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Dual-Temperature Photothermal Tandem Catalysis for CO₂ Conversion to OlefinsZiyang Li^a, Yuhang Qia^{a*}, Lipu Han^a, Chu Zhang^a, Zhenwen Yang^a, Zhike Wu^a, Shanshan Xu^{b,c*}Received 00th January 20xx,
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We report a dual-temperature photothermal tandem system that overcomes this temperature mismatch and enables efficient CO₂ hydrogenation to light olefins in a single reactor. A graphene-modified In₂O₃ top bed drives the RWGS reaction and provides strong broadband absorption for efficient photothermal heating, thereby achieving higher temperatures for CO generation. The CO then feeds into a downstream K-doped Fe₂O₃ bed maintained at a lower temperature suitable for FTS to produce light olefins. This vertically arranged, dual-temperature configuration spatially separates and couples RWGS and FTS, effectively synchronising the tandem pathway. As a result, the system achieves a light-olefin production rate of 52.1 μmol g⁻¹ h⁻¹ with 69% selectivity at 0.05 MPa under photothermal conditions without external heating.

The increasing atmospheric CO₂ level is aggravating climate change and energy insecurity, highlighting the urgent need for sustainable carbon-recycling technologies.^{1, 2} Among the various strategies, photothermal CO₂ reduction has attracted growing attention because it does not require external electrical input and enables full-spectrum solar utilization to activate inert CO₂ molecules.^{3, 4} Significant progress has been achieved in CO₂ hydrogenation to C₁ products such as CH₄, CO, and CH₃OH.^{5, 6} However, overcoming the kinetic barriers associated with C–C bond formation to directly produce C₂⁺ hydrocarbons remains highly challenging.^{7, 8}

For thermocatalytic CO₂ hydrogenation towards C₂⁺ products, two tandem strategies are most frequently explored. One approach involves coupling CO₂-to-methanol with a subsequent methanol-to-olefins (MTO) process.^{9, 10} Although this route can yield light olefins, methanol selectivity is highly pressure-dependent. It typically requires elevated pressures to suppress methanol decomposition, which is unfavourable for scalable operations under photothermal conditions. Another

widely studied route converts CO₂ to CO via the reverse water–gas shift (RWGS) reaction, followed by Fischer–Tropsch synthesis (FTS) to form hydrocarbons.^{11–13} Integrating RWGS and FTS in a single reactor is intrinsically difficult because the two steps requires different optimal temperature windows: RWGS is endothermic and favours high temperatures at > 400 °C for efficient CO formation,^{14, 15} whereas FTS requires moderate temperatures (200–350 °C) to promote C–C coupling and avoid excessive methane formation.^{16, 17} This fundamental temperature mismatch is a key limitation to achieving direct CO₂-to-olefins conversion in a single photothermal reactor.

Compared with the recently reported single-reactor tandem photothermal system,¹⁸ this work employs a dual-bed reactor in which the RWGS and FTS functions are spatially separated. This design enables cascade heat utilization, suppresses local overheating, and allows independent optimization of each catalytic bed for improved flexibility in tuning target products. The top layer, made of graphene-modified In₂O₃ (G-In₂O₃), exhibits excellent photothermal conversion and efficiently reduces CO₂ to CO while simultaneously heating the gas phase and transferring heat to the downstream bed via combined radiative and convective heat transfer. This controlled heat flow determines the lower temperature of the potassium-doped Fe₂O₃ (K-Fe₂O₃) bed, where CO undergoes FTS to produce light olefins with high selectivity. The vertically stacked configuration establishes a well-defined temperature gradient, enabling the RWGS and FTS stages to operate within their respective optimal temperature ranges. Under low pressure (0.05 MPa), the tandem system achieves a light-olefin production rate of 52.1 μmol g⁻¹ h⁻¹ with 69% selectivity, demonstrating that engineering a temperature gradient is an effective strategy to overcome the inherent temperature mismatch in tandem photothermal catalysis.

The G-In₂O₃ composite in the top bed was prepared by using the hydrothermal method. The average particle size of In₂O₃ particles is 50–100 nm and is uniformly dispersed on the graphene sheets (Figure S1). The high-resolution transmission electron microscopy (HRTEM) image (Figure 1a) shows a distinct interface between graphene and In₂O₃. The corresponding fast Fourier transform patterns provide atomic-resolution structural

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information: the cubic lattice spots can be assigned to In_2O_3 , while the hexagonal pattern arises from the characteristic six-fold symmetry of graphene. Raman spectroscopy further confirms the formation of the G- In_2O_3 composite (Figure 1b). The D band at $\sim 1300\text{ cm}^{-1}$ and G band at $\sim 1400\text{ cm}^{-1}$ are characteristic of graphene, whereas the peaks at 307, 366, 494, and 628 cm^{-1} are aligned with the In–O vibrations of InO_6 structural units in the body-centred cubic In_2O_3 structure.

Potassium modification of Fe_2O_3 in the lower catalyst bed was verified by SEM and elemental mapping, which shows that K is homogeneously distributed on the Fe_2O_3 (Figure 1c). TEM and HRTEM analyses indicate that the lattice spacings of K- Fe_2O_3 are 0.251, 0.208, and 0.295 nm, corresponding to the (311), (400), and (220) crystal planes of $\gamma\text{-Fe}_2\text{O}_3$ (Figure S2). As shown in the X-ray diffraction (XRD) patterns (Figure 1d), all diffraction peaks can be indexed to Fe_2O_3 (JCPDS No. 39-1346) and In_2O_3 (JCPDS No. 71-2194),^{19, 20} indicating that the crystalline structures of both oxides are preserved after graphene and K modification.

In the catalyst selection for this dual-bed reactor, In_2O_3 was chosen because it is an efficient catalyst for the RWGS reaction and can produce CO with nearly 100% selectivity. The generated CO acts as the substrate for the FTS reaction, thereby initiating the tandem reaction. Fe_2O_3 was selected instead of Fe_5C_2 because it offers advantages such as facile preparation, low cost, and good structural stability. In addition, Fe_2O_3 can be transformed into the active Fe_5C_2 phase through in situ phase conversion under reaction conditions.

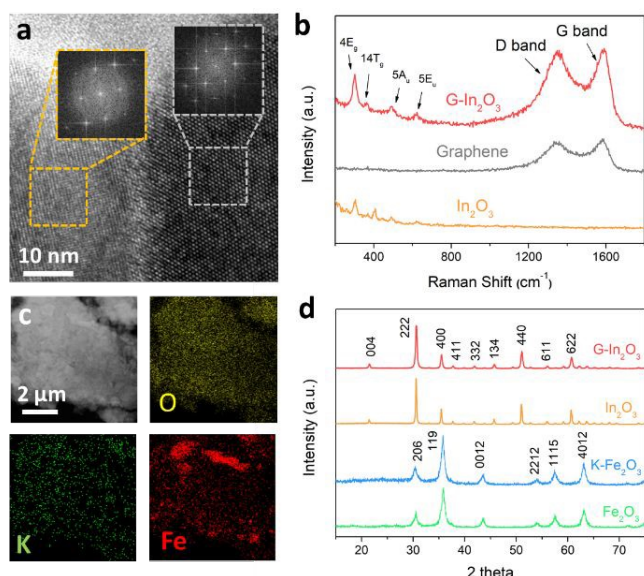


Figure 1. (a) The HRTEM image of G- In_2O_3 -10% and corresponding Fast Fourier transform, (b) The Raman spectrum of graphene, In_2O_3 and G- In_2O_3 -10%, (c) The element mapping of K- Fe_2O_3 -5%, (d) The XRD pattern of In_2O_3 , G- In_2O_3 -10%, Fe_2O_3 and K- Fe_2O_3 -5%.

To determine the optimal composition for CO_2 conversions, we evaluated G- In_2O_3 composites with various graphene loadings in the photothermal RWGS reaction (Figure S3) and K-modified Fe_2O_3 with different K contents in FTS (Figure S4). As shown in Figure S5, all the G- In_2O_3 composites produce CO as the only product, confirming that graphene incorporation does

not alter the reaction pathway. The CO production rate increases progressively, reaching a maximum for G- In_2O_3 -10%, but drops when the loading is further increased to 15 wt%. At moderate loadings (<10 wt%), graphene improves the dispersion of In_2O_3 and its specific surface area (Figure S6), and enhances CO_2 adsorption and activation (Figure S7).²¹ The strong broadband absorption of graphene raises the catalyst surface temperature under illumination, thereby accelerating the endothermic RWGS reaction (Figure S8).²² Graphene also suppresses non-radiative recombination of photogenerated carriers in In_2O_3 , resulting in higher photothermal conversion efficiency (Figure S9).^{23, 24} Such effects contribute to the enhanced CO production at the optimal 10 wt% loading. However, excessive graphene loadings will block In_2O_3 active sites and reduce accessible surface area for gas adsorption, leading to reduced CO production.

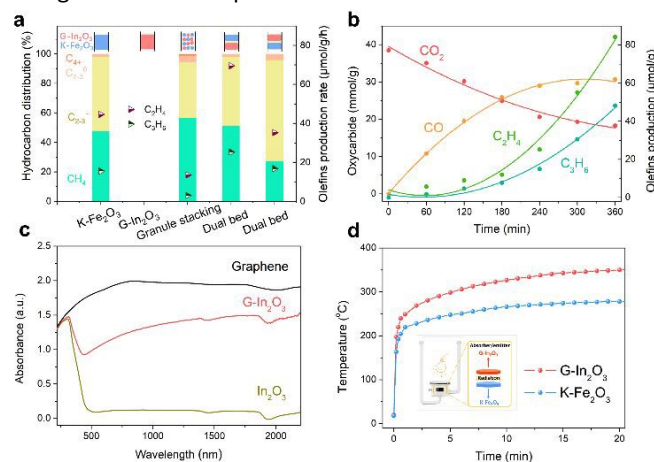


Figure 2. (a) Effect of the integration mode of G- In_2O_3 -10% and K- Fe_2O_3 -5% (mass ratio 3:5) on hydrocarbon distribution and light-olefin production rate under identical photothermal CO_2 hydrogenation conditions. (b) Time profiles of CO_2 , CO, C_2H_4 and C_3H_6 over the dual-bed G- In_2O_3 -10%/K- Fe_2O_3 -5% catalyst under continuous illumination. (c) The UV-Vis-NIR spectrum of graphene, In_2O_3 and G- In_2O_3 -10%, (d) Time evolution of the surface temperature of the G- In_2O_3 -10% and K- Fe_2O_3 -5% beds.

In the following FTS reaction, introducing potassium significantly changes both hydrocarbon distribution and light-olefin productivity. As shown in Figure S10, the K- Fe_2O_3 catalyst containing 2.5 wt% K exhibits the highest light-olefin production rate (i.e., 0.55 mmol/g/h for C_2H_4 and 0.37 mmol/g/h for C_3H_6). In contrast, although the overall olefin production rate declines at higher K loadings, the K- Fe_2O_3 with 5 wt% K catalyst shows the highest selectivity toward C_{2-3} olefins (51%), accompanied by a suppressed formation of CH_4 . A moderate amount of potassium improves the catalytic performance by slightly reducing the photoluminescence intensity of Fe_2O_3 (Figure S11), and enhancing CO adsorption and activation (Figure S12). The broad desorption peak around $450\text{ }^\circ\text{C}$ corresponds to the recombinative desorption of dissociatively adsorbed CO. However, excessive K loading further neutralises surface acidity and suppresses H_2 chemisorption, thus lowering the FTS reaction rate and shifting the product distribution away from CH_4 and toward heavier hydrocarbons and a higher olefin fraction, which agrees with the trends in Figure S10.^{25, 26} Furthermore, the photothermal FTS performance of K- Fe_2O_3



was evaluated under identical conditions while varying the light intensity, as detailed in Figure S13.

After optimising the compositions of G-In₂O₃ and K-Fe₂O₃, we evaluated tandem photothermal CO₂ hydrogenation over the G-In₂O₃-10% and K-Fe₂O₃-5% catalyst. The hydrocarbon selectivity and product distribution depend strongly on the integration configuration of the two components. The single K-Fe₂O₃ component shows low activity and poor olefin selectivity (i.e., 50%) because it cannot accumulate sufficiently high CO concentrations, allowing the Sabatier reaction to proceed and producing CH₄ (i.e., 48%) as the product (Figure 2a and S14). In contrast, single In₂O₃ primarily generates CO as a product with negligible hydrocarbons, as the low-pressure and high-temperature conditions are unfavourable for methanol or olefin formation. Simply physical mixing of two components (granule stacking) shows only a minor tandem effect, indicating that excessive proximity does not enable optimal photothermal CO₂-to-olefins performance.

Therefore, we adopt a dual-bed configuration, with G-In₂O₃-10% in the upper layer and K-Fe₂O₃-5% in the lower layer, separated by an inert screen (Figure 2a). When K-Fe₂O₃-5% is positioned beneath G-In₂O₃-10%, the light-olefin production rate achieved 52.1 $\mu\text{mol g}^{-1} \text{h}^{-1}$ with a selectivity of 69%. In contrast, reversing the order—placing K-Fe₂O₃-5% above G-In₂O₃-10%—resulted in a significantly lower olefin productivity, confirming that efficient tandem CO₂ reduction and F-T coupling requires the correct thermal and reactant-flow sequence. This improvement arises from the temperature distribution and reaction dynamics revealed in Figure 2b. In the dual-bed configuration, the upper G-In₂O₃-10% layer, operating at a higher photothermal temperature, rapidly converts CO₂ to CO and steadily increases the CO concentration throughout the reaction (Figure 2b). As CO continues to accumulate, the concentration surpasses the threshold required to trigger chain-growth reactions in the downstream FTS bed. Olefin production begins to rise significantly after 120 min and continues to increase over time. Meanwhile, the CO formation rate gradually approaches a quasi-steady state, while CO₂ is continuously consumed. These results demonstrate that the upper G-In₂O₃ bed must supply CO with high selectivity and rate to initiate and sustain FTS in the lower K-Fe₂O₃ bed, thereby enabling efficient tandem photothermal CO₂-to-olefins conversion. Detailed analysis of the stability test is provided in Figure S15 of the Supporting Information.

As a result, the photothermal conversion efficiency of the upper G-In₂O₃ layer is crucial. The light source primarily irradiates the top bed containing G-In₂O₃. G-In₂O₃ was selected because the tandem process is initiated by light-driven RWGS, which produces CO as the key intermediate for the subsequent FTS reaction in the lower bed. Positioning In₂O₃ in the upper bed promotes the formation of a CO-rich stream under continuous unidirectional gas flow. Moreover, UV-Vis-NIR spectra (Figure 2c) show that pristine In₂O₃, as a wide-band-gap semiconductor (3.2 eV, Figure S16), shows weak absorption across visible and near-infrared regions. Graphene endows G-In₂O₃ with strong, nearly broadband light absorption, enabling efficient photothermal conversion. The heat generated in the upper bed

can also be transferred to the downstream K-Fe₂O₃ bed through thermal radiation and gas-phase convection, helping to maintain the temperature required for the tandem reaction. The resulting thermal distribution in the dual-bed configuration is confirmed under photothermal operation (Figure 2d): the upper G-In₂O₃ layer rapidly reaches $\sim 360^\circ\text{C}$ within 10 min, while the lower K-Fe₂O₃ layer stabilises at $\sim 260^\circ\text{C}$. The respective temperatures also precisely correspond to the optimal adsorption range (Figure S17).

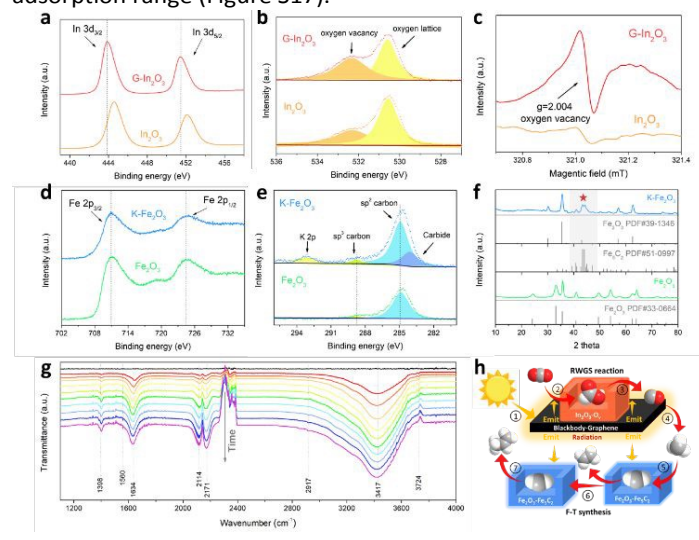


Figure 3. The XPS spectra of (a) In 3d and (b) O 1s, (c) The EPR of G-In₂O₃-10% and In₂O₃, The XPS spectra of (d) Fe 2p and (e) C 1s, (f) The XRD pattern of K-Fe₂O₃-5% and Fe₂O₃, (g) The in situ FTIR of G-In₂O₃-10% and K-Fe₂O₃-5%, (h) The advance and process of the photothermal CO₂ reduction to light olefin.

To identify the active sites in G-In₂O₃ for CO₂ reduction, XPS and EPR measurements were carried out. In comparison, the corresponding peaks of G-In₂O₃ shift to lower binding energies, which can be attributed to electron transfer at the graphene/In₂O₃ interface and an increased concentration of oxygen vacancies (Figure 3a). The O 1s spectra (Figure 3b) can be deconvoluted into two contributions: the peak at 529.8 eV is assigned to lattice oxygen in In–O–In, whereas the peak at 531.4 eV is associated with oxygen vacancies.²⁸ Furthermore, EPR spectra exhibit a signal at $g = 2.004$ (Figure 3c), which is characteristic of electrons trapped at oxygen vacancy sites.²⁹ The stronger EPR signal for G-In₂O₃ shows it has a higher oxygen-vacancy level, aligning with XPS results. This suggested that carbon at the graphene/In₂O₃ interface lowers the energy needed to create oxygen vacancies in the In₂O₃ lattice.

Because the oxygen vacancy concentration evolves under photothermal reaction conditions, we compared the XRD, PL, EPR and UV-vis-NIR spectra of G-In₂O₃ before and after the reaction (Figure S18–21). Detailed characterization and analysis are provided in the Supporting Information.

Regarding the active phase of K-Fe₂O₃ catalyst in the FTS reaction, XPS measurements were carried out to determine the surface elemental composition and chemical states (Figure 3d). In the Fe 2p spectrum, upon K promotion, the Fe 2p_{3/2} peak shifts to lower binding energy, indicating a more reduced Fe environment and thus facilitating the transformation of Fe species into Fe₅C₂, which is widely recognised as the active



phase for FTS in Fe-based catalysts.^{30–32} In the C 1s spectrum (Figure 3e), three C 1s components together with an additional K 2p signal are observed. Compared with pristine Fe₂O₃, the K-Fe₂O₃ sample exhibits new carbide-related C 1s features, aligning with the leftward shift of the Fe 2p_{3/2} peak.³³ XRD patterns of Fe₂O₃ and K-Fe₂O₃ (Figure 3f) further reveal that additional reflections in K-Fe₂O₃ can be indexed to the main diffraction peaks of Fe₅C₂, with no Fe₃C phase detected (Detailed analysis is provided in Figure S22 of the Supporting Information.).^{34, 35} K doping plays multiple roles in the Fe-based catalyst by enhancing CO adsorption and activation, increasing chain-growth probability, and suppressing the secondary hydrogenation of olefins, thereby improving light-olefin selectivity. In addition, K facilitates the formation and stabilization of the Fe₅C₂ active phase, which contributes to the enhanced FTS performance.³⁶

Although In₂O₃ is widely recognized as an effective catalyst for CO₂ hydrogenation to methanol under relatively high pressures, the reaction in this work was performed at 0.5 atm, which is less favorable for methanol formation.³⁷ To elucidate the CO₂ activation pathway, in situ FTIR spectroscopy was conducted (Figure 3g). The spectra show a band at 1398 cm⁻¹ assigned to carbonate species and a band at 1560 cm⁻¹ attributed to COOH* intermediates. Since COOH* species are generally associated with the RWGS pathway to CO rather than the methanol pathway via HCOO*, these results indicate that CO is the predominant product in the present system. The feature at 1634 cm⁻¹ originates from adsorbed water, and the bands at 2114 and 2171 cm⁻¹ are ascribed to linearly and bridge-bonded adsorbed CO*, respectively. The band at 2917 cm⁻¹ is characteristic of CH₃* species, and the broad absorptions at 3417 and 3724 cm⁻¹ are associated with surface hydroxyl groups and adsorbed water molecules.³⁸ Accordingly, these surface intermediates provide direct spectroscopic evidence for a tandem RWGS–FTS mechanism, wherein CO₂ is first reduced to CO via RWGS, and then resulting CO is subsequently hydrogenated through CH_x* intermediates to form light hydrocarbons.^{39–41}

In summary, we have developed a tandem dual-temperature catalytic configuration that enables controlled multi-stage temperature distribution and efficient energy utilisation for the photothermal hydrogenation of CO₂ into high-value hydrocarbons. In the upper bed, G–In₂O₃ drives the RWGS reaction. The absorbed light energy is converted into heat and transmitted to the underlying K–Fe₂O₃ bed via both radiative and convective heat transfer. In the lower bed, K–Fe₂O₃ drives the FTS process (Figure 3h). By synergistically engineering the dual-bed catalysts with optimal operational temperatures in a single photothermal reactor, we achieve direct CO₂-to-light-olefin conversion with a production rate of 52.1 μmol g⁻¹ h⁻¹ and a selectivity of 69%, demonstrating a promising strategy for solar-driven CO₂ valorisation.

Author contributions

L.H., J.H., and Y.Q. Conceived and wrote this project. L.H. conducted a software simulation and collected and analysed

experimental data. C.L. and Z.L. assisted with collecting experimental data and offered suggestions for revising the paper. S.X revised and conceptualized the manuscript.

Conflicts of interest

There are no conflicts to declare.

Data availability

The data supporting this article have been included as part of the SI. See DOI: <https://doi.org/10.1039/d5cc04650e>.

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Data availability

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