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# 2D V<sub>2</sub>C MXene/2D g-C<sub>3</sub>N<sub>4</sub> nanosheet heterojunctions constructed *via* a one-pot method for remediating water pollution through high-efficient adsorption together with *in situ* photocatalytic degradation†

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With the development of modern industry, the problems of water pollution have become increasingly serious. There is a strong need to develop highly efficient and environmentally friendly technologies to address water pollution. In this work, a novel 2D V<sub>2</sub>C MXene/2D g-C<sub>3</sub>N<sub>4</sub> nanosheet heterojunction was constructed *via* a one-pot method. The obtained composite materials displayed excellent purifying capacity for dye pollutants, with removal ratios for crystal violet (CV), Rhodamine B (RhB) and methylene blue (MB) of 99.5%, 99.5%, and 95% within 80 min (including an adsorption process for 50 min and photodegradation process for 27 min), respectively. The extraordinary purifying capacity was accomplished through high-efficient adsorption together with *in situ* photocatalytic degradation within the unique 2D/2D heterojunction structure. The successful exploitation of 2D V<sub>2</sub>C MXene/2D g-C<sub>3</sub>N<sub>4</sub> nanosheet heterojunctions provided a simple method to efficiently remedy water pollution.

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## 1. Introduction

Nowadays, water pollution has become an extremely serious issue in modern society, greatly threatening economic growth and ecosystem health. Industrial water, especially dye waste water, which is primarily discharged by textile, printing, and electroplating industries, has attracted considerable attention due to its massive quantity and the difficulty in remediation.<sup>1,2</sup> These high toxicity organic dyes can lead to carcinogenic, teratogenic, and mutagenesis effects on humans and aquatic organisms.<sup>3</sup> In consequence, it is urgently required to eliminate contaminants from dye wastewater before it is discharged.

In the past decades, many water treatment technologies have been developed by researchers, including adsorption, flocculation, membrane separation, biological degradation, photocatalysis, and Fenton reaction.<sup>4–6</sup> Among them, photocatalysis has attracted abundant attention due to the sustainable utilization of solar energy, which enables the conversion of solar energy into chemical energy to address water pollution issues.<sup>7</sup> In recent years, semiconductors including BiVO<sub>4</sub>, TiO<sub>2</sub>, CdS, Bi<sub>2</sub>WO<sub>4</sub> and non-metallic graphitic carbon nitride (g-C<sub>3</sub>N<sub>4</sub>) have

been extensively researched in various fields.<sup>8–11</sup> g-C<sub>3</sub>N<sub>4</sub>, with a 2D sheet-like structure formed by the regular arrangement of tris-*s*-triazine units, displays numerous extraordinary features, such as nontoxicity, non-metallic composition and a narrow band gap.<sup>12</sup> However, bulk g-C<sub>3</sub>N<sub>4</sub>, fabricated through the thermal condensation of organic precursors containing both carbon and nitrogen, is restricted by low light adsorption and slow charge transfer rate of electron–hole pairs.<sup>13</sup> Researchers working in this field have proposed many strategies to improve the photocatalysis activity, including introducing heteroatoms or nitrogen vacancies, coupling with other semiconductors, and controlling the morphology.<sup>14</sup>

Delaminating multi-layered bulk g-C<sub>3</sub>N<sub>4</sub> to obtain g-C<sub>3</sub>N<sub>4</sub> nanosheets is one of the most popular strategies for controlling morphology to improve the photocatalytic activity.<sup>15</sup> Common methods to exfoliate bulk g-C<sub>3</sub>N<sub>4</sub> include ultrasonic thinning, thermal oxidation peeling and acid-based chemical peeling.<sup>16</sup> For instance, Lin *et al.* exfoliated bulk g-C<sub>3</sub>N<sub>4</sub> into g-C<sub>3</sub>N<sub>4</sub> nanosheets through an ultrasound method in ethanol/water, isopropanol/water and dimethylformamide/water with a concentration of 0.1–3 mg mL<sup>−1</sup>. The specific surface area of the obtained g-C<sub>3</sub>N<sub>4</sub> nanosheets was as high as 59.4 m<sup>2</sup> g<sup>−1</sup>, which was five times that of bulk g-C<sub>3</sub>N<sub>4</sub>.<sup>17</sup> Wan *et al.* exfoliated bulk g-C<sub>3</sub>N<sub>4</sub> into ultrathin protonated g-C<sub>3</sub>N<sub>4</sub> (PCN) nanosheets under the assistance of H<sub>2</sub>SO<sub>4</sub>, which exhibited improved photocatalytic activity.<sup>18</sup> However, regulation and control of the structure of single-phase material to enhance the photocatalyst

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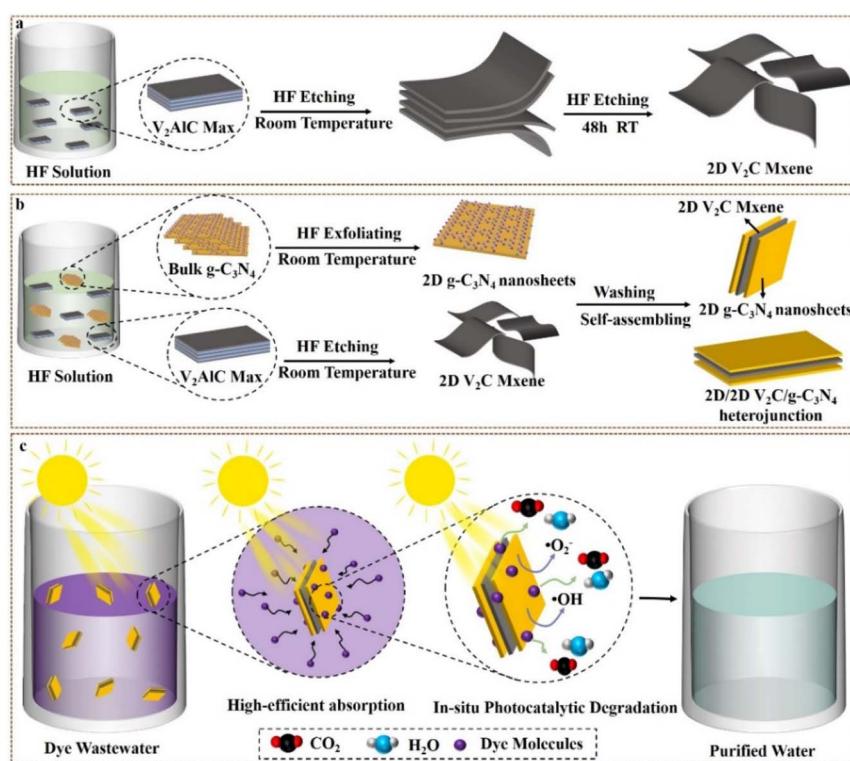
activity was finite, attributed to the recombination between electrons and holes within the single-phase material, thus largely impeding the photocatalytic reaction.<sup>19</sup>

Establishing heterojunction photocatalysts enables to settle these problems through accelerating the separation of electrons and holes and advancing the capacity of light adsorption.<sup>19,20</sup> Zhu *et al.* constructed a novel  $\text{Bi}_2\text{Sn}_2\text{O}_7/\text{g-C}_3\text{N}_4$  heterojunction through an ultrasound-assisted hydrothermal method, which decomposed 94% norfloxacin in the experimental solution and eliminated 89% of norfloxacin after five cycles, exhibiting outstanding stability and cyclability of photocatalytic activity.<sup>21</sup> Zhao *et al.* synthesized a 2D/2D  $\text{BiPO}_4/\text{g-C}_3\text{N}_4\text{-B}$  nanosheet heterojunction photocatalyst with 97.3% degradation of RhB within 25 min under visible light.<sup>22</sup> These works demonstrated that 2D layered material coupled with 2D  $\text{g-C}_3\text{N}_4$  could improve the charge carrier separation efficiency, which is ascribed to the cations in the layered structure providing the charge trapping ability.<sup>23</sup>

MXenes, as a novel family of 2D nanomaterials comprising transition metal carbides, nitrides, and carbonitrides, have stimulated considerable attentions in advancing the photocatalytic activity of semiconductor materials.<sup>24</sup>  $\text{V}_2\text{C}$  MXene, the newly explored layered material, manifests numerous distinguished properties including excellent electrical conductivity, high carrier mobility, and various active groups at the surface terminus, which possesses a greater specific surface area and more active centers compared to other MXenes, leading to promising application as a cocatalyst for pollutant

photodegradation.<sup>6</sup> Sherrya *et al.* exploited the novel  $\text{V}_2\text{C}$  MXenes-coupled  $\text{g-C}_3\text{N}_4$  2D/2D nanohybrids showing the remarkable yield of  $\text{H}_2$  with a maximal rate of  $360 \text{ mmol g}^{-1} \text{ h}^{-1}$ , which was four times than that of pristine  $\text{g-C}_3\text{N}_4$ .<sup>25</sup> Tahir designed  $\text{V}_2\text{C}$  MXene combined with exfoliated  $\text{g-C}_3\text{N}_4$ , which enabled the reduction of  $\text{CO}_2$  to  $\text{CO}$  with a yield rate of  $9289 \text{ mmol g}^{-1} \text{ h}^{-1}$ .<sup>26</sup> These published works convincingly demonstrated that constructing heterojunction photocatalysts enabled a prominent enhancement of the photocatalytic activity.

In this work, 2D  $\text{V}_2\text{C}$  MXene/2D  $\text{g-C}_3\text{N}_4$  heterojunctions were successfully developed *via* a one-pot method in HF solution at room temperature, which almost completely purified the dye waste water under visible light through high-efficiency adsorption together with *in situ* photocatalytic degradation (Scheme 1). Compared to pristine  $\text{g-C}_3\text{N}_4$ , 2D  $\text{V}_2\text{C}$  MXene/2D  $\text{g-C}_3\text{N}_4$  heterojunctions displayed higher charge transfer rate, better separation efficiency of photogenerated  $e^- - h^+$  pairs, narrower band gap and greater adsorption ability of visible light, promoting the photodegradation of RhB, CV, and MB. The removal ratio (including adsorption and photodegradation processes) onto CV, RhB and MB reached 99.5%, 99.5%, and 95%, respectively, demonstrating that 2D  $\text{V}_2\text{C}$  MXene is a perfect cocatalyst with excellent performance to modify 2D  $\text{g-C}_3\text{N}_4$  nanosheets. As we know, there is no report on the preparation of 2D  $\text{V}_2\text{C}$  MXene/2D  $\text{g-C}_3\text{N}_4$  heterojunctions *via* the one-pot strategy. This work provided a profoundly simple path to synthesize 2D  $\text{V}_2\text{C}$  MXene/2D  $\text{g-C}_3\text{N}_4$  heterojunctions with outstanding photocatalytic



Scheme 1 Schematic of the preparation process of (a)  $\text{V}_2\text{C}$  MXene and (b) 2D/2D  $\text{V}_2\text{C}/\text{g-C}_3\text{N}_4$  heterojunction; (c) schematic of the purification process of dye wastewater using 2D/2D  $\text{V}_2\text{C}/\text{g-C}_3\text{N}_4$  heterojunction.



activity, tremendously inspiring the preparation of high-activity photocatalysts. Meanwhile, this work is also important for remediating water pollution through high-efficiency adsorption together with *in situ* photocatalytic degradation.

## 2. Results and discussion

### 2.1 Morphological and structural analysis

**2.1.1 X-Ray diffraction (XRD).** The crystalline structure and existence of  $\text{g-C}_3\text{N}_4$  components in the nanocomposites were confirmed by XRD. As shown in Fig. 1a, the typical peaks of  $\text{V}_2\text{AlC}$  Max at  $13.4^\circ$ ,  $27.1^\circ$ ,  $35.5^\circ$ ,  $41.1^\circ$ ,  $45.1^\circ$ ,  $55.1^\circ$ ,  $63.8^\circ$ ,  $75^\circ$  and  $78.8^\circ$  belonged to (002), (004), (101), (103), (104), (106), (110), (109) and (116) planes, respectively, demonstrating the hexagonal crystal structure of  $\text{V}_2\text{AlC}$  Max.<sup>27–29</sup> The diffraction peaks at  $13.4^\circ$ ,  $35.5^\circ$ ,  $41.1^\circ$ ,  $55.5^\circ$ ,  $63.8^\circ$ ,  $75^\circ$  and  $78.8^\circ$ , attributed to (002), (101), (103), (106), (110), (109) and (116), were observed in the XRD pattern of exfoliated  $\text{V}_2\text{C}$  MXene, in which  $13.4^\circ$  and  $41.1^\circ$  were weakened after etching by HF solution for 48 h, verifying the incomplete removal of Al atoms, while  $75^\circ$  and  $78.8^\circ$  coincided with the (109) and (116) crystal planes of  $\text{V}_2\text{AlC}$  Max, respectively, illustrating that  $\text{V}_2\text{C}$  MXene nanosheets were fabricated from  $\text{V}_2\text{AlC}$  Max.<sup>30,31</sup> Besides, the appearance of new characteristic peaks at  $9.2^\circ$  and  $11.8^\circ$  further confirmed the successful synthesis of  $\text{V}_2\text{C}$  MXene.<sup>32</sup> The distinctive peaks at  $12.8^\circ$  and  $27.5^\circ$  in the  $\text{g-C}_3\text{N}_4$  XRD pattern assigned to (100) and (002) crystalline planes indicated the  $\text{g-C}_3\text{N}_4$  aromatic systems' interlayer structural packing and the distinctive interplanar stacking peak, verifying the successful production of  $\text{g-C}_3\text{N}_4$  from urea precursor by thermal polycondensation.<sup>33,34</sup> A slight shift from  $27.1^\circ$  to  $27.6^\circ$  (belonging to  $\text{V}_2\text{C}$  MXene) was observed after compositing with  $\text{g-C}_3\text{N}_4$ , attesting the variation of interlayer spacing, while the primary characteristic peaks all appeared without distinct shifting, demonstrating the efficient synthesis of both nanocomposite  $\text{V}_2\text{C}/\text{g-C}_3\text{N}_4$  heterojunction, as evidenced by their effective interaction.<sup>35</sup>

**2.1.2 Fourier transform infrared (FT-IR).** FT-IR was carried out to identify the molecular structures of the  $\text{V}_2\text{C}/\text{g-C}_3\text{N}_4$  heterojunction and the interactions between  $\text{V}_2\text{C}$  MXene and  $\text{g-C}_3\text{N}_4$  nanosheets. As shown in Fig. 1b, the characteristic peak in the FT-IR spectrum of  $\text{g-C}_3\text{N}_4$  at  $814\text{ cm}^{-1}$  was assigned to the

triazine ring system, and the peaks in the region from  $1100$  to  $1700\text{ cm}^{-1}$  originated from trigonal C–N(–C)–C (full condensation) and bridging C–NH–C units (partial condensation).<sup>13,36</sup> For pristine  $\text{V}_2\text{C}$  MXene, the adsorption peaks at  $983\text{ cm}^{-1}$ ,  $1594\text{ cm}^{-1}$  and  $618\text{ cm}^{-1}$  were attributed to –C–F, –C=O and V–O vibrations, respectively. Besides, the characteristic peak at  $2353\text{ cm}^{-1}$  belonging to  $\text{V}_2\text{C}$  was also observed in the spectrum.<sup>6,37,38</sup> Notably, all the characteristic peaks belonging to both  $\text{g-C}_3\text{N}_4$  and  $\text{V}_2\text{C}$  MXene that appeared in the FT-IR spectrum of  $\text{V}_2\text{C}/\text{g-C}_3\text{N}_4$  heterojunction with a slight shift were ascribed to the strong interactions between  $\text{V}_2\text{C}$  MXene and  $\text{g-C}_3\text{N}_4$ . All the discoveries concluded that  $\text{V}_2\text{C}$  MXene was successfully incorporated into the nanocomposite to yield the heterojunction.

**2.1.3 Raman spectroscopy.** Raman spectroscopy analysis is capable of determining rotational, vibrational and other low frequency patterns of heterojunctions. As shown in Fig. 1c, the Raman spectrum of  $\text{V}_2\text{C}$  MXene displayed distinct peaks at  $141$ ,  $195$ ,  $280$ ,  $404$  and  $986\text{ cm}^{-1}$ , which were ascribed to Raman-active  $\text{V}_2\text{C}$  MXene vibration mode. Additionally, the appearance of the peaks at  $986\text{ cm}^{-1}$ , attributed to the enhancement of interlayer spacing in  $\text{V}_2\text{C}$  MXene and  $404\text{ cm}^{-1}$  arising from the active vibration modes of the terminated  $\text{V}_2\text{C}$  MXene confirmed the successful synthesis of  $\text{V}_2\text{C}$  MXene.<sup>35,39,40</sup> However, the resolution of the recorded Raman spectra of  $\text{g-C}_3\text{N}_4$  was fairly poor due to the fluorescence interference, while some weak characteristic peaks appeared at  $207$ ,  $353$ ,  $482$ ,  $576$ ,  $739$ ,  $961$ ,  $1233$ ,  $1307\text{ cm}^{-1}$  for bulk  $\text{g-C}_3\text{N}_4$  corresponding to the vibrational modes of CN heterocycles (Fig. S1†).<sup>35,40,41</sup> For the  $\text{V}_2\text{C}/\text{g-C}_3\text{N}_4$  heterojunction, the distinct peaks weakened for  $\text{V}_2\text{C}$  MXene compared to that of  $\text{V}_2\text{C}$  MXene, which was disturbed by the fluorescence of  $\text{g-C}_3\text{N}_4$  on it. However, this phenomenon verified the successful combination of  $\text{g-C}_3\text{N}_4$  and  $\text{V}_2\text{C}$  MXene to establish the  $\text{V}_2\text{C}/\text{g-C}_3\text{N}_4$  heterojunction.

**2.1.4 X-ray photoelectron spectra (XPS).** The elemental states of  $\text{V}_2\text{C}/\text{g-C}_3\text{N}_4$  heterojunction,  $\text{g-C}_3\text{N}_4$  and  $\text{V}_2\text{C}$  MXene were determined by XPS, as shown in Fig. 2 S2 and S3,† respectively. As shown in Fig. S2,† obvious C, N and O peaks were exhibited in the XPS spectrum of  $\text{g-C}_3\text{N}_4$ , where the O peak was probably attributed to  $\text{H}_2\text{O}$ /oxidation-generated oxides adsorbed on the sample surface. C 1s spectra showed three

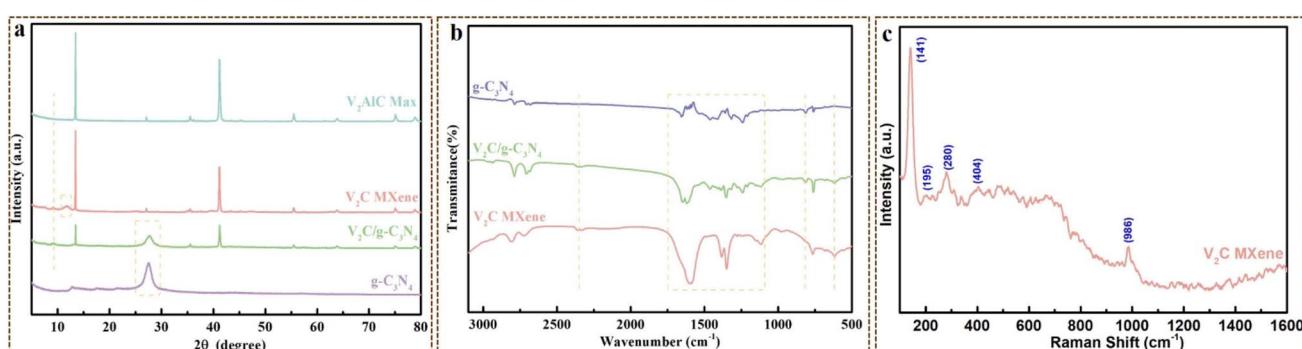


Fig. 1 (a) XRD patterns of  $\text{V}_2\text{AlC}$  Max,  $\text{V}_2\text{C}$  MXene, bulk  $\text{g-C}_3\text{N}_4$  and  $\text{V}_2\text{C}/\text{g-C}_3\text{N}_4$ ; (b) FT-IR spectra of  $\text{g-C}_3\text{N}_4$ ,  $\text{V}_2\text{C}$  MXene and  $\text{V}_2\text{C}/\text{g-C}_3\text{N}_4$ ; Raman spectra of (c)  $\text{V}_2\text{C}$  MXene.



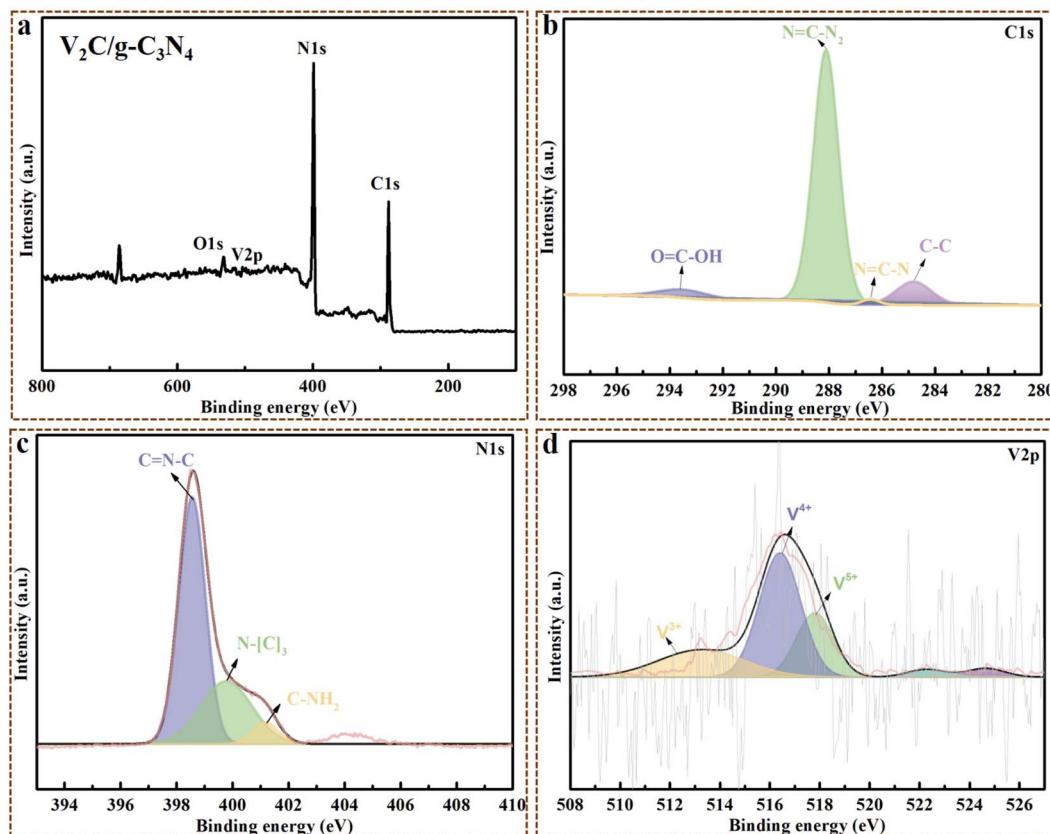


Fig. 2 XPS spectra of (a)  $\text{V}_2\text{C}/\text{g-C}_3\text{N}_4$ ; (b) C 1s; (c) N 1s; (d) V 2p.

different chemical states corresponding to  $\text{sp}_3$  C–N=C, O–CN and C–C at 288.1 eV, 286.2 eV and 284.8 eV, respectively. The N 1 s spectrum displayed the typical features of g-C<sub>3</sub>N<sub>4</sub>, with 398.6 eV (C=N=C), 401.0 eV (C–NH<sub>2</sub>) and 404.6 eV ascribed to  $\text{sp}^2$ -hybridized N, amino, and excitation, respectively.<sup>41,42</sup> For pristine V<sub>2</sub>C MXene, C, V, O and Al peaks were distinctly displayed in the XPS spectra, where Al peaks were weak, certifying that Al were almost completely etched away from V<sub>2</sub>AlC Max (Fig. S3†).<sup>43</sup> The five peaks that appeared in the V 2p spectrum correspond to different oxidation states of V, where 513.3 eV, 514.2 eV, 516.6 eV, 521.2 eV and 522.8 eV were contributed by V<sup>2+</sup>, V<sup>3+</sup>, V<sup>4+</sup>, V–C bond and their doublets, respectively.<sup>26,44</sup> The spectrum in the C 1s region well fitted the peaks of 282.3 eV, 284.8 eV, 286.3 eV and 288.2 eV that originated from C–V, C–C, C–O and O=C=O, respectively, while the peaks in the O 1 s region located at 529.6 eV, 531.7 eV and 533.8 eV originated from V–O, V–C–O and OH<sup>−</sup>, respectively.<sup>44</sup> For the V<sub>2</sub>C/g-C<sub>3</sub>N<sub>4</sub> heterojunction, C, N, V, and O peaks were all observed in the XPS spectrum (Fig. 2a), where the O peaks were possibly attributed to the presence of oxygen vacancies and hydroxyl group. As shown in Fig. 2b, the remarkable peaks at 284.8 eV, 286.4 eV, 288.1 eV and 293.6 eV were derived from C–C, N=C–N, N=C–N<sub>2</sub> and O=C–OH, respectively, which was in accordance with the C 1s spectrum of g-C<sub>3</sub>N<sub>4</sub> and V<sub>2</sub>C MXene without a distinct shift. Three binding energy peaks in the N 1 s spectrum (Fig. 2c) at 398.5 eV (C=N=C), 399.8 eV (N–C<sub>3</sub>) and 401.1 eV (C–NH<sub>2</sub>) were ascribed to  $\text{sp}^2$ -hybridized N, graphitic N

and amino, respectively, which was highly consistent with the XPS results of g-C<sub>3</sub>N<sub>4</sub> and V<sub>2</sub>C MXene. The V 2p spectra displayed five peaks at 513.2 eV, 516.4 eV, and 517.8 eV belonging to V<sup>3+</sup>, V<sup>4+</sup>, and V<sup>5+</sup> with their doublets at 522.3 eV and 524.6 eV (Fig. 2d). All the XPS results of the V<sub>2</sub>C/g-C<sub>3</sub>N<sub>4</sub> heterojunction, g-C<sub>3</sub>N<sub>4</sub> and V<sub>2</sub>C MXene revealed that the 2D/2D V<sub>2</sub>C MXene/g-C<sub>3</sub>N<sub>4</sub> nanosheets heterojunctions were successfully developed in this work without damaging the pristine elemental states of g-C<sub>3</sub>N<sub>4</sub> and V<sub>2</sub>C MXene.

**2.1.5 SEM and EDX analysis.** The microstructures of g-C<sub>3</sub>N<sub>4</sub>, V<sub>2</sub>AlC Max, V<sub>2</sub>C MXene and V<sub>2</sub>C/g-C<sub>3</sub>N<sub>4</sub> heterojunction are displayed in Fig. 3 and S4† surveyed by SEM. As shown in Fig. S4a,† the sheets-like structure of g-C<sub>3</sub>N<sub>4</sub> was observed while the sheets exhibited distinct agglomeration. However, after etching by HF solution, the bulk g-C<sub>3</sub>N<sub>4</sub> was exfoliated to nanosheets, displaying the 2D nanostructure. The tight layered structures of V<sub>2</sub>AlC Max constituted by boards and compacted sheets is shown in Fig. S4b,† which was linked through metallic bonds, generating the infinitesimal interlayer spacing. V<sub>2</sub>C MXene showed a 2D structure after etching with HF solution for 48 h, exhibiting the unconsolidated layered structure which enabled to establish the interactions between other 2D materials such as 2D g-C<sub>3</sub>N<sub>4</sub> (Fig. 3a). Intriguingly, the successful synthesis of 2D/2D V<sub>2</sub>C/g-C<sub>3</sub>N<sub>4</sub> heterojunctions through the one-pot etching method was certified by the SEM image, in which 2D g-C<sub>3</sub>N<sub>4</sub> nanosheets growing on 2D V<sub>2</sub>C MXene increased the interfacial contact area for charge carrier migration (Fig. 3c).

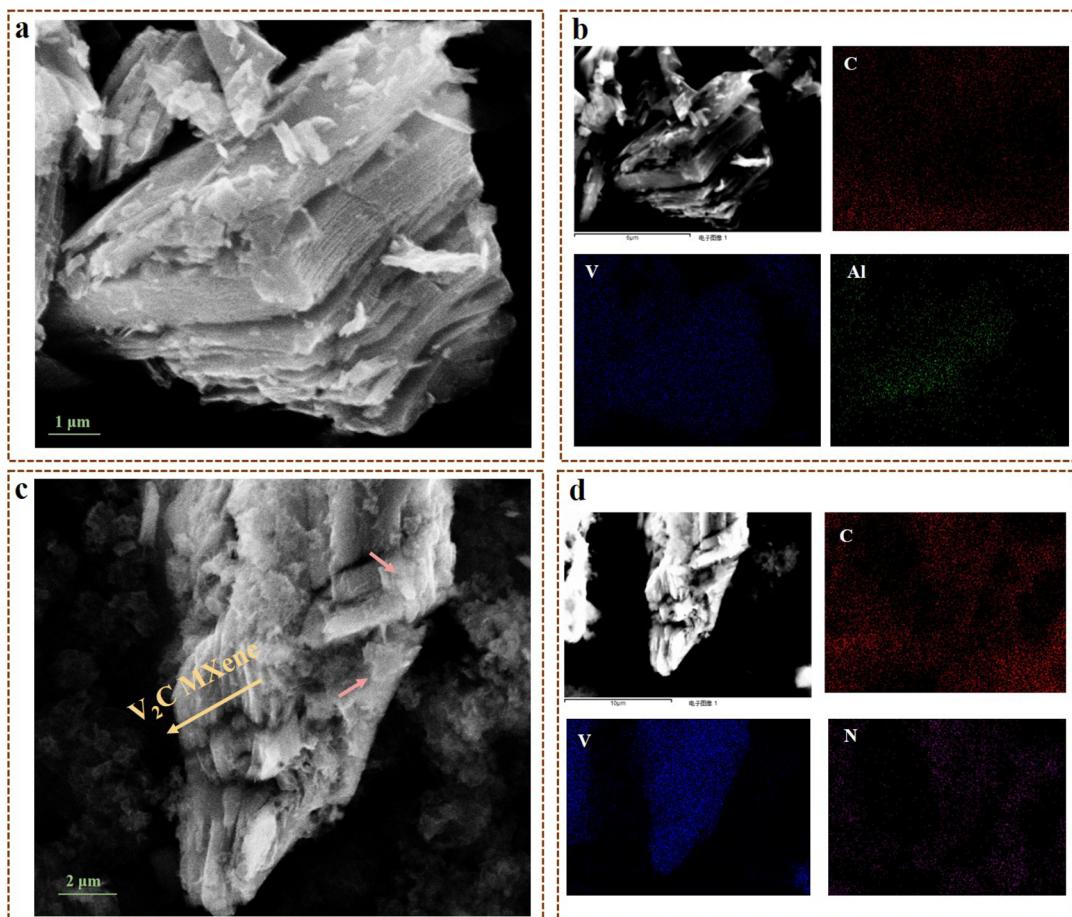


Fig. 3 (a) SEM image of  $V_2C$  MXene and (b) corresponding EDX mapping of  $V_2C$  MXene; (c) SEM image of  $V_2C$  MXene  $V_2C/g-C_3N_4$  heterojunction and (d) corresponding EDX mapping of the  $V_2C$  MXene  $V_2C/g-C_3N_4$  heterojunction.

The compact contact between 2D  $g-C_3N_4$  nanosheets and 2D  $V_2C$  MXene led to the production of more electron transport routes, contributing to electron transport and separation.

EDX mapping analysis was performed to investigate the distribution of elements in the samples (Fig. 3b, d, S4b and d†). The presence of all elements in  $g-C_3N_4$ ,  $V_2C$  MXene and  $V_2C/g-C_3N_4$  heterojunctions was confirmed by the EDX plots in Fig. S5.† V and Al were observed in the plots of  $V_2C$  MXene and  $V_2C/g-C_3N_4$  heterojunctions, while N appeared in the plots of  $g-C_3N_4$  and  $V_2C/g-C_3N_4$  heterojunctions. The existence of main elements in the samples was also supported by the EDX mapping images. In  $g-C_3N_4$ , the main elements were C, N, and O, while the appearance of Au was attributed to the gold-spraying process on the sample (Fig. S4 and S5†). C, V, Al, and F were observed in  $V_2C$  MXene, verifying the etching process on  $V_2AlC$  by HF to remove Al, generating  $V_2C$  MXene. All the elements in both  $g-C_3N_4$  and  $V_2C$  MXene appeared in the EDX mapping of the  $g-C_3N_4/V_2C$  heterojunction, which determined the hybrid of  $g-C_3N_4$  and  $V_2C$  MXene. Finally, the  $g-C_3N_4/V_2C$  heterojunction was synthesized with a high degree of purity due to the presence of the predicted elements in  $V_2C/g-C_3N_4$  photocatalysts.

**2.1.6 BET analysis.** The specific surface area of  $V_xG_y$  was verified by Brunauer–Emmett–Teller (BET) analysis. As shown

in Fig. 4a, the  $N_2$  adsorption–desorption isotherms of  $V_2AlC$  Max,  $V_2C$  MXene,  $g-C_3N_4$  and  $V_2C/g-C_3N_4$  heterojunction are presented, which showed type IV isotherms with distinct  $H_3$  hysteresis loop, according to IUPAC classification.<sup>45</sup> These phenomena coincided with the characteristic of solids with micro- and mesopores and orderly macropores containing narrow mesopores interconnecting channels, certifying the mesoporous structure of the as-prepared materials.<sup>46</sup> Compared to  $V_2C$  MXene, the hysteresis loop area of the  $V_2C/g-C_3N_4$  heterojunction greatly magnified, which is attributed to the production of abundant mesopores generated from the recombination of  $V_2C$  MXene and  $g-C_3N_4$ .<sup>47</sup> Besides, the specific surface area of  $V_2AlC$  Max,  $V_2C$  MXene,  $g-C_3N_4$  and  $V_2C/g-C_3N_4$  heterojunction was calculated as 0.102, 7.135, 115.636 and 70.122  $m^2 g^{-1}$ , respectively (Table S1†). The average pore diameter of  $V_2AlC$  Max,  $V_2C$  MXene,  $g-C_3N_4$  and  $V_2C g^{-1}C_3N_4$  heterojunction was calculated through the Barrett–Joyner–Halenda (BJH) method as 10.86, 13.94, 9.09 and 14.64 nm, respectively (Fig. S6 and Table S1†), demonstrating the existence of mesopores on these materials. Remarkably, the specific surface area and the pore volume of the  $V_2C/g-C_3N_4$  heterojunction was much higher than that of the  $V_2C$  MXene, which indicated that  $g-C_3N_4$  was successfully intercalated in  $V_2C$  MXene, thus enlarging the interlamellar spacing and providing



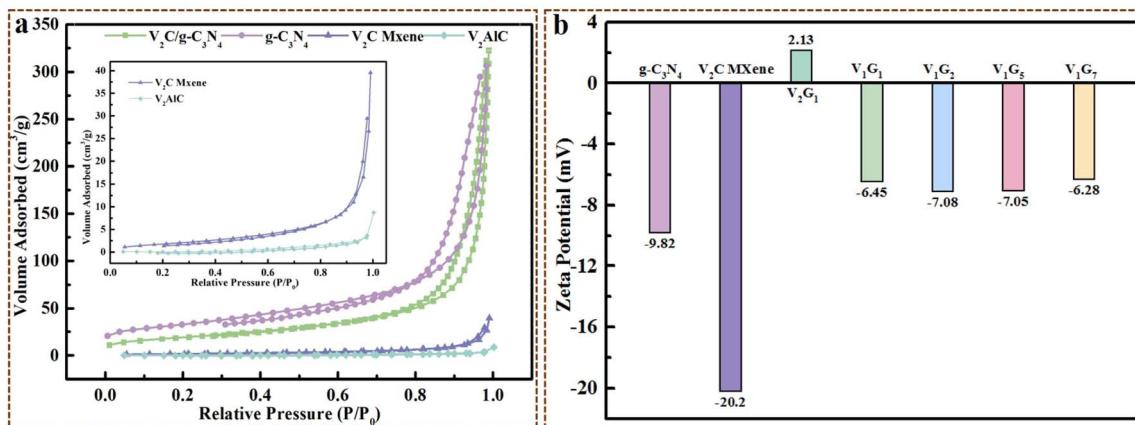


Fig. 4 (a) N<sub>2</sub> adsorption–desorption isotherms of V<sub>2</sub>AlC Max, V<sub>2</sub>C MXene, g-C<sub>3</sub>N<sub>4</sub> and V<sub>2</sub>C/g-C<sub>3</sub>N<sub>4</sub> heterojunction; (b) zeta Potential of g-C<sub>3</sub>N<sub>4</sub>, V<sub>2</sub>C MXene and V<sub>x</sub>G<sub>y</sub> heterojunctions.

affluent active sites to enhance the photocatalytic activity.<sup>34</sup> Thus, the intercalation of g-C<sub>3</sub>N<sub>4</sub> in the V<sub>2</sub>C MXene highly advanced the specific surface area and pore volume, which was vital to boost the photocatalytic property.

**2.1.7 Zeta potential.** The investigation of the surface electrical behaviour of materials is crucial to understand the adsorption behaviour onto dye molecules. Zeta potential studies enabled to reflect the surface charge performance of the materials to parse the interactions between g-C<sub>3</sub>N<sub>4</sub> and V<sub>2</sub>C MXene and comprehend the adsorption behaviour of different dye pollutants. As shown in Fig. 4b, the zeta potential of bulk g-C<sub>3</sub>N<sub>4</sub> and V<sub>2</sub>C MXene was measured to be  $-9.8$  mV and  $-20.2$  mV, respectively, demonstrating that the materials carried the negative charges on their surface. Many published works demonstrated that g-C<sub>3</sub>N<sub>4</sub> would be protonized through etching by acid, even the surface charge of which would transform to positive.<sup>48,49</sup> The different surface charge performance illustrated that g-C<sub>3</sub>N<sub>4</sub> and V<sub>2</sub>C MXene bound with each other through electrostatic interaction. More crucially, the zeta potential of V<sub>2</sub>G<sub>1</sub> heterojunction was  $2.13$  mV, showing electropositivity. It was supposed that the appropriate conjunction ratio within V<sub>2</sub>G<sub>1</sub> between protonized g-C<sub>3</sub>N<sub>4</sub> and V<sub>2</sub>C MXene transformed the surface potential to positive. However, V<sub>1</sub>G<sub>1</sub>, V<sub>1</sub>G<sub>2</sub>, V<sub>1</sub>G<sub>5</sub> and V<sub>1</sub>G<sub>7</sub> heterojunctions were negative, and the results of their zeta potential are very close.

## 2.2 Band gap and optoelectronic characterization

**2.2.1 Photoluminescence analysis.** The optical characteristics of V<sub>2</sub>C/g-C<sub>3</sub>N<sub>4</sub> heterojunction were discerned by photoluminescence (PL) analysis, which enabled to elucidate the surface oxygen vacancies and the separation of photogenerated charges to evaluate the segregation and transfer performance of photogenerated charge carriers. The peaks were observed with visual wavelengths between 400 and 500 nm for g-C<sub>3</sub>N<sub>4</sub> and V<sub>2</sub>C/g-C<sub>3</sub>N<sub>4</sub> heterojunction, while the PL peak of V<sub>2</sub>C MXene barely appeared, attributed to its dark color (nearly black) reducing electron generation, indicating that the wavelength of both g-C<sub>3</sub>N<sub>4</sub> and V<sub>2</sub>C/g-C<sub>3</sub>N<sub>4</sub> heterojunction has remarkable effects on PL spectral emission (Fig. 5a).<sup>50</sup> Moreover, the lower intensity

peak of V<sub>2</sub>C MXene was observed, ascribed to its metallic property, which could barely generate electron–hole pairs.<sup>51</sup> Generally, lower density and depressed emission peak manifested the efficient separation of e<sup>-</sup> and h<sup>+</sup>, resulting in better photocatalytic activity. As shown in Fig. 5a, g-C<sub>3</sub>N<sub>4</sub> displayed the highest PL peak owing to the band–band PL signal phenomenon generated by excitonic PL caused by n–π\* electronic transitions containing lone pairs of nitrogen atoms in g-C<sub>3</sub>N<sub>4</sub>, verifying the existence of sub gap flaws in g-C<sub>3</sub>N<sub>4</sub>.<sup>51</sup> However, after coupling with V<sub>2</sub>C MXene, the intensity of PL peaks sharply decreased, contributing to the more excellent charges separation proficiency, which meant more photocatalytic activity, ascribed to the introduction of V<sub>2</sub>C that traps electron, facilitating charge transport over the interface between 2D V<sub>2</sub>C MXene and 2D g-C<sub>3</sub>N<sub>4</sub> nanosheets and resulting in the decreasing rate of recombination.<sup>52,53</sup> More importantly, the superb separation efficiency of photogenerated e<sup>-</sup>–h<sup>+</sup> pairs created by the interfaces between 2D V<sub>2</sub>C MXene and 2D g-C<sub>3</sub>N<sub>4</sub> nanosheets gave rise to the lowest excitation intensity of the V<sub>2</sub>C/g-C<sub>3</sub>N<sub>4</sub> heterojunction, illustrating that the metal-like nature of V<sub>2</sub>C MXene as a mediator coupled with g-C<sub>3</sub>N<sub>4</sub> dramatically accelerated the separation of photogenerated charge carriers and alleviated the photogenerated recombination e<sup>-</sup>–h<sup>+</sup> pairs, which was beneficial to boost the photo-degradation efficiency of dye pollutants.

**2.2.2 Electrochemical impedance spectra.** Electrochemical impedance spectra (EIS) of g-C<sub>3</sub>N<sub>4</sub>, V<sub>1</sub>G<sub>5</sub> heterojunction and V<sub>1</sub>G<sub>7</sub> heterojunction obtained under visible light and dark environment are shown in Fig. 5b and S7,† respectively. Notoriously, the smaller arc radius means the smaller impedance of charge transfer, leading to a higher charge transfer rate of the materials.<sup>54–56</sup> As shown in Fig. 5b, under visible light, the V<sub>1</sub>G<sub>5</sub> heterojunction exhibited the smallest arc radius, while g-C<sub>3</sub>N<sub>4</sub> showed the largest arc radius, which illustrated that the addition of V<sub>2</sub>C MXene largely improved the efficiency of photogenerated carrier separation and the interfacial charge transfer potential. It was demonstrated that the extraordinary 2D/2D heterojunction structure showed high charge separation capacity, which remarkably advanced the efficiency of electron/

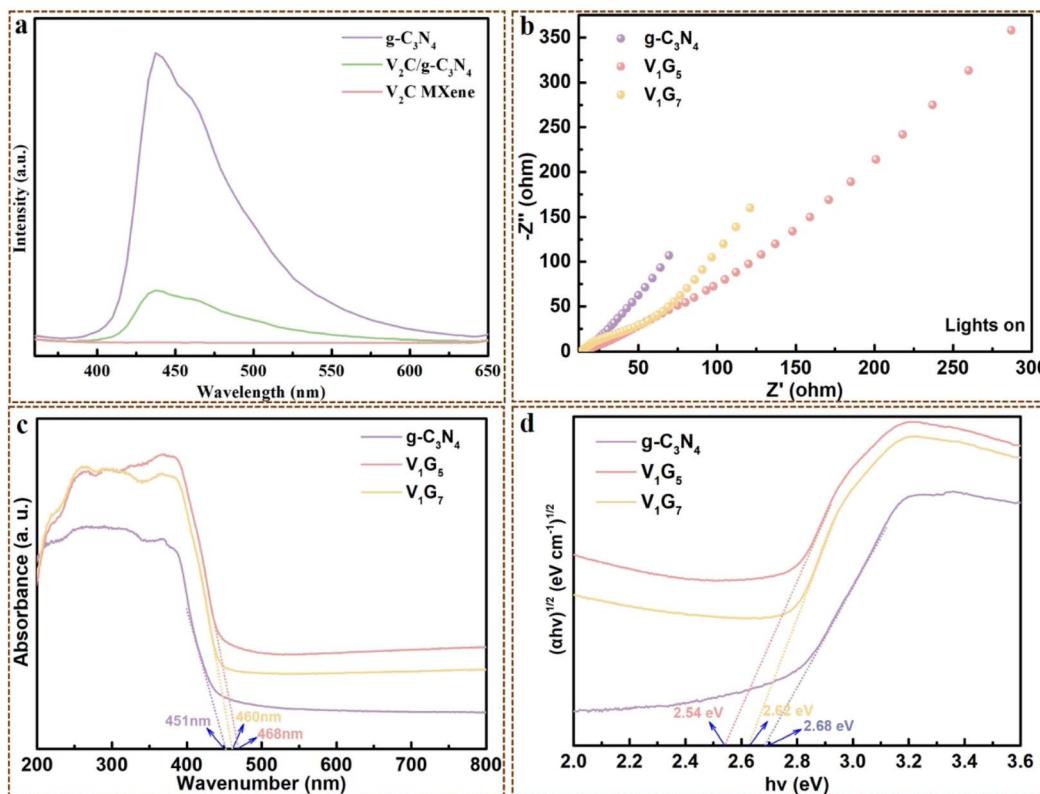


Fig. 5 (a) PL spectra of  $\text{V}_2\text{C}$  MXene,  $\text{g-C}_3\text{N}_4$  and  $\text{V}_2\text{C}/\text{g-C}_3\text{N}_4$  heterojunction; (b) electrochemical impedance spectra (under visible light); (c) UV-vis diffuse reflectance spectra (DRS); (d) Tauc-function band gap simulations of  $\text{g-C}_3\text{N}_4$ ,  $\text{V}_1\text{G}_5$  and  $\text{V}_1\text{G}_7$  heterojunctions.

hole pair separation and transfer within the  $\text{V}_1\text{G}_5$  heterojunction. However, the excessive addition of  $\text{V}_2\text{C}$  MXene to the heterojunction enlarged the arc radius, obstructing the charge transfer. Interestingly, under a dark environment, with the increase in the  $\text{V}_2\text{C}$  MXene contents, the arc radius lessened, which is probably owing to the MXene that enabled to trap electrons.<sup>57-59</sup> Moreover, the PL spectra further verified that the  $\text{V}_x\text{G}_y$  heterojunction manifested significant photocatalyst activity after exposing to visible light (450–800 nm), which corresponded with the results of electrochemical impedance spectroscopy.

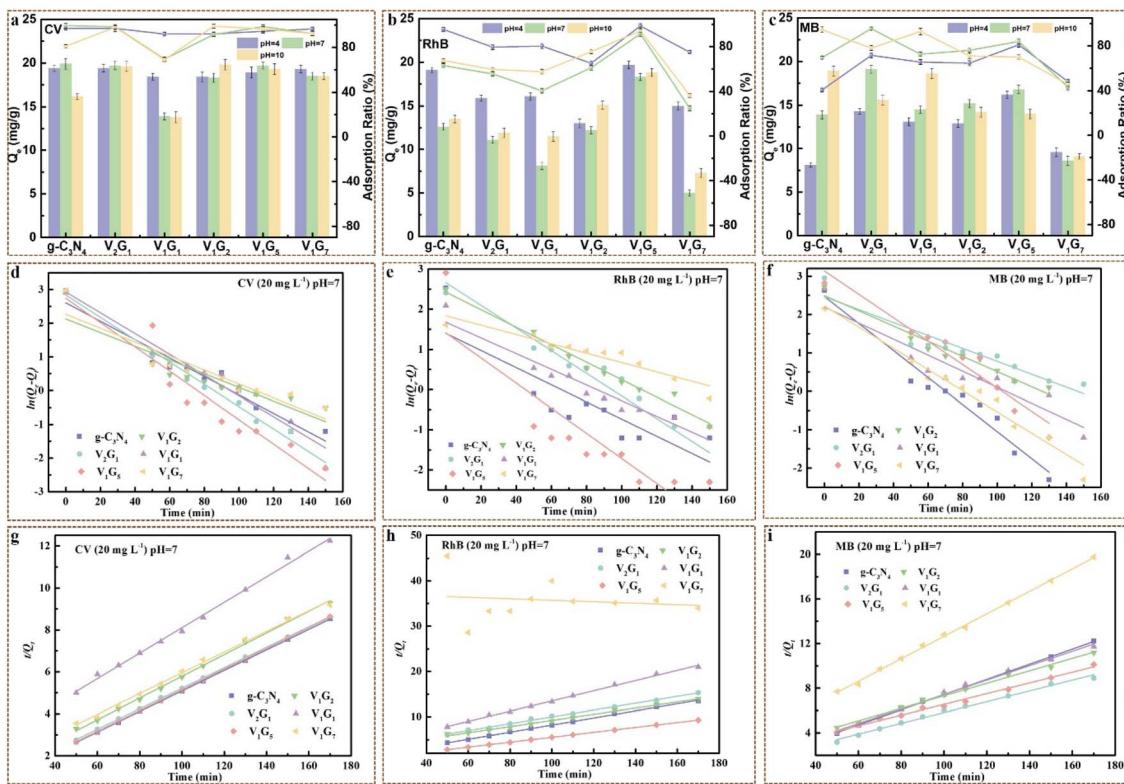
**2.2.3 UV-vis diffuse reflectance spectra.** UV-vis diffuse reflectance spectra (DRS) were carried out to understand the light absorption behaviour of the samples. The adsorption spectra of  $\text{g-C}_3\text{N}_4$ ,  $\text{V}_1\text{G}_5$  and  $\text{V}_1\text{G}_7$  heterojunctions are shown in Fig. 8c. The absorption edges of  $\text{g-C}_3\text{N}_4$ ,  $\text{V}_1\text{G}_5$  and  $\text{V}_1\text{G}_7$  were apparently observed at about 450 nm, which was in the blue region of the visible spectrum. Compared with  $\text{g-C}_3\text{N}_4$ ,  $\text{V}_1\text{G}_5$  and  $\text{V}_1\text{G}_7$  heterojunctions displayed enhanced light absorption behavior in both ultraviolet and visible regions, ascribed to the broadband adsorption of  $\text{V}_2\text{C}$  MXene across the solar spectrum.<sup>6</sup> Moreover, the absorbance of the  $\text{V}_1\text{G}_5$  heterojunction was almost 10 times than that of  $\text{g-C}_3\text{N}_4$  within the visible light region (450–800 nm) (Fig. 5c). The enhancement of light absorption capacity offered a higher photothermal effect to foster the photocatalytic reaction.<sup>51,60</sup> However, the absorbance of the  $\text{V}_1\text{G}_7$  heterojunction across the solar spectrum was slightly weaker than that of the  $\text{V}_1\text{G}_5$  heterojunction, which

demonstrated that the excessive proportion  $\text{V}_2\text{C}$  MXene in the  $\text{V}_x\text{G}_y$  heterojunctions was detrimental to the absorption capacity for solar light. Further analysis of energy bandgap was performed to evaluate the optical performances of these samples. The bandgaps of  $\text{g-C}_3\text{N}_4$ ,  $\text{V}_1\text{G}_5$  heterojunction and  $\text{V}_1\text{G}_7$  heterojunction simulated by Tauc-function model were 2.68 eV, 2.54 eV and 2.62 eV, respectively (Fig. 5d). Thereinto, the  $\text{V}_1\text{G}_5$  heterojunction possesses the narrowest bandgap (2.54 eV), which means it enables the absorption of the greatest range of wavelengths in the visible spectrum, leading to the best photocatalytic activity.

### 2.3 Adsorption behaviour

The adsorption behaviour of  $\text{V}_x\text{G}_y$  was explored through the adsorption tests in different dye solutions (CV, RhB and MB) with the same original content of  $20 \text{ mg L}^{-1}$  at different pH values (pH = 4, 7 and 10). The adsorption capacity ( $Q_t$ ,  $\text{mg g}^{-1}$ ) of  $\text{V}_x\text{G}_y$  to all kinds of dyes at different pH values rapidly increased in the first 60 min and reached equilibrium after 100 min (Fig. S8†). As shown in Fig. 6a, the adsorption ratios (AR) of  $\text{V}_x\text{G}_y$  to CV were above 90% within 170 min, which was much higher than that of  $\text{V}_x\text{G}_y$  to RhB and MB, attributed to the smaller molecules of CV. With the increase in the  $\text{g-C}_3\text{N}_4$  contents, the AR to CV ratio slightly changed in acidic environment, presenting the tendency to decline and then rise, while dramatic fluctuations of AR were observed in the neutral and alkaline environment, showing the same trend as that in





**Fig. 6**  $Q_e$  and the adsorption ratio of (a) CV, (b) RhB and (c) MB at equilibrium in different pH environment onto  $V_xG_y$  heterojunctions; Adsorption kinetics for the adsorption of (d) CV, (e) RhB and (f) MB at pH value of 7 onto  $V_xG_y$  heterojunctions fitted by the pseudo-first-order kinetic model; adsorption kinetics for the adsorption of (g) CV, (h) RhB and (i) MB at pH value of 7 onto  $V_xG_y$  heterojunctions fitted by the pseudo-second-order kinetic model.

acidic environment. According to the results of zeta potential (Fig. 4b),  $V_xG_y$  protonized in acid solution, while more ionized in basic solution, which means that the adsorption ratios of  $V_xG_y$  in basic solution would be much better than that in acidic solution. However, the results of the adsorption experiments did not coincide with the supposition. For RhB, the AR in the solution with pH value of 4 was distinctly higher than that in the solution with pH value of 7 and 10. Thus, the pKa of RhB was highly related to the adsorption results. It is known that the pKa of RhB is 3.2. When the pH value was low,  $-COOH$  on RhB barely ionized, leading to a positive charge on RhB, which was beneficial to the adsorption process. With increasing pH value,  $-COOH$  on RhB gradually ionized until it was transformed to a zwitterion establishing dimer RhB molecules, which hardly entered the  $V_xG_y$ , thus impeding the adsorption process.<sup>61</sup> On the other hand, the discrepancy of AR was highly dependent on the contents of  $g-C_3N_4$  owing to the surface potential and occupation of adsorption sites of  $V_xG_y$ . Interestingly, the AR of  $V_1G_5$  to CV, RhB and MB was much higher than that of other samples and barely affected by the pH value of the dye solutions, which is probably attributed to the appropriate conjunction ratio between  $V_2C$  MXene and  $g-C_3N_4$  nanosheets, leading to the exposure of more adsorption sites and feasible surface potential.

The adsorption data of CV, RhB and MB by  $V_xG_y$  heterojunctions at different pH environment were analyzed by pseudo-

first-order kinetic model and pseudo-second-order kinetic model, respectively. The linear fitting lines and the corresponding kinetic parameters are shown in Fig. 6d–i, S9–S10 and Table S2–S10.<sup>†</sup> Remarkably, the adsorption process of CV, RhB and MB by the  $V_xG_y$  heterojunctions at different pH environment were all well simulated by the pseudo-second-order kinetic model with most of the correlation coefficients ( $R^2$ )  $\geq 0.99$ , while the  $R^2$  of the pseudo-first-order kinetic model was below 0.95 at even about 0.6 (Tables S2–S10<sup>†</sup>), illustrating that the adsorption processes were dominantly depended on chemisorption. For the adsorption process of CV by  $V_xG_y$  heterojunctions at pH 4, most of the  $R^2$  values of the pseudo-second-order kinetic model were above 0.99, while the  $R^2$  of the pseudo-second-order kinetic model adsorbed by  $V_1G_5$  was about 0.95, possibly affected by the interactions between  $V_2C$  MXene and  $g-C_3N_4$ . When the pH value improved to 7, all the  $R^2$  of pseudo-second-order kinetic model were above 0.99, while the  $R^2$  of pseudo-second-order kinetic model were above 0.99 at pH value of 10, except that of  $V_1G_1$ , further demonstrating that the ratio of  $V_2C$  MXene and  $g-C_3N_4$  and pH value of the environment simultaneously impacted the surface potentials of  $V_xG_y$  heterojunctions, leading to the transformation of predominance of adsorbing type and mechanism. Interestingly, the  $R^2$  of pseudo-second-order kinetic model was above 0.99 during the adsorption process by  $V_xG_y$  heterojunctions when the dye solutions (CV, RhB and MB) were adsorbed at pH 7,

illustrating that the adsorption processes by  $V_xG_y$  heterojunctions were dominated by chemisorption when the influences of acidity and alkalinity were eliminated. The chemisorption including electrostatic and hydrogen bonds between adsorbent and adsorbates guaranteed that the dye molecules are hardly desorbed from  $V_xG_y$  heterojunctions rinsed in pure water. The electrostatic interaction was easily influenced by the pH of the dye solutions, further confirming the characteristics of the adsorption behaviours of  $V_xG_y$  heterojunctions at different pH values.

On the other hand, the original concentrations of dye solutions were also highly relevant to the adsorption behavior of  $V_xG_y$  heterojunctions. As shown in Fig. 7, with the increase in the original contents of dye solutions (CV, RhB and MB), the adsorption capacities at equilibrium increased, while the adsorption ratios decreased.  $V_1G_5$  exhibited excellent adsorption property with adsorption ratio onto CV and RhB of 99.6% and 94%, respectively, while the adsorption ratio onto MB was only 66%. This was because MB has the lowest  $pK_a$  among the three dyes, leading to the largest differentials between it and the pH value (pH = 7), which means the least number of cationic functional groups on MB.<sup>48,62</sup> Notably, the original contents of dye solutions greatly influenced the adsorption capacity at equilibrium. When the original content of CV was 15 mg L<sup>-1</sup>,

the  $Q_e$  of CV onto  $V_1G_5$  was 14.4 mg g<sup>-1</sup>, while the  $Q_e$  of CV onto  $V_1G_5$  improved to 19.7 and 24.9 mg g<sup>-1</sup> when the original contents of CV solution increased to 20 mg L<sup>-1</sup> and 25 mg L<sup>-1</sup>, respectively. The same phenomena were also observed in RhB and MB solutions, verifying the excellent adsorption property of  $V_xG_y$  heterojunctions.

The adsorption data onto CV, RhB and MB with different original contents onto  $V_xG_y$  heterojunctions were fitting by the pseudo-first-order kinetic model and pseudo-second-order kinetic model (Fig. 7d-i and S12†). Analogously, the  $R^2$  of the pseudo-second-order kinetic model was much higher than that of the pseudo-first-order kinetic model during the adsorption process by  $V_xG_y$  heterojunctions when the dye solutions (CV, RhB and MB) had different original contents (15, 20, 25 mg L<sup>-1</sup>) at a pH value of 7, further confirming that the adsorption process is primarily based on chemisorption (Tables S11–S16†). It was clear that the  $R^2$  of both pseudo-first-order kinetic model and pseudo-second-order kinetic model is barely related to the original contents of the dye solution. Notably, with increasing original contents of dye solutions, the adsorption rate was prominently boosted probably owing to the higher differential concentration. In addition,  $Q_{e, exp}$  obtained through adsorption experiments and  $Q_{e, cal}$  calculated through (E-7) were close to each other, illustrating that the adsorption processes were

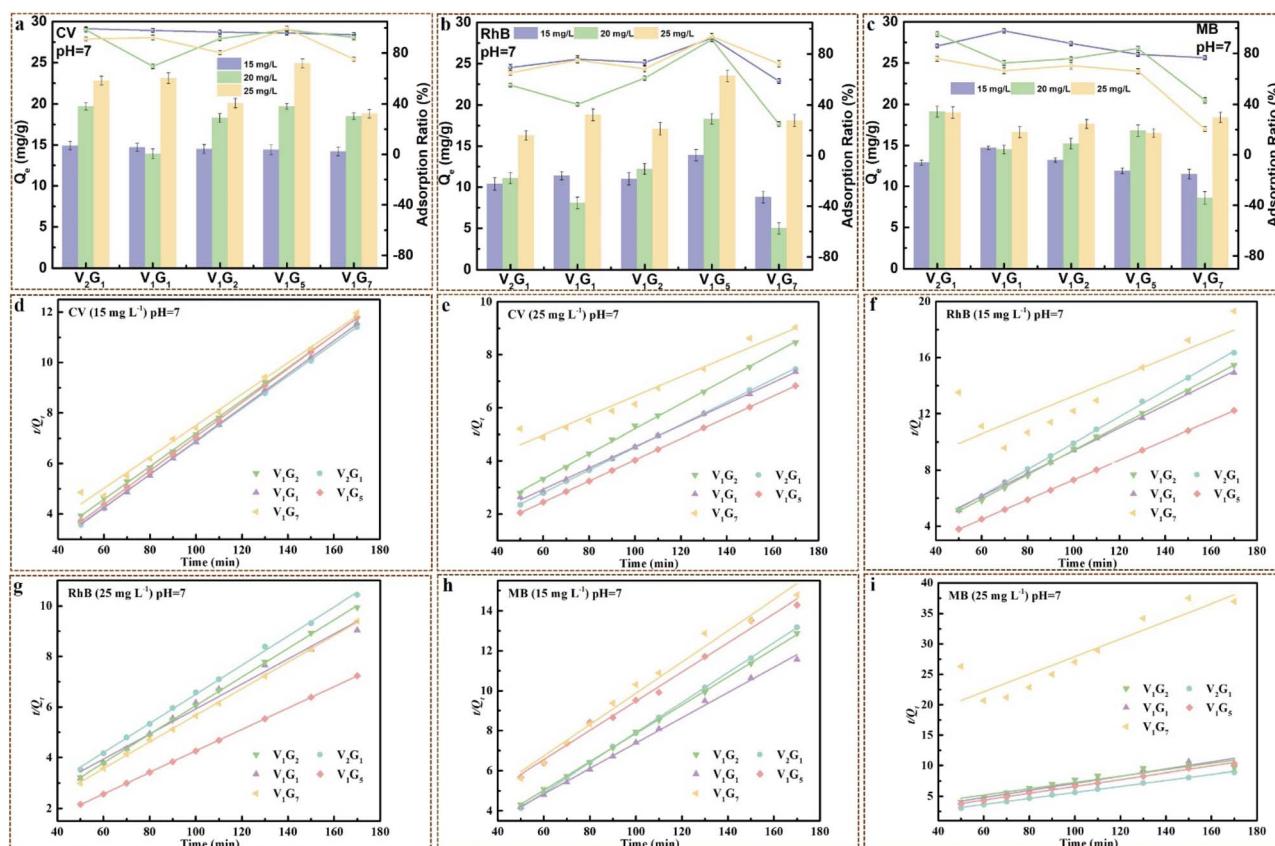


Fig. 7  $Q_e$  and the adsorption ratio of (a) CV, (b) RhB and (c) MB at equilibrium with different original contents at pH 7 onto  $V_xG_y$  conjunctions; adsorption kinetics on the adsorption of CV with original contents of (d) 15 mg L<sup>-1</sup> and (e) 25 mg L<sup>-1</sup>; RhB with original contents of (f) 15 mg L<sup>-1</sup> and (g) 25 mg L<sup>-1</sup>; MB with original contents of (h) 15 mg L<sup>-1</sup> and (i) 25 mg L<sup>-1</sup> at pH 7 onto  $V_xG_y$  heterojunctions fitted by the pseudo-second-order kinetic model.



wonderfully simulated by pseudo-second-order kinetic model (Tables S11–S16†). For instance, the  $Q_e$ , exp of  $V_1G_5$  onto CV (25 mg L<sup>-1</sup>, pH = 7) was 24.9 mg g<sup>-1</sup>, while  $Q_e$ , cal of  $V_1G_5$  onto CV (25 mg L<sup>-1</sup>, pH = 7) was 25.05 mg g<sup>-1</sup>.

#### 2.4 Photodegradation performance

The photodegradation performances of  $V_xG_y$  heterojunctions were investigated for CV, RhB and MB at different pH values with different initial concentrations under the visible light supplied by a metal halide lamp (50 W). As shown in Fig. 8a–c, the removal ratios of  $V_1G_5$  heterojunctions for CV at pH value of 4, 7 and 10 were 95%, 99.5% and 98%, respectively, within 80 min (adsorption in dark environment for 50 min and then irradiation under visible light for 27 min), which was much higher than that of pure  $g\text{-}C_3N_4$ . In addition, the removal ratios of the  $V_1G_5$  heterojunction for RhB at different pH values were all above 90%, while for MB, they were just about 80%. On the other hand, the removal ratios of the  $V_1G_5$  heterojunction for CV with different original contents of 15, 20 and 25 mg L<sup>-1</sup> at pH 7 were 98%, 99.5% and 96%, respectively, illustrating that the original contents of CV barely influence the purifying capacity of heterojunctions (Fig. S13†). Remarkably, the removal ratios of the  $V_1G_5$  heterojunction for RhB with different original contents of 15, 20 and 25 mg L<sup>-1</sup> at pH 7 were 96.6%, 92% and 98.8%, respectively, further certifying the extraordinary

purifying capacity of the  $V_1G_5$  heterojunction for dye wastewater (Fig. S13†). However, the removal ratios of heterojunctions for MB were much lower than that of CV and RhB within 80 min, ascribed to the final removal ratio dependent on the two processes of adsorption and photodegradation, whereas the adsorption ratios of MB onto the heterojunctions were much lower than that of CV and RhB, resulting in the same surface potential between the heterojunctions and MB. However, the removal ratios of the  $V_2G_1$  heterojunction for MB reached 99.3% when the irradiation time was prolonged to 90 min (total time of 140 min), which means that the photodegradation process enabled to nearly completely remove MB molecules. The adsorption and photocatalysis by the  $V_1G_5$  heterojunction conjointly disposed dye wastewater, exhibiting the high-efficiency disposal capacity, which enabled the almost thorough removal of dye molecules within 80 min including an irradiation time of only 27 min.

### 3 Proposed mechanism for purifying water pollution

#### 3.1 Proposed mechanism for adsorption behaviour

Based on the characteristics of adsorption of  $V_xG_y$  heterojunctions onto different dyes and combined with the results of BET analysis, zeta potential, adsorption behaviour and

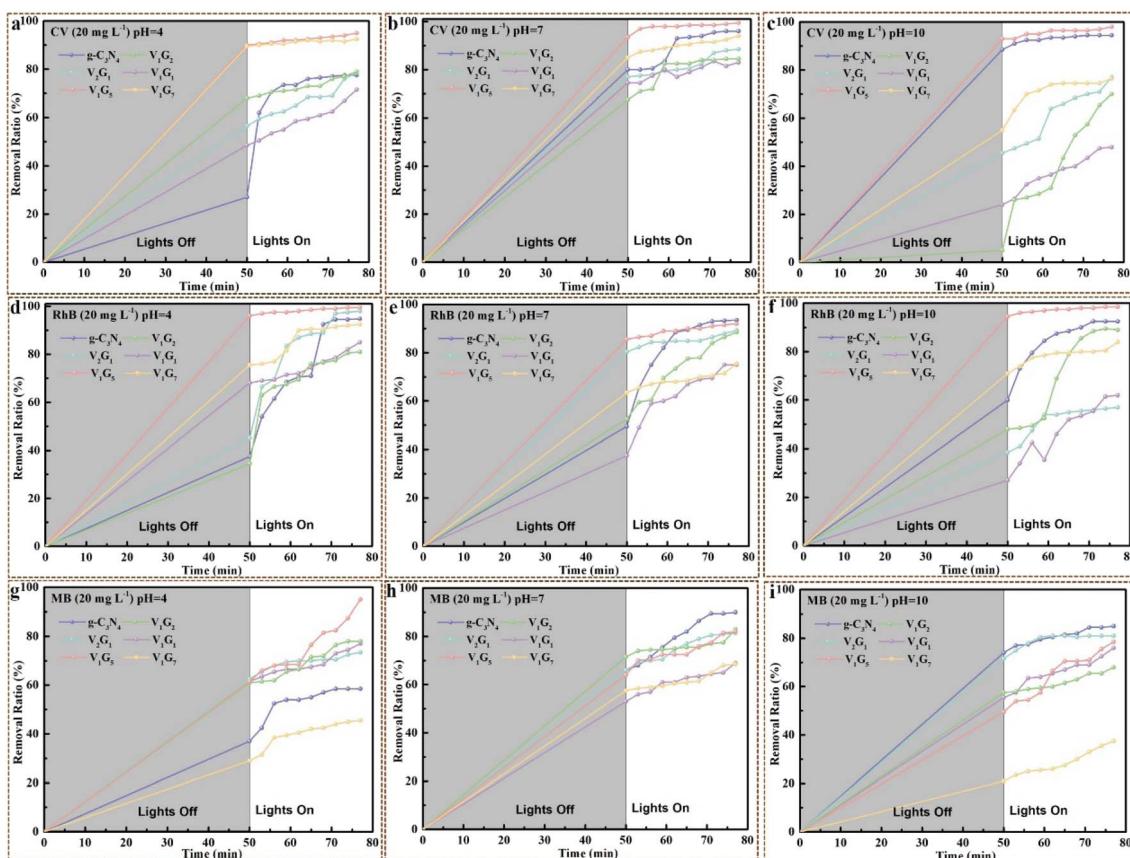


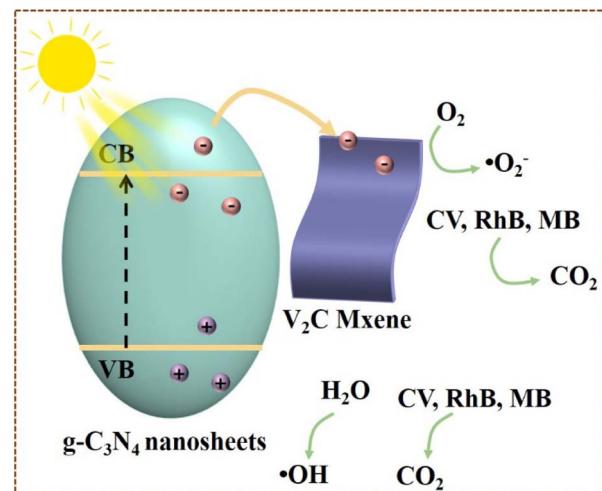
Fig. 8 Removal ratio of  $V_xG_y$  heterojunctions for CV (original content of 20 mg L<sup>-1</sup>) at (a) pH = 4, (b) pH = 7, (c) pH = 10; for RhB (original content of 20 mg L<sup>-1</sup>) at (d) pH = 4, (e) pH = 7, (f) pH = 10; for MB (original content of 20 mg L<sup>-1</sup>) at (g) pH = 4, (h) pH = 7, (i) pH = 10.



adsorption kinetics fittings, the mechanism of adsorption behaviour of  $V_xG_y$  heterojunctions was proposed in this work. According to the results of BET analysis,  $g\text{-C}_3\text{N}_4$  possessed the largest specific surface area among all the samples while the  $Q_e$  of  $g\text{-C}_3\text{N}_4$  onto dyes is not the highest one. Obviously, the specific surface area of the  $V_2\text{C}/g\text{-C}_3\text{N}_4$  heterojunction was smaller than that of  $g\text{-C}_3\text{N}_4$ , while the  $Q_e$  and adsorption ratio at equilibrium for the  $V_2\text{C}/g\text{-C}_3\text{N}_4$  heterojunction were much higher than that of pristine  $g\text{-C}_3\text{N}_4$ , illustrating that the adsorption process scarcely depended on physical adsorption through mesopores within the materials. It was also confirmed by the fitting results of the first-order kinetic model (Fig. 6 and 7). The results of zeta potential further corroborated this supposition. As shown in Fig. 4b, all the samples exhibited electronegativity except the  $V_2\text{G}_1$  heterojunction, while CV, RhB and MB were all cationic dyes, which means that chemisorption dominated the whole adsorption process. This supposition is also corroborated by the coinciding fitting results of the second-order kinetic model,  $R^2$  is above 0.99 (Fig. 6, 7 and Tables S2–S10†).

### 3.2 Proposed mechanism for photocatalytic degradation

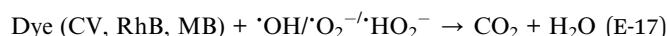
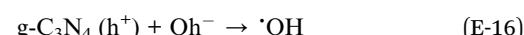
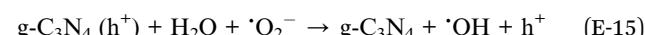
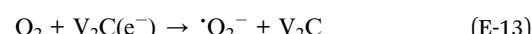
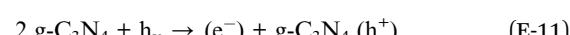
Based on the series characterizations and experiments, the photocatalytic mechanism was proposed to interpret the photocatalytic process of  $V_2\text{C}/g\text{-C}_3\text{N}_4$  heterojunctions on CV, RhB and MB. PL and DRS analysis revealed that  $V_2\text{C}$  MXene could barely generate  $e^-$  and  $h^+$  and hardly recombine  $e^-$  and  $h^+$  pairs. These findings indicated that  $g\text{-C}_3\text{N}_4$  was the main photocatalyst responsible for the electron–hole pairs generation, while  $V_2\text{C}$  MXene worked as a metallic conductor to expedite the photoreaction. This was the possible mechanism of charge transfer of heterojunctions that we supposed. According to the reported work, eqn (E10)–(E17) display the conversion and establishment of different active groups of  $V_2\text{C}/g\text{-C}_3\text{N}_4$  heterojunctions throughout photocatalytic degradation onto dyes.<sup>63,64</sup> Scheme 2 describes the process of generating photocarriers by  $g\text{-C}_3\text{N}_4$ . The  $e^-$  and  $h^+$  were generated at the VB of  $g\text{-C}_3\text{N}_4$  under visible light. The charge separation of photogenerated charges occurred, where  $e^-$  are excited to CB while  $h^+$  remained in the VB (E-11). The shift of C 1s, N 1s and V 2p in  $V_2\text{C}/g\text{-C}_3\text{N}_4$  heterojunctions (XPS spectra) further testified the transfer of  $e^-$  from  $g\text{-C}_3\text{N}_4$  (electron-rich) to  $V_2\text{C}$  MXene (electron-deficient). The  $e^-$  trapping centre was established within  $V_2\text{C}/g\text{-C}_3\text{N}_4$  heterojunctions, ascribed to the difference in the work function and the band alignment, generating a built-in internal electric field.<sup>51</sup>  $V_2\text{C}$  MXene as cocatalyst with higher metallic electrical conductivity coupled with  $g\text{-C}_3\text{N}_4$  would accept the electrons, leading to less recombination of electron–hole pairs. Moreover, the heterojunctions caused charge redistribution, hindering the backward flow of  $e^-$  at the  $V_2\text{C}$  MXene from returning to  $g\text{-C}_3\text{N}_4$  and recombining with  $h^+$ . Therefore, more reactions with protons occurred to degrade dye molecules because of the more available  $e^-$  at the reduction sites. This was one of the most important reason to enhance the photocatalytic activity after coupling with  $V_2\text{C}$  MXene, which was determined by the PL peak depression in  $V_2\text{C}/g\text{-C}_3\text{N}_4$  heterojunctions.<sup>51</sup> The  $h^+$  at VB



Scheme 2 Photocatalytic degradation mechanism by  $V_2\text{C}/g\text{-C}_3\text{N}_4$  heterojunction under visible light.

reacted with water (oxidation reaction) to produce protons and oxygen. Notably, with the increase of recombination rates of  $e^-$  and  $h^+$ , the number of available  $e^-$  and  $h^+$  participating in redox reactions is reduced.

On the other hand, (E-13) and (E-14) expounded the production of  $\cdot\text{OH}$ . Specifically,  $O_2$  was sequentially reduced and protonated by  $e^-$  at the CB of  $g\text{-C}_3\text{N}_4$ . The negative CB of  $g\text{-C}_3\text{N}_4$  donated  $e^-$  to capture  $O_2$  and reduce them to superoxide anion ( $\cdot O_2^-$ ). Then,  $\cdot O_2^-$  were protonated by  $H^+$  to generate  $\cdot\text{HO}_2^-$  and decomposed by  $e^-$  to give  $\cdot\text{OH}$ . These photo-generated electrons ( $\cdot\text{OH}$ ,  $\cdot\text{HO}_2^-$  and  $\cdot O_2^-$ ) would react with dye molecules (CV, RhB, MB) to finally produce  $\text{CO}_2$  and  $\cdot\text{OH}$ .



## 4. Conclusions

In summary, the  $V_xG_y$  heterojunctions were synthesized through a one-pot method. The as-prepared  $V_xG_y$  heterojunctions exhibited excellent capacity for remedying water pollution. The adsorption ratios of  $V_1\text{G}_5$  heterojunctions onto CV and RhB ( $25 \text{ mg L}^{-1}$ ,  $\text{pH} = 7$ ) were as high as 99.6% and 94%, respectively, within 170 min at room temperature in dark



environment. After adsorbing for 50 min, the dye solution equipped with  $V_xG_y$  heterojunctions was exposed to visible light for 30 min (the intensity of light irradiation was  $100 \text{ W m}^{-2}$ ), displaying the final removal ratio onto CV, RhB and MB of 99.5%, 99.5%, and 95%, respectively, which is much higher than that of the published works under such low intensity of light irradiation.<sup>65-69</sup> The results of DRS, PL, and band structure illustrated that the metal-like nature of  $V_2C$  MXene as a mediator coupled with  $g\text{-C}_3\text{N}_4$  dramatically accelerated the separation of photogenerated charge carriers and alleviated the photogenerated recombination  $e^- - h^+$  pairs. In addition, the extraordinary 2D/2D heterojunction structure showed high charge separation capacity, which remarkably advanced the efficiency of electron/hole pair separation and transfer within  $V_xG_y$  heterojunctions. The one-pot approach to fabricate the 2D  $V_2C$  MXene/2D  $g\text{-C}_3\text{N}_4$  nanosheet heterojunctions proposed in this paper provides a new idea to synthesize high-efficiency composite photocatalysts, which is significant for purifying dye wastewater.

## Data availability

The data supporting this article are available from the corresponding author upon reasonable request. The data supporting this article have been included as part of the ESI.†

## Author contributions

Conceptualization: Shishan Xue, Dengliang He, Shuxin Liu, Ning Chen; data curation: Herong Zhang, Yuning Zhang, Yu Wang, Yurong Zeng; funding acquisition: Shishan Xue, Shuxin Liu, Ning Chen; investigation: Yu Wang, Yurong Zeng, Herong Zhang; project administration: Shishan Xue, Dengliang He; writing – original draft: Shishan Xue, Dengliang He; writing – review & editing: Shishan Xue, Dengliang He.

## Conflicts of interest

There are no conflicts to declare.

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