



## Introduction

Hydrogen energy, as a high-density, zero-carbon emission clean energy source, produces only water as a combustion product. It is expected to replace fossil fuels, thereby reducing carbon emissions and air pollution at the source. Thus, it represents a critical pathway to achieving carbon neutrality [1]. Current hydrogen production technologies primarily include hydrogen production from fossil fuels [2], electrocatalytic hydrogen production [3], and photocatalytic hydrogen production [4]. Among the widely adopted technologies for hydrogen production from fossil fuels, hydrogen is generated from coal, natural gas, and other sources through steam reforming and partial oxidation processes, which emit carbon dioxide and cause severe environmental pollution. Hydrogen production via water electrolysis is a green hydrogen production technology that decomposes water into hydrogen and oxygen using electrode catalysts. This method features simple technical processes and easy maintenance, but it suffers from extremely high electricity consumption and elevated hydrogen production costs. Photocatalytic hydrogen production, as a novel green hydrogen generation technology, has become a research hotspot in the field of energy catalysis due to its unique advantage of directly utilizing solar energy to drive water splitting.

The core objective of this study is to address key challenges in existing photocatalytic hydrogen production research, including the narrow visible-light absorption range of photocatalytic materials, low light utilization efficiency, high system costs, and the inability to provide enhanced and stable sunlight exposure. To address these issues, a solar-powered photocatalytic hydrogen production system based on  $\text{Ti}_3\text{C}_2(\text{OH})_2/\text{porous g-C}_3\text{N}_4$  materials was designed and fabricated. Its practical application value is demonstrated in three aspects. First, the application of non-tracking concentrator technology reduces the cost and maintenance complexity of the photocatalytic hydrogen production system, enhances solar energy density in the reaction system, and thereby improves solar energy utilization efficiency. Second, the photocatalytic activity of  $\text{Ti}_3\text{C}_2(\text{OH})_2/\text{porous g-C}_3\text{N}_4$  materials was enhanced by adjusting their preparation to broaden the visible-light absorption range and improve charge separation efficiency.

## Experimental setup

### System Description:

Fig.1 shows a solar-driven photocatalytic hydrogen production system. The system is mainly composed of core components, including a non-tracking trough concentrator, a transparent vacuum glass tube, and a gas collection bottle. Based on the concentrating theory of geometrical optics, the non-tracking trough concentrator can directionally focus incident parallel sunlight. Its theoretical concentration intensity is approximately equivalent to 5 standard suns, which can significantly improve the incident light energy density in the photocatalytic reaction zone [18] and provide sufficient energy input for the photocatalytic reaction. As the core component of the photocatalytic reaction, the transparent vacuum glass tube exhibits excellent light transmittance and thermal insulation properties. Excited by the concentrated solar energy, the photocatalytic material undergoes photo-excitation to generate photogenerated electron-hole pairs, which participate in the water redox reaction, thereby enabling hydrogen production. The gas collection bottle is used for the collection and temporary storage

of the product hydrogen. The system features a simple structure and low operational energy consumption, providing a feasible miniaturized experimental and application model for the engineering implementation of solar photocatalytic hydrogen production technology.

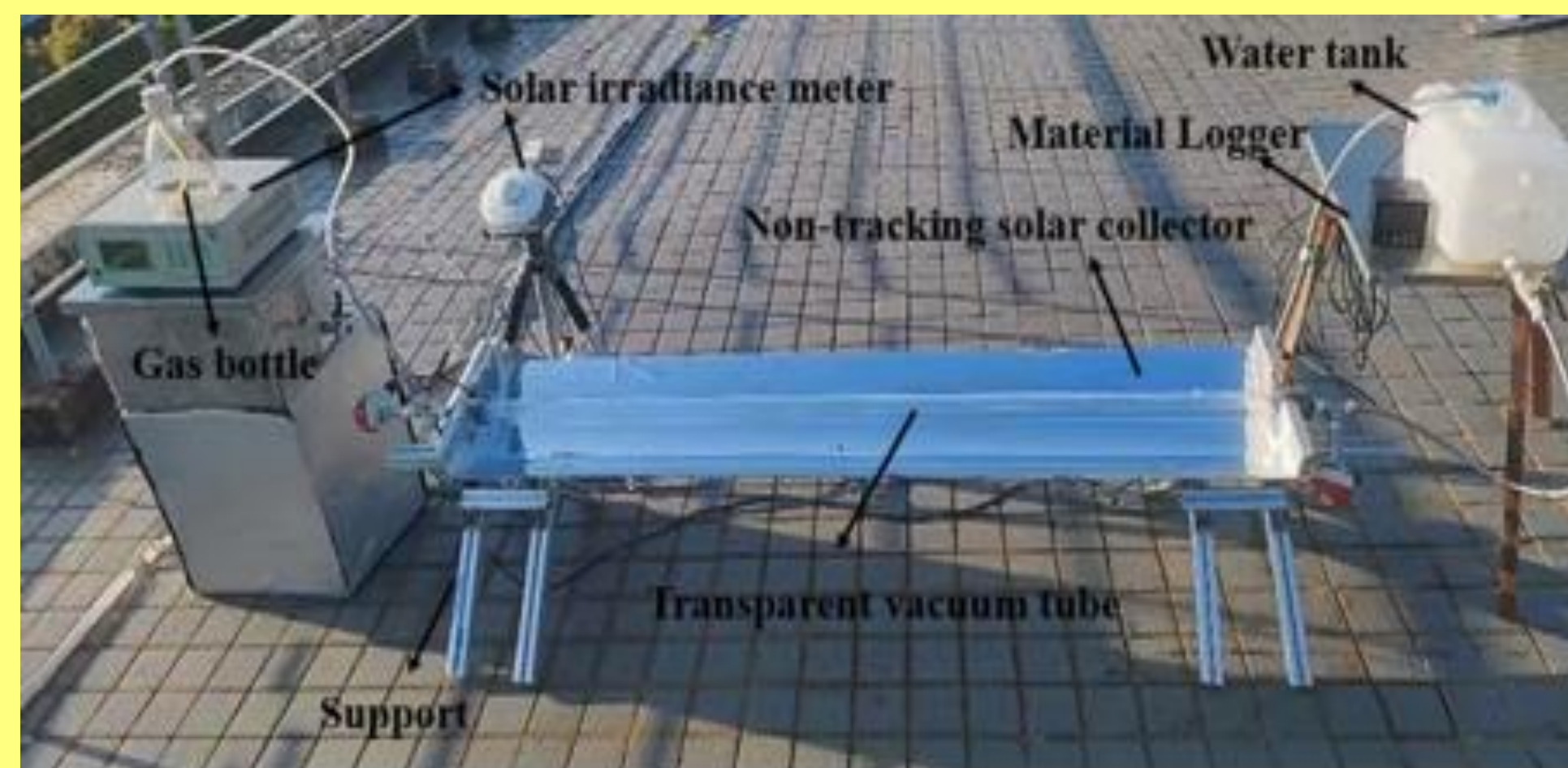


Fig. 1. Photograph of the solar-driven photocatalytic hydrogen production system.

## Experiment

### Preparation of $\text{Ti}_3\text{C}_2(\text{OH})_2/\text{Porous g-C}_3\text{N}_4$

The preparation of porous  $\text{g-C}_3\text{N}_4$  materials is illustrated in Fig.2. In this study, the precursor was regulated via a hydrothermal method, and porous  $\text{g-C}_3\text{N}_4$  was synthesized by thermal polymerization and calcination. First, 6 g urea and 8 g melamine were dispersed separately into 40 mL deionized water. After uniform dispersion, the two solutions were mixed, followed by addition of 20 mg KOH. The mixture was stirred at room temperature for 1 hour. Subsequently, the mixed solution was transferred into a 100 mL Teflon-lined autoclave for a 24 h hydrothermal reaction at 200 °C. After cooling, the product was collected by vacuum filtration and dried at 80 °C to obtain the  $\text{g-C}_3\text{N}_4$  precursor. The  $\text{g-C}_3\text{N}_4$  precursor was placed in a crucible and calcined in a muffle furnace at 550 °C for 4 h with a heating rate of 5 °C/min. After the furnace cooled to room temperature, pale yellow  $\text{g-C}_3\text{N}_4(\text{CN})$  was obtained. Subsequently, 2 g of CN was mixed with 1.23 g of LiCl, 1.5 g of KCl, and a photosensitizer(x mg), followed by thorough grinding to ensure homogeneity. The mixture was then heated in a muffle furnace at 550 °C for 4 h with a heating rate of 5 °C/min to obtain CN-G. Finally, the resulting CN-G was dispersed in 100 mL of deionized water and stirred overnight. The product was collected via centrifugation and filtration to remove residual KCl and LiCl, and then dried at 60 °C to yield pure CN-G powder.

To prepare  $\text{Ti}_3\text{C}_2(\text{OH})_2$  material. 30 mL of concentrated hydrochloric acid solution, 10 mL of deionized water, and 3.2 g of LiF were added to the reactor. The mixture was stirred thoroughly at 35 °C to achieve uniformity. Subsequently, 2 g of  $\text{Ti}_3\text{AlC}_2$  powder was added in small portions, and the mixture was stirred vigorously at 35 °C for 48 h to obtain a black suspension. The resulting product was collected by centrifugation and then dried overnight at 60 °C under vacuum to obtain multilayer  $\text{Ti}_3\text{C}_2\text{Tx}$ . Finally,  $\text{Ti}_3\text{C}_2(\text{OH})_2$  was synthesized via KOH washing. 1 g of  $\text{Ti}_3\text{C}_2\text{Tx}$  powder was dispersed in 200 mL of 2 mol/L KOH solution. After heating and stirring at 80 °C for 12 h, the mixture was washed multiple times with DI water to remove residual KOH. The mixture was then dried overnight at 80 °C under vacuum to obtain  $\text{Ti}_3\text{C}_2(\text{OH})_2$ .

Preparation of  $\text{TiCOH}/\text{CN-G-x}$  composites:  $\text{Ti}_3\text{C}_2(\text{OH})_2$  and CN-G-x were uniformly mixed in 30 mL deionized water at mass ratios of 5% and 10%, respectively. After stirring for 12 h, the mixture was centrifuged and filtered. The collected material was dried under vacuum at 60 °C for 12 h to obtain the  $\text{TiCOH}/\text{CN-G-x}$  composite.

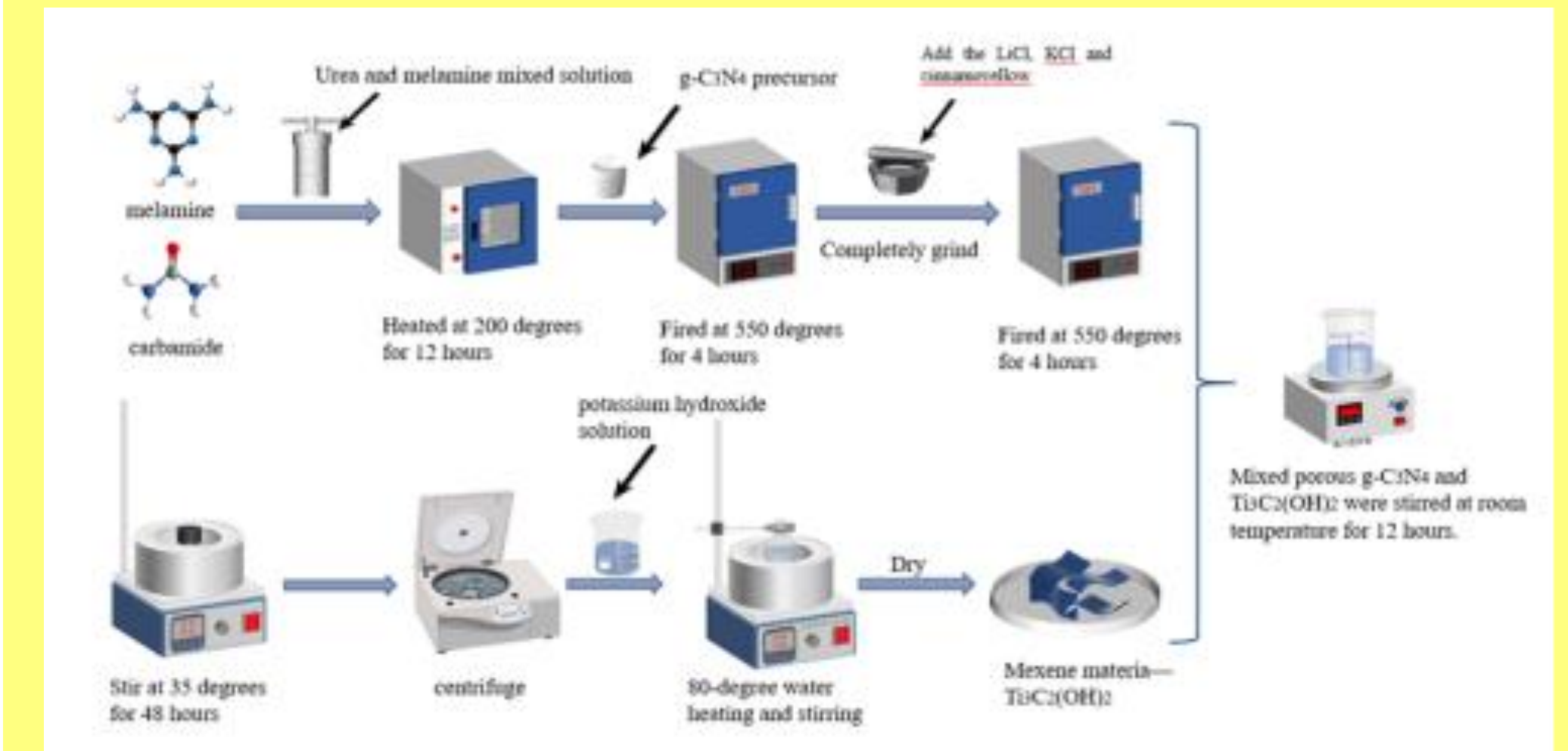


Fig.2. a schematic diagram of preparation of  $\text{TiCOH}/\text{CN-G-x}$ .

## Results

Fig 3 shows the cumulative hydrogen production of the sample material over time.

To systematically evaluate the photocatalytic hydrogen evolution performance of the as-prepared materials, hydrogen production tests were carried out in a triethanolamine (TEOA) sacrificial agent system. In the left of Fig.3 shows the hydrogen evolution performance curves of 3 mg of catalyst in 40 mL of deionized water. In the right of Fig.3 show the  $\text{TiCOH-5\%}/\text{CN-G-3}$  photocatalyst under real sunlight, an outdoor photocatalytic hydrogen production platform was constructed based on a non-tracking trough concentrator and a transparent vacuum glass tube. A continuous 5 h real-time hydrogen production test was conducted

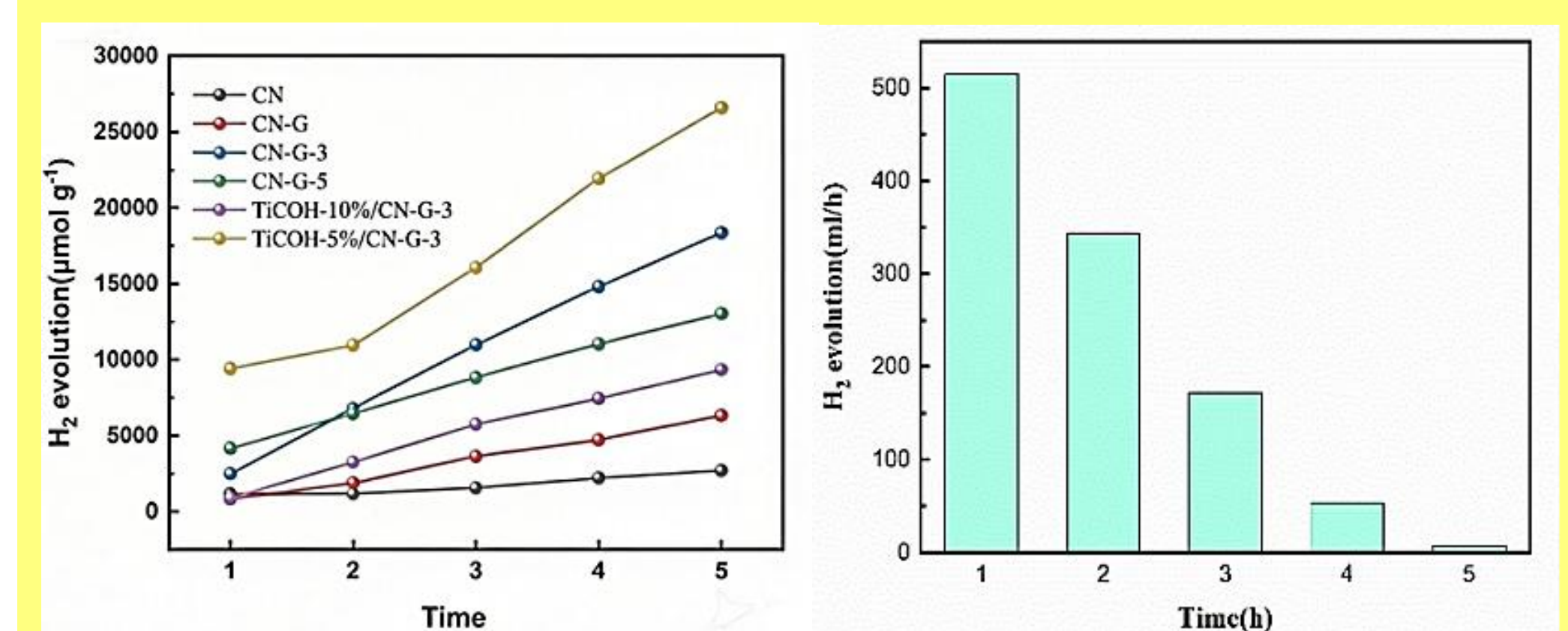


Fig. 3. Time-dependent hydrogen evolution performance.

## Conclusions

The main conclusions drawn from all these work are:

- Accuracy of the evaluation depends on the care taken when performing the test;
- A synergistic modification strategy combining structural regulation and  $\text{TiCOH}$  cocatalysis, this study designed a solar-driven photocatalytic hydrogen production system;
- The practical application of low-cost, trackless centralized photocatalytic hydrogen production technology.

## References

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