Enhanced Gas Sensing Performance of ZnO/Ti$_3$C$_2$T$_x$ MXene Nanocomposite

Qui Thanh Hoai Ta 1,†, Deepika Thakur 2,† and Jin-Seo Noh 1,*

1 Department of Physics, Gachon University, 1342 Seongnam-daero, Sujeong-gu, Seongnam-si 13120, Korea
2 Department of Agriculture, Forestry and Bioresources, Seoul National University, 1 Gwanak-ro, Gwanak-gu, Seoul 08826, Korea
* Correspondence: jinseonoh@gachon.ac.kr; Tel.: +82-317505611
† These authors contributed equally to this work.

Abstract: A representative of titanium carbide MXene, Ti$_3$C$_2$T$_x$ is a promising candidate for high performance gas sensing and has attracted significant attention. However, MXene naturally has a multilayer structure with low porosity, which prevents its gas-sensing activity. Zinc oxide (ZnO) has long been utilized as a gas detector. Despite its good response to multiple gases, high operation temperature has limited its widespread use as a gas-sensing material. In this study, a room-temperature toxic gas sensor was prepared from ZnO/Ti$_3$C$_2$T$_x$ MXene nanocomposite consisting of 2D few-layered MXene and 1D ZnO nanoparticles. A simple technique for synthesizing the nanocomposite was established. The physicochemical properties of the nanocomposite were fine-controlled with more active sites and higher porosity. The sensitivity and gas-selectivity of the sensing material were closely examined. The nanocomposite showed enhanced response and recovery behaviors to toxic gases, which outperformed pure Ti$_3$C$_2$T$_x$ MXene and pure ZnO. This study offers a practical strategy by which to increase the gas-sensing performance of Ti$_3$C$_2$T$_x$ MXene, and expands comprehensive understanding of the gas-sensing process of ZnO/Ti$_3$C$_2$T$_x$ p-n heterostructure.

Keywords: Ti$_3$C$_2$T$_x$ MXene; ZnO; nanocomposite; gas sensor

1. Introduction

MXenes, also known as two-dimensional (2D) carbides, nitrides, and carbonitrides, have gained huge attention due to their unique surface properties, morphology, and potential applications [1–3]. MXenes are represented by the general formula: M$_{n+1}$X$_n$T$_x$ (n = 1–4), where M stands for transition metals, X stands for carbon or nitrogen, and T$_x$ holds for a functional group such as fluorine, hydroxyl, and oxygen [4–6]. Mostly, MXenes have been researched for gas sensing due to the ease of tuning the surface termination groups. Among various MXenes, titanium carbide (Ti$_3$C$_2$T$_x$ MXene) has been studied the most, due to its high electron density and sensitive interactions with toxic gas molecules [2,7,8].

However, while MXenes have proven to be useful, their poor stability in humid environments and loss in surface area after hydration limit its potential applications [9–11]. In addition, low-porosity and high operating temperatures have caused various complications during their synthesis process [12,13]. This is why there is a need for the incorporation of other materials in combination with pure MXenes.

Some efforts have been made by scientists to sensitize Ti$_3$C$_2$T$_x$ MXene [14–17]. In our previous study, we synthesized Si-doped TiO$_2$/Ti$_3$C$_2$T$_x$ hybridstructure using a diffusion process of Si atom at 1000 °C for enhancing NO$_2$ sensing activity [18]. Zhao et al. explored PANI/Ti$_3$C$_2$T$_x$ nanocomposite to enhance ethanol sensing properties [19]. In 2021, Yang et al. studied crumpled ZnO/Ti$_3$C$_2$T$_x$ sphere, using ultrasonic spray pyrolysis technique to improve the NO$_2$ sensing performance [20]. In fact, in these previous works, a certain unilateral parameter of the sensor was enhanced. The comprehensive sensing activity of the response and recovery values should therefore be further improved [21,22].
In this work, we have highlighted the enhanced gas sensing performance of ZnO/Ti3C2Tx MXene nanocomposite at room-temperature. To increase the surface area of the material, we incorporated 1D ZnO nanoparticles on the surface of 2D few-layered MXene. The ZnO/Ti3C2Tx MXene nanocomposite significantly improved the performance for toxic gas sensing. In addition, it was found that the response and recovery time of the ZnO/Ti3C2Tx MXene nanocomposite outperformed the pure ZnO NPs and Ti3C2Tx MXene. Furthermore, the gas-sensing process of ZnO/Ti3C2Tx p-n heterostructure revealed better stability and performance than Ti3C2Tx MXene in gas sensing.

2. Materials and Methods

2.1. Materials and Chemicals

Ti3AlC2 powder was provided by 11 Technology Company (China). Zinc acetate dihydrate (Zn(CH3COO)2•2H2O), potassium hydroxide (KOH), and methanol (CH3OH) were obtained from Sigma-Aldrich (St. Louis, MO, USA). Hydrofluoric acid (HF, 48–51%) was bought from Fisher Scientific (Ward Hill, MA, USA). Ethyl alcohol (C2H5OH) was purchased from Daeung Chem (Gyeonggi-do, Korea). All the chemicals were used as provided without any further treatment.

2.2. Preparation of Ti3C2Tx MXene and ZnO/Ti3C2Tx Nanocomposites

First, multilayered Ti3C2Tx MXenes were prepared by selectively etching Al layers from the pristine Ti3AlC2 powder. Typically, a 2.0 g of pristine Ti3AlC2 MAX phase was gradually immersed into 56 mL of a concentrated HF under continuous stirring at 300 rpm in an ice bath to avoid heat generation, and the mixed solution was transferred to an oil bath at 50 °C [23,24]. After one day of reaction, the MXene multilayer structures were collected and washed several times with DI water. Subsequently, a sample of few-layered Ti3C2Tx MXene was prepared by ultrasonic probe sonicator (Sonics & Materials INC, Newtown, CT, USA) for around 30 min using multilayered MXene. Then, the obtained precipitate was dried overnight using a freeze-drying system (iShinBioBase Co. Ltd., Gyeonggi-do, Korea) for further experiments.

ZnO/Ti3C2Tx nanocomposites were synthesized by a simple technique, as schematically shown in Figure 1. Briefly, varying amounts (100, 200, and 300 mg) of the few-layered Ti3C2Tx MXene powders were added to 25 mL of Zn2+ solution. A total of 340 mg of KOH was dispersed in 15 mL of methanol under heat [25]. After 30 min of reaction, the obtained precipitates were continuously washed to remove the byproducts and named following real ratio as ZT1 (163:100), ZT2 (163:200), and ZT3 (163:300), respectively. Moreover, the pure ZnO nanoparticles were prepared for comparison in the same parameters without any Ti3C2Tx MXene.

![Figure 1. Schematic synthesis procedure of ZnO/Ti3C2Tx heterostructure.](image)

2.3. Characterizations and Gas Sensing Measurements

The X-ray diffraction (XRD) patterns of as-prepared samples were obtained using a high resolution X-ray diffractometer (Rigaku, SmartLab, Tokyo, Japan) equipped with 3 kW Cu Kα radiation. The morphology of all samples was collected using scanning electron microscopy (FESEM, Hitachi, S-4700, Tokyo, Japan) and high resolution transmission electron microscopy (HR-TEM, Tecnai, Hillsboro, OR, USA). The bonding states and surface characteristics of selected samples were analyzed by X-ray photoelectron spectroscopy (XPS, Kα plus, Thermo Fisher Scientific, Waltham, MA, USA). The Brunauer-Emmett-Teller (BET)
analysis was measured using a nitrogen adsorption-desorption system (Micromeritics, ASAP 2020, Norcross, GA, USA).

To achieve gas sensing properties, concentrated suspensions of the as-prepared samples were covered on a glass substrate (10 × 10 mm). The gas sensing measurements were measured in a stainless-steel chamber (682 cm$^3$) at room temperature. The real-time resistance of the samples, when exposed to the toxic target gas ($R_g$) and synthetic air ($R_a$), was measured through Au wires using a digital source measure unit (SMU, Keithley 2450) [26]. Five target gases (CO$_2$, H$_2$, CH$_4$, NH$_3$, and NO$_2$) were used as the tested gases at high and low humidity ambient. The different gas concentrations of the toxic gas were produced by mixing it with synthetic air using a flow and pressure controller (GMC 1200). The gas sensor response value of the nanocomposite was expressed using the following equation [27]:

$$\text{Response} \, (\%) = \frac{(R_g - R_a)}{R_a} \times 100$$

3. Results and Discussion

3.1. Microstructures and Structural Components

Observation of morphological surfaces of the Ti$_3$AlC$_2$ MAX phase, Ti$_3$C$_2$Tx MXene, pure ZnO nanoparticles, and ZnO/Ti$_3$C$_2$Tx nanocomposite were analyzed using FESEM with high- and low-magnification (FESEM, Hitachi, S-4700, Tokyo, Japan). As displayed in Figure 2a, the pristine MAX phase has terraced microstructures with an average grain below 8 µm. After etching the MAX phase in concentrated HF solution, the micromorphology of multilayered Ti$_3$C$_2$Tx MXene accounts for an accordion-like sandwich, which exhibited successful Al-selective etching (Figure 2b). After sonicating the MXene multilayer for 30 min, a few-layered Ti$_3$C$_2$Tx MXene were regularly obtained. It can be observed from the inset magnified image in Figure 2c that the thickness of the Ti$_3$C$_2$Tx MXene was around 150–350 nm, which could be due to the few-layered Ti$_3$C$_2$Tx MXene. The as-synthesized ZnO displayed a particle shape with a diameter of 60–100 nm, as shown in Figure 2d. Compared to the pure ZnO nanoparticles, the few-layered Ti$_3$C$_2$Tx MXene might suppress the growth and accumulation of ZnO nanoparticles. The heterostructure could support, not only faster, but also excess diffusion paths for toxic target gases.

By focusing in further, the SEM image of ZnO/Ti$_3$C$_2$Tx nanocomposite clearly showed that the ZnO exhibited particle morphology, and featured a side anchored freely on the MXene two sides, suggesting efficient assembly between few-layered Ti$_3$C$_2$Tx MXene sheets and ZnO particles during the treatment process (Figure 2e,f). It was estimated that a single-layer Ti$_3$C$_2$Tx flake (2 nm thick) and few-layer (2–5 nm thick) flakes. The SEM and elemental mapping measurements of ZnO/Ti$_3$C$_2$Tx heterostructure (ZT2) suggested the presence of C, O, Ti, and Zn components in the nanocomposite. They displayed a highly homogeneous distribution of C, O, Ti, and Zn (Figure 3a). Moreover, the EDX spectrum also exhibited obvious C, O, Ti, and Zn elements, which were valuable for the stability and performance of the nanocomposite. As presented in Figure 3b,c, the HR-TEM images demonstrated the modification between the two interfaces, where both Ti$_3$C$_2$Tx MXenes and ZnO were cross-linked to each other. This local communication could enhance the interaction between electrons and gas molecules.
Figure 2. SEM images of (a) Ti$_3$AlC$_2$ MAX phase, (b) Ti$_3$C$_2$Tx MXene multilayer, (c) Ti$_3$C$_2$Tx MXene few-layer, (d) pure ZnO nanoparticles, and (e,f) ZnO/Ti$_3$C$_2$Tx hybrid structure. The insets show low-magnification SEM images.

Figure 3. (a) SEM images and EDX spectrum with EDX element maps of Ti, C, O, Zn, and (b,c) HR-TEM images of ZnO/Ti$_3$C$_2$Tx heterostructure.
The XRD patterns of prepared samples are displayed in Figure 4a. Well-defined diffraction peaks can be seen at 2θ = 9.5°, 19.1°, 36.7°, 38.7°, 38.9°, and 60.1°, which are indexed to the planes (002), (004), (101), (104), (105), (110), respectively (JCPDS card No.52-0875) [16]. After the etching development, a remarkable peak (002) in pure Ti₃C₂Tx MXene exhibits that Al was successfully corroded from the Ti₃AlC₂ MAX phase. The chemical bonding between each layer was decreased by increasing the c parameter. Meanwhile, ZnO showed preferred growth in the (101) orientation in the nanocomposite, and prominent ZnO peaks were also observed at 2θ = 31.7° and 34.4°, which were indexed to (100) and (002) planes, respectively. These peaks corresponded with the pure ZnO and the wurtzite structure (JCPDS card No.36-1451) [28]. The absence of any TiO₂ peaks revealed that oxidation seemed to not occur in the combination procedure. Thus, the XRD results of synthesized samples were completely expected. As displayed in Figure 4b, both samples illustrated a type IV isotherm plot with a relative pressure from 0.42 to 1.0, indicating H₃-type hysteresis circles [29]. Moreover, the BET-specific surface area (SSA) of ZnO/Ti₃C₂Tx nanocomposite was calculated to be 29.7 m²/g, which is a 3.9-fold increase compared with the SSA of the Ti₃C₂Tx MXene (7.5 m²/g). The results evidenced that the SSA of ZnO/Ti₃C₂Tx nanocomposite was significantly improved owing to ultra-sonication and decoration, which were expected to improve the active sites and enhance the transportation of electrons.

![Figure 4. (a) XRD patterns of the Ti₃AlC₂ MAX phase, Ti₃C₂Tx MXene, pure ZnO, and ZnO/Ti₃C₂Tx nanocomposite. (b) N₂ adsorption-desorption isotherms for Ti₃C₂Tx MXene, and ZnO/Ti₃C₂Tx nanocomposite.](image)

3.2. Chemical Binding States

The surface chemical and binding energy of ZnO/Ti₃C₂Tx nanocomposite was observed in detail using XPS measurement for Ti 2p, C 1s, Zn 2p, and O 1s. The results are displayed in Figure 5. The overall XPS survey spectrum of ZnO/Ti₃C₂Tx heterostructure is illustrated in Figure 5a, indicating the existence of Zn 2p, O 1s, Ti 2p, and C 1s. Meanwhile, Figure 5b–e reveals the various focused XPS spectra of individual elements. Figure 5b, showing a Ti 2p XPS spectra, indicates peaks at 454.9, 458.8, and 461.08 eV, corresponding to Ti-C, Ti-C and Ti-O, which may originate from C-Ti³⁺-Tx and C-Ti²⁺-Tx with Tx = -OH, -O₂-, -F, respectively [30]. Figure 5c reveals the C 1s high-resolution spectrum, fitted using two main components at 284.7 and 281.2 eV, which could originate from graphitic C=C, and C-Ti with a surface termination group, respectively [31]. Additionally, two main electronic states of Zn 2p, shown in Figure 5d, spotted at 1045.9 and 1023.0 eV by a spin separation of 22.9 eV, are assigned to Zn 2p₁/₂ and Zn 2p₃/₂, respectively [20]. As
shown in Figure 5e, the O 1s high-resolution spectrum of the nanocomposite exhibited that the two prominent binding energy peaks at 530.2 and 532.2 eV could be assigned to O-Ti and C-Ti-Tx (Tx = -OH), respectively [32]. Therefore, the above results could deduce that the optimized nanocomposite exhibited an enhanced gas sensing performance.

![Image](image_url)

**Figure 5.** (a) Overall XPS spectrum of ZnO/Ti3C2Tx nanocomposite (ZT2). Focused XPS spectra of (b) Ti 2p, (c) C 1s, (d) Zn 2p, and (e) O 1s, respectively.

### 3.3. Gas Sensing Performance and Sensing

In order to find the performance of the excellent gas-sensitive and selective activity of ZnO/Ti3C2Tx nanocomposite, the gas sensing activities of pure MXene, pure ZnO, and ZnO/Ti3C2Tx with different MXene weight ratios were also recorded and shown in Figure 6. Figure 6a,b illustrates the response of ZT1, ZT2, ZT3, and pure MXene sensors to NO2 concentrations at 5 ppm and 10 ppm, respectively. The response signal of pure ZnO did not detect at room temperature, indicating semiconductor characteristics. The response signals of all the sensors were enhanced with the increase of NO2 concentration, while the response of pure Ti3C2Tx MXene was not well developed with noise signals, due to metallic behavior. The responses to 5 and 10 ppm NO2 were calculated at 35% and 54%, respectively. The NO2 response and recovery properties of ZT2 at room temperature were more substantial than those of ZT1 and ZT3, which were inadequate (Figure 6b).

In addition, the saturation time of the ZnO/Ti3C2Tx heterostructure to NO2 was recorded. For this evaluation, the ZT2 sensor was exposed to NO2 at a time interval of 5 min. After 10 min, the sensing signal seemed to be saturated, as shown in Figure 6c. For further clarification, Figure 6d displays the sensing response to 10ppm of various gases (e.g., H2, NH3, CH4, NO2, CO2) based on the ZnO/Ti3C2Tx sensor. It can be observed that the ZT2 nanocomposite had the significant response to NO2 in comparison with other gases at low and high humidity.

In order to further explore the sensing activities of ZnO/Ti3C2Tx heterostructure to NO2 gas molecules, the ZT2 sensor was exposed to various NO2 concentrations ranging from 5 to 30 ppm. As illustrated in Figure 6e, the result increased with the rising concentration of NO2, exhibiting that the proposed ZT2 sensor was a remarkable NO2 molecule was logarithmic with the linear regression (R2) of 0.992 was acquired. Moreover, the repeatable cycle to 5–30 ppm NO2 molecules of the ZT2 sensor, provided in Figure 6f, reveals that the sensor possesses a repeatable characteristic. The gas sensing performance of Ti3C2Tx-based sensors is summarized in Table 1. Although our ZnO/Ti3C2Tx sensor did not show a remarkable NO2 response compared with other sensors, it is a promising candidate for NO2 detection at room temperature.
response compared with other sensors, it is a promising candidate for NO₂ detection at room temperature.

Table 1. The comparison of NO₂ sensing in other materials, including Ti₃C₂Tx MXene, was explored in previous literature.

| Material                        | Temp. (°C) | Concentration (ppm) | Response (%) |
|---------------------------------|------------|---------------------|--------------|
| 2D MoS₂/Ti₃C₂Tx [16]           | RT         | 10                  | 25           |
| 3D crumpled Ti₃C₂Tx/ZnO [20]    | RT         | 20                  | 22.5         |
| 2D/2D/2D Ti₃C₂Tx@TiO₂@MoS₂ [31] | RT         | 50                  | 55           |
| ZnO/Ti₃C₂Tx nanocomposite *     | RT         | 10                  | 54           |

* This work.

The improved NO₂ sensing activity of the nanocomposite was attributable to the creation of a heterogeneous p-n junction. When the ZnO/Ti₃C₂Tx MXene nanocomposite sensor was exposed to the air, oxygen molecules were covered and captured by the electrons on the material layers, as shown in Figure 7. At room temperature, most adsorbed oxygen molecules tend to create negative oxygen species (O₂⁻, O₂²⁻, and O⁻) at defects and active sites of the sensor layers, which play a predominant part in improved gas sensing [33–35].
During the checking of NO\textsubscript{2} sensing properties, it was expected that the NO\textsubscript{2} gas would react with O\textsuperscript{2−} species to form NO\textsubscript{3}− species, owing to the oxygen having lower electronegativity than the target gas \cite{20,36,37}. Simultaneously, NO\textsubscript{2} gas collected electrons and enhanced the depletion layer, thus improving the response and recovery values. Moreover, NO\textsubscript{2} gas can be absorbed on the surface of p-type Ti\textsubscript{3}C\textsubscript{2}T\textsubscript{x} MXene due to its oxidizing and electrophilic properties, resulting in reduced resistance \cite{38}. Essentially, ZnO nanoparticles with an n-doping effect were decorated with a number of negative oxygen species (O\textsuperscript{2−}) on the surface and supported an important role in the absorption of NO\textsubscript{2} \cite{39}. Under ambient humidity, the hydrogen bonding between NO\textsubscript{2} and H\textsubscript{2}O molecules further promoted the chemical adsorption of NO\textsubscript{2} \cite{40}. The electron donation reaction can be described as follows \cite{20,41,42}:

\[
\begin{align*}
O_2 (g) & \rightarrow O_2 (ads) \quad (2) \\
O_2 (ads) + e^- & \rightarrow O_2^- (ads) \quad (3) \\
O_2^- (ads) + e^- & \rightarrow 2O^- (ads) \quad (4) \\
O^- (ads) + e^- & \rightarrow O^2- (ads) \quad (5) \\
NO_2 (g) & \rightarrow NO_2 (ads) \quad (6) \\
NO_2 (ads) + e^- & \rightarrow NO_2^- (ads) \quad (7) \\
2NO_2 (ads) + O_2^- (ads) + 2e^- & \rightarrow 2NO_3^- (ads) \quad (8)
\end{align*}
\]

4. Conclusions

In summary, the preparation of ZnO/Ti\textsubscript{3}C\textsubscript{2}T\textsubscript{x} MXene nanocomposite by effective ultrasonication was investigated. In comparison with pure ZnO nanoparticles and Ti\textsubscript{3}C\textsubscript{2}T\textsubscript{x} MXene, the optimized nanocomposite achieved sensitivity and selectivity to NO\textsubscript{2} gas sensing. The response to 5 ppm NO\textsubscript{2} was calculated at 35% with a speedy recovery and long-term stability at room temperature. The high sensitivity NO\textsubscript{2} sensing activity of ZnO/Ti\textsubscript{3}C\textsubscript{2}T\textsubscript{x} MXene nanocomposite was ascribed to the active sites and defects, which were the p-n heterointerface contacts between ZnO nanoparticles and Ti\textsubscript{3}C\textsubscript{2}T\textsubscript{x} MXene. The results of this combination structure pave the way for the chemical sensing of Ti\textsubscript{3}C\textsubscript{2}T\textsubscript{x} MXene-based materials, thus providing a reference by which to understand the fundamentals of Ti\textsubscript{3}C\textsubscript{2}T\textsubscript{x} MXene gas sensing.

Author Contributions: Conceptualization, methodology, validation, formal analysis, investigation, resources, data curation, Q.T.H.T.; writing—original draft preparation, D.T.; writing—review and editing, visualization, supervision, project administration, funding acquisition, J.-S.N. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by a National Research Foundation of Korea (NRF) grant funded by the Korean government (MSIT) (No. 2019R1A2C1008746).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.
Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

References
1. Nguyen, T.P.; Nguyen, D.M.T.; Le, H.K.; Vo, D.-V.N.; Lam, S.S.; Varma, R.S.; Shokouhimehr, M.; Nguyen, C.C.; Van Le, Q. MXenes: Applications in Electrocatalytic, Photocatalytic Hydrogen Evolution Reaction and CO2 Reduction. *Mol. Catal.* 2020, 486, 110850. [CrossRef]
2. Asanori, B.; Gogotsi, Ü.G. *2D Metal Carbides and Nitrides (MXenes)*; Springer: Berlin, Germany, 2019; ISBN 3030190269.
3. Thanh, Q.; Ta, H.; Tran, N.M.; Noh, J. Rice Crust-Like ZnO/Ti3C2Tx MXene Hybrid Structures for Improved Photocatalytic Activity. *Catalysts* 2020, 10, 1140. [CrossRef]
4. Ta, Q.T.H.; Tran, N.M.; Noh, J.S. Pressureless Manufacturing of Cr2A1C Compound and the Temperature Effect. *Mater. Manuf. Process.* 2021, 36, 200–208. [CrossRef]
5. Anasori, B.; Lukatskaya, M.R.; Gogotsi, Y. 2D Metal Carbides and Nitrides (MXenes) for Energy Storage. *Nat. Rev. Mater.* 2017, 2, 16098. [CrossRef]
6. Thi, Q.V.; Patil, S.A.; Katkar, P.K.; Rabani, I.; Patil, A.S.; Ryu, J.; Kolekar, G.; Tung, N.T.; Sohn, D. Electrochemical Performance of Zinc-Based Metal-Organic Framework with Reduced Graphene Oxide Nanocomposite Electrodes for Supercapacitors. *Synth. Met.* 2022, 290, 117155. [CrossRef]
7. Li, X.; Bai, Y.; Shi, X.; Su, N.; Nie, G.; Zhang, R.; Nie, H.; Ye, L. Applications of MXene (Ti3C2Tx) in Photocatalysis: A Review. *Mater. Adv.* 2021, 2, 1570–1594. [CrossRef]
8. Thi, Q.V.; Nguyen, N.Q.; Oh, I.; Hong, J.; Koo, C.M.; Tung, N.T.; Sohn, D. Thorny Trunk-like Structure of Reduced Graphene Oxide/HKUST-1 MOF for Enhanced EMI Shielding Capability. *Ceram. Int.* 2021, 47, 10027–10034. [CrossRef]
9. Yan, L.; Luo, X.; Yang, R.; Dai, F.; Zhu, D.; Bai, J.; Zhang, L.; Lei, H. Highly Thermoelectric ZnO@MXene (Ti3C2Tx) Composite Films Grown by Atomic Layer Deposition. *ACS Appl. Mater. Interfaces* 2022, 14, 34562–34570. [CrossRef]
10. Lee, Y.; Kim, S.J.; Kim, Y.J.; Lim, Y.; Chae, Y.; Lee, B.J.; Kim, Y.T.; Han, H.; Gogotsi, Y.; Ahn, C.W. Oxidation-Resistant Titanium Carbide MXene Films. *J. Mater. Chem. A* 2020, 8, 573–581. [CrossRef]
11. Ta, Q.T.H.; Cho, E.; Sreedhar, A.; Noh, J.-S. Mixed-Dimensional, Three-Level Hierarchical Nanostructures of Silver and Zinc Oxide for Fast Photocatalytic Degradation of Multiple Dyes. *J. Catal.* 2019, 371, 1–9. [CrossRef]
12. Sreedhar, A.; Ta, Q.T.H.; Noh, J.-S. Advancements in the Photocatalytic Activity of Various Bismuth-Based Semiconductor/Ti3C2 MXene Interfaces for Sustainable Environmental Management: A Review. *J. Ind. Eng. Chem.* 2022, 115, 26–47. [CrossRef]
13. Shahzad, F.; Alhabeb, M.; Hatter, C.B.; Anasori, B.; Man Hong, S.; Koo, C.M.; Gogotsi, Y. Electromagnetic Interference Shielding with 2D Transition Metal Carbides (MXenes). *Science* 2016, 353, 1137–1140. [CrossRef]
14. Van Ngoc, H.; Pham, K.D. First-Principles Study on N2, H2, O2, NO, NO2, CO, CO2, and SO2 Gas Adsorption Properties of the Sc2Cx CF2 Monolayer. *Phys. E Low-Dimens. Syst. Nanostruct.* 2022, 141, 115162. [CrossRef]
15. Nguyen, T.D.; Lee, J.S. Electrospinning-Based Carbon Nanofibers for Energy and Sensor Applications. *Appl. Sci.* 2022, 12, 6048. [CrossRef]
16. Ta, Q.T.H.; Tri, N.N.; Noh, J.-S. Improved NO2 Gas Sensing Performance of 2D MoS2/Ti3C2Tx MXene Nanocomposite. *Appl. Surf. Sci.* 2022, 604, 154624. [CrossRef]
17. Mohammadi, A.V.; Rosen, J.; Gogotsi, Y. The World of Two-Dimensional Carbides and Nitrides (MXenes). *Science* 2021, 372, eabf1581. [CrossRef]
18. Ta, Q.T.H.; Tran, N.M.; Tri, N.N.; Sreedhar, A.; Noh, J.-S. Highly Surface-Active Si-Doped TiO2/Ti3C2Tx Heterostructure for Gas Sensing and Photodegradation of Toxic Matters. *Chem. Eng. J.* 2021, 425, 131437. [CrossRef]
19. Zhao, L.; Wang, K.; Wei, W.; Wang, L.; Han, W. High-performance Flexible Sensing Devices Based on Polyaniline/MXene Nanocomposites. *InfoMat* 2019, 1, 407–416. [CrossRef]
20. Yang, Z.; Jiang, L.; Wang, J.; Liu, F.; He, J.; Liu, A.; Lv, S.; You, R.; Yan, X.; Sun, P. Flexible Resistive NO2 Gas Sensor of Three-Dimensional Crumpled MXene Ti3C2Tx/ZnO Spheres for Room Temperature Application. *Sens. Actuators B Chem.* 2021, 326, 128828. [CrossRef]
21. Thi, Q.V.; Ko, J.; Jo, Y.; Joo, Y. Ion-Incorporative, Degradable Nanocellulose Crystal Substrate for Sustainable Carbon-Based Electronics. *ACS Appl. Mater. Interfaces* 2022, 14, 43538–43546. [CrossRef]
22. Wang, R.; Li, M.; Sun, K.; Zhang, Y.; Li, J.; Bao, W. Element-Doped Mxenes: Mechanism, Synthesis, and Applications. *Small* 2022, 18, 2201740. [CrossRef]
23. My Tran, N.; Thanh Hoai Ta, Q.; Sreedhar, A.; Noh, J.S. Ti3C2Tx MXene Playing as a Strong Methylene Blue Adsorbent in Wastewater. *Appl. Surf. Sci.* 2021, 537, 148006. [CrossRef]
24. Sreedhar, A.; Ta, Q.T.H.; Noh, J.-S. Rational Engineering of Morphology Modulated Ti-ZnO Thin Films Coupled Monolayer Ti3C2 MXene for Efficient Visible Light PEC Water Splitting Activity. *J. Electroanal. Chem.* 2022, 921, 116703. [CrossRef]
25. Bahng, J.H.; Yeon, B.; Wang, Y.; Tung, S.O.; Hoff, J.D.; Kotoy, N. Anomalous Dispersion of “hedgehog” Particles. *Nature* 2015, 517, 596–599. [CrossRef]
26. Tran, N.M.; Ta, Q.T.H.; Noh, J.-S. RGO/Ti3C2Tx Heterostructures for the Efficient, Room-Temperature Detection of Multiple Toxic Gases. *Mater. Chem. Phys.* 2021, 273, 125087. [CrossRef]
27. Ta, Q.T.H.; Namgung, G.; Noh, J.S. Synthesis of Ag@rGO/g-C$_3$N$_4$ Layered Structures and Their Application to Toxic Gas Sensors: Effect of Ag Nanoparticles. *Electron. Mater. Lett.* 2019, *15*, 750–759. [CrossRef]

28. Thanh Hoai Ta, Q.; Park, S.; Noh, J.-S. Ag Nanowire/ZnO Nanobush Hybrid Structures for Improved Photocatalytic Activity. *J. Colloid Interface Sci.* 2017, *505*, 437–444. [CrossRef]

29. Kim, J.-H.; Ta, Q.T.H.; Noh, J.-S. Highly Strain-Tolerant, Disposable Gas Sensor on Plastic Wrap Using Two-Dimensional Hybrid Nanomaterials. *Mater. Chem. Phys.* 2022, *292*, 126820. [CrossRef]

30. Nguyen, N.T.A.; Kim, H. Ag$_3$PO$_4$-Deposited TiO$_2$@Ti$_3$C$_2$ Petals for Highly Efficient Photodecomposition of Various Organic Dyes under Solar Light. *Nanomaterials* 2022, *12*, 2464. [CrossRef]

31. Liu, Z.; Lv, H.; Xie, Y.; Wang, J.; Fan, J.; Sun, B.; Jiang, L.; Zhang, Y.; Wang, R.; Shi, K. A 2D/2D/2D Ti$_3$C$_2$T_x@TiO$_2$@MoS$_2$ Heterostructure as an Ultrafast and High-Sensitivity NO$_2$ Gas Sensor at Room-Temperature. *J. Mater. Chem. A* 2022, *10*, 11980–11989. [CrossRef]

32. Bui, H.T.; Van Thuan, D.; Huong, P.T.; Nguyen, K.D.; Nguyen, M.V.; Chu, T.H.; Van Le, Q.; Jitae, K.; Devanesan, S.; AlSalhi, M.S. Enhanced Photocatalytic H$_2$ Evolution and Photodegradation of Antibiotic Tetracycline in Wastewater by TiO$_2$@Ti$_3$C$_2$. *Int. J. Hydrogen Energy*, 2022, in press. [CrossRef]

33. Ji, H.; Zeng, W.; Li, Y. Gas Sensing Mechanisms of Metal Oxide Semiconductors: A Focus Review. *Nanoscale* 2019, *11*, 22664–22684. [CrossRef]

34. Li, Z.; Yao, Z.; Haidry, A.A.; Plecenik, T.; Xie, L.; Sun, L.; Fatima, Q. Resistive-Type Hydrogen Gas Sensor Based on TiO$_2$: A Review. *Int. J. Hydrogen Energy* 2018, *43*, 21114–21132. [CrossRef]

35. Zhu, Y.; Ma, Y.; Wu, D.; Jiang, G. Preparation and Gas Sensing Properties of ZnO/MXene Composite Nanomaterials. *Sensors Actuators A Phys.* 2022, *344*, 113740. [CrossRef]

36. Rzaij, J.M.; Abass, A.M. Review on: TiO$_2$ Thin Film as a Metal Oxide Gas Sensor. *J. Chem. Rev.* 2020, *2*, 114–121. [CrossRef]

37. Wu, M.; He, M.; Hu, Q.; Wu, Q.; Sun, G.; Xie, L.; Zhang, Z.; Zhu, Z.; Zhou, A. Ti$_3$C$_2$ MXene-Based Sensors with High Selectivity for NH$_3$ Detection at Room Temperature. *ACS Sens.* 2019, *4*, 2763–2770. [CrossRef]

38. Kim, S.J.; Koh, H.-J.; Ren, C.E.; Kwon, O.; Maleski, K.; Cho, S.-Y.; Anasori, B.; Kim, C.-K.; Choi, Y.-K.; Kim, J. Metallic Ti$_3$C$_2$T_x MXene Gas Sensors with Ultrahigh Signal-to-Noise Ratio. *ACS Nano* 2018, *12*, 986–993. [CrossRef]

39. Doan, T.H.P.; Ta, Q.T.H.; Sreedhar, A.; Hang, N.T.; Yang, W.; Noh, J.-S. Highly Deformable Fabric Gas Sensors Integrating Multidimensional Functional Nanostructures. *ACS Sens.* 2020, *5*, 2255–2262. [CrossRef]

40. Bhati, V.S.; Kumar, M.; Banerjee, R. Gas Sensing Performance of 2D Nanomaterials/Metal Oxide Nanocomposites: A Review. *J. Mater. Chem. C* 2021, *9*, 8776–8808. [CrossRef] [PubMed]

41. Nazemi, H.; Joseph, A.; Park, J.; Emadi, A. Advanced Micro-and Nano-Gas Sensor Technology: A Review. *Sensors* 2019, *19*, 1285. [CrossRef] [PubMed]

42. Thi, Q.V.; Park, S.; Jeong, J.; Lee, H.; Hong, J.; Koo, C.M.; Tung, N.T.; Sohn, D. A Nanostructure of Reduced Graphene Oxide and NiO/ZnO Hollow Spheres toward Attenuation of Electromagnetic Waves. *Mater. Chem. Phys.* 2021, *266*, 124530. [CrossRef]