



2D/2D composites based on graphitic carbon nitride and MXenes for photocatalytic reactions: a critical review

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Abstract

The combination of the two-dimensional (2D) materials g-C₃N₄ and MXenes in photocatalysis offers several advantages. The g-C₃N₄ can serve as a visible light-absorbing material, while MXenes can enhance the charge separation and transfer processes leading to improved photocatalytic efficiency. A critical review of 77 already published articles in the field of photocatalytic reactions using g-C₃N₄ and MXenes, such as hydrogen evolution, the reduction of carbon dioxide, the degradation of organic compounds, the redox reactions of nitrogen, was conducted. For the purpose of greater objectivity, the published results were analysed by non-parametric Mann–Whitney, Kolmogorov–Smirnov, and Mood's median tests and visualised by box and whisker plots. It was found that MXenes can significantly improve the photocatalytic activity of g-C₃N₄. Adding other co-catalysts to the MXene/g-C₃N₄ composites does not bring a significant improvement in the photocatalytic performance. Promising results were obtained especially in the fields of hydrogen evolution and the reduction of carbon dioxide. Since the MXenes are relatively a new class of materials, there is still a big challenge for finding new photocatalytic applications and for the enhancement of existing photocatalytic systems based on g-C₃N₄, especially in terms of the MXenes and g-C₃N₄ surface and in the heterojunction engineering.

Keywords g-C₃N₄ · MXene · Photocatalysis

1 Introduction

Graphitic carbon nitride is a metal free polymeric semiconductor (a band gap energy of 2.7 eV) which has been intensively studied since 2009 when Wang et al. published their paper about water splitting using g-C₃N₄ [1]. It has noteworthy physico-chemical properties, such as high thermal, physical, chemical, and photochemical stability. These properties were reviewed in many comprehensive papers, for example, [2–10].

MXenes are a novel group of materials which have been studied since a paper of Naguib and co-authors was

published in 2011 [11]. MXenes are formed by the etching of MAX phases, where M is metal (Ti, V, Nb, Ta, Mo etc.), A is III or IV element and X is carbon and/or nitrogen, forming materials with a general formula of M_{n+1}X_nT_x (n = 1–4), where T is a surface functional group, such as O, OH, and F. MXenes are important materials due to their high conductivity and specific surface area, thermal and mechanical stability, hydrophilicity, and tuneable surface chemistry. The various MXene properties and possible applications were reviewed in the literature, for example, [12–19].

The combination of g-C₃N₄ and MXenes for photocatalysis began to be interesting for scientists since 2017 when Shao et al. [20] referred to the hydrogen evolution reaction (HER) in the presence of these materials. Both g-C₃N₄ and MXenes are 2D materials and their benefits are the following: (1) the absorption of light can be controlled by their thickness, (2) the migration path of charge carries to the surface is short, and (3) these materials have a high surface area and provide a lot of active sites for redox reactions [21]. In addition, composite 2D/2D materials have a high coupling interface which can accelerate the charge migration at their interface.

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On the whole, the combination of g-C₃N₄ and MXenes in photocatalysis offers several advantages. G-C₃N₄ can serve as a visible light-absorbing material generating photoinduced electrons and holes, while MXenes can enhance the charge separation and its transfer processes, leading to improved photocatalytic efficiency. The large surface area of MXenes provides abundant active sites for catalytic reactions, further enhancing the overall performance of the photocatalytic system.

The aim of this review is to summarize the already published photocatalytic applications of the g-C₃N₄ and MXenes composites and to critically assess the synergy of both materials and their suitability for various reactions. For this purpose, the non-parametric Mann–Whitney and Kolmogorov–Smirnov, and Mood’s median tests [22] were used. The reaction data were visualised by box and whisker plots.

2 A review of literature data

Peer reviewed research articles reporting the photocatalytic application of the g-C₃N₄ and MXene composites were mostly taken from journals indexed in the Web of Science. In total, only 77 articles containing results suitable for the statistical assessment were used in this study. In fact, more articles have been published so far but they did not contain the necessary reaction data. The non-parametric Mann–Whitney, Kolmogorov–Smirnov, and Mood’s median tests, without making any assumptions of the statistical distributions, were used for this purpose. A hypothesis, if there is a significant difference in the distributions of two data groups, was tested at a significance level $\alpha=0.05$ (OriginPro 2018, OriginLab Corporation, Northampton, MA, USA). In Fig. 1, it is visible that the g-C₃N₄ and MXene composites were mostly applied for the photocatalytic degradation of organic compounds (31%) and the hydrogen evolution (30%). A smaller part about 19% was devoted to

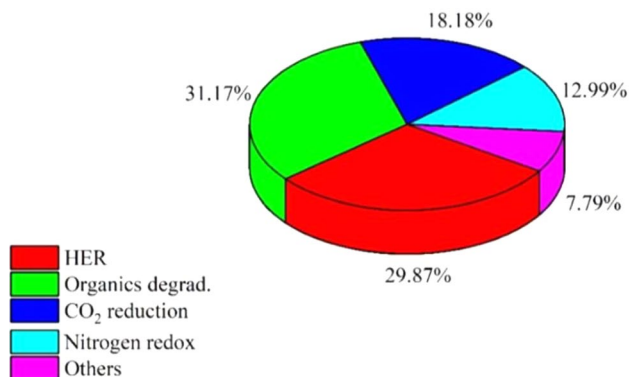
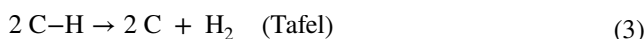
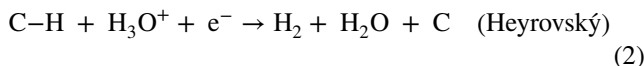
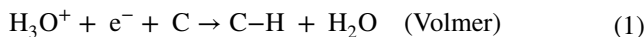


Fig. 1 An overview of the works published on the g-C₃N₄ and MXene composites for various photocatalytic reactions

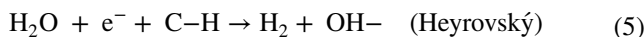
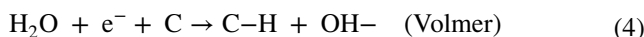
the reduction of carbon dioxide. The rest of the applications was devoted to the nitrogen redox reactions and other ones.

3 Hydrogen evolution reaction

It is widely accepted, e.g. [23], that the catalytic hydrogen evolution mechanism consists of several steps in acidic conditions



and in alkaline conditions



where C refers to a vacant surface site on a catalyst and C–H means an adsorbed hydrogen atom on the catalyst.

The first paper on the application of the composite g-C₃N₄ and Ti₂C for the hydrogen evolution by water splitting was published in 2017 by Shao et al. [20]. For this purpose, a standard AM 1.5 solar simulator was used. The highest production of hydrogen of 47.5 $\mu\text{mol h}^{-1}$ (recalculated at 950 $\mu\text{mol h}^{-1} \text{g}^{-1}$) was reached using a Ti₂C loading of 0.4 wt%. Moreover, Ti₂C itself also showed the evolution of hydrogen because it formed TiO₂ and Ti₂CO₂. The production of bare g-C₃N₄ was 66.0 $\mu\text{mol h}^{-1} \text{g}^{-1}$. The apparent quantum efficiency (AQE) was 4.3%. Triethanolamine (TEOA) was used as a hole scavenger.

The Ti₃C₂ MXene and a Pt co-catalyst in combination with g-C₃N₄ were used for HER by An et al. in 2018 [24]. The hydrogen production of 5.1 $\text{mmol h}^{-1} \text{g}^{-1}$ was achieved under the irradiation of a 300 W xenon lamp. The AQE was about 3.1% at 420 nm. A synergy effect of Ti₃C₂ and Pt nanoclusters was ascribed to the efficient separation of the photoinduced electrons and holes. It was found that more -O surface terminations improve the HER performance. The holes were scavenged by TEOA.

The composite of g-C₃N₄ and Ti₃C₂ with oxygen terminating surface groups was prepared by Sun et al. [25]. The best hydrogen production performance reached 88 $\mu\text{mol h}^{-1} \text{g}^{-1}$ and the AQE was 1.27% (using TEOA). DFT calculations demonstrated that the oxygen terminations were covered by hydrogen atoms.

Zhang et al. [26] mixed delaminated Ti₃C₂ with the nanosheets of g-C₃N₄ and calcinated these mixture at 350 °C for 1 h to obtain a Ti₃C₂/TiO₂/g-C₃N₄ composite.

The composite was used for HER under an Xe lamp with a 420 nm cut-off filter. The hydrogen production reached $1.62 \text{ mmol h}^{-1} \text{ g}^{-1}$ and the AQE was 4.16% at 420 nm (with TEOA).

Hexagonal Ni_3C nanoparticles were coupled with $\text{g-C}_3\text{N}_4$ by a simple grinding method [27]. A resulting nanocomposite containing 15 wt% Ni_3C produced $304 \text{ } \mu\text{mol h}^{-1} \text{ g}^{-1}$ corresponding to the AQE of 0.40% at 420 nm. The Xe arc lamp (350 W) with an UV cut-off filter ($> 420 \text{ nm}$) was used as a source of irradiation.

Ti_3C_2 quantum dots from Ti_3AlC_2 powders were synthesized by Li et al. [28]. The quantum dots in combination with $\text{g-C}_3\text{N}_4$ reached the hydrogen production rate of $5111.8 \text{ } \mu\text{mol g}^{-1} \text{ h}^{-1}$ which is 26 times higher than that of bare $\text{g-C}_3\text{N}_4$ ($196.8 \text{ } \mu\text{mol h}^{-1} \text{ g}^{-1}$). The highest AQE was 3.654% under simulated solar irradiation produced by a 300 W Xe arc lamp equipped with an AM-1.5 filter.

The synthesis of Ti_3C_2 and O-doped $\text{g-C}_3\text{N}_4$ composites was reported by Lin et al. [29]. The composite $\text{Ti}_3\text{C}_2/\text{O-doped g-C}_3\text{N}_4$ (see Fig. 2) produced hydrogen at the rate of $25,124 \text{ } \mu\text{mol g}^{-1} \text{ h}^{-1}$ in comparison to $13,745 \text{ } \mu\text{mol g}^{-1} \text{ h}^{-1}$ and $15,573 \text{ } \mu\text{mol g}^{-1} \text{ h}^{-1}$ provided by single O-doped $\text{g-C}_3\text{N}_4$ and $\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$, respectively. The AQE values were measured at 17.59% and 6.53% for 405 nm and 420 nm, respectively (with TEOA). The Schottky junction between the MXene and graphitic carbon nitride was based on their electrostatic assembly and resulted in a short charge transport distance from $\text{g-C}_3\text{N}_4$ and Ti_3C_2 . Oxygen doping was found to improve the separation of the electrons and holes.

The synthesis of $\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ composites based on their electrostatic self-assembly was also described by Su

et al. [30]. Ti_3C_2 was found to be -O terminated. The optimized composite reached the hydrogen performance of $72.3 \text{ } \mu\text{mol g}^{-1} \text{ h}^{-1}$ (using TEOA) under a Hg lamp equipped with a cut-off filter for 400 nm.

The $\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ composites were prepared by the mechanical mixing of sonicated $\text{g-C}_3\text{N}_4$ and plasma treated Ti_3C_2 . Hydrogen was evolved with the maximum rate of $17.8 \text{ } \mu\text{mol h}^{-1} \text{ g}^{-1}$ under a 350 W Xe arc lamp (a cut-off filter for 400 nm, TEOA) [31]. The improved photocatalytic activity was explained by the presence of Ti^{4+} due to the plasma treatment, which could capture photo-induced electrons from $\text{g-C}_3\text{N}_4$ and thus improved their separation from the holes. The plasma treatment modified the surface functional groups of Ti_3C_2 , and the increase of Ti-O and the reduction of Ti-C, Ti-F and Ti-OH were observed. Bare $\text{g-C}_3\text{N}_4$ provided only $0.7 \text{ } \mu\text{mol h}^{-1} \text{ g}^{-1}$ of hydrogen.

In the work of Han et al. [32] the MXene Ti_3C_2 served for the synthesis of $\text{C-TiO}_2/\text{g-C}_3\text{N}_4$ by the heating of a mixture of Ti_3C_2 and $\text{g-C}_3\text{N}_4$ at $450 \text{ }^\circ\text{C}$ for 4 h. In fact, Ti_3C_2 was designed of C-doped TiO_2 , forming a $\text{C-TiO}_2/\text{g-C}_3\text{N}_4$ photocatalyst. The hydrogen production rate was $1409 \text{ } \mu\text{mol h}^{-1} \text{ g}^{-1}$. The production using $\text{g-C}_3\text{N}_4$ was $174 \text{ } \mu\text{mol h}^{-1} \text{ g}^{-1}$ using TEOA for scavenging the holes.

Zhang et al. [33] synthesized rod-like $\text{g-C}_3\text{N}_4$ decorated by Mo_2C for the hydrogen evolution. The highest rate of $507 \text{ } \mu\text{mol h}^{-1} \text{ g}^{-1}$ was reached with an AQE of 3.74% (420 nm) using TEOA. The decorating Mo_2C co-catalysts on the surface of $\text{g-C}_3\text{N}_4$ led to an improved visible light absorption and promoted charge separation.

The 3D hollow spheres of Ti_3C_2 and $\text{g-C}_3\text{N}_4$ were synthesized by Kang et al. [34]. A 2D heterostructure shortened the electron migration distance and formed the Schottky

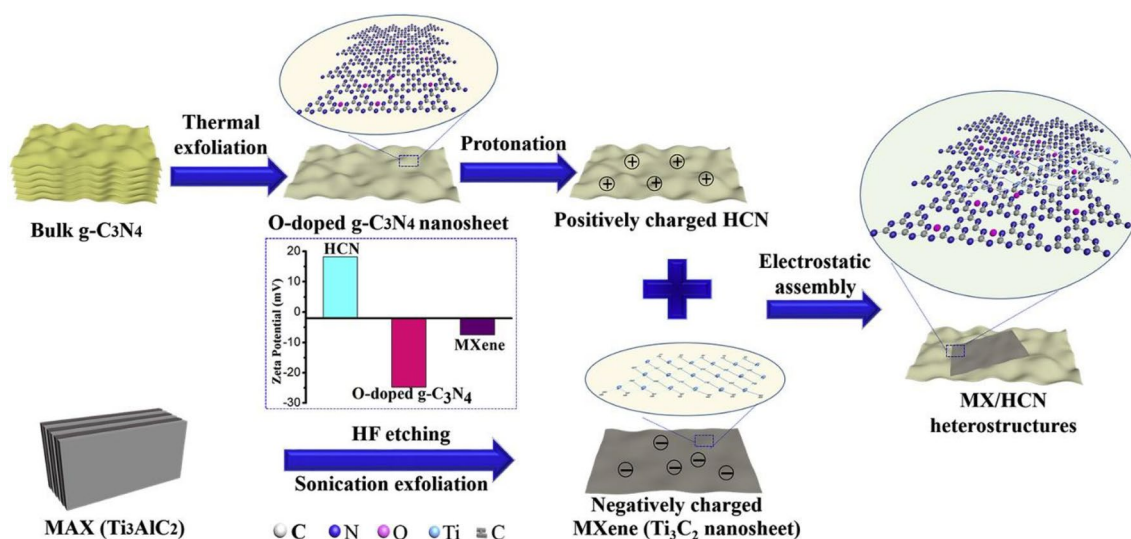


Fig. 2 The synthetic process of the 2D/2D $\text{Ti}_3\text{C}_2/\text{O-g-C}_3\text{N}_4$ Schottky-junction. Reproduced from Ref. [29]. Copyright (2019), with permission from Elsevier

junction to facilitate the separation and transfer of the charge carriers. The hydrogen production rate of the hollow spheres was $982.8 \mu\text{mol h}^{-1} \text{g}^{-1}$ while that of $\text{g-C}_3\text{N}_4$ was $1279.5 \mu\text{mol h}^{-1} \text{g}^{-1}$ using TEOA.

The in-situ fabrication of the 2D/3D $\text{g-C}_3\text{N}_4/\text{Ti}_3\text{C}_2$ heterojunction as the Schottky catalyst was referred by Li et al. [35]. A hydrogen evolution rate of $116.2 \mu\text{mol h}^{-1} \text{g}^{-1}$ (using TEOA) was reported. Using pure $\text{g-C}_3\text{N}_4$ the rate was $17.5 \mu\text{mol h}^{-1} \text{g}^{-1}$. The MXene -F termination was removed or replaced by terminal -O during the synthetic process. The experimental and theoretical studies confirmed that the ability of conductive Ti_3C_2 greatly restrained the electron–hole recombination on the $\text{g-C}_3\text{N}_4$ and numerous functional groups on the surface of Ti_3C_2 provided sufficient active sites for the HER.

Dong et al. [36] synthesised the 2D/2D $\text{g-C}_3\text{N}_4/\text{Ti}_3\text{C}_2$ composites providing the hydrogen evolution rate of $534 \mu\text{mol h}^{-1} \text{g}^{-1}$ in comparison with $\text{g-C}_3\text{N}_4$ providing the rate of $250 \mu\text{mol h}^{-1} \text{g}^{-1}$. The AQE was 1.61% using a 300 W Xe arc lamp equipped with a cut-off filter ($\lambda > 420 \text{ nm}$, 150 mW cm^{-2}). TEOA was used as the scavenger.

A 2D/2D/2D $\text{Bi}_2\text{WO}_6/\text{g-C}_3\text{N}_4/\text{Ti}_3\text{C}_2$ composite was synthesized by a one-step method [37]. The highest rate of $54.4 \mu\text{mol h}^{-1} \text{g}^{-1}$ was reached under a 300 W Xenon lamp and with a TEOA scavenger. The AQE was 1.6% at 420 nm. Ti_3C_2 was found not only to accelerate a Z-scheme charge transfer but also enhance the visible light absorption and redox ability of the $\text{Bi}_2\text{WO}_6/\text{g-C}_3\text{N}_4$ composite.

Crystalline carbon nitride was anchored on Ti_3C_2 as the ordered Schottky heterojunction photocatalyst for the enhanced visible-light hydrogen evolution [38]. Under the irradiation of 420 nm the hydrogen evolution rate was $4225 \mu\text{mol h}^{-1} \text{g}^{-1}$ and the AQE was 14.6% (TEOA). For comparison, the hydrogen rate in the presence of $\text{g-C}_3\text{N}_4$ was $513 \mu\text{mol h}^{-1} \text{g}^{-1}$. The photocatalytic enhancement was attributed to the synergistic effects of the crystallization of $\text{g-C}_3\text{N}_4$, the high conductivity of Ti_3C_2 and the well-constructed Schottky heterojunction.

An ultrathin Ti_3C_2 was applied to accelerate the charge transfer in an ultrathin metal-free 0D/2D black phosphorus/ $\text{g-C}_3\text{N}_4$ heterojunction for the hydrogen production [39]. The optimized composites exhibited the production rate of $18,420 \mu\text{mol h}^{-1} \text{g}^{-1}$ which was 47.2 times more than that of bare $\text{g-C}_3\text{N}_4$. The irradiation was produced by a 300 W Xe lamp equipped with a 420 nm cut-off filter.

The Schottky heterojunction of protonated $\text{g-C}_3\text{N}_4$ and Ti_3C_2 was constructed by Xu et al. [40]. Strong interfacial interactions highly improved the hydrogen evolution rate at $2181 \mu\text{mol h}^{-1} \text{g}^{-1}$ in comparison with bare $\text{g-C}_3\text{N}_4$ ($393 \mu\text{mol h}^{-1} \text{g}^{-1}$) and protonated $\text{g-C}_3\text{N}_4$ ($816 \mu\text{mol h}^{-1} \text{g}^{-1}$). The charge-regulated surfaces and the accelerated charge transport over the 2D/2D Schottky heterojunction interface are the main reasons for the higher

hydrogen evolution performance of the composite photocatalyst. The AQE at 420 nm was 8.6%. Isopropanol was used as the scavenger.

Hieu et al. [41] synthesized ternary $\text{TiO}_2/\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ heterojunction photocatalysts for the hydrogen evolution. The ternary composite was prepared by the annealing of $\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ in the air at 550 °C for 4 h. The highest evolution rate reached $1150 \mu\text{mol h}^{-1} \text{g}^{-1}$ while bare $\text{g-C}_3\text{N}_4$ provided $150 \mu\text{mol h}^{-1} \text{g}^{-1}$ (TEOA was used as the scavenger).

2D/2D $\text{Mo}_2\text{C}/\text{g-C}_3\text{N}_4$ composites were prepared by Liu et al. [42]. The hydrogen production rate of the photocatalyst at the optimal ratio was $675.27 \mu\text{mol g}^{-1} \text{h}^{-1}$. The enhanced photocatalytic activity was explained so that Mo_2C can rapidly transfer photoinduced electrons from $\text{g-C}_3\text{N}_4$ to its surface and thus prevent their recombination with the holes.

3D interconnected $\text{g-C}_3\text{N}_4$ hybridized with 2D Ti_3C_2 MXene nanosheets were synthesized by Liu et al. [43]. The MXene termination by -F and -O was identified. This provided a more negative Fermi level than $\text{g-C}_3\text{N}_4$ (about -0.22 V) which is favourable for the electrons to migrate to Ti_3C_2 . The HER performance of the composite was $1948 \mu\text{mol h}^{-1} \text{g}^{-1}$ compared with $118.1 \mu\text{mol h}^{-1} \text{g}^{-1}$ of pure $\text{g-C}_3\text{N}_4$ (with TEOA). The AQE at 420 nm was determined at 3.83%. The high specific surface area ($85 \text{ m}^2 \text{g}^{-1}$) and highly efficient charge migration led to the improved hydrogen generation.

In the work of Huang et al. [44], Pt nanoparticles were employed to modify few-layer Ti_3C_2 sheets which were further used as efficient co-catalysts to enhance the photocatalytic hydrogen evolution of $\text{g-C}_3\text{N}_4$. The Ti–O bonds were observed on the MXene surface. Using several sacrificial agents, such as methanol, TEOA, lactic acid, and $\text{Na}_2\text{S}/\text{Na}_2\text{SO}_3$, the maximal HER performance of $2644 \mu\text{mol h}^{-1} \text{g}^{-1}$ was achieved in the case of a 5% composite of Pt (with $\text{Na}_2\text{S}/\text{Na}_2\text{SO}_3$). The AQE values at 365 nm and 420 nm were 12.9% and 8.3%, respectively. When methanol was used the HER performance of $2308 \mu\text{mol h}^{-1} \text{g}^{-1}$ and $243 \mu\text{mol h}^{-1} \text{g}^{-1}$ were observed for the 5% Pt composites.

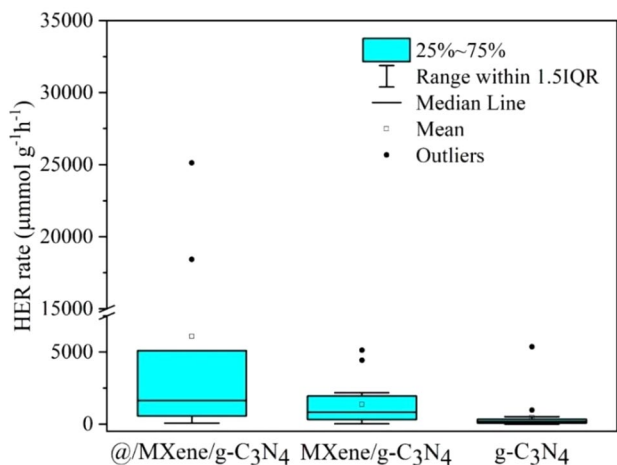
Phosphorus-doped $\text{g-C}_3\text{N}_4$ and Ti_3C_2 composites were tested for the HER [45] as well. The presence of -O terminations and TiO_2 was found. The highest achieved performance under the 300 W Xe lamp was $565 \mu\text{mol h}^{-1} \text{g}^{-1}$ in comparison with $107 \mu\text{mol h}^{-1} \text{g}^{-1}$ of $\text{g-C}_3\text{N}_4$ and $184 \mu\text{mol h}^{-1} \text{g}^{-1}$ of P-doped $\text{g-C}_3\text{N}_4$. The AQE at 420 nm was 5.9%. Methanol was used as the scavenger.

3.1 The quantum efficiency of HER

Quantum efficiency is an important factor of the photocatalytic HER. However, only little information is available in the literature. Only 15 articles reported these data. The basic summary is given in Table 1 where the quantum efficiency was expressed as the AQE. Since the AQE strongly

Table 1 AQE at 420 nm of all kinds of MXene and g-C₃N₄ composites

| Statistics | AQE (%) | Statistics | AQE (%) |
|--------------|---------|--------------------|---------|
| Number | 15 | Median | 3.83 |
| Minimum | 0.4 | 3rd Quartile | 6.22 |
| Maximum | 14.6 | Mean | 4.77 |
| 1st quartile | 2.36 | Standard deviation | 3.65 |

**Fig. 3** Box plots of the HER rate of @/MXene/g-C₃N₄ (left), MXene/g-C₃N₄ (centre), and g-C₃N₄ (right)

depends on the irradiation wavelength, only those measured at 420 nm are given here. Their values varied from 0.4 to 14.6%.

3.2 The performance of HER

All the above-mentioned articles are summarized in Table S1. When comparing the results of the photocatalytic reactions, it is possible to take into account that the same kinds of reactions (HER) were performed under different experimental conditions, such as the types of reactors and their geometry, the positions of the irradiation sources, irradiation intensity, photocatalyst concentrations, reaction temperatures, etc. These different experimental conditions can be visible when the photocatalytic rates obtained using the same photocatalyst, that is bare g-C₃N₄, are compared. The HER rates were visualised by the box and whisker lots as shown in Fig. 3.

The composites of MXene, g-C₃N₄ and other co-catalysts were labelled as @/MXene/g-C₃N₄. It is clearly visible that two HER rates are outlying. The basic descriptive statistics are given in Table S2.

The highest (outlying) HER rate of 25,124 μmol h⁻¹ g⁻¹ and 5366 μmol h⁻¹ g⁻¹ corresponding to the Ti₃C₂ and

O-doped g-C₃N₄ composite and g-C₃N₄, respectively were obtained by Lin et al. [29]. The next very high HER rates of 18,420 μmol h⁻¹ g⁻¹ and 950 μmol h⁻¹ g⁻¹ (also outlying) measured for the black phosphorus/Ti₃C₂/g-C₃N₄ composite and g-C₃N₄ were obtained by Song et al. [39]. The calculated medians of the HER rates of @/MXene/g-C₃N₄, MXenes/g-C₃N₄, and g-C₃N₄ were 1620 μmol h⁻¹ g⁻¹, 813 μmol h⁻¹ g⁻¹ and 162 μmol h⁻¹ g⁻¹. The Mann–Whitney (M–W), Kolmogorov–Smirnov (K–S) and Mood’s median (MM) confirmed the difference between MXenes/g-C₃N₄ and g-C₃N₄ (p_{MW} = 0.007, p_{KS} = 0.006, and p_{MM} 0.005) but the difference between @/MXene/g-C₃N₄ and MXenes/g-C₃N₄ was not confirmed (p_{MW} = 0.270, p_{KS} = 0.314, and p_{MM} = 0.147). All the reviewed articles referred to the significant improvement of the photocatalytic activity of g-C₃N₄ by adding the MXenes which was confirmed by this testing. However, adding some co-catalysts to MXenes/g-C₃N₄ does not lead to a further significant increase of the HER performance. It indicates that the surface (vacancy) and heterojunction engineering of the MXenes/g-C₃N₄ heterojunction is likely more important than the synthesis of the more complex @/MXene/g-C₃N₄ composites.

The HER performance was also compared with other works in which g-C₃N₄ was used for the fabrication of some composites, see Table 2. According to the used statistical tests these results are similar to those obtained by all the @/MXene/g-C₃N₄ and MXene/g-C₃N₄ composites (p_{MW} = 0.654, p_{KS} = 0.885, and p_{MM} = 0.740). In addition, the AQE values obtained with all the composites (@/MXene/g-C₃N₄ and MXene/g-C₃N₄) are also similar to those listed in Table 2 (p_{MW} = 0.184 and p_{KS} = 0.386, and p_{MM} = 0.273).

4 The degradation of the organic compounds

The photocatalytic degradation of the organic compounds is based on the direct reaction of the photoinduced holes with the organic molecules and on the reactions of the radicals formed by the various reaction mechanisms of the electrons and oxygen dissolved in an aqueous phase [61] as follows

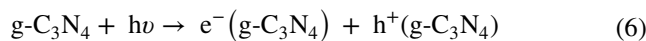
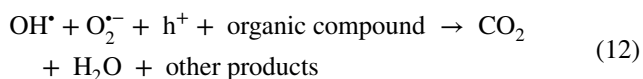


Table 2 The HER performance of the other g-C₃N₄ based photocatalysts

| Photocatalysts/statistics | Rate ($\mu\text{mol g}^{-1} \text{h}^{-1}$) | AQE (%) at 420 nm | Hole scavenger | References |
|--|---|-------------------|----------------|------------|
| CdS/g-C ₃ N ₄ | 3670 | 2.03 | Methanol | [46] |
| CeO ₂ /g-C ₃ N ₄ | 203.6 | 8.94 | TEOA | [47] |
| CoO/g-C ₃ N ₄ | 262.8 | 4.93 | Ethanol | [48] |
| Co ₃ (PO ₄) ₂ /g-C ₃ N ₄ | 375.6 | 1.32 | None | [49] |
| Fe ₂ O ₃ /g-C ₃ N ₄ | 38.2 | 2.1 | TEOA | [50] |
| Pt-CdS/ g-C ₃ N ₄ -MnO _x | 924.4 | 1.75 | None | [51] |
| N doped g-C ₃ N ₄ | 3579 | 27.8 | Lactic acid | [52] |
| Ni(OH) ₂ /g-C ₃ N ₄ | 21 | 2.78 | TEOA | [53] |
| ZnIn ₂ S ₄ /g-C ₃ N ₄ | 1650 | 18.2 | TEOA | [54] |
| CoS/ g-C ₃ N ₄ /NiS | 1930 | 16.4 | TEOA | [55] |
| 1TMoS ₂ /g-C ₃ N ₄ | 4426 | 13.55 | TEOA | [56] |
| NiS/g-C ₃ N ₄ | 1346 | 7.67 | TEOA | [57] |
| Ru-CoP/g-C ₃ N ₄ | 1173 | 3.49 | TEOA | [58] |
| Co(Mo-Mo ₂ C)/g-C ₃ N ₄ | 11.29 | 6.7 | TEOA | [59] |
| C ₃ N ₅ /g-C ₃ N ₄ | 3330 | 20.6 | TEOA | [60] |



The whole degradation reaction of an organic compound can be written as



Most of the reviewed articles evaluated the degradation of the organic compounds as the first-order reaction and in some cases reaction rate constants were published. One of the first articles on this topic reported Ag-decorated g-C₃N₄ and Ti₃C₂ ternary plasmonic photocatalysts for the degradation of aniline [62]. The highest degradation performance of 81.8% was reached after 8 h. The highest rate constant was 0.172 min⁻¹; the rate constant for g-C₃N₄ was 0.039 min⁻¹. The Ag clusters were prepared by photodeposition. The source of irradiation was a 300 W Xenon lamp with a 420 nm cut-off filter.

The composite Ti₃C₂/g-C₃N₄ was investigated for the degradation of ciprofloxacin under a light source with a 400 nm cut-off filter [63]. The ciprofloxacin (CIP) was completely decomposed after 150 min. The rate constant was 0.035 min⁻¹ for the composite and 0.016 min⁻¹ for pure g-C₃N₄.

The photocatalytic activity of a TiO₂@Ti₃C₂/g-C₃N₄ ternary heterostructure was studied by Ding et al. [64]. The O/OH-terminated Ti₃C₂ and the by-product TiO₂ could act as excellent supporters for migrating electrons. A photocatalysis was performed in terms of the degradation of aniline and Rhodamine B (RhB). After 8 h, 76.4%

of the aniline was removed. The majority (98.2%) of RhB was removed after 60 min. For this purpose, a 300 W Xe lamp (100 mW cm⁻²) was used.

Also, the nanotubes of the TiO₂@Ti₃C₂/g-C₃N₄ composites were prepared by Ti meshes by anodization, the chemical vapor deposition method, and in-situ growth. They were used for the degradation of RhB and tetracycline (TC) with the degradation rates of 0.0370 min⁻¹ and 0.0091 min⁻¹, respectively [65]. 96.04% of RhB was removed after 90 min and 85.12% of the tetracycline was removed after 3 h.

He et al. [66] referred to using the composite of the 2D/2D heterostructure of Ti₃C₂/g-C₃N₄ to activate peroxy-monosulfate for the removal of diclofenac. The optimal degradation rate of 0.21 min⁻¹ was achieved when pure g-C₃N₄ nanosheets provided a rate of 0.071 min⁻¹. Due to the synergistic effect of the photocatalysis and peroxy-monosulfate, ¹O₂ was generated as a primary reactive species. Almost 95% of diclofenac was degraded in 30 min. The photocatalytic experiments were performed under a 300W Xe lamp simulating solar irradiation.

Dong et al. [36] synthesized the 2D/2D Ti₃C₂/g-C₃N₄ composites in a semi-closed system and tested for the degradation of tetracycline hydrochloride. The highest degradation performance reached 84.4% during 120 min while single g-C₃N₄ reached only 29.0%. The largest rate constant was 0.0141 min⁻¹ for the composite and 0.0028 min⁻¹ for g-C₃N₄.

Wu et al. [37] fabricated the 2D/2D/2D Bi₂WO₆/g-C₃N₄/Ti₃C₂ composites. The well-matched band structure favours the formation of a Z-scheme heterojunction. This composite was used for the photocatalytic degradation of CIP. The rate

constant was 0.058 min^{-1} ; 87.7% of the CIP was degraded within 70 min.

$\text{TiO}_2/\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ heterojunction photocatalysts were also fabricated by Luo et al. [67]. The degradation of methylene orange achieved the maximal value of 93.1%. The highest rate constant was 0.02095 min^{-1} in comparison with 0.00578 min^{-1} of pure $\text{g-C}_3\text{N}_4$. The degradation of the best photocatalyst was also confirmed for the degradation of RhB (90.47%) and methylene blue (MB) (91.14%).

$\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ composites were prepared by the intercalation of the Ti_3C_2 layers into $\text{g-C}_3\text{N}_4$ layers to obtain an efficient heterojunction. The photocatalysts were used for the oxidation of pyridine and thiophene for fuels [68]. Dissolved oxygen (from the atmosphere) formed superoxide radicals and assisted in the oxidation of pyridine to nitrate and thiophene to sulphate. The band gap of Ti_3C_2 was determined at 1.09 eV.

A 2D/2D $\text{Ti}_3\text{C}_2/\text{porous g-C}_3\text{N}_4$ van der Waals heterostructure photocatalyst was synthesized by Liu et al. [69]. A porous $\text{g-C}_3\text{N}_4$ was prepared by a hard template method. Packed silica spheres (50 nm) were employed as a templating agent. It was showed that a 98% phenol removal during the day and even a 32% degradation during the night was reached which was explained by the storage of photoinduced electrons and holes under solar irradiation. It was simulated by a 500 W Xenon lamp ($\lambda > 400 \text{ nm}$).

Cationic dyes MB and RhB were photodegraded in the presence of hybrid chitosan and $\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ nanosheets [70]. The presence of the $-\text{OH}$ terminal groups of Ti_3C_2 was confirmed. The $-\text{OH}$ termination, providing high conductivity and outstanding hydrophilicity, is highly efficacious in the removal of organic molecules. The degradation efficiency was 99.1% for MB in 60 min and 98.5% for RhB in 40 min. The degradation pathways were studied in detail. The photocatalytic experiments were performed under a 250 W Xe-lamp (400–800 nm). The MB and RhB results were verified by the degradation of other dyes, such as Methyl Orange, Malachite Green, and Orange Green.

Embedding few-layer Ti_3C_2 into alkalinized $\text{g-C}_3\text{N}_4$ nanosheets was reported by Yi et al. [71]. The photocatalytic degradation was tested on tetracycline with an efficiency of 77% during 60 min. The amine groups in $\text{g-C}_3\text{N}_4$ were supposed to interact with the carbon layers and the $-\text{OH}$ groups of Ti_3C_2 , building a van der Waals heterostructure between the two constituents of the composite. Moreover, AgNO_3 was added into the reaction mixture. Electrons trapped with the MXene possibly reduced the Ag^+ ions to Ag particles forming another Schottky junction that suppresses the charge recombination process and slightly improved the photocatalytic degradation.

A high-energy ball milling technology was applied for a one-step synthesis of the $\text{TiO}_2/\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ composites

for the visible light degradation of Methyl Orange [72]. This ternary composite exhibited a 90.55% performance after 120 min. Ti_3C_2 acted as a transport medium for this Z-type heterojunction structure. TiO_2 promoted the separation of the photocatalytic carriers and thus the photocatalytic reaction.

Samarium-doped $\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ was synthesized by the prepolymerization of $\text{g-C}_3\text{N}_4$ and the solid mixture calcination by Yu et al. [73]. The degradation of CIP under visible irradiation achieved > 99% within 60 min. The Sm-doping was supposed to improve the transfer of photoinduced electrons and Ti_3C_2 broadened the light absorption and improved the charge carrier migration efficiency.

The already mentioned work of Liu et al. [43] about the $\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ nanosheets for the HER was also tested for the degradation of RhB. The highest degradation rate was 0.194 min^{-1} and with a degradation efficiency of 78.5%.

The $\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ nanosheets were tested by Liu et al. [74] for their photocatalytic redox capacity. The degradation of levofloxacin reached 72% using a composite with a 1 wt% of Ti_3C_2 . A photocatalytic improvement was achieved by the faster separation of the photoinduced electrons and holes, the higher light response ability, and the more active sites. The successful formation of the 2D/2D Schottky junction and the Ti–N bonds between $\text{g-C}_3\text{N}_4$ and Ti_3C_2 were observed.

Atrazine was phototactically degraded by using a $\pi-\pi$ stacked pyromellitic dimide (PDI) $\text{TiO}_2/\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ photocatalyst [75]. The degradation was activated by peroxy-monosulphate. The $\pi-\pi$ interaction in PDI/ $\text{g-C}_3\text{N}_4$ induced the delocalization of the photoelectrons and thus promoted their migration. The highest degradation efficiency reached 75%. The degradation was performed under visible irradiation (the Xe lamp) with a 420 nm cut-off filter.

The tetracycline, ciprofloxacin, bisphenol A (BPA), and RhB degradation under visible light irradiation was performed in the presence of graphene layers anchored on $\text{TiO}_2/\text{g-C}_3\text{N}_4$ which was synthesized from Ti_3C_2 by a one-step in-situ calcination [76]. The obtained degradation rates were 0.02442 min^{-1} for TC, 0.01675 min^{-1} for CIP, 0.01935 min^{-1} for BPA, and 0.05586 min^{-1} for RhB. The best removal efficiencies were 83.5% (TC in 80 min), 61.7% (CIP in 60 min), 79.5% (BPA in 70 min), and 98.0% (RhB in 50 min).

Tetracycline together with dyes, such as Congo red (CR) and Trypan blue (TB), were removed by composite membranes made of $\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ and polyethersulphone [77]. The presence of $-\text{F}$ and $-\text{O}$ terminations was confirmed. The membranes were irradiated by a 70 W golden lamp for 8 h. The achieved removal efficiency was 98% for CR, 96% for TB, and 86% for TC.

The fabrication of the carbonized cellulose nanofibrils/ $\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ was reported by Zu et al. [78]. This

heterojunction composite was then applied for the photocatalytic degradation of MB, RhB, and TC with an efficiency 96.5%, 95.4%, and 86.5% respectively. The irradiation source was a 300 W Xe arc lamp with a 420 nm filter. The photocatalytic experiments took 210 min.

The $\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ Schottky junction photocatalyst was prepared to be used for the degradation of arbidol hydrochloride (ABLH) as a drug against COVID-19. Ti_3C_2 was terminated by the $-\text{F}$ and $-\text{O}$ species. The composite containing 0.5% of Ti_3C_2 was able to remove 99.2% of the ABLH during 150 min under visible irradiation (the 300 W Xe lamp). The highest rate constant of the composite was 0.02959 min^{-1} while that of $\text{g-C}_3\text{N}_4$ was 0.01111 min^{-1} .

Graphitic carbon nitride coupled with the Ti_3C_2 derived amorphous Ti-peroxo heterojunction for the photocatalytic degradation of rhodamine B and tetracycline was prepared by Tu et al. [79]. The RhB highest rate constant of the composite containing 35% of $\text{g-C}_3\text{N}_4$ was measured at 0.051 min^{-1} and that for bare $\text{g-C}_3\text{N}_4$ was 0.0029 min^{-1} using a 270 W Xe lamp ($\lambda > 420 \text{ nm}$). In the case of TC, the best performing composite containing 25% of $\text{g-C}_3\text{N}_4$ demonstrated a rate constant of 0.0208 min^{-1} . A constant of 0.0003 min^{-1} was found for $\text{g-C}_3\text{N}_4$.

Zhou et al. [80] synthesized the 2D/2D $\text{Mo}_2\text{C}/\text{g-C}_3\text{N}_4$ Van der Waals heterojunction composites. Their photocatalytic performance was tested by the degradation of TC. The highest rate constant of 0.066 min^{-1} was obtained for the composite with 2% of Mo_2C while the constant of bare $\text{g-C}_3\text{N}_4$ was 0.017 min^{-1} . The Mo_2C enhanced the light absorption capacity and served as an electron trap to elongate the lifetime of the charge carriers.

Methylene blue was also degraded by $\text{g-C}_3\text{N}_4$ on which the Ti_3C_2 particles were loaded as reported by Nasri et al. [81]. A 1 wt% MXene/ $\text{g-C}_3\text{N}_4$ heterostructure photocatalyst achieved the 60% degradation of methylene blue during 180 min. A 500 W halogen lamp was used as the irradiation source. The band gap of this most active photocatalyst was 2.53 eV.

$\text{BiOBr}/\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ Z-scheme heterostructure photocatalysts were tested for the TC degradation [82]. The Xe lamp with 400–800 nm cut-off filters was applied for this purpose. TC was completely removed after 30 min and the best rate constant was 0.204 min^{-1} . The optimal BiOBr and $\text{g-C}_3\text{N}_4$ composite was investigated. The proposed mechanism of the charge transfer is demonstrated in Fig. 4.

A novel $\text{MnFe}_2\text{O}_4/\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ composite was fabricated by Hou et al. [83] and used for the peroxymonosulfate-assisted photocatalytic degradation of naphthalene. Almost 100% was removed during 45 min. The highest rate constant of 0.09984 min^{-1} was achieved. There existed both free radical and non-radical pathways for the naphthalene degradation, in which singlet oxygen was identified as the main reactive oxygen species as a result of the Mn–Fe

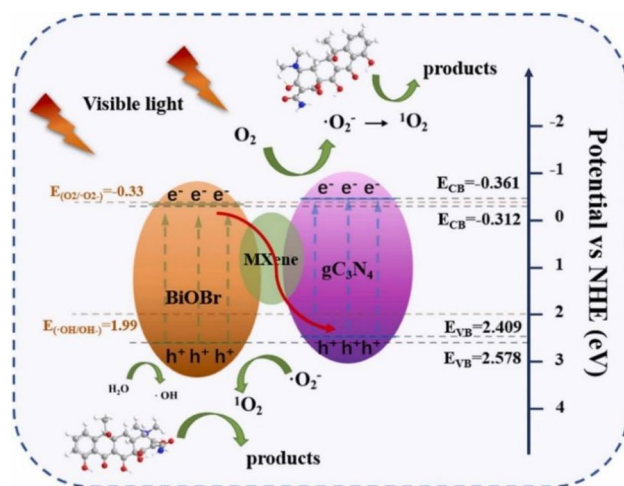


Fig. 4 The proposed mechanism of the charge transfer and separation on the surface of $\text{BiOBr}/\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$ for TC degradation under visible light illumination. Reproduced from Ref. [82]. Copyright (2023), with permission from Elsevier

valence transformation process. The degradation pathway of naphthalene was studied.

4.1 The performance of the organic compound degradation

The degradation efficiency of the organic compounds strongly depends on their types (their chemical structures) and on experimental conditions. The articles included in this work refer to the degradation of various dyes, pharmaceutical products, aniline, naphthalene etc. In general, the decomposition reactions are mostly described by the first-order equation as

$$\frac{c}{c_0} = e^{-kt} \quad (13)$$

where t is time, k is the rate constant, c and c_0 are concentrations at $t=t$ and $t=0$, respectively.

A half-life time, which is the time taken for a given concentration to decrease to half of its value, was used as a measure of how the degradation performance of $\text{BiOBr}/\text{Ti}_3\text{C}_2/\text{g-C}_3\text{N}_4$, MXenes/ $\text{g-C}_3\text{N}_4$, and $\text{g-C}_3\text{N}_4$ improved. For the half-life time, it is valid that

$$t_{1/2} = \frac{\ln 2}{k} \quad (14)$$

The half-life time of the first-order reaction remains constant throughout the reaction and is independent of the concentrations. In order to compare the photocatalytic degradation of the various organic compounds at various concentrations, the ratios (r) of the half-times for the individual

organic compounds decomposed in the presence of the MXene based photocatalysts (2) and g-C₃N₄ (1) were calculated as follows

$$r = \frac{t_{1/2}(1)}{t_{1/2}(2)} = \frac{k(2)}{k(1)} \quad (15)$$

The kinetic constants were taken from the reviewed articles if they were published. The ratio is supposed to be dependent on the catalysts used, not on the experimental conditions. The ratios were statistically processed by the already used statistics.

The box and whisker plots are shown in Fig. 5. The basic statistical characteristics were calculated, see Table S3. The outlying ratio of 69.3 (Fig. 5) corresponds to the degradation of tetracycline using the composite of g-C₃N₄ and Ti₃C₂ with the Ti-peroxo heterojunction [79]. The medians were 3.369 and 2.425 for @/MXene/g-C₃N₄ and MXenes/g-C₃N₄ respectively.

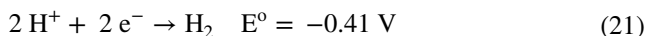
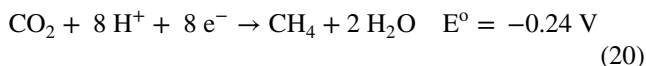
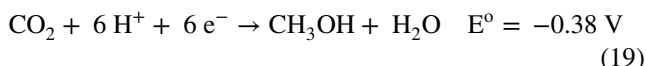
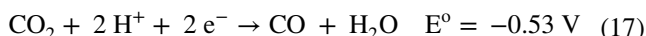
These median values (higher than 2 and 3) tell us that the photocatalytic activity of g-C₃N₄ was highly improved by adding the MXenes. On the contrary, it was found that some co-catalysts added to MXene/g-C₃N₄ do not improve its photocatalytic degradation ($p_{MW} = 0.751$, $p_{KS} = 0.869$, and $p_{MM} = 0.414$) which means the surface and heterojunction engineering are more important.

A comparison with other photocatalysts based on g-C₃N₄ was not performed because the degradation efficiency depends on the individual organic compounds as already mentioned. Their redox potentials have different positions toward the valence and the conduction bands of g-C₃N₄ which means that the photoinduced holes are not always

involved in the photocatalytic reactions, see Eq. (12). In addition, the organic compounds differ in their adsorption properties to the photocatalysts.

5 The reduction of carbon dioxide

MXenes have been also used as co-catalysts for the photocatalytic reduction of carbon dioxide. Some redox reactions (versus NHE at pH = 7) are summarized below [84]:



The TiO₂/Ti₃C₂/g-C₃N₄ S-scheme photocatalysts were constructed by He et al. [85]. Ti₃C₂ MXene quantum dots were deposited on the 2D/2D van der Waals heterojunction of TiO₂ and g-C₃N₄. 2D mesoporous TiO₂ nanosheets were synthesized by a hydrothermal induced solvent-confined monomicelle self-assembly. Reduction products, such as CO and CH₄, reached their maxima at 4.39 and 1.20 μmol g⁻¹ h⁻¹. The S-scheme heterojunction at the TiO₂/g-C₃N₄ interface and the Schottky heterojunction at the g-C₃N₄ and the quantum dots interface mostly affected the photocatalytic activity.

Tahir and Tahir [86] constructed a 2D/2D/2D O-C₃N₄/bentonite/Ti₃C₂ heterojunction composite. The bentonite/Ti₃C₂ composite demonstrated high light absorption and fast charge separation and transportation. The CO₂ reduction to CH₄ and CO was performed under a 35 W HID lamp with a maximum intensity at 420 nm (20 mW cm⁻²). The CO evolution rate was 365 μmol g⁻¹ h⁻¹ (76 μmol g⁻¹ h⁻¹ with pure g-C₃N₄) and the CH₄ one was 955 μmol g⁻¹ h⁻¹ (33 μmol g⁻¹ h⁻¹ with g-C₃N₄). The AQE values were measured at 1.203% for CO and 3.146% for CH₄ (Fig. 6).

The photocatalytic reduction of carbon dioxide to carbon monoxide and methane was investigated by Tang et al. [87] using g-C₃N₄ decorated with alkalized Ti₃C₂. The highest production rates were 11.21 μmol g⁻¹ h⁻¹ for CO and 0.269 μmol g⁻¹ h⁻¹ for CH₄. The AQE values were estimated at 0.0099% for both CO and CH₄ (at 420 nm). The optimal content of Ti₃C₂ was 5 wt%. The adsorption of CO₂

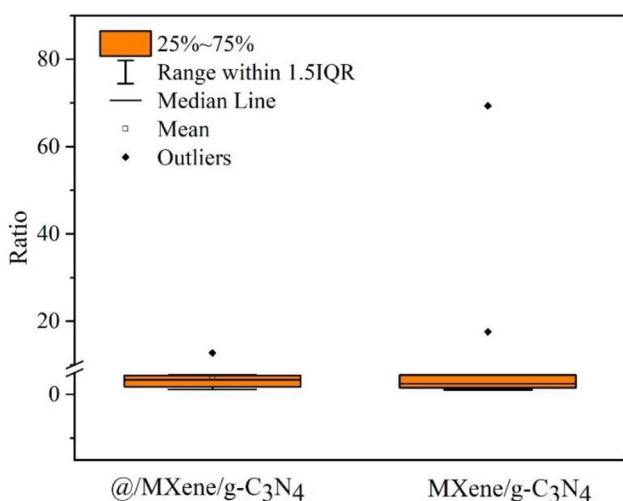


Fig. 5 Box plots of ratios of the half-life times of organic compounds photocatalytically degraded using @/MXene/g-C₃N₄ and MXene/g-C₃N₄ composites

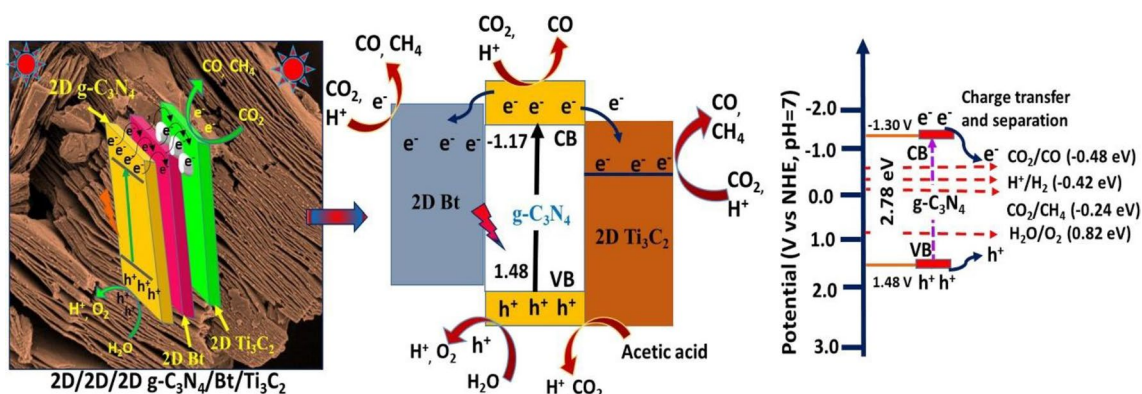


Fig. 6 Proposed mechanism for the photocatalytic CO₂ conversion to CO and CH₄ over the g-C₃N₄/bentonite/Ti₃C₂ heterojunction system under visible light. Reproduced from Ref. [86]. Copyright (2020), with permission from Elsevier

was attributed to the Ti₃C₂ surface modified with the -OH groups.

The Ti₃C₂/g-C₃N₄ heterojunction with a double role of urea was synthesized by Yang et al. [88]. The urea was used for the synthesis of g-C₃N₄ and also for the exfoliation of Ti₃C₂. The optimal sample (10 wt% of Ti₃C₂) possessed the best photocatalytic performance with the rates of 5.19 μmol h⁻¹ g⁻¹ for CO and 0.044 μmol h⁻¹ g⁻¹ for CH₄. The -F terminal groups were fully substituted with -OH and -O ones which was beneficial for Ti₃C₂ to trap the photoinduced electrons.

Similar Ti₃C₂/g-C₃N₄ photocatalysts were fabricated by Hu et al. [89]. The Ti₃C₂ was anchored on g-C₃N₄ via NH_x-Ti bonds. The optimized materials containing 2 wt% of Ti₃C₂ exhibited a CH₄ production rate of 0.99 μmol h⁻¹ g⁻¹ under visible irradiation (λ > 420 nm). This high performance was mainly attributed to the synergistic effects of the close interfacial interaction, the Schottky junction, and the remarkable conductivity of Ti₃C₂.

The ultrathin S-scheme heterojunction photocatalyst, based on few-layer g-C₃N₄ and monolayer Ti₃C₂, was constructed by Yang et al. [90]. An ultrathin heterojunction g-C₃N₄/TiO₂/C was synthesized through electrostatic self-assembly and calcination. CO and CH₄ were the main products of the CO₂ reduction. The highest achieved reduction rates using this kind of photocatalyst were 25.96 μmol h⁻¹ g⁻¹ for CO and 3.70 μmol h⁻¹ g⁻¹ for CH₄. The CO₂ reduction was performed in an airtight reactor with a certain amount of water under Xe arc lamp irradiation.

The already mentioned authors, Liu et al. [74], also tested their Ti₃C₂/g-C₃N₄ composite for the reduction of CO₂. The highest rate for the CO evolution was 10.67 μmol h⁻¹ g⁻¹ and that for the CH₄ evolution was 2.64 μmol h⁻¹ g⁻¹. The 2D/2D Schottky junction and a new Ti-N bond between g-C₃N₄ and Ti₃C₂ were formed.

Boron-doped g-C₃N₄ was combined with few-layer Ti₃C₂ by electrostatic self-assembly and used for the CO₂ reduction under visible irradiation (a 300 W Xe lamp with λ > 420 nm, 175 mW cm⁻²) [91]. The observed products were carbon monoxide, methane, and hydrogen. The highest CO₂ reduction yields were 14.4 and 0.80 μmol h⁻¹ g⁻¹ for CO and CH₄, respectively. The AQE for CO and CH₄ was measured to be 0.0117% at 420 nm.

A 2D/2D Ti₃C₂T_{A/R} MXene coupled g-C₃N₄ heterojunction with the in-situ growth of anatase/rutile TiO₂ nucleates was developed by Khan and Tahir [92]. Etching by high concentrated HF enabled the better conversion of Ti₃C₂ to TiO₂. The composite provided the hydrogen and CO rates of 51.24 and 87.34 μmol h⁻¹ g⁻¹, respectively. The photoreactor was equipped with a 35 W Xe lamp with the wavelength of 420 nm (20 mW cm⁻²). The highest AQE values were 0.732% for CO and 0.430% for H₂.

Mesoporous Ti₃C₂/g-C₃N₄ photocatalysts were fabricated by Li et al. [93] for the CO₂ reduction. Mesoporous structures possess abundant absorption sites for the CO₂ molecules and reduces the recombination of the electrons and holes. The methane production rate was 2.117 μmol h⁻¹ g⁻¹, which was 2.4 times more than the bare mesoporous g-C₃N₄. The CO rate was 3.98 μmol h⁻¹ g⁻¹ (all with a 300 W Xe lamp).

A 2D Ti₃C₂ decorated Z-scheme BiOIO₃/g-C₃N₄ heterojunction was fabricated by Hong et al. [94] and used for the CO₂ reduction. The optimal photocatalytic efficiency of 5.88 and 1.55 μmol h⁻¹ g⁻¹ was reached for CO and CH₄, respectively, under the visible irradiation of the 300 W Xe lamp with a cut-off filter for 420 nm.

The composite consisting of 2D V₂C nanosheets coupled 2D with porous g-C₃N₄ was designed by Madi et al. [95]. The highest performance of 37.75 and 51.25 μmol h⁻¹ g⁻¹ of CO and CH₄, respectively, was reached with the composite containing 15% of V₂C. These values are 6.7 and 1.3 times

higher than using bare $g\text{-C}_3\text{N}_4$. An irradiation source was a 35 W HID Xe lamp. (20 mW cm^{-2}).

Carbon vacancy-mediated exciton dissociation in the $\text{Ti}_3\text{C}_2/g\text{-C}_3\text{N}_4$ Schottky heterojunction photocatalysts was synthesized by Song et al. [96]. The synergy between the vacancy engineering and the Schottky junction was studied. The optimal photocatalyst contained 20% of Ti_3C_2 . Graphitic carbon nitride with C vacancies was prepared for this purpose. The highest CO yield was $20.54 \mu\text{mol h}^{-1} \text{g}^{-1}$ which was 7.3 times higher than that of bare $g\text{-C}_3\text{N}_4$. The 300 W Xe arc lamp was equipped with a 400 nm cut-off filter.

A novel hybrid $g\text{-C}_3\text{N}_4/\text{ZnO}/\text{Ti}_3\text{C}_2$ was recently synthesized by Li et al. [97]. In the interface of $g\text{-C}_3\text{N}_4/\text{ZnO}$, $g\text{-C}_3\text{N}_4/\text{Ti}_3\text{C}_2$ and $\text{ZnO}/\text{Ti}_3\text{C}_2$, an internal electric field was generated. The electron transfer directions were from $g\text{-C}_3\text{N}_4$ to ZnO, from ZnO to Ti_3C_2 and from $g\text{-C}_3\text{N}_4$ to Ti_3C_2 . Due to the metallic properties of Ti_3C_2 (electrons migrate to Ti_3C_2 more easily), the CO production rate was increased up to $6.41 \mu\text{mol h}^{-1} \text{g}^{-1}$ and the CH_4 rate up to $0.27 \mu\text{mol h}^{-1} \text{g}^{-1}$. The AQEs were measured at 0.22% for CO and 0.06% for CH_4 at 350 nm.

5.1 The performance of the CO_2 reduction

All the above-mentioned information is summarized in Table S4. The CO results were analysed by the box and whisker plots and the highest (outlying) values were the CO rates of $365 \mu\text{mol h}^{-1} \text{g}^{-1}$ for $\text{O-C}_3\text{N}_4/\text{bentonite}/\text{Ti}_3\text{C}_2$ and $76 \mu\text{mol h}^{-1} \text{g}^{-1}$ for $g\text{-C}_3\text{N}_4$ [86], see Fig. 7.

The calculated medians of the CO rates of $@/\text{Ti}_3\text{C}_2/g\text{-C}_3\text{N}_4$, $\text{Ti}_3\text{C}_2/g\text{-C}_3\text{N}_4$, $g\text{-C}_3\text{N}_4$ were $5.88 \mu\text{mol h}^{-1} \text{g}^{-1}$, $9.93 \mu\text{mol h}^{-1} \text{g}^{-1}$ and $1.69 \mu\text{mol h}^{-1} \text{g}^{-1}$. For other statistics see Table S5. A significant improvement of photocatalytic activity by adding MXenes to $g\text{-C}_3\text{N}_4$ is obvious

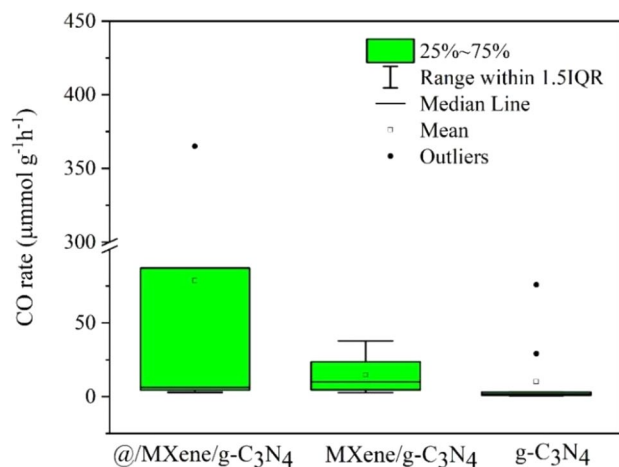


Fig. 7 Box plots of the CO rate of $@/\text{MXene}_2/g\text{-C}_3\text{N}_4$ (left), $\text{MXene}/g\text{-C}_3\text{N}_4$ (centre), and $g\text{-C}_3\text{N}_4$ (right)

($p_{\text{MW}}=0.023$ and $p_{\text{KS}}=0.009$, and $p_{\text{MM}}=0.006$). Comparing the $@/\text{MXene}/g\text{-C}_3\text{N}_4$ and $\text{MXene}/g\text{-C}_3\text{N}_4$ data by the statistical tests ($p_{\text{MW}}=0.948$, $p_{\text{KS}}=0.784$, and $p_{\text{MM}}=0.280$) shows that no difference between these two datasets was found which means that the added co-catalysts did not play an important role in the photocatalytic performance.

The same analysis was performed for the CH_4 production rate. The highest (outlying) value of $955 \mu\text{mol h}^{-1} \text{g}^{-1}$ obtained using the $\text{O-C}_3\text{N}_4/\text{bentonite}/\text{Ti}_3\text{C}_2$ complex was detected [86]. The calculated medians of the CH_4 rates measured in the presence of the $@/\text{MXene}/g\text{-C}_3\text{N}_4$, $\text{MXene}/g\text{-C}_3\text{N}_4$, $g\text{-C}_3\text{N}_4$ composites were $1.20 \mu\text{mol h}^{-1} \text{g}^{-1}$, $0.990 \mu\text{mol h}^{-1} \text{g}^{-1}$ and $0.200 \mu\text{mol h}^{-1} \text{g}^{-1}$. Other statistic parameters are shown in Table S6. The already used tests ($p_{\text{MW}}=0.871$, $p_{\text{KS}}=0.997$, and $p_{\text{MM}}=0.558$) indicated that the CH_4 rates corresponding to the $@/\text{MXene}/g\text{-C}_3\text{N}_4$ and $\text{MXene}/g\text{-C}_3\text{N}_4$ composites were not found to be statistically different. Similarly, as in the CO production, the added co-catalysts did not influence the CH_4 production.

However, the adding of MXene to $g\text{-C}_3\text{N}_4$ did not show the important photocatalytic effect ($p_{\text{MW}}=0.174$ and $p_{\text{KS}}=0.294$, and $p_{\text{MM}}=0.147$). Based on the used nonparametric statistical testing results, the reduction of CO_2 to CH_4 was not found to be significantly affected by the heterojunction of $g\text{-C}_3\text{N}_4$ with MXenes.

The CO_2 reduction performance was compared with other works using some $g\text{-C}_3\text{N}_4$ based composites. The data in Table 3 indicate that concerning all the $@/\text{MXene}/g\text{-C}_3\text{N}_4$ and $\text{MXene}/g\text{-C}_3\text{N}_4$ composites the CO rates are comparable with the others based on $g\text{-C}_3\text{N}_4$ ($p_{\text{MW}}=0.234$, $p_{\text{KS}}=0.289$, and $p_{\text{MM}}=0.180$). In the case of CH_4 , the statistical testing confirmed that the MXene and $g\text{-C}_3\text{N}_4$ composites provided different (lower) performance than the other $g\text{-C}_3\text{N}_4$ based composites ($p_{\text{MW}}=0.011$ and $p_{\text{KS}}=0.003$, $p_{\text{MM}}=0.001$), see Figure S1. The AQE values were not compared with those in Table 3 because only four articles reported these data.

The findings concerning the CO_2 reduction to CH_4 can be explained by the low adsorption of CO on the MXenes. Since the reduction of CO_2 takes place in a step-wise process [113], after its reduction to CO this oxide is mostly released [114] and its further reduction to CH_4 is limited. It also explains the lower rates of CH_4 in comparison with the rates of CO, see Figs. 7 and 8.

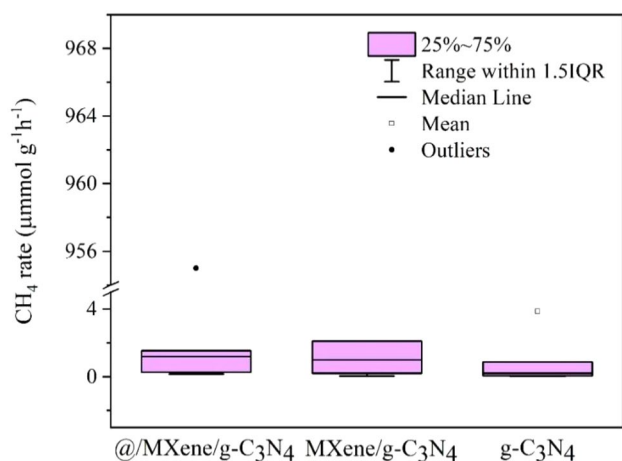
6 The redox reactions of nitrogen

6.1 Nitrogen reduction reaction

The nitrogen reduction reaction (NRR) to ammonia performed by MXene-derived $\text{TiO}_2@C/g\text{-C}_3\text{N}_4$ was reported by Liu et al. [115]. This composite was prepared via a facile one step calcination of a mixture of Ti_3C_2 and melamine.

Table 3 CO₂ reduction performance of some g-C₃N₄ based photocatalysts

| Photocatalysts/statistics | Rate CO ($\mu\text{mol g}^{-1} \text{h}^{-1}$) | Rate CH ₄ ($\mu\text{mol g}^{-1} \text{h}^{-1}$) | AQE (%) at 420 nm | References |
|--|--|---|-------------------|------------|
| NiS ₂ /g-C ₃ N ₄ | 10.68 | | 2.03 | [98] |
| SnO ₂ /B-P-g-C ₃ N ₄ | | 30.6 | 2.02 | [99] |
| Phosphorylated/g-C ₃ N ₄ | 7.7 | 14.6 | 0.86 | [100] |
| Eu(III)/g-C ₃ N ₄ | | 3.8 | 1.6 | [101] |
| Ag/g-C ₃ N ₄ | 179.6 | | 0.25 | [102] |
| WO ₃ /g-C ₃ N ₄ | 48.7 | | 2.21 | [103] |
| Au/TiO ₂ /g-C ₃ N ₄ | 21.7 | 37.4 | 1.91 | [104] |
| NiAl-LDH/rGO/g-C ₃ N ₄ | 2.6 | 20 | 0.99 | [105] |
| TiO ₂ /C/Pt/g-C ₃ N ₄ | 14.7 | 65.6 | 5.67 | [106] |
| FeV ₂ O ₄ /g-C ₃ N ₄ | 5.79 | | 5.79 | [107] |
| BiO _{(1-x)Br} /g-C ₃ N ₄ | 13.11 | | 0.098 | [108] |
| Nb ₂ O ₅ /g-C ₃ N ₄ | 253.34 | 68.11 | 4.95 | [109] |
| Bi ₃ O ₄ Cl/g-C ₃ N ₄ | 6.6 | 1.9 | 0.68 | [110] |
| Bi ₂ O ₃ /Bi ₂ WO ₆ /g-C ₃ N ₄ | 29.3 | | 1.3 | [111] |
| Bi ₂ S ₃ /g-C ₃ N ₄ | 54.74 | 12.52 | 2.31 | [112] |

**Fig. 8** Box plots of the CH₄ rate of @/MXene/g-C₃N₄ (left), MXene/g-C₃N₄ (centre), and g-C₃N₄ (right)

The formation of C–Ti–O bonds was confirmed. The NH₃ production rate of 250.6 $\mu\text{mol h}^{-1} \text{g}^{-1}$ was achieved under visible irradiation (the 300 W Xe lamp, $\lambda > 420 \text{ nm}$). Methanol was used as the scavenging reagent. The AQE was 0.14% at 420 nm.

Nb₂O₅/C/Nb₂C/g-C₃N₄ heterojunction photocatalysts were constructed by Jiang et al. [116]. The photocatalysts were prepared by growing Nb₂O₅ on Nb₂C and then forming g-C₃N₄ nanosheets in situ on Nb₂O₅/C/Nb₂C. The optimized photocatalyst with a Nb₂O₅/C/Nb₂C g-C₃N₄ ratio of 1:1 exhibited the NRR rate of 365 $\mu\text{mol h}^{-1} \text{g}^{-1}$. With the optimized pH 9 adjusted by a NaOH solution, the NRR rate increased to 927 $\mu\text{mol h}^{-1} \text{g}^{-1}$. Methanol was used as the scavenger as well.

The already mentioned work of Liu et al. [74] about the Ti₃C₂/g-C₃N₄ nanosheets also reported the production of NH₃ with the rate of 601 $\mu\text{mol L}^{-1} \text{h}^{-1} \text{g}^{-1}$, which is 3.64 times higher than that of g-C₃N₄ (203 $\mu\text{mol L}^{-1} \text{h}^{-1} \text{g}^{-1}$).

Sun et al. [117] synthesized a 2D/2D Ti₃C₂/N-defect g-C₃N₄ heterostructure composite for the NRR. The heterostructure was constructed by filling the -O terminating groups of Ti₃C₂ in the N-defects of g-C₃N₄ forming C-O-Ti interactions. The defects were prepared by the acid-assist thermal polymerization reaction of melamine and citric acid. The highest reported reaction rate was 341 $\mu\text{mol h}^{-1} \text{g}^{-1}$.

The Schottky junction photocatalysts were synthesized from hollow g-C₃N₄ decorated with the partly reduced quantum dots of Ti₃C₂ [118]. The surface of Ti₃C₂ was rich in Ti³⁺ sites and oxygen vacancies which facilitated the capture and activation of the nitrogen molecules for the subsequent ammonia formation. The highest NH₃ production rate was 328.9 $\mu\text{mol h}^{-1} \text{g}^{-1}$ under white light (300 mW cm⁻², $\lambda = 300\text{--}780 \text{ nm}$). Pristine g-C₃N₄ had the rate of 124.2 $\mu\text{mol h}^{-1} \text{g}^{-1}$.

6.2 NO oxidation reaction

Ti₃C₂ modified g-C₃N₄ was used for the photocatalytic NO removal [119]. The formation of a built-in electron field could lead to the photo-induced electrons accumulation on Ti₃C₂ to promote the activation of O₂, which promoted the generation of radical species and the subsequent photocatalytic surface reaction of NO. The photocatalytic oxidation was performed under visible irradiation ($\lambda > 420 \text{ nm}$) and NO was oxidized by oxygen derived radicals to nitrite and nitrate.

The heterojunction photocatalyst $\text{Ti}_3\text{C}_2@\text{TiO}_2/\text{g-C}_3\text{N}_4$ was constructed by Zhang et al. [120] and applied for the nitric oxide removal as well. The two-dimensional Ti_3C_2 was partially oxidized to form $\text{TiO}_2\text{-Ti}_3\text{C}_2$ nanosheets by controlled oxidation conditions. A removal efficiency of 28.9% was reached. A small amount of NO_2 (18.75 ppb), as a reaction product, was determined. The other reaction species were nitrate and nitrite.

The oxidation removal of NO using Ti_3C_2 -derived $\text{TiO}_2@\text{C}$ coupled with $\text{g-C}_3\text{N}_4$ was also published by Wang et al. [121]. The Z-scheme heterojunction enabled the enhanced photocatalytic NO removal. These authors reached a 94% removal efficiency of the nitric oxide using atomized H_2O_2 under a 500 W Xenon lamp (a 420 nm cut-off filter), see Fig. 9.

Ternary $\text{g-C}_3\text{N}_4/\text{TiO}_2/\text{Ti}_3\text{C}_2$ MXene S-scheme heterojunction photocatalysts were fabricated by Hu et al. [122] and tested for the oxidation of NO. $\text{TiO}_2/\text{Ti}_3\text{C}_2$ was prepared by the partial oxidation of Ti_3C_2 . $\text{g-C}_3\text{N}_4$ polymerized from melamine on the surface of $\text{TiO}_2/\text{Ti}_3\text{C}_2$. The removal efficiency was 66.3% after 30 min. The other reactions products were NO_2 and nitrate.

The similar reaction with H_2O_2 but using a 2D/0D/2D $\text{g-C}_3\text{N}_4/\text{TiO}_2@\text{C}$ aerogel was published by the same group of authors [123]. The removal efficiency of 90.7% was achieved during 50 min. The main reaction product was nitrate.

There are not enough data in the literature concerning the MXene and $\text{g-C}_3\text{N}_4$ composites for both the nitrogen reduction reaction and the NO oxidation reaction yet. Therefore, the statistical testing was not performed for these photocatalytic reactions.

7 Miscellaneous

A $\text{Ti}_3\text{C}_2/\text{porous g-C}_3\text{N}_4$ interfacial Schottky junction photocatalyst was employed for the production of hydrogen peroxide by Yang et al. [124]. The highest H_2O_2 production rate of $2.20 \mu\text{mol L}^{-1} \text{min}^{-1}$ under visible light irradiation ($\lambda > 420 \text{ nm}$) was 2.1 times higher than that in the presence of pristine porous $\text{g-C}_3\text{N}_4$. The interfacial Schottky junction was fabricated by an electrostatic self-assembly process.

Ti_3C_2 quantum dot-modified defective inverse opal $\text{g-C}_3\text{N}_4$ via a facile electrostatic self-assembly method was prepared by Lin et al. [125]. The opal $\text{g-C}_3\text{N}_4$ contained carbon vacancies. The Schottky junction in the interface realized the spatial separation of the electrons and holes. The highest H_2O_2 yield reached $560.7 \mu\text{mol L}^{-1} \text{h}^{-1}$, which is 9.3 times higher than that of $\text{g-C}_3\text{N}_4$ under visible light irradiation.

A work of Wang et al. [126] was devoted to the already described $\text{Ti}_3\text{C}_2/\text{porous g-C}_3\text{N}_4$ composite for the oxidation of 5-hydroxymethylfurfural to 2,5-formylfuran. The composite containing 6% Ti_3C_2 provided a yield higher than

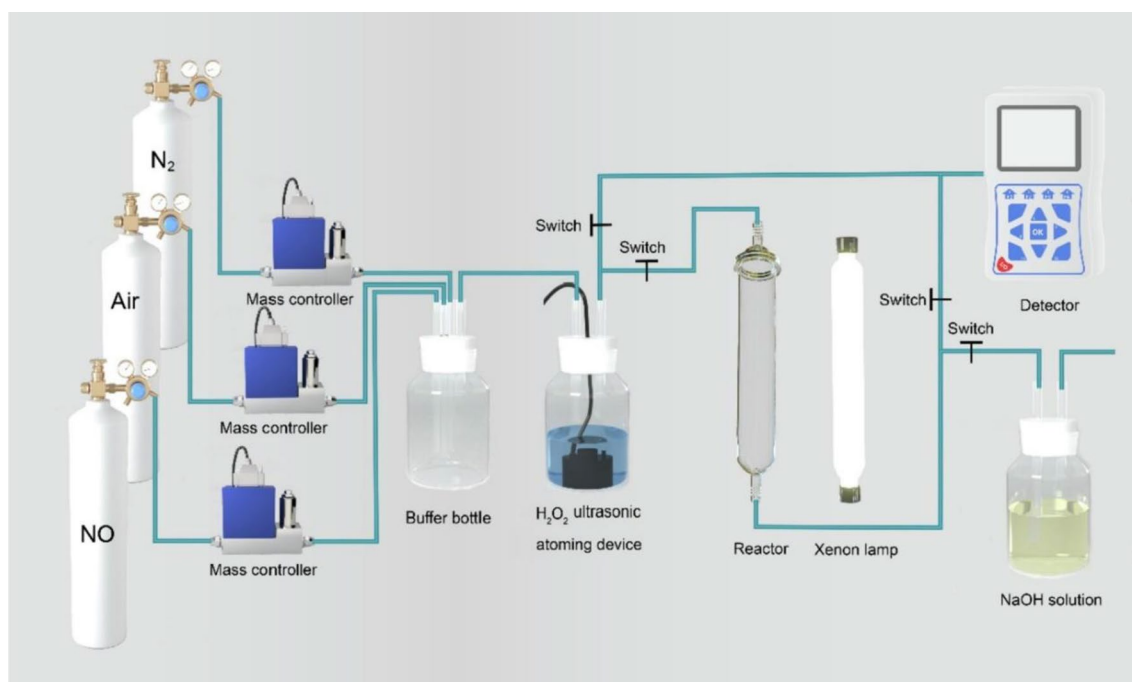


Fig. 9 Schematic diagram of the experimental apparatus for the photocatalytic oxidation method of NO. Reproduced from Ref. [121]. Copyright (2022), with permission from Elsevier

90% during 10 h under visible irradiation. The selectivity was 97%.

Zr₂CO₂/g-C₃N₄ heterostructure photocatalysts were designed, modelled, and tested for the volatile organic compound degradation [127]. The first principal calculations were confirmed by the degradation of formaldehyde, acetone, benzene, and trichloro ethylene. A spectroscopic analysis indicated the Z-scheme formation providing the photocatalytic activity as well as a direct band gap. The theoretical calculations were also performed for other MXenes, such as Ti₂CO₂, Hf₂CO₂, and Sc₂CO₂.

The Ti₃C₂/g-C₃N₄ was also tested for the reduction of U(VI) by Li et al. [128]. U(VI) was reduced to U(IV) forming UO_{2+x}, where $x < 0.25$. The main reducing species was the superoxide radical. The optimal photocatalytic reduction rate for U(VI) was 0.267 min⁻¹ under a 500 W Xe lamp. The optimal photocatalyst was able to remove all U(VI) during 15 min.

The authors Makola et al. [129] designed and fabricated a 2D-2D Nb₂CT_x@g-C₃N₄ MXene-based Schottky-heterojunction. They experimentally verified that this material is suitable for photocatalytic applications, however, no photocatalytic tests were performed.

8 Conclusion

The research articles (77 in total) about the photocatalytic application of the MXene and g-C₃N₄ composites were reviewed in this work. The MXene and g-C₃N₄ composites were mostly tested for the photocatalytic degradation of organic compounds (31%), the hydrogen evolution (30%), and the reduction of carbon dioxide (18%). The remaining 21% of chemical reactions were devoted to the redox reactions of nitrogen (13%) and other applications (8%). The CO₂ reduction results were only carbon monoxide and methane.

The majority of articles were devoted to Ti₃C₂ based composites with g-C₃N₄. Only a few articles described the results obtained with the Ni₃C, Nb₂C, Zr₂CO₂, Mo₂C, and V₂C MXenes. The basic mechanism of the MXene/g-C₃N₄ composites' photocatalytic activity is in the combination of the ability of g-C₃N₄ to absorb visible light and the excellent electron transport properties of Ti₃C₂. The heterojunction formed between both compounds enables the effective separation of electrons and holes photoinduced in g-C₃N₄. Both charge carriers can then take part in various photocatalytic reactions without their recombination.

A critical review of the cited articles was performed based on the statistical processing of the reported results. Besides the photocatalytic reduction of CO₂ to CH₄, it was found that MXenes significantly improve the photocatalytic activity of bare graphitic carbon nitride. In addition, it was

found that adding some co-catalysts to these MXene/g-C₃N₄ composites do not lead to a further increase in photocatalytic activity. The surface (vacancy) and heterojunction engineering of the MXenes and g-C₃N₄ is likely more important.

The MXene and g-C₃N₄ composites were also compared with the other photocatalysts based on g-C₃N₄, especially in terms of the hydrogen evolution and the reduction of CO₂ to CO. Ongoing research should aim to optimize the properties and performance of these materials and explore their potential in photocatalytic applications, such as the higher HER performance and the reduction of CO₂ to other more useful products. The degradation of organic compounds is also a challenge especially in terms of the application and recycling of the photocatalysts in real conditions. The described materials were obtained and tested in laboratories, but still there is a long way to their practical applications.

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Data availability There are no available data.

Declarations

Conflict of interest The author declares no conflict of interest.

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