during the submission to Nature Synthesis and are honored to have received your positive acknowledgment at that time. In this revised version, we have added additional data to further strengthen the manuscript for submission to Nature Communications. We sincerely hope to receive your continued support and guidance. Thank you again for your thoughtful and valuable

feedback on our work.

Q1. In Figure S13, only very limited or negligible amount of H₂O₂ evolution is detected in the gas phase, although the authors claim that H₂O₂ generation was obtained in liquid and gas phases. Thus, it is recommended to report the UV-vis results for H₂O₂ quantification and the calibration curves for H₂O₂ generation.

Response: Thank you for your guidance, which has helped us further improve this section. We have now added a detailed description of the calibration curve used in the quantification of H₂O₂ production: To quantify the production of H₂O₂, the Ce⁴⁺ back-titration method was employed. Standard solutions of ceric sulfate (Ce(SO₄)₂) with concentrations of 0.1, 0.2, 0.3, 0.4, and 0.5 mM were prepared and added to sulfuric acid (H₂SO₄, 0.5 mM). The calibration curve was then measured at 316 nm using a UV-Vis spectrophotometer (UH5300, Hitachi). As shown in **Figure R1e** and **R1f**, a linear calibration curve was established based on the relationship between concentration and absorbance, with a correlation coefficient (R^2) of 0.996, confirming the reliability of the calibration. In addition, the absorbance values of the samples added to 100 mL of the standard solution (0.5 mM Ce(SO₄)₂) were measured. The Cu₍₉₎/AgBr₍₁₀₎/TiO₂ and Cu₍₉₎/TiO₂ samples showed absorbances of 1.353

and 2.285, respectively (**Figure R1g** and **R1h**), corresponding to H_2O_2 concentrations of 0.1097 and 0.0135 mM. The calculated H_2O_2 production rates are 683.6 and 83.4 $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$. The corresponding revision and discussion have been added in manuscript (line 300-321).

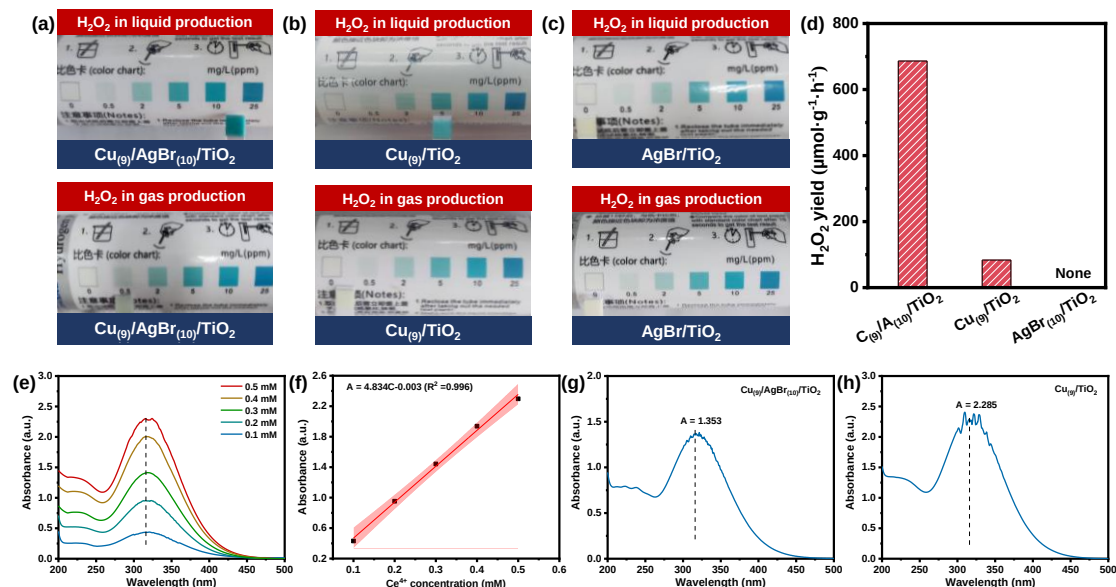


Figure R1. Images of H_2O_2 test strips used for both gas-phase and liquid-phase product detection over (a) $\text{Cu}_9/\text{AgBr}_{10}/\text{TiO}_2$, (b) Cu_9/TiO_2 , and (c) $\text{AgBr}_{10}/\text{TiO}_2$. (d) H_2O_2 production rates of the as-prepared samples. (e) UV-vis absorption spectra of Ce^{4+} solution in different concentrations (0.1-0.5 mM). (f) The concentration standard curve of Ce^{4+} solution. UV-vis absorption spectra of Ce^{4+} solution (0.5 mM) mixed with the generated H_2O_2 over (g) $\text{Cu}_9/\text{AgBr}_{10}/\text{TiO}_2$ and (h) Cu_9/TiO_2 .

Q2. What are the H_2O_2 evolution results after 72 h reaction?

Response: We appreciate your valuable feedback, which has enabled us to provide a more complete dataset. We have now added the quantitative results of the three independent 24-hour cycling experiments. By measuring the H₂O₂ yield in each cycle, the absorbance values obtained after adding the samples to 200 mL of the standard solution (0.5 mM Ce(SO₄)₂) were 0.561, 0.769, and 0.973, respectively (**Figure R2**). These values correspond to H₂O₂ concentrations of 0.1917, 0.1701, and 0.1490 mM. In total, 102.2 μ mol of H₂O₂ were produced over 72 hours using 2 mg of catalyst. The corresponding revision and discussion have been added in manuscript (line 337-342).

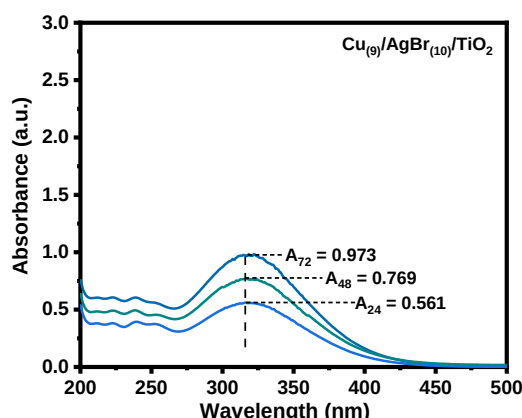


Figure R2. UV-vis absorption spectra of Ce⁴⁺ solution (0.5 mM) mixed with the generated H₂O₂ over Cu₍₉₎/AgBr₍₁₀₎/TiO₂ after 72 h reaction.

3. In Figure 5a and b, there are no peaks related to the generation of H₂O₂. It is recommended that authors provide more evidence on this through in situ IR technique.

Response: We sincerely appreciate your guidance, which has greatly improved the depth and clarity of our mechanistic analysis. Thank you for sharing your valuable experience, which has helped us clarify the H₂O₂ formation process in our system. We previously overlooked the critical role of in-situ IR spectroscopy in elucidating the molecular evolution pathways of H₂O₂. After expanding the investigated spectral range, we identified substantial differences in the H₂O₂ formation pathways on the surfaces of AgBr₍₁₀₎/TiO₂ and Cu₍₉₎/AgBr₍₁₀₎/TiO₂. As shown in **Figure R3a** and **R3b**, both samples exhibit surface *OH formation peaks, indicating that oxidation reactions can effectively generate *OH species on AgBr₍₁₀₎/TiO₂ and Cu₍₉₎/AgBr₍₁₀₎/TiO₂. Notably, on AgBr₍₁₀₎/TiO₂, we observed pronounced downward-shifting O-O dissociation-related bands, including O₂⁻ (1162 cm⁻¹), O-O (891 cm⁻¹), *O-OH (1053 cm⁻¹), and H₂O₂ (875 cm⁻¹).^{1,2} These features suggest that AgBr₍₁₀₎/TiO₂ lacks sufficient O-O coupling sites to promote efficient H₂O₂ formation. In contrast, Cu₍₉₎/AgBr₍₁₀₎/TiO₂ exhibits distinct O-O formation signals, most prominently the generated characteristic peaks assigned to *O-OH (1053 cm⁻¹) and H₂O₂ (875 cm⁻¹). This clearly indicates that the presence of Cu sites significantly enhances the O-O coupling process and thereby facilitates effective H₂O₂ production. The corresponding revision and discussion have been added in manuscript (line 406-420).

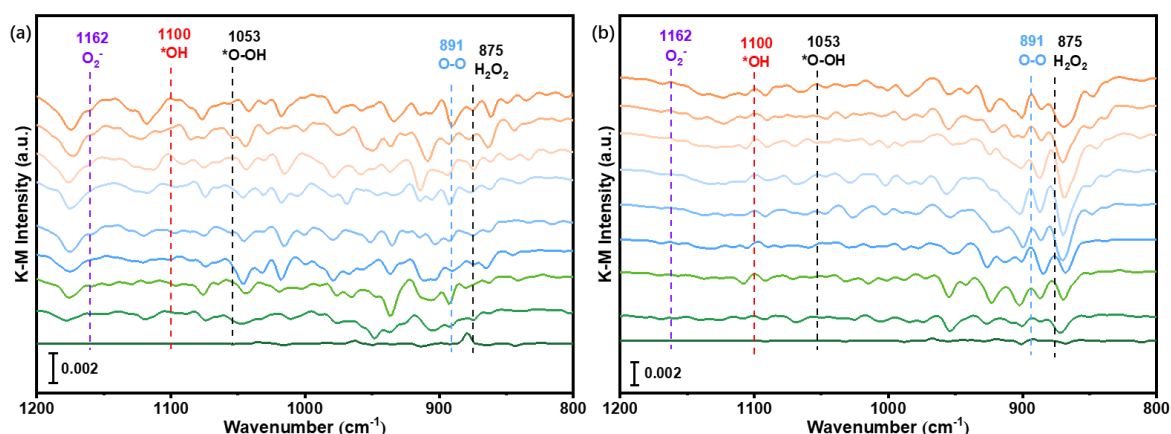


Figure R3. *In situ* DRIFTS spectra of AgBr₍₁₀₎/TiO₂ (a) and Cu₍₉₎/AgBr₍₁₀₎/TiO₂ (b).

[1] Chen, H. et al. Integration of plasmonic Ag(I) clusters and Fe(II) porphyrinates into metal-organic frameworks for efficient photocatalytic CO₂ reduction coupling with photosynthesis of pure H₂O₂. *Angew. Chem. Int. Ed.* **63**, e202412553 (2024).

[2] Zou, W. et al. Metal-free photocatalytic CO₂ reduction to CH₄ and H₂O₂ under non-sacrificial ambient conditions. *Angew. Chem. Int. Ed.* **62**, e202313392 (2023).

4. It is recommended that the quenching test results for generation of H₂O₂ and ethylene can be provided.

Response: Thank you for your valuable suggestion. We have added a clearer description of the quenching test and the quantification procedure for H₂O₂ generation. The Ce⁴⁺ back-titration method is based on the

reaction between H_2O_2 and Ce^{4+} , in which Ce^{4+} is reduced to Ce^{3+} , accompanied by a color change from yellow to colorless. Standard solutions of ceric sulfate ($\text{Ce}(\text{SO}_4)_2$) with concentrations of 0.1, 0.2, 0.3, 0.4, and 0.5 mM were individually prepared and mixed with sulfuric acid (H_2SO_4 , 0.5 mM). The calibration curve was recorded at 316 nm using a UV-Vis spectrophotometer (UH5300, Hitachi). As shown in **Figure R4a** and **R4b**, a linear calibration curve was established based on the relationship between concentration and absorbance, with a correlation coefficient (R^2) of 0.996, confirming the reliability of the calibration. Furthermore, the samples were added to 100 mL of the standard solution (0.5 mM $\text{Ce}(\text{SO}_4)_2$). The measured absorbance values for $\text{Cu}_{(9)}/\text{AgBr}_{(10)}/\text{TiO}_2$ and $\text{Cu}_{(9)}/\text{TiO}_2$ were 1.353 and 2.285, respectively (**Figure R4c** and **R4d**), corresponding to H_2O_2 concentrations of 0.1097 and 0.0135 mM. The calculated production rates are 683.6 and 83.4 $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$.

The quenching test results for ethylene generation also provided. First, a gas chromatography calibration curve was established by adjusting the amount of standard gas (**Figure R5a**). The calibration curve for C_2H_4 exhibited a correlation coefficient (R^2) of 0.997, indicating that the retention time and integrated peak area obtained from gas chromatography allow accurate qualitative and quantitative analysis of the C_2H_4 product. Moreover, during continuous photocatalytic reactions, the peak area

showed a consistently stable increasing trend, further confirming the reliability of the detection (**Figure R5b**). Time-dependent *in situ* diffuse reflectance Fourier transform infrared spectroscopy (DRIFTS) was also conducted to track the concrete reactive intermediates during the constant irradiation (**Figure R5c**). The obvious peaks at 1510, and 1552 cm^{-1} are assigned to $^*\text{OCCO}$ intermediate, indicating the accessible capacities for $^*\text{CO}$ coupling on $\text{AgBr}_{(10)}/\text{TiO}_2$ and $\text{Cu}_{(9)}/\text{AgBr}_{(10)}/\text{TiO}_2$. Moreover, the peaks at 1585 and 1309 are ascribed to $^*\text{OCCOH}$ and $^*\text{OCCHOH}$, which further implies the C_2H_4 production pathway. To further investigate the origin of the C_2H_4 , the photocatalytic isotope labeling experiment using $^{13}\text{CO}_2$ was conducted, and the resulting products were analyzed by gas chromatography-mass spectrometry (GC-MS). In **Figure R5d**, the total ion chromatogram (TIC) shows characteristic peak for C_2H_4 at 6.03. By employing the Selected Ion Monitoring (SIM) method to analyze the peak, the mass spectra signals at $m/z = 30$ (**Figure R5e**) is corresponded to $^{13}\text{C}_2\text{H}_4$, implying that C_2H_4 is derived from the conversion of CO_2 . Therefore, the multiple quenching test results provide detailed evidence for elucidating the C_2H_4 formation process.

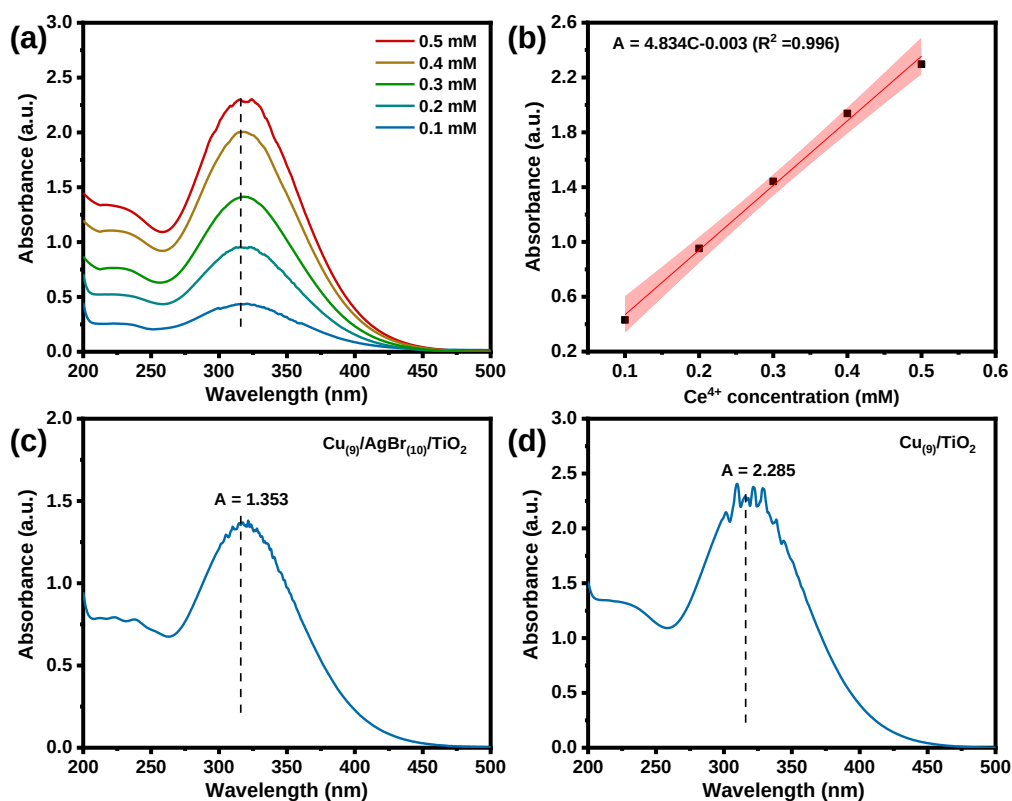


Figure R4. (a) UV-vis absorption spectra of Ce^{4+} solution in different concentrations (0.1-0.5 mM). (b) The concentration standard curve of Ce^{4+} solution. UV-vis absorption spectra of Ce^{4+} solution (0.5 mM) mixed with the generated H_2O_2 over (c) $\text{Cu}_{(9)}/\text{AgBr}_{(10)}/\text{TiO}_2$ and (d) $\text{Cu}_{(9)}/\text{TiO}_2$.

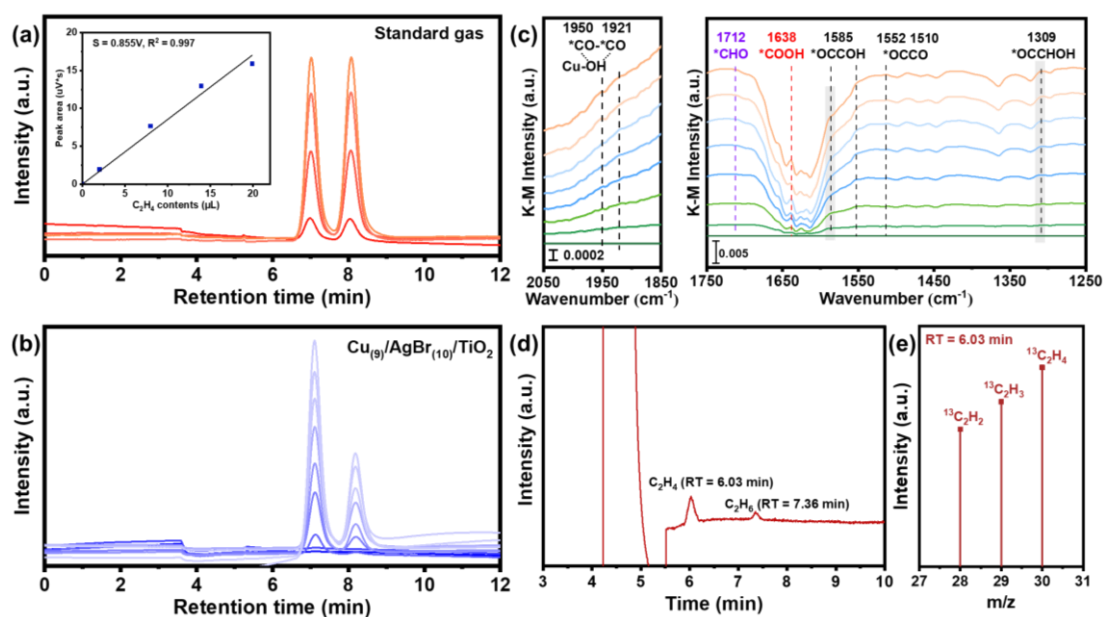


Figure R5. (a) Gas chromatogram of the C_2H_4 standard gas. (b) Gas chromatogram of the C_2H_4 product generated by $Cu_{(9)}/AgBr_{(10)}/TiO_2$ under continuous illumination. (c) *In situ* DRIFTS spectra of $AgBr_{(10)}/TiO_2$ (a) and $Cu_{(9)}/AgBr_{(10)}/TiO_2$. (d) GC spectra of C_2H_4 and C_2H_6 . (e) MS spectra of C_2H_4 .

5. How about the structure and composition change after the 72-hour reaction for $Cu_{(9)}/AgBr_{(10)}/TiO_2$? It is recommended that the authors provide some results on the structure/composition change.

Response: Thank you for your valuable input on our research. We conducted three separate 24-hour stability tests on our most efficient catalyst, and even after a total reaction time of 72 hours, its performance remained at approximately 80% of the initial value (**Figure R6a**), demonstrating outstanding catalytic stability. Furthermore, structural analyses of the post-reaction catalyst were performed using conventional XRD, HRTEM, and EDS mapping methods. The XRD results confirmed that the crystal structure of $Cu_{(9)}/AgBr_{(10)}/TiO_2$ photocatalyst remained unchanged (**Figure R6b**). In addition, HRTEM and EDS mapping analyses confirmed that the ternary heterojunction structure—comprising Cu nanoclusters anchoring AgBr nanoparticles on a TiO_2 support—retained its integrity (**Figure R7 and R8**). These findings underscore the excellent

structural stability of the $\text{Cu}_{(9)}/\text{AgBr}_{(10)}/\text{TiO}_2$ during photocatalytic process.

The corresponding revision and discussion have been added in manuscript (line 342-351).

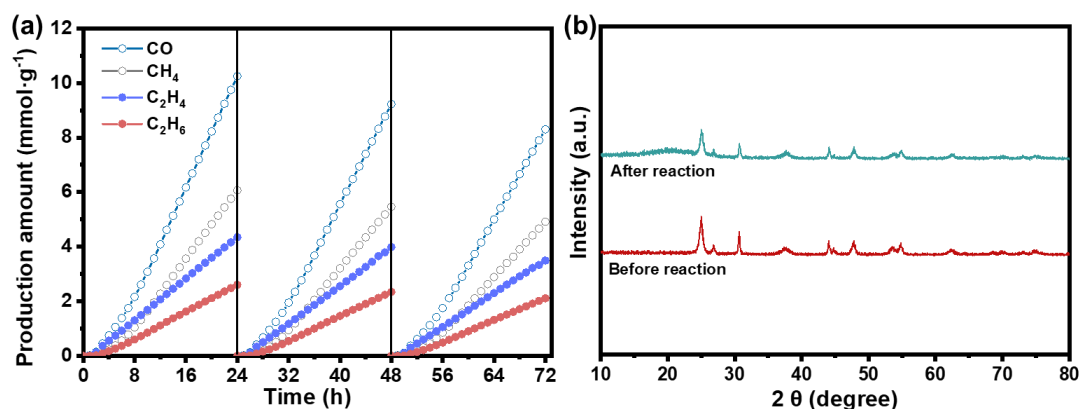


Figure R6. (a) Time-dependent products evolution over 24 h irradiation for 3 times repeated tests. (b) XRD patterns of $\text{Cu}_{(9)}/\text{AgBr}_{(10)}/\text{TiO}_2$ before and after cycling tests.

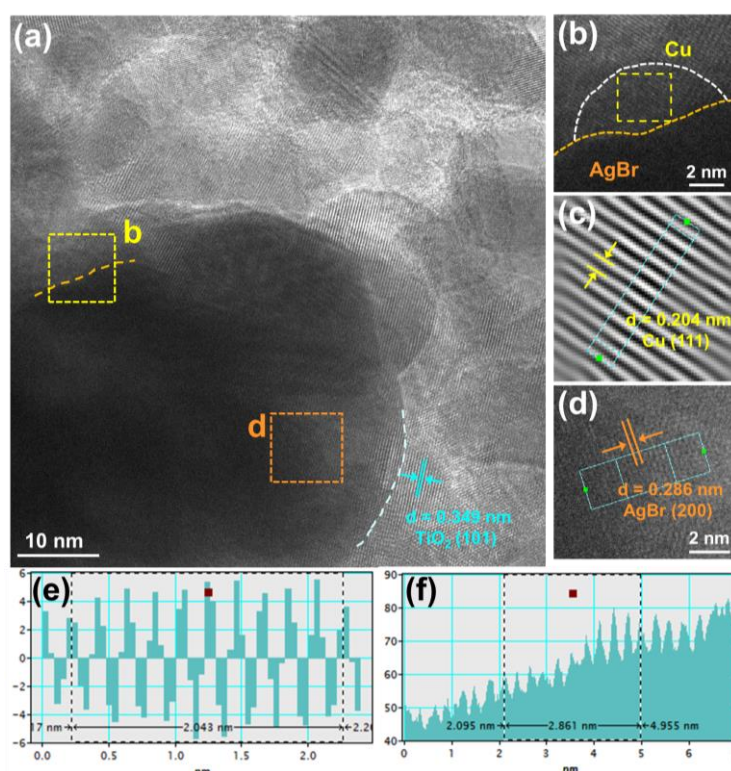


Figure R7. (a) HRTEM of $\text{Cu}_{(9)}/\text{AgBr}_{(10)}/\text{TiO}_2$ after 24 h irradiation for 3

times repeated tests. (b) Magnifying area in (a). (c) Fast fourier transform (FFT) of the yellow area in (b). (d) Magnifying area in (a). (e) Brightness intensity of the blue line profile in (c). (f) Brightness intensity of the blue line profile in (d).

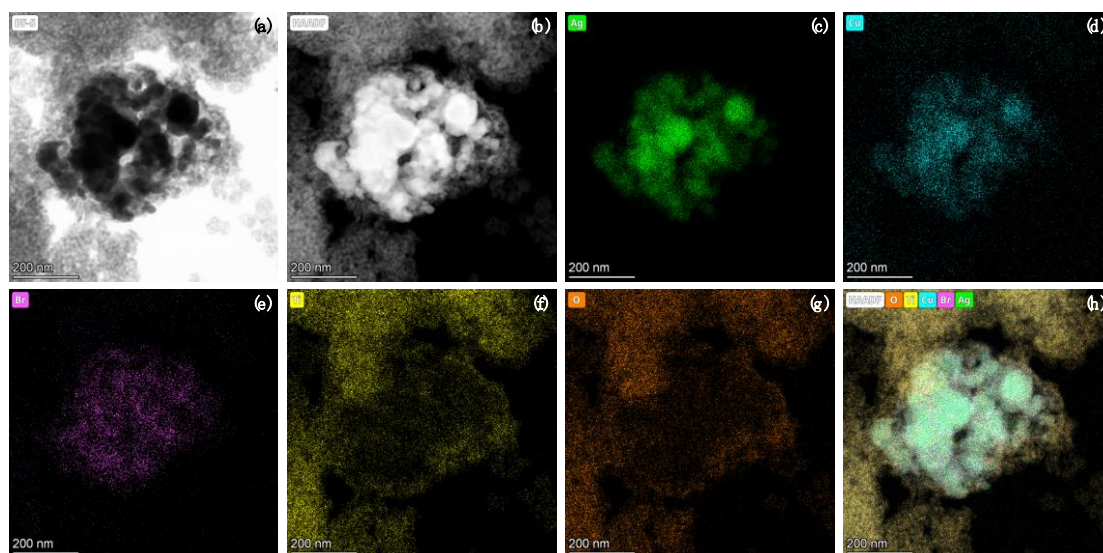


Figure R8. (a) TEM, (b) HAADF-STEM image, and (c-h) the corresponding EDS element mappings of Cu₉/AgBr₁₀/TiO₂ after 24 h irradiation for 3 times repeated tests.

6. Authors claim that the Cu²⁺ of the catalyst are reduced under light irradiation. How about the status of Cu element after the reaction without light irradiation?

Response: Thank you for your thoughtful suggestion regarding the examination of possible structural changes in the catalyst before and after illumination. We subjected the Cu₉/AgBr₁₀/TiO₂ catalyst to 1 hour of

light irradiation and analyzed the valence state of Cu before and after the reaction. The results show that during illumination, a portion of the surface Cu^{2+} species was reduced due to the strong reducing power of the photogenerated electrons (**Figure R9**). After the light was turned off, the surface Cu species returned to the Cu^{2+} state. This indicates that Cu^{2+} remains intrinsically stable and is only reduced under illumination, further demonstrating the excellent stability of the photocatalyst.

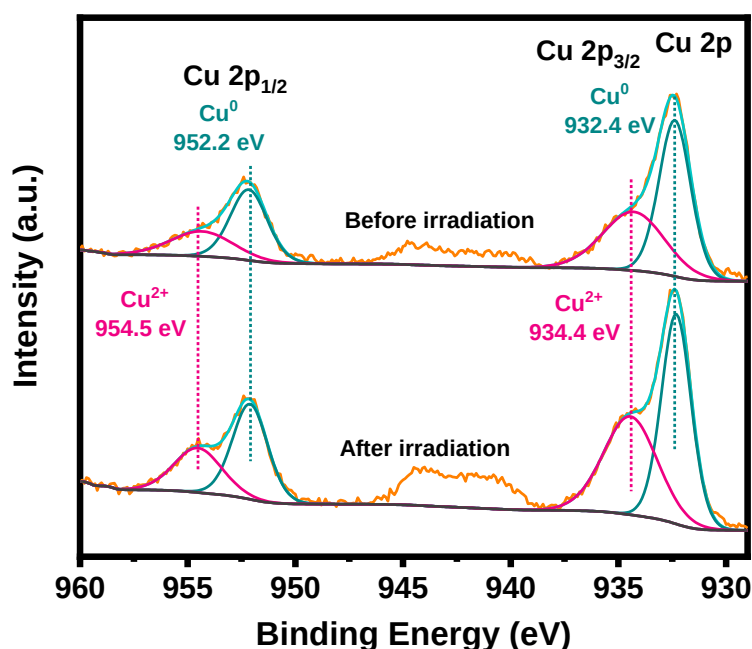


Figure R9. XPS spectra for Cu 2p of $\text{Cu}_{(9)}/\text{AgBr}_{(10)}/\text{TiO}_2$ before and after irradiation.

I have reviewed the Review 3 comment and the response from the authors.

There are still some issues to be resolved:

Response to comments: Thank you very much for your careful

re-evaluation of the comments and our responses. We sincerely appreciate your continued attention to the remaining issues. We will carefully address each point you raised and further revise and clarify the manuscript to ensure that all concerns are fully resolved. Your constructive feedback is invaluable to improving the quality and rigor of our work.

Q1. In Figure 6e, the deconvolution of Br 3d peak in the dark is not correct. The peak area ratio of Br 3d_{3/2} peak to Br 3d_{5/2} peak ought to be 2 : 3.

Response: Thank you for your careful guidance. We have adjusted the Br 3d peak area ratio between the Br 3d_{3/2} and Br 3d_{5/2} peaks to the correct value of 2:3. As shown in **Figure R1**, the binding energies of the Br 3d_{3/2} and Br 3d_{5/2} peaks exhibit almost no change before and during illumination, whereas a significant increase in surface electrons is observed on Ag. This indicates that electrons are continuously replenished from TiO₂ to Br atoms and subsequently transferred to the Ag active sites. These results further support the presence of an S-scheme charge transfer pathway at the AgBr/TiO₂ heterointerface.

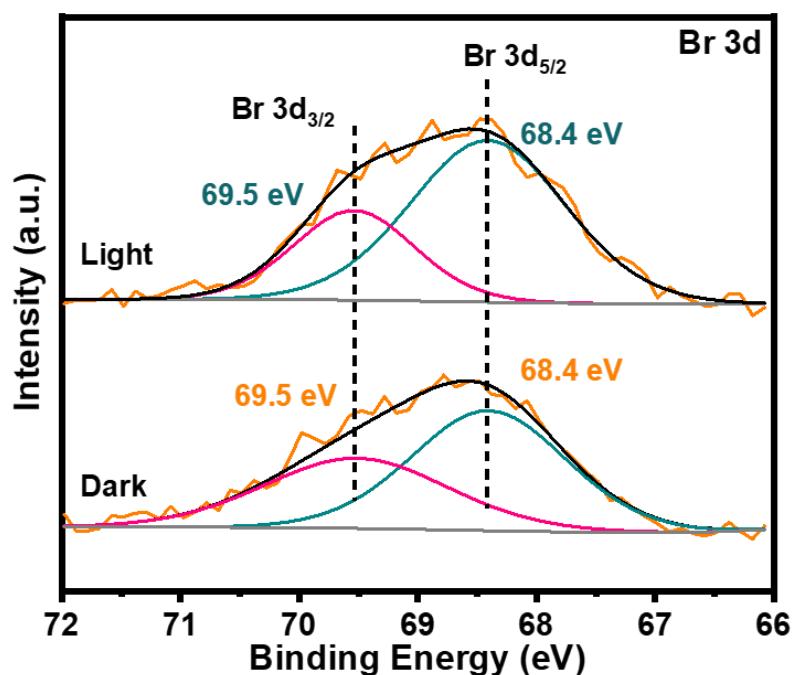


Figure R1. *In situ* irradiated XPS spectra for Br 3d of Cu₍₉₎/AgBr₍₁₀₎/TiO₂ in the dark or under irradiation.

Q2. In Figure S22e and f, the line 1 and 2 potential values are ~750 and ~600 mv, respectively, corresponding to the potential values on the surface of Cu-modified AgBr and TiO₂ particles. However, according to the mechanism in Figure 7b and c, the photo-generated electrons remain at Cu-modified AgBr and the photo-generated holes remain at TiO₂ particles, respectively. Thus, the claims and the AFM-KPFM test results are contradictory.

Response: Thank you for your thoughtful review and constructive suggestions, which have helped improve the quality of this work. We sincerely apologize for the imprecise region selection in the previous

Kelvin probe force microscopy (KPFM) measurements. After realizing that the originally selected areas predominantly covered TiO₂-aggregated regions, we have now reselected new regions that include both Cu-modified AgBr and TiO₂ to provide a clearer contrast in surface potential under light excitation. Since the contact potential difference (CPD) in KPFM is closely related to surface charge distribution, the CPD analysis of TiO₂ under dark and illuminated conditions shows a substantial increase, rising from 551.7 mV in the dark to 795.2 mV under illumination (Figure R2a-c). This pronounced enhancement indicates that light excitation effectively promotes the migration of photogenerated holes from the bulk to the TiO₂ surface. In contrast, for the Cu₍₉₎/AgBr₍₁₀₎/TiO₂ composite catalyst, a significant decrease in CPD was observed, dropping from -4.3 mV in the dark to -58.1 mV under illumination (Figure R2d-f). This notable shift in surface potential demonstrates the substantial modulation introduced by Cu-modified AgBr due to Fermi level equilibration, and further indicates that a large number of photogenerated electrons accumulate on the Cu-modified AgBr surface under light irradiation. These findings collectively support the presence of an S-scheme charge-transfer pathway. The corresponding revision and discussion have been added in manuscript (line 540-557).

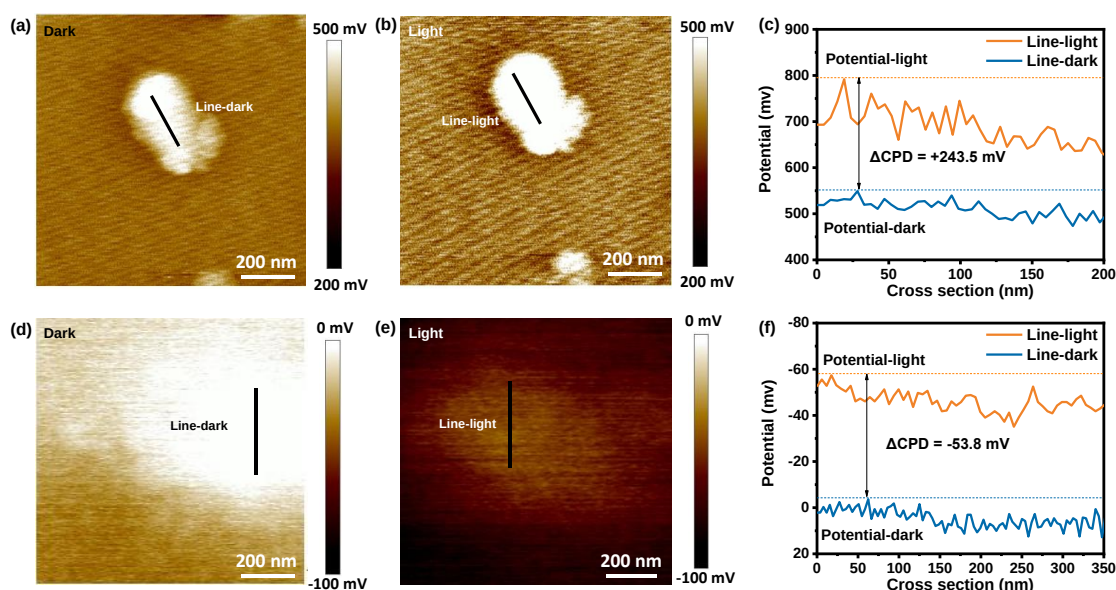


Figure R2. (a) KPFM potential image of TiO₂ under illumination under dark. (b) KPFM potential image of TiO₂ under illumination. (c) Cross-sectional SPV of TiO₂ acquired from KPFM potential images under dark and illumination. (d) KPFM potential image of Cu₉/AgBr₁₀/TiO₂ under illumination under dark. (e) KPFM potential image of Cu₉/AgBr₁₀/TiO₂ under illumination. (f) Cross-sectional SPV of Cu₉/AgBr₁₀/TiO₂ acquired from KPFM potential images under dark and illumination.

Q3. In Figure S14, the XRD results of the reacted catalyst exhibits the clear reduced peak intensity and the appearance of apparent bump at ~ 20 degrees, obviously indicating the corrosion and reduced crystallinity of the reacted catalyst and also the appearance of amorphous structure in the reacted catalyst. All these results indicate the stability of catalyst is not that good. Authors ought to revise the claim.

Response: Thank you for your careful examination. We have revised the corresponding descriptions in the manuscript. The observed differences may mainly arise from variations in testing techniques and sample preparation, as well as the fact that the structural stability of the catalyst still requires improvement. Before the reaction, a relatively large amount of catalyst was used for XRD measurement, ensuring uniform sample preparation, and the glass substrate was fully covered. In contrast, the amount of catalyst collected after the reaction was much smaller, leaving parts of the glass substrate exposed during XRD testing, which resulted in a weak broad peak near 20 degrees and a slight decrease in crystallinity. In addition, long-term reactions may induce structural changes in the catalyst, such as the oxygen vacancies formation, surface carbon impurities adsorption, or partial migration and aggregation of metal atoms. Therefore, in our future work, we will further focus on simultaneously improving both the stability and activity of the catalyst to promote continued advancement in photocatalytic technology.

Q4. In Figure 6b, there are sub-peaks in the XPS results of Ag 3d_{3/2} and Ag 3d_{5/2} peaks. However, the authors might try to deconvolute the peaks.

Response: Thank you for your valuable comments. Based on your suggestion, we have performed peak fitting for the Ag XPS results. Without

irradiation, Ag 3d exhibits two independent characteristic peaks of Ag⁺ at 367.8 and 373.8 eV, corresponding to Ag 3d_{5/2} and Ag 3d_{3/2} (**Figure R3**). Under illumination, the binding energies of Ag 3d_{5/2} and Ag 3d_{3/2} shift negatively by 0.6 eV, further indicating that the photogenerated electrons tend to transfer from TiO₂ to AgBr, driven by the built-in electric field at the AgBr/TiO₂ heterointerface.

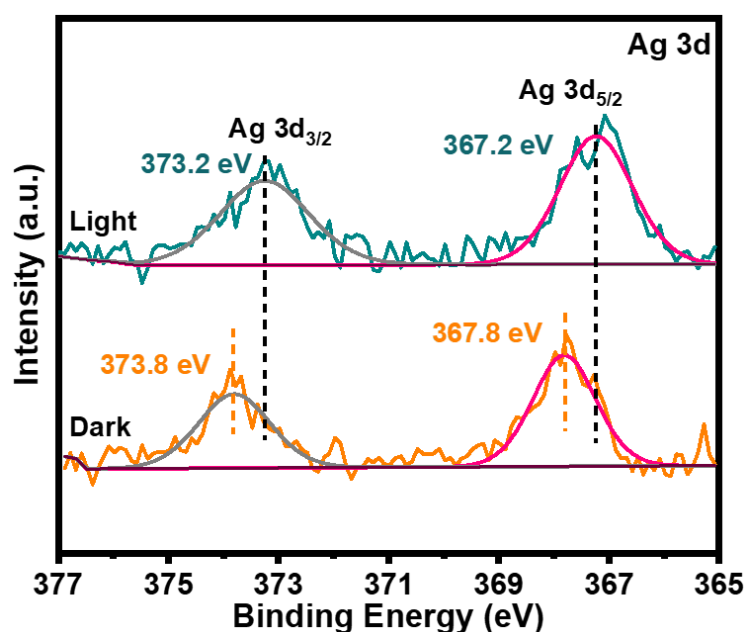


Figure R3. *In situ* irradiated XPS spectra for Ag 3d of Cu₍₉₎/AgBr₍₁₀₎/TiO₂ in the dark or under irradiation.

Reviewer #2 (Remarks to the Author):

The authors have thoroughly addressed all major concerns from the previous review, and the manuscript has been significantly improved. I find that its quality now meets the high standards required by Nature

Communications. Therefore, I recommend acceptance.

Response to comments: Thank you sincerely for your continuous support and constructive guidance throughout the review process, from Nature Synthesis to Nature Communications. Your insightful comments have greatly contributed to improving the rigor, clarity, and overall quality of our work. We truly appreciate your positive evaluation and recommendation for acceptance.