



# MXene-based designer nanomaterials and their exploitation to mitigate hazardous pollutants from environmental matrices

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## ABSTRACT

MXenes are a rapidly expanding and large family of two-dimensional (2D) materials that have recently garnered incredible research interests for diverse applications domains in various industrial sectors. Owing to unique inherent structural and physicochemical characteristics, such as high surface area, biological compatibility, robust electrochemistry, and high hydrophilicity, MXenes are appraised as a prospective avenue for environmental-clean-up technologies to detect and mitigate an array of recalcitrant hazardous contaminants from environmental matrices. MXene-based nanoarchitectures are thought to mitigate inorganic pollutants via interfacial chemical transformation and sorption, while three different mechanisms, including i) surface complexation and sorption (ii) catalytic activation and removal and (iii) radical's generation-based photocatalytic degradation, are involved in the removal of organic contaminants. Considering the application performance of MXenes on the incessant rise to expansion, in this review, we discuss the wide-spectrum applicability of diverse MXenes-based hybrid nanocomposites in environmental remediation. A brief description related to environmental pollutants, structural properties, chemical abilities, and synthesis route of MXenes is delineated at the start. Afterwards, the adsorption and degradative robustness of MXene-based designer nanomaterials for various contaminants including organic dyes, toxic heavy metals, pesticide residues, phenolics, antibiotics, radionuclides, and many others are thoroughly vetted to prove their potentiality in the arena of wastewater purification and remediation. Lastly, challenges and trends in assessing the wide-range applicability and scalability of MXenes are outlined. Seeing encouraging outcomes in plenty of reports, it can be concluded that MXenes-based nanostructures could be considered the next-generation candidates for water sustainability.

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## 1. Introduction

MXenes, a new and large family of two-dimensional (2D) materials that come under the category of early transition metal carbides and carbonitrides have gained special research interests in recent years. Owing to their unique structural, physical, chemical, and functional characteristics, MXenes are considered designer nanomaterials for a range of applications in different industrial sectors (Al-Hamadani et al., 2020). Among different applied perspectives, MXenes have been the focus of supreme interest in mitigating hazardous pollutants from environmental matrices. For instance, several ongoing studies, around

the globe have spotlighted the environmental-related applications of MXenes either in the form of novel adsorbents to mitigate organic and inorganic based contaminants of high concern (Vakili et al., 2019; Jeon et al., 2020; Kim et al., 2021), multifunctional nanocomposites for robust bio-catalysis and/or bio-sensing of numerous toxic elements and related hazardous pollutants (Fanourakis et al., 2020; Jun et al., 2020a, 2020b, 2020c; Im et al., 2021), nanomembranes for wastewater filtration or simply water purification (Karahhan et al., 2020; Liu et al., 2020), nanoconstructs for energy storage and conversion (Kannan et al., 2020; Nan et al., 2021) and so on. Though each of the above-given applications of MXenes has its own merits and demerits that should be considered with care, most of them are not comparable to each other due to the application dependency criteria. Aiming to avoid or limit the generality, herein, we only focused on the MXenes as designer candidate materials and their notable potentialities

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to mitigate an array of emerging contaminants from environmental matrices.

With ever-increasing industrial growth, the water consumption rate is also proportionally increasing in different units of multiple industrial sectors globally. Consequently, all those industrial units are generating a wasteful amount of water in the form of effluents loaded with multiple polluting agents (Kausar et al., 2020; Rashid et al., 2021; Sher et al., 2021). Regardless of the astonishing industrial revolution, the current industrialization and urbanization practices, at a fast pace, have signified the environmental pollution issues by releasing an array of harmful pollutants (Bilal and Iqbal, 2020; Parveen et al., 2020; Qamar et al., 2020). Further to this, the controlled or uncontrolled discharge of industrial wastewater is contaminating different water matrices of the environment. For instance, the release or leach of pesticidal residues, pharmaceutically related active residues, antibiotics, toxic elements, toxic phenols, and gaseous pollutants has been intensified and accelerated the environmental pollution issues than ever (Yoon et al., 2010; Bilal et al., 2019, 2020; Parra-Saldivar et al., 2020; Américo-Pinheiro et al., 2021; Rasheed et al., 2021). All adversely affect the entire living ecosystem with their direct/indirect free movement in water matrices (Bilal et al., 2020; Morsi et al., 2020; Rasheed et al., 2020; Mishra et al., 2020; Zeb et al., 2020). Once entered the water matrices, many of them get bioaccumulated and thus biomagnified in several aquatic organisms, subject to type and concentration, and cause worse situations due to their biotoxicity and carcinogenicity attributes (López-Pacheco et al., 2019, 2021; Jeon et al., 2020). This phenomenon of bioaccumulation and biomagnification of various hazardous pollutants and their adverse effects on living organisms are shown in Fig. 1 (López-Pacheco et al., 2019). In order to avoid such direct or indirect adverse effects on living organisms, there is a dire need to develop and deploy an efficient and sustainable remediation treatment to sustain the green environment. So far, numerous treatment methods and techniques have been proposed, developed, and employed to remove a broader spectrum of emerging contaminants from environmental matrices. For instance, chemical, physical, physiochemical, and biological methods have been used to mitigate organic and inorganic based polluting agents.

Considering the above critiques, this review focuses on the wide-spectrum applicability of diverse MXenes-based hybrid nanocomposites in environmental remediation. A brief description related to environmental pollutants, structural properties, chemical abilities, and synthesis route of MXenes is delineated at the start. Afterwards, the adsorption and degradative robustness of MXene-based designer nanomaterials for various contaminants including organic dyes, toxic heavy metals, pesticide residues, phenolics, antibiotics, radionuclides, and many others are thoroughly vetted to prove their potentiality in the arena of wastewater purification and remediation. Lastly, challenges and trends

in assessing the wide-range applicability and scalability of MXenes are outlined.

## 2. Structural characteristics, synthesis route and catalytic properties of MXenes

### 2.1. Structural attributes of MXenes

Modeling and synthesis route of MXenes played an important role to understand its structure and properties. The etching of acid involves in synthesis makes surface-functionalized with multiple functionalities such as hydroxyl, oxygen and fluorinated functional group. In fact, the structure of the first OH-terminated titanium carbide ML-MXenes was proposed as a layered structure based on modeling through density functional theory (DFT) (Naguib et al., 2011). However, the XRD data obtained from geometry-optimized and experimentally synthesized hydroxylated MXene was too close and in good agreement with each other. Although, there is a close match of c-parameter appeared, whereas XPS analysis shows the generation of O, F, and OH functionalities. This show that there is an incomplete or mixed termination is present at their surfaces. It is also most probable; some water molecules are intercalated into the layered structure of MXenes. Through XRD pattern, absence of  $Ti_3AlC_2$  MAX phase and loss of crystallinity was confirmed after etching with HF acid (Mashtalir et al., 2013).

In general, a closely packed hexagonal (HCP) structure of MXenes was observed. Moreover, the sequence of M atoms might be different with different combinations. Such as, in M2X, M exhibit HCP pattern like (ABABAB), whereas in M3X2 and M4X3, preferred face center cubic pattern (ABCABC) (Anasori et al., 2017). This arrangement is suitable for MXenes to involve M atom having HCP pattern in bulk like molybdenum carbide and chromium carbide. This truth was disclosing from DFT that the HCP pattern of carbide of molybdenum is more stable than its counterparts in FCC. However, in some cases, it was noted from energy formation some MXenes are unstable due to M atoms arrangement variation. To stabilize this kind of MXenes, Titanium is inserted with carbon to stabilize their bond and produce an ordered double M element known as 2D carbides (Anasori et al., 2015).

The structure of free-standing and hydroxylated  $Ti_2C(OH)_2$  and  $Ti_3C_2(OH)_2$  were studied and analyzed by Enyashin and Ivanovskii, (2012) with three different configurations of the hydroxyl group with hollow space (A), situated at the top of carbon atom (B) or a mixture of these two configurations (C). It was noted that configuration A has more stability than B and C with free-standing and functionalized structure. On the other hand, B configuration shows the least stability and less preferred toward configuration adoption by different functionalities due to its maximum steric repulsion among T and C. The maximum stability of configuration A can be explained due to the formation of the ABCABC ordered pattern because of the distinct position of MX and T (Fig. 2) (Tang et al., 2012).

### 2.2. Synthesis route of MXenes

The synthesis of MXenes based on the selective etching of element a layer from the parent MAX phase at room temperature. For this purpose, powder form of layered MAX phase is mix with aqueous hydrofluoric acid at room temperature. As a result, a layer gets separated from MAX by replacing the strong metallic bond with weak bonds among A and MX layer selectively. Through this weak interaction, other functionalities such as fluorine, oxygen and hydroxyl get attached at the surface of ML-MXene (Naguib et al., 2012). Furthermore, in order to get a few-layer MXene having a number of layers less than 5, centrifugation and filtration of the solid carried out to get the isolated form of supernatant after washing with deionized water having a pH range from 4 to 6 (Naguib et al., 2014). The MXenes layer structure mainly depends on the pH of the solution, i.e., a decrease in pH breaks down

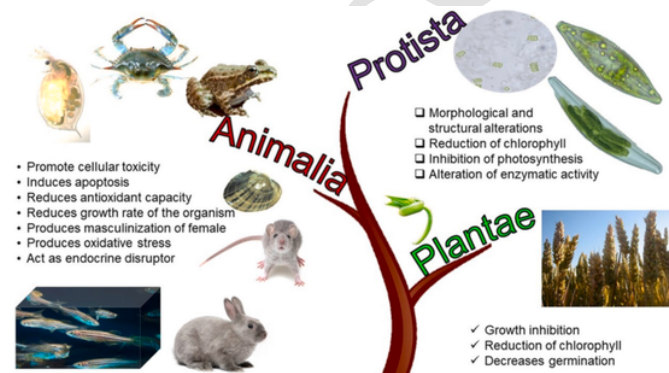
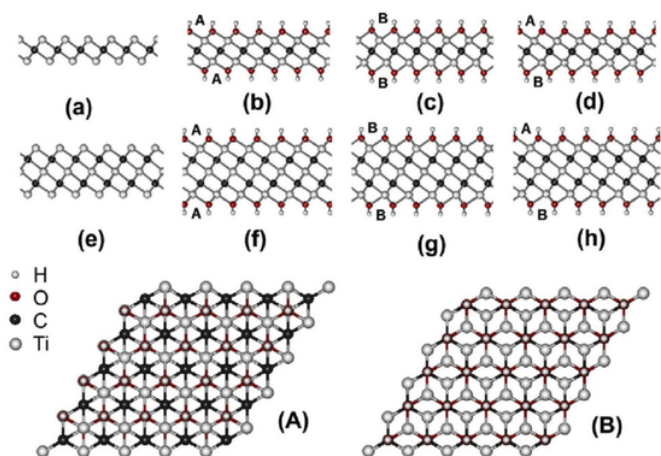


Fig. 1. Representation of bioaccumulation and biomagnification of various hazardous pollutants and their adverse effects on living organisms. Reprinted from López-Pacheco et al. (2019) with permission from Elsevier. License Number: 5,086,000,685,924.



**Fig. 2.** Atomic ordering of bare (a)  $\text{Ti}_2\text{C}$  and (e)  $\text{Ti}_3\text{C}_2$  and their hydroxylated forms, (b–d)  $\text{Ti}_2\text{C}(\text{OH})_2$  and (f–h)  $\text{Ti}_3\text{C}_2(\text{OH})_2$  with three configurations according to the placement of OH groups. (A) and (B) are the top views of configuration a and b respectively. Reprinted from Enyashin and Ivanovskii (2012) with permission from Elsevier. License Number: 5,086,000,868,212.

the layers. This crumpling mostly happens in the  $\text{Ti}_3\text{C}_2\text{Tx}$  MXene case (Natu et al., 2018). MXene can be synthesized from a non-Max precursor by etching additional parts like  $\text{Mo}_2\text{CTx}$  could be synthesized by etching of Ga from a non MXene precursor  $\text{Mo}_2\text{Ga}_2\text{C}$ . They contain two layers of A, in this case, it is Ga; one of them will be etched out and secondly will remain with the MAX phase (Halim et al., 2016). A similar case is in  $\text{Zr}_3\text{Al}_3\text{C}_5$ , in which aluminium carbide  $\text{Al}_3\text{C}_3$  layer etched instead of aluminum (Al) layer (Zhou et al., 2016).

A high-temperature etching method can also be employed to synthesize MXenes. In 2016, nitride-based MXene was experimentally synthesized by molten fluoride salt mixture at  $550^\circ\text{C}$  and inert atmosphere. At these conditions, Al layer etches from  $\text{TiAlN}_3$  powder and further delaminated by tetrabutylammonium hydroxide to generate monolayers of  $\text{Ti}_4\text{N}_3\text{Tx}$  MXene (Urbankowski et al., 2016). More interestingly, MXene can also be synthesized by sublimating some layers at  $800^\circ\text{C}$  in a vacuum. For example,  $\text{TiCx}$  can be prepared from  $\text{Ti}_2\text{InC}$  by the sublimation of In and Si layers elimination is carried out from  $\text{Ti}_3\text{SiC}_2$  at  $960^\circ\text{C}$  from molten cryolite (Barsoum et al., 2002). The carbide obtained on these conditions are 3D cubic rather than 2D due to certain treatment conditions. In addition, metal carbides are stable only below  $800^\circ\text{C}$  in accordance with their phase diagram. (Gusev and Rempel, 1999). Thus, synthesis procedures and heat treatments of MXenes should be below  $800^\circ\text{C}$ , otherwise, there will be the possibility of the 3D structure appeared.

Different methodologies, such as migration-enhanced encapsulated growth technique; scalable salttemplated synthesis approach, are employed to synthesized metal nitrides of GaN and MoN, respectively (Xiao et al., 2017). The nitrides such as  $\text{V}_2\text{N}$  and  $\text{W}_2\text{N}$  were synthesized by utilizing the procedure same as where 2D nitrides were obtained by ammoniated hexagonal oxides. In 2017, Urbankowski et al. synthesized 2D  $\text{V}_2\text{N}$  and  $\text{Mo}_2\text{N}$  through ammoniation of their 2D precursor's carbide MXenes  $\text{V}_2\text{CTx}$  and  $\text{Mo}_2\text{CTx}$ , respectively. This process involves the replacement of C atoms by N atoms by ammoniation at  $600^\circ\text{C}$ . Even after obtaining these 2D nitrides through the same method, i.e., ammoniation, the resulting crystals show different structures (Urbankowski et al., 2017). Until now, the obtained 2D transition metal carbides have some defects with functionalities present on their surfaces with a maximum lateral size of up to 10 mm. This problem was further solved by Xu et al. (2015) by the synthesis of high quality and defect-free MoC crystals through the vapor deposition method (CVD). In this method, molybdenum carbide with lateral size up to or higher than 100 mm was synthesized at low temperature and

having 2D superconductivity. A copper foil was placed as a substrate on the top of molybdenum foil, and methane gas was provided as a source of carbon. The whole process was carried out at  $1085^\circ\text{C}$ , which is above the melting temperature of copper. This produces an alloy of Cu and Mo at their interface, and molybdenum reacts with carbon produce after the disintegration of methane gas after passing through the interface, as shown in figure Z(A). This technique is an analogue of the CVD method for the production of high-quality graphene. The  $\text{Mo}_2\text{C}$  crystals obtained in this work show a minimum thickness of 3 nm, as shown in figure Z(B). The monolayer MXene from the top is still under discussion and yet not be demonstrated. The versatility of this method can be seen in Fig. 3C and D by comparing the optical micrograph and high-resolution transmission electron microscopy results of tungsten carbide crystals. In the near future, CVD and salt-templated synthesis will open up a new foundation to generate novel 2D carbides, carbonitrides and nitrides (Fig. 3) (Huang et al., 2019; Wang et al., 2018a, 2018b).

### 2.3. Properties of MXenes

The outstanding mechanical properties, metallic conductivity, and hydrophobicity nature of MXene make it a useful material for various applications. The mobility of electron in MXene is around  $10^6 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  or higher than this value, which is somehow more effective than graphene (Shao et al., 2017). The conductivity of  $\text{Ti}_3\text{C}_2\text{Tx}$  is around  $6500 \text{ S cm}^{-1}$  above than other 2D materials, such as CNTs and graphene. In this regard, MXenes are leading 2D material like clays, graphene, graphene oxide (Ling et al., 2014; Dillon et al., 2016). They also have very good bending rigidity, about 49.55 eV (Borysiuk et al., 2018) higher than graphene 2.3 eV (Kang and Lee, 2013), and  $\text{MoS}_2$  has 9.61 eV (Jiang et al., 2013). The stiffness and strength of  $\text{M}_2\text{X}$  MXenes are better than that of the  $\text{M}_3\text{X}_2$  structure and  $\text{M}_4\text{X}_3$  structure (Borysiuk et al., 2015). MXenes with the highest Young's modulus of 0.33 TPa compared to graphene oxide 0.21 TPa and  $\text{MoS}_2$  0.27 TPa was reported in a recent publication (Lipatov et al., 2018).

Ultrathin films of MXenes and their derivative are transparent in nature and shows transmittance about 97% for  $\text{Ti}_3\text{C}_2$ . This transmittance value varies with the thickness of films, and it normally reduces with higher thickness. The layer thickness also responsible for the electrochemical properties of MXenes, as the thickness enhanced restacking of layers occur and create a barrier, which renders the mobility of ions and their transportation as a result of capacitance decrease. In recent work, it is concluded that the arrangement of layers can control the electrochemical performance of MXenes. As in the case of  $\text{Ti}_3\text{C}_2\text{Tx}$ , when the layers are arranged vertically through mechanical shear instead of horizontal arrangement, a perpendicular array formed between the layers and current collector, which enhanced the mobility of ions (Xia et al., 2018).

The effect of storage conditions is also studied by the scientist. Like, when Ti-based MXenes are stored under a normal atmosphere, their degradation occurs within 5 days due to the interaction of water and air. Whereas MXene found very stable when stored in an argon atmosphere, it shows no oxidation or hydroxylation towards dissolved oxygen (Zhang et al., 2017). Later on, it was noted that water plays a vital role in the degradation of MXenes rather than oxygen (Huang et al., 2019). In contrast with graphene, MXenes show less thermal stability and renders their applications in nanoelectronics systems. Titanium-based MXenes shows a contact angle of about  $21.5^\circ$  and very well stable even with vigorous shaking with water. This property makes MXene used as water purification membranes. This happened due to the very small interlayer distance of MXene only allows ions with smaller radius to pass through it (Ren et al., 2015). The abovementioned qualities of MXenes make them useful material in energy storage, electrochemical biosensors, water purification, electrocatalysts, electromag-

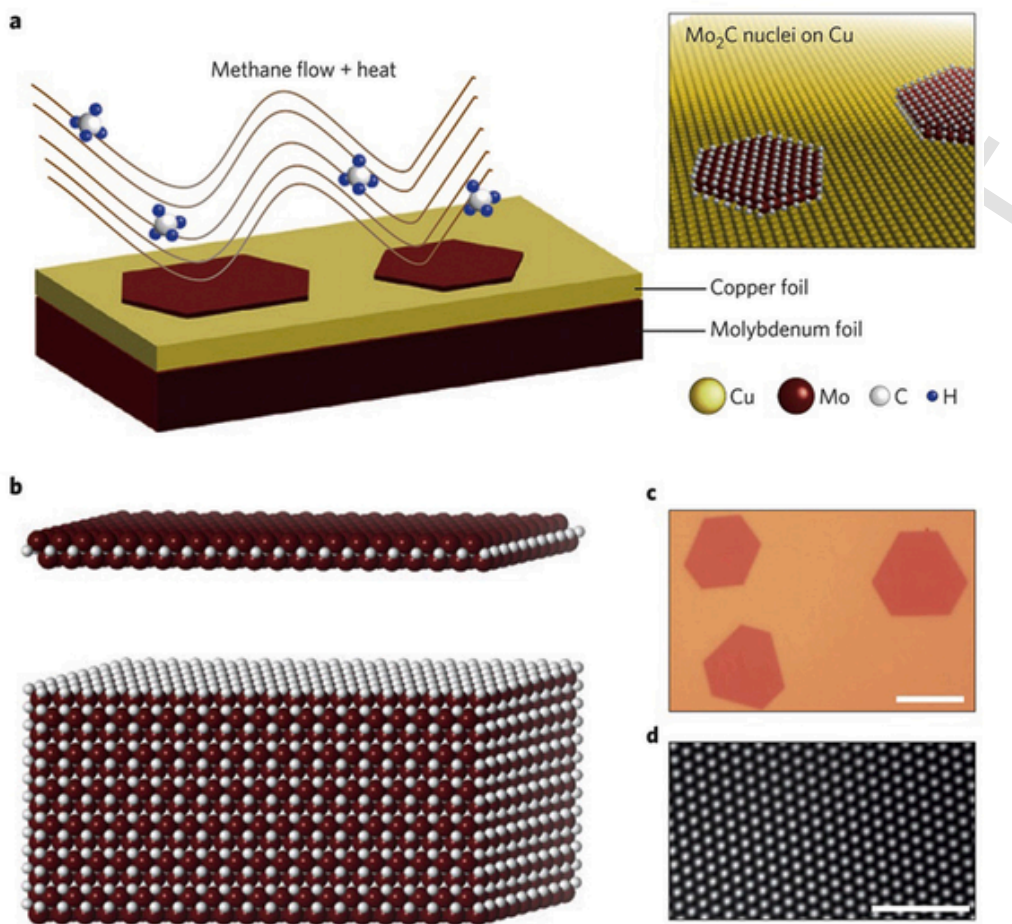


Fig. 3. 2D TMC crystals grown by CVD process. (a) Schematic of fabrication of Mo<sub>2</sub>C crystal at 1085 °C with Cu foil as substrate and methane gas as carbon source. Mo atoms diffuse through copper and react with methane to form α-Mo<sub>2</sub>C (shown in inset). (b) Monolayer Mo<sub>2</sub>C (top) and experimental α-Mo<sub>2</sub>C crystal of 3 nm thickness (bottom). (c) Optical image (10 μm scale) and (d) HR-TEM (2 nm scale) of WC crystals grown by this method. Reprinted from Gogotsi (2015) with permission. Copyright © 2015, Nature Publishing Group, a division of Macmillan Publishers Limited. License Number: 5,086,001,290,431.

netic absorption, gas sensors, conductive coatings, antibacterial membranes, structural composites and hydrogen storage (Choi et al., 2018; Weng et al., 2018; Pang et al., 2019; Tunesi et al., 2021).

### 3. Environmental applications of MXene-based nanocomposites

#### 3.1. Removal of dye pollutants

Dyes are a group of organic substances that have been extensively applied in different industrial sectors, such as textile, printing, cosmetics, and pharmaceuticals (Ali et al., 2020a, 2020b, 2020c; Munir et al., 2020; Ahmad et al., 2020; Nawaz et al., 2020; Ali et al., 2021; Muhammad et al., 2021; Yang et al., 2021). Among various dyes, methylene blue is challenging to degrade due to its complicated structure, thus hindering photocatalysis-based treatment approaches (Feng et al., 2020). In a recent report, Yao et al. (2021) fabricated porous p-MXene/SWCNTs nanocomposite using a scalable and facile technique and employed for organic dyes sorption in wastewater samples (Fig. 4). The synthesized p-MXene/SWCNTs film displayed excellent efficiency (1068.8 mg g<sup>-1</sup>) for methylene blue adsorption. It also showed spectacular recycling ability retaining 95.2% of adsorption efficiency after five repeated cycles. High dye adsorption was attributed to the substantial specific surface area of MXene-based free-standing film and potent hydrogen bonding and electrostatic interaction effect.

Preparation of functionalized cellulose/MXene bio-nano composite aerogel using oxidative dopamine hydrochloride self-polymerization and freeze-drying showed noteworthy adsorptive efficiency for methyl-

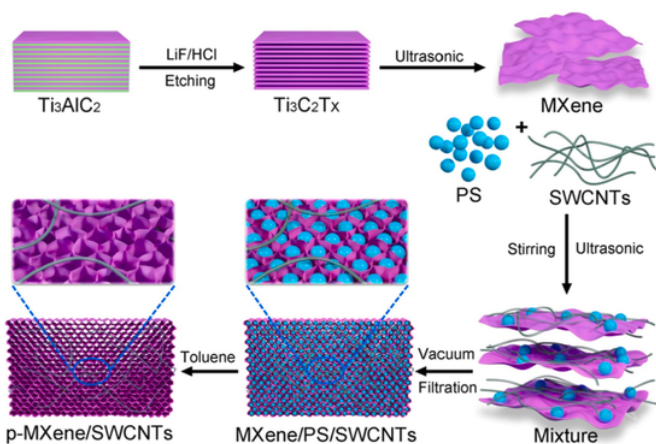


Fig. 4. Schematic illustration for the p-MXene/SWCNTs film. Reprinted from Yao et al. (2021) with permission from Elsevier. License Number: 5,086,010,040,364.

ene blue, reaching 168.93 mg g<sup>-1</sup>. The composite aerogel indicated good stability and maintained over 84% of MB removal efficiency after five consecutive cycles. SEM analysis revealed the good preservation of 3D interconnected absorbent structure after absorption of the dye in multiple cycles. The adsorption process was better fitted to Langmuir isotherm model. The appearance of a typical absorption peak at 1593 cm<sup>-1</sup> after dye adsorption might be ascribed to the stretching vi-

bration of methylene blue aromatic rings (Wei et al., 2017). Fig. 5 represents the schematic depiction of the adsorption mechanism of methylene blue dye by P-M/MX-50. Weak interactions like  $\pi$ - $\pi$  interaction, electrostatic interaction and hydrogen bonding determine the adsorption of dye. Given the concerted role of all these forces, the nanocomposite aerogel has endowed to exhibit superior adsorptive capability. Retention of superior adsorption ability in salt-containing water (over 3% NaCl) recommends applicability for effective dyes removal in the high-salted environment (Zhang et al., 2021).

Methyl orange is a commonly employed organic dye that can cause several undesirable health problems, such as vomiting, tachycardia, cyanosis, jaundice, and tissue necrosis. Karthikeyan et al., (2021) reported efficient adsorption of methyl orange dye and Cr(VI) from the aqueous matrix using  $Ti_3C_2T_x$  MXene nanosheets. The MXenes-driven removal rate of MO and Cr(VI) was very rapid, achieving 94.8 and 104 mg/g for MO and Cr(VI), respectively. Langmuir isotherm was found well appropriate to describe the sorption isotherm. The  $Ti_3C_2T_x$  MXene nanosheets can be successfully regenerated and recycled in multiple reactions. Chemical adsorptions, including complexation, electrostatic adsorption, ion exchange and surface interactions, were determined to be the main mechanisms involved in up taking MO and Cr(VI) by the MXenes nanosheet.

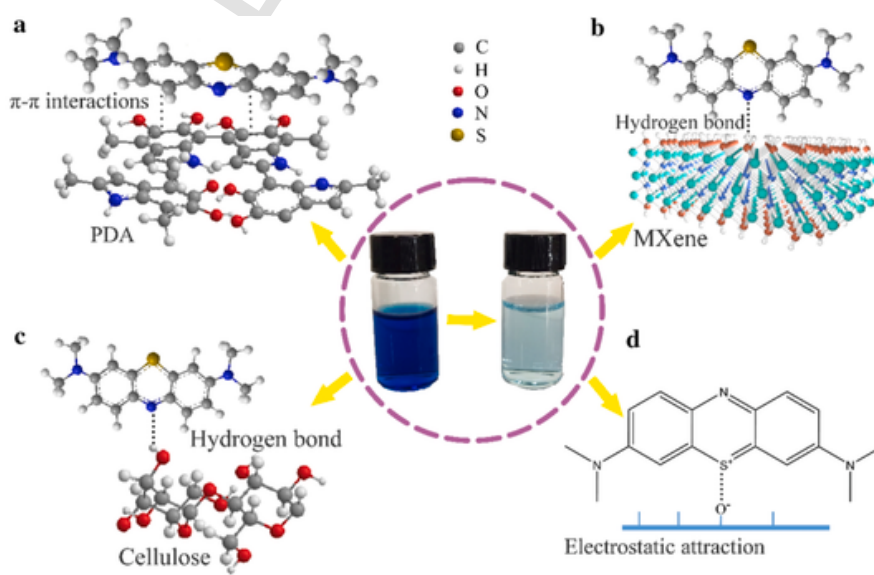
### 3.2. Degradation of pesticides

Pesticides are chemicals used to control and kill pests and weeds and, therefore, help protect agricultural production. Based on their chemical structures, pesticides can be categorized as organophosphate (OPs), organochlorines, carbamate, substituted urea, and biopesticides (Rani and Shanker, 2018; Bagheri et al., 2020a,b). Several pesticides and their active residues/metabolites pose adverse impacts on human health and severely damage ecological sustainability and integrity (Cai et al., 2018a; Henry et al., 2015; Cheng et al., 2018). Due to environmental prevalence and high toxicity, the development of low-cost, effective, and environmentally friendly approaches is direly needed to solve their remediation issues. Highly conductive surface of MXene, provoked researchers and scientists to scrutinize pesticides and PPCPs abatement from an aqueous environment via catalytic activation and sorption mechanisms.

Three different pathways, including complexation and sorption, peroxymonosulfate or other reagents activated catalytic degradation, and photocatalytic decontamination through in-situ radical formation, have been reported to eliminate pesticides from the aqueous solutions (Wu et al., 2016; Luo et al., 2020). Ding et al. (2019) developed a new  $Fe_2CoTi_3O_{10}$ -MXene (FCT-M) nanocomposite-based heterogeneous catalyst to catalyze the oxidative removal of 2,4-dichlorophenoxyacetic acid (2,4-D) in the presence of peroxymonosulfate (PMS) as an activating agent. The  $Fe_2CoTi_3O_{10}$  nanocatalyst presented enhanced catalytic activity resulting in almost complete degradation of 2,4-D degraded in 15 min under optimized conditions of PMS (0.2 g/L), FCT-M-2 composite (0.2 g/L), and 2,4-D (20 mg/L) at a reaction pH of 7.0. Radical scavenger tests and electron paramagnetic resonance spectroscopy confirmed the co-existence of hydroxyl radicals ( $\cdot OH$ ) and sulfate radicals ( $SO_4^{\cdot -}$ ), and  $SO_4^{\cdot -}$  radicals were detected to be the major reacting species in this catalysis system. The profound PMS activated FCT-M performance was ascribed to outstanding synergetic effects of 3 transition metals (Co, Fe, and Ti) on the surface of the catalyst. In another investigation, the same group carried out the degradation of 2,4-D using  $Fe_2CoTi_3O_{10}$ -MXene nanocomposite in the presence of PMS as an activating agent. Experimental results revealed more than 98% 2,4-D degradation by the simultaneous inclusion of FCT-M composite and PMS with promising recycling ability for up to 4 consecutive reactions. The addition of humic acid inhibited the degradation rate by competition with free radicals generation or masking the reactive sites. Redox pairing of 3 different metals, such as Co(II)/Co(III), Fe(II)/Fe(III), and Ti(II)/Ti(III) led to augmented pesticides degradation on the surface of composite (Ding et al., 2020).

### 3.3. Removal of phenolics

Phenolic compounds are highly toxic organic contaminants, which are commonly detected in a few ppm to several grams per litre in an extensive range of various industrial effluents, like plastic, rubber, dyeing, printing, pharmaceutical, textile, wood processing, cosmetics, and petrochemical (Mohammadi et al., 2015). Due to their toxic, mutagenic, carcinogenic, and teratogenic effects, the phenolics have been declared as pollutants of top-priority concerns by the US and European Union. Therefore, the development of effective approaches is important



**Fig. 5.** Schematic illustration of adsorption mechanism (a  $\pi$ - $\pi$  interactions of MB and PDA; b Hydrogen bond of MB and MXene; c Hydrogen bond of cellulose; d Electrostatic attraction of MB and adsorbent) (Zhang et al., 2021). Reprinted from Zhang et al. (2021) with permission. Copyright © 2021, The Author(s), under exclusive licence to Springer Nature B.V. License Number: 5,086,010,234,683.

to eliminate these phenolic compounds from water streams before liberation (Cao et al., 2021). Given a diverse number of surface moieties, MXene-based nano constructs, and nanocomposites, such as metal/MXene (i.e., Au/Ti<sub>3</sub>C<sub>2</sub>Tx, Ag/Ti<sub>3</sub>C<sub>2</sub>Tx and Rh/Ti<sub>3</sub>C<sub>2</sub>Tx), metal oxide/MXene (i.e., Fe<sub>3</sub>O<sub>4</sub>/Ti<sub>3</sub>C<sub>2</sub>Tx, α-Fe<sub>2</sub>O<sub>3</sub>/Ti<sub>3</sub>C<sub>2</sub>Tx, TiO<sub>2</sub>/Ti<sub>3</sub>C<sub>2</sub>Tx, Co<sub>3</sub>O<sub>4</sub>/Ti<sub>3</sub>C<sub>2</sub>Tx), and oxysalt/MXene (i.e., Ag<sub>3</sub>PO<sub>4</sub>/Ti<sub>3</sub>C<sub>2</sub>) have been widely employed for the photocatalytic degradation of numerous organic compounds. For example, Cai et al. (2018b) synthesized a high-performance Ag<sub>3</sub>PO<sub>4</sub>/Ti<sub>3</sub>C<sub>2</sub> Schottky catalyst (Fig. 6) that catalyzed 2.5 and 10-times greater removal of 2,4-Dinitrophenol compared to Ag<sub>3</sub>PO<sub>4</sub>/RGO and Ag<sub>3</sub>PO<sub>4</sub>, respectively. It also showed elevated stability degrading more than 65% of tetracycline after eight continuous reaction cycles, while only Ag<sub>3</sub>PO<sub>4</sub>/RGO and Ag<sub>3</sub>PO<sub>4</sub> retained only 36.2 and 7.8% catalytic performance, respectively under the same conditions. Based on the experimental investigations, the MXene-based composites assisted degradation process of phenolic compounds was attributed to electrostatic attraction, H-bonding, and reductivity.

Chlorophenols are toxic pollutants that show potent bioaccumulation ability and acute toxicity (Jin et al., 2009). Therefore, their degradation is a pressing subject owing to growing environmental concern. Ming et al. (2017) fabricated regularly dispersed Rh/alk-Ti<sub>3</sub>C<sub>2</sub>X<sub>2</sub> adopting a simple synthesis approach via removing Al layers of Ti<sub>3</sub>AlC<sub>2</sub> followed by alkalization treatment. The as-fabricated Rh/alk-Ti<sub>3</sub>C<sub>2</sub>X<sub>2</sub> stable catalyst presented substantially improved hydrochlorination of 4-chlorophenol, which might be ascribed to the numerous active components from small Rh nanoscale particles. Such superior efficiency of Rh/alk-Ti<sub>3</sub>C<sub>2</sub>X<sub>2</sub> catalysts was attributed to improved capacity for hydrogen uptake.

### 3.4. Removal of antibiotics

Antibiotics are a particular class of pollutants of utmost concern that have drawn increasing attention over the last two decades. Various antibiotics, such as tetracyclines, sulfonamides, lincomycin, fluoroquinolones, are extensively applied to animal husbandry, medicals, and poultry metabolites as medicines to treat and cure bacterial infections (Valdés et al., 2021). Abuse and overuse of antibiotics, environmental persistence, and the emergence of antimicrobial resistance have appeared major health problems worldwide (Carvalho and Santos, 2016). As an explicit antibacterial agent, the excessive utilization of sulfonamides in various food and medical fields has severe environmental consequences (Chen et al., 2019; Li et al., 2019a, 2019b). Due to water solubility, persistence, complex structure, and pro-

nounced chemical stability, sulfonamides and their active metabolic residues pose a considerable menace to humans health and environmental matrices (Esplugas et al., 2007; Chen et al., 2019; Payan et al., 2019; Li et al., 2019a, 2019b). Hence, the design of high-performance photo-assisted for their efficient removal is the major research arena in environmental sciences.

In order to overcome the antibiotics problems, Cao et al. (2020) designed a new CuFe<sub>2</sub>O<sub>4</sub>/MXene (CFO/Ti<sub>3</sub>C<sub>2</sub>) heterojunction catalyst for visible light assisted photocatalytic degradation of sulfamethazine. They extended the photoinduced carriers' lifetime by loading Ti<sub>3</sub>C<sub>2</sub> and interfering with the photogenerated electron holes reorganization. Results revealed substantial synergistic degradation of as-fabricated photocatalyst, reaching about 60% removal of sulfamethazine. Moreover, SMZ molecules were found to be adsorbed or attacked by the active material in the entire photocatalytic process. The proposed removal mechanism points out the generation of small-molecular weight organic substances (aminobenzenes, methylpyrimidines, etc.) by nanocomposite material following pyrimidine NeS bond cleavage hydroxylation, deamination, aniline oxidation, etc. (Figs. 7 and 8). The intermediate products were finally converted into CO<sub>2</sub>, H<sub>2</sub>O, and some other low weight molecules.

Wu et al. (2020) synthesized Ti<sub>3</sub>C<sub>2</sub>-modulated MIL-125-NH<sub>2</sub>-based novel nano hybrids using a one-step solvothermal method for photocatalytic removal of antibiotics. Under visible light illumination, the optimally fabricated NH<sub>2</sub>-MIL-125(Ti) (TiO<sub>2</sub>)/Ti<sub>3</sub>C<sub>2</sub> hybrid nanocatalyst achieved 1.65- and 11.5-times greater production of H<sub>2</sub>O<sub>2</sub> and tetracycline removal compared to pristine MIL-125-NH<sub>2</sub>. Based on the characterization analysis, enhanced photocatalysis was predominantly assigned to the dual-heterojunction in the newly developed nano hybrid, which results in enhanced carrier density, and accelerated the separation of interfacial charge. During photocatalytic activities, h<sup>+</sup> and •OH were detected to be the major reactive species analyzed by ESR spectra and radical trapping experiments. Thus, the development of MIL-125-NH<sub>2</sub>(TiO<sub>2</sub>)/Ti<sub>3</sub>C<sub>2</sub> nano hybrid offers a novel approach with adjustable morphology, design, and optimized property to address energy and environment issues.

### 3.5. Removal of radionuclides

As a carbon-free energy source, nuclear power is likely to contribute a major role in meeting global energy requirements given extensive energy density and lack of greenhouse gas emissions (Kim and Yoo, 2021). Nevertheless, nuclear power results in the generation of massive

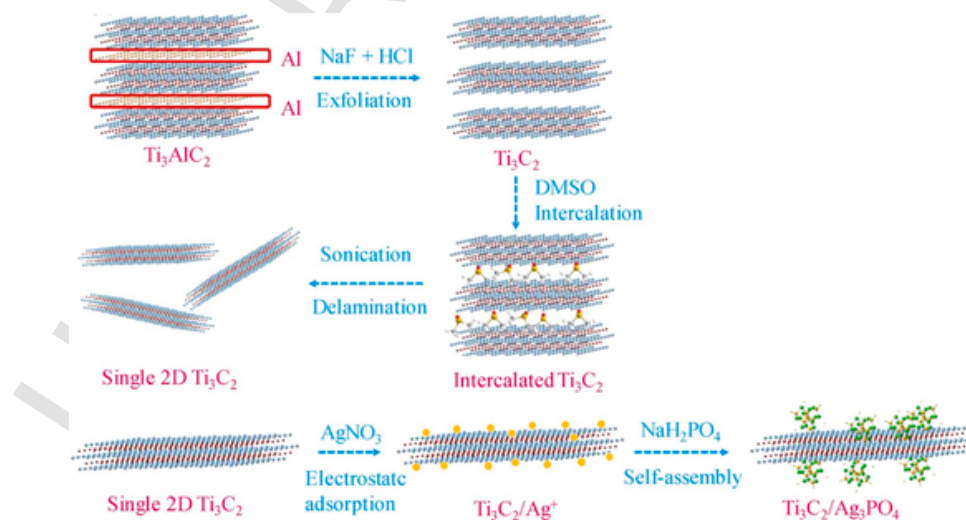


Fig. 6. Schematic representation of single 2D Ti<sub>3</sub>C<sub>2</sub> sheets and Ag<sub>3</sub>PO<sub>4</sub>/Ti<sub>3</sub>C<sub>2</sub> synthesis. Reprinted from Cai et al. (2018b) with permission from Elsevier. License Number: 5,086,010,419,858.

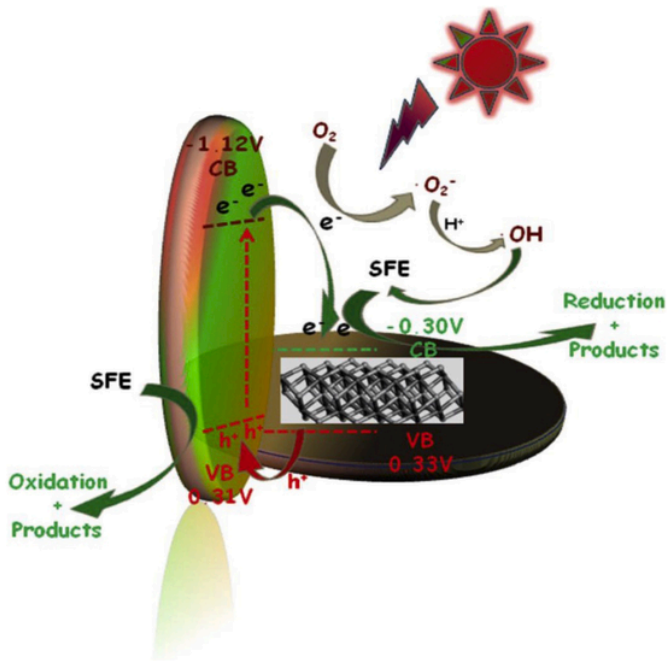


Fig. 7. Possible degradation mechanism of CFO/Ti<sub>3</sub>C<sub>2</sub> for degrading SMZ (Cao et al., 2020). Reprinted from Cao et al. (2020) with permission from Elsevier. License Number: 5,086,010,865,492.

amount of radioactive wastes (i.e., <sup>232</sup>Th, <sup>235</sup>U, <sup>90</sup>Sr, and <sup>137</sup>Cs) as the byproduct. This waste is considered extremely harmful materials because of potent radioactivity, longer half-lives, high chemical toxicity, and mobility (Zhou et al., 2020; Song et al., 2019). Benign

management of this radionuclide waste is a burgeoning issue to develop sustainable nuclear energy (Zhong et al., 2020; Qiu et al., 2020). Thus, seeking and designing eco-friendly and highly efficient adsorbent candidates is urgently needed for exterminating these radioactive waste materials from the natural ecosystem. Despite plenty of reports on developing emerging nanostructured materials for radionuclides removal (Li et al., 2019a, 2019b; Wang et al., 2020), very scarce data are available to mitigate radionuclides by exploiting MXene-based designer composite materials.

Due to chemical toxicity and radiological behavior, uranium is regarded as an environmental pollutant of emerging concern. Hexavalent uranium, explicitly soluble UO<sub>2</sub><sup>2+</sup>, tends to readily released into environmental matrices following spent fuel reprocessing, milling, and nuclear wastes disposal. Recently, various MXenes, like chromium carbide, vanadium carbide, and titanium carbide, have been widely examined for U(VI) mitigation using spectroscopic, batch, and theoretical methods. In a study, titanium carbide MXene (470 mg/g) (Fig. 9A) (Wang et al., 2018a, 2018b) exhibited a significantly higher removal capacity of U(VI) compared to vanadium carbide counterpart (174 mg/g) (Wang et al., 2016). Such as uptake capacity of titanium carbide MXene might be attributed to electrostatic attraction and surface complexation. Moreover, MXene-based nanocomposites presented encouraging regeneration towards U(VI). Experimental outcomes demonstrated titanium-assisted MXene as potential adsorbing candidates to eliminate radionuclides from aqueous systems (Wang et al., 2018a, 2018b).

Barium from oil and gas industries pose adverse impacts on aquatic creatures and human health, like swelling of the brain, liver injury, heart rhythm and muscle feebleness (Celebi et al., 2009) WHO set the highest level of barium to be less than 0.7 mg/L (Edition, 2011). The application of 2DTi<sub>3</sub>C<sub>2</sub>Tx nanosheets catalyzed over 90% removal

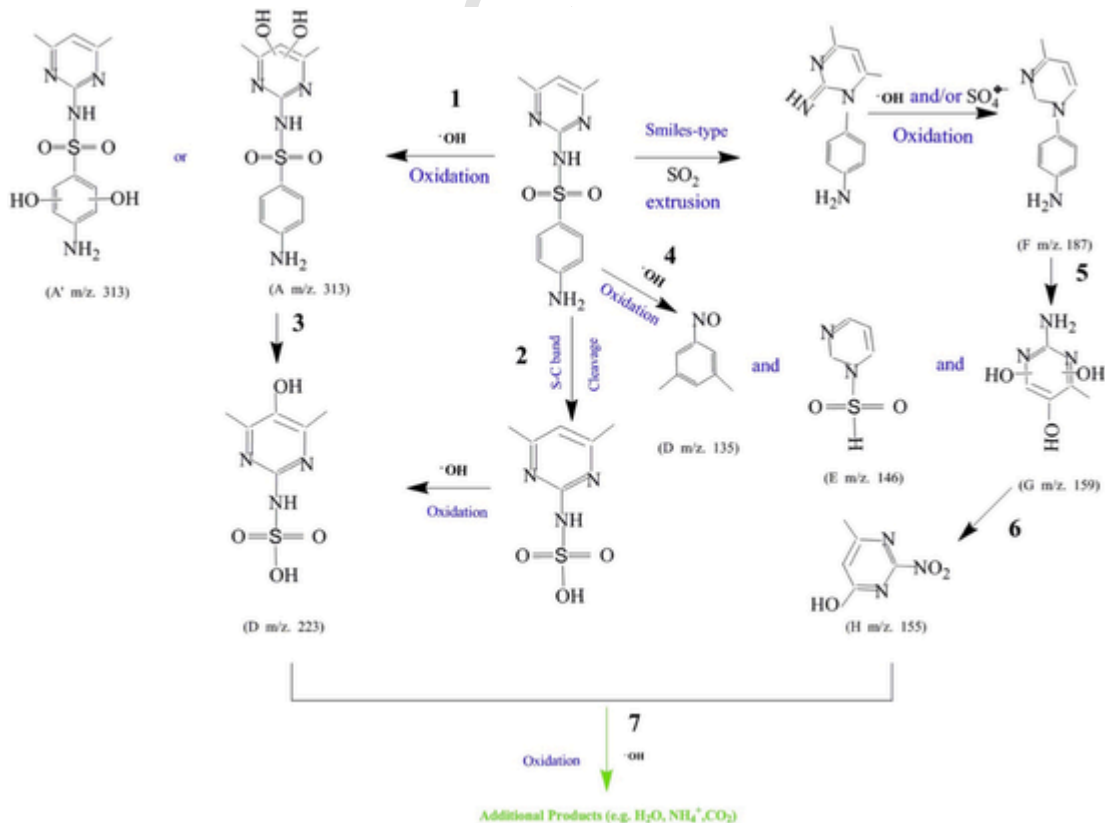


Fig. 8. Analysis of degradation intermediates and possible degradation pathways of SMZ. Reprinted from Cao et al. (2020) with permission from Elsevier. License Number: 5,086,010,865,492.

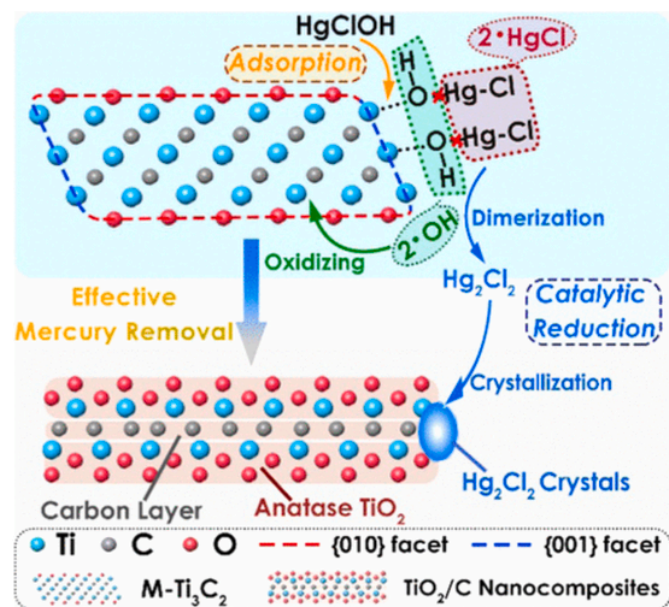


Fig. 9. Removal of Hg(II) from water Using  $\text{Ti}_3\text{C}_2\text{O}_x$  MXene via adsorption coupled with catalytic reduction mechanism. Reprinted from Fu et al. (2020) with permission. Copyright © 2020 American Chemical Society.

of Ba(II) (Fard et al., 2017). Jun et al. (2020b) reported that  $\text{Ti}_3\text{C}_2\text{Tx}$  showed a noteworthy adsorptive ability (180 mg/g), good recycling efficiency and quick equilibrium kinetics. It is found that pH markedly influenced the Ba(II) adsorption by influencing the ionization of Ba(II) and the surface charge of MXenes. A superior Ba(II) adsorption at basic and neutral and pH values was mainly ascribed to the  $\text{Ba}(\text{OH})_2$  precipitates formation and electrostatic attraction, respectively (Shahzad et al., 2017). Characterization analyses unveiled that effective Ba(II) adsorption onto MXene was due to the Ba(II) intercalation into MXene interlayers and surface coordination with  $-\text{F}$  and  $-\text{OH}$  groups on the surface of MXene (Jun et al., 2020a, 2020b, 2020c).

In addition, 2D  $\text{Ti}_2\text{CTx}$  MXene have also been applied to remediate many other radionuclides, including Eu(III), Sr(II), Th(IV), Cs(I), and Pd(II). For example, elevated Re(VII) removal performance (363 mg/g) of  $\text{Ti}_2\text{CTx}/\text{poly}$  (diallyl dimethyl-ammonium chloride) was due to its potent reducing activities (Wang et al., 2019b). Inner sphere surface complexation was found to be involved for the enhanced Th(IV) adsorption (213.2 mg/g) by  $\text{Ti}_2\text{CTx}$  MXene in the presence of various competing metal ions (Li et al., 2019a, 2019b). In conclusion, various parameters, such as nature of radionuclides (surface charge, functional groups on surface), MXene nanocomposites attributes (i.e., specific surface area, shape, size, surface charge, and functional groups) and water quality aspects (temperature, pH and concentration) influence the mitigation performance of MXene-based composites for radionuclides. It is worth noting that due to high compatibility, superior thermal resistance, and outstanding stability to radiation-induced impairment, MXene-assisted nanostructured composites are known to effectively catalyze the removal of diverse radionuclides and regarded as prospective adsorbents for radionuclides treatment in real environmental settings.

### 3.6. Removal of toxic metals

The electrolysis, manufacturing, metallurgy, metal electroplating, ceramics and mineral extraction industries generate an enormous amount of wastewater containing various metals (i.e., Zn, As, Pb, Hg, Cu, Ni, Cd, and Cr), which are non-degradable, carcinogenic, and extremely toxic (Qu et al., 2013; Wang et al., 2019a, 2019b;

Homaieghar, 2020). These metal ions after irrigation tend to enter freshwater via the soil, causing severe health problems to animals, aquatic plants, and humans (Zhao et al., 2019). MXene's appear as high-performance adsorbents in removing metals, and the explosion of recent reports are published in the scientific literature related to fabrication and applying MXene for environmental cleanup (Zhan et al., 2020). The adsorption capacity of MXenes for metal ions is primarily attributed to the surface moieties, which are generated during the alkalization-intercalation process.

Lead is regularly produced by various agricultural and industrial processes, like mining, batteries, metal plating, paint manufacturing and fertilizers. It is among the most common environmental contaminants and considered hazardous toxins (class 2 hazardous compound) (Moore, 1977). A high level of lead may result in renal, reproductive, cardiovascular, gastrointestinal, and central nervous system disorders. Dong et al. (2019) developed a novel type of MXene/alginate biocomposites to remove copper and lead ions from wastewater samples. The as-fabricated hybrid composites enhanced the chelation of copper and lead ions as well as improved the ion transport efficacy. Integrated benefits of rapid equilibrium time and superior adsorption capacity constitute MXene/alginate hybrid nanocomposites to adsorb 87.6 and 382.7  $\text{mg g}^{-1}$  of  $\text{Cu}^{2+}$  and  $\text{Pb}^{2+}$ , respectively and achieving the adsorptive equilibrium in just 15 min. MXene ( $\text{Ti}_3\text{C}_2\text{Tx}$ ) was applied to eliminate Pb(II) from aqueous media by adsorption in the presence of powder activated carbon as a control agent of MXene. Despite the low surface area than powder activated carbon, MXene showed superior adsorption capacity owing to its high negative charge on the surface. MXene-assisted adsorption of Pb(II) was better elucidated by the Freundlich isotherm models and pseudo-second-order kinetic. Based on the experimental outcomes attained using solutions of various ionic strengths, pH and humic acid levels, the electrostatic attraction was determined to be the main Pb(II) adsorptive mechanism. In addition, MXene demonstrated encouraging recyclability for four consecutive adsorption/desorption cycles (Jun et al., 2020a, 2020b, 2020c).

Chromium produced by different industrial process, like electroplating, metallurgy, refractory, and leather tanning, is one of the toxic metals (Gu et al., 2019). In wastewater, chromium ions can exist either in hexavalent Cr(VI) or trivalent Cr(III) forms. It is reported that all forms of chromium can cause diarrhea, asthma and damage the renal, liver, and male reproductive system (GracePavithra et al., 2019), necessitating its complete removal on an urgent basis. By in situ (3-aminopropyl)triethoxysilane polymerization onto multilayer  $\text{Ti}_3\text{C}_2\text{Tx}$  the surface, Kong et al. (2021) constructed amino-functionalized MXenes ( $\text{NH}_2\text{-Ti}_3\text{C}_2\text{Tx}$ ) for the removal of Cr(VI) ions in an aqueous environment.  $\text{Ti}_3\text{C}_2\text{Tx}$  nanosheets and amino groups presented cooperative roles in reducing and adsorption of Cr(VI) anions. Using the Langmuir model, the highest  $\text{NH}_2\text{-Ti}_3\text{C}_2\text{Tx-0.5}$  catalyzed adsorption was calculated to be 107.4 mg/g for Cr(VI). The synthesized  $\text{NH}_2\text{-Ti}_3\text{C}_2\text{Tx-0.5}$  nanosheets displayed potential reusability and superior selectivity for removing Cr(VI). A series of new MXene-chitosan organic-inorganic green nanocomposites were constructed by the solution reaction technique and employed to treat Cr(VI)-loaded polluted water. With an increase in MXene-chitosan dose level from 0.02 to 0.12 g/L, the Cr(VI) elimination efficiency was enhanced from 12.9% to 40.5%. The adsorption phenomenon was best explained by the pseudo-second-order kinetics that indicates the occurrence of chemisorption. In summary, MXene-based composite materials can be regarded as promising adsorption alternatives for chromium ions from wastewater.

Mercury ions are volatile, toxic, and easily accumulated leading to serious health and environmental pollution issues (Rasheed et al., 2018, 2019a,b; Bilal et al., 2018). The adverse mercury impacts on the neurons and nervous system are well recognized. It can also affect biological creatures, where it reacts with hemoglobin, albumin and sulphur-containing amino acids, and thus declaring as among the top 10



most hazardous metal ions (Guzzi et al., 2021). In a recent study, titanium carbide MXene core ( $\text{Ti}_3\text{C}_2\text{T}_x$ ) shell aerogel spheres (MX-SA) were fabricated to remove mercuric ions. The synthesized MX-SA<sub>4:20</sub> spheres showed excellent adsorption capacity for  $\text{Hg}^{2+}$  (932.84 mg/g). It achieved 100% removal of  $\text{Hg}^{2+}$  along with over 90% efficiency for 5 other toxic metal ions. Under extreme pH environments (0.5–1.0 M  $\text{HNO}_3$ ), the nanomaterials were found to be highly efficacious for  $\text{Hg}^{2+}$  eradication and possess satisfactory reusable features. Interestingly, the spherical shape and micro-size enabled MX-SA<sub>4:20</sub> for applying in column-packed reactors (Shahzad et al., 2019). Of most recent, Fu et al. (2020) prepared multilayered oxygen-functionalized  $\text{Ti}_3\text{C}_2$  (M- $\text{Ti}_3\text{C}_2$ ) nanosheets for removing mercury from aqueous solutions. The newly constructed M- $\text{Ti}_3\text{C}_2$  exhibited impressive adsorption capacity (4806 mg  $\text{g}^{-1}$ ), ultrafast adsorption kinetics, broad pH working behavior (pH 3.0 to 12) and high selectivity. According to the experimental outcomes and DFT calculations, this exceptional removal of  $\text{Hg}(\text{II})$  was ascribed to distinct interactive coordination like synergistic adsorption and catalytic reduction (Fig. 9). Over 95% of  $\text{Hg}_2\text{Cl}_2$  could be recovered using an easy thermal strategy. M- $\text{Ti}_3\text{C}_2$  was oxidized to  $\text{TiO}_2/\text{C}$  nanocomposites due to the adsorbed  $\bullet\text{OH}$  and release of energy during interactions, and these nanocomposites led to superior photocatalytic performance to degrade organic contaminants compared to Degussa P25. Efficient degradation together with recyclable properties render M- $\text{Ti}_3\text{C}_2$  an emerging designer candidate for quick removal and recovery of  $\text{Hg}(\text{II})$ .

Copper ion,  $\text{Cu}(\text{II})$ , is another toxic metal that is widely discharged by various industrial activities such as mining, fertilizer synthesis, electronics and batteries production, and paper and pharmaceutical manufacturing (Krstić et al., 2018). Excessive exposure to copper can result in liver and kidney damage, intravascular bleeding, and gastrointestinal injury. The copper intoxication manifestations are identical to food poisonings, such as vomiting, nausea, diarrhea, and headache (Saravu et al., 2007). Studies have reported the mitigation of copper using MXenes and their conjugated materials. Shahzad et al. (2017) evaluated the efficiency of  $\text{Ti}_3\text{C}_2\text{Tx}$  MXene nanosheets in the adsorptive removal of copper from aqueous solutions.  $\text{Ti}_3\text{C}_2\text{Tx}$  presented outstanding Cu removal performance due to hydrophilicity, substantial surface area, and distinctive surface functional attributes. Characterization analyses revealed that oxygenated moieties in the MXene layered structure are likely to facilitate the reduction of  $\text{Cu}^{2+}$  to form  $\text{CuO}$  and  $\text{Cu}_2\text{O}$  species. The utmost adsorption was recorded to be 78.45 mg  $\text{g}^{-1}$ , with about 80% adsorption of metal ions within 1 min. Freundlich adsorption isotherm and pseudo-second-order kinetic model precisely explained the highest Cu uptake and equilibrium time, respectively. Compared to activated carbon, DL- $\text{Ti}_3\text{C}_2\text{Tx}$  displayed 2.7-times higher adsorption capacity, illustrating the promise of MXene nanosheets in removing toxic copper from water samples.

#### 4. Challenges of MXenes exploitation and perspectives

Notwithstanding the significant progress in the design and synthesis of MXenes, several challenges like less biocompatibility, insufficient durability, high aggregation tendency, and inadequate recyclability remain to be addressed for effective exploitation of MXenes-based composites in removing hazardous pollutants from environmental matrices. In addition, the rational fabrication of MXene with controlled surface properties and well-defined structures is another major challenge. Though over 30 different kinds of MXene-based compounds have been reported, most of these are prophesied theoretically; thus intensive research is required to experimental fabrication of various functionalized MXene with suitability for water decontamination. In addition, the biocompatibility and cytotoxicity aspects of MXenes compounds should be inspected in detail before their deployment in the remediation of wastewater. Based on the literature assessment, it was observed that most of the earlier studies mainly emphasized the use of  $\text{Ti}_3\text{C}_2\text{Tx}$  for

water treatment without considerable efforts in designing other new MXenes. Nevertheless, minimum interlayer spacing and restack tends in aqueous systems hinders the large-scale application of these MXenes in removing pollutants. The surface attributes, reusability, and stability of MXenes might be significantly improved by integrating with abundant, cheaper, and stable polymers, as well as other carbon materials with interesting physicochemical characteristics and substantial-high surface. Therefore, additional investigations are needed to deeply investigate the application of MXene-based nanocomposites in mitigating a vast number of toxic contaminants from actual wastewater samples. Moreover, comparative analysis between MXenes and a range of other nanoadsorbents, like graphene, MOFs, and carbon nanotubes, should be assessed. Undoubtedly, MXene-based products are likely to appear in the commercial market soon for various applications. Considering encouraging outcomes in plenty of reports, it can be concluded that MXenes-based nanostructures could be considered the next-generation candidates for water remediation.

#### 5. Conclusions

In summary, MXene and MXene-based nanoarchitectures have emerged as highly promising candidates for the adsorptive mitigation of an array of environmentally related pollutants from water and wastewater. Targeted polluting agents examined for removal include organic dyes, toxic heavy metals, pesticide residues, phenolics, antibiotics, radionuclides, and many others. MXenes-based nanomaterials are progressing continuously to prove their potentiality in the arena of wastewater purification and remediation. Given wider functionality, elevated surface area, and surface tunability, MXene-based nanoarchitectures are thought to mitigate inorganic pollutants via interfacial chemical transformation and sorption, while three different mechanisms including i) surface complexation and sorption (ii) catalytic activation and removal and (iii) radical's generation-based photocatalytic degradation are involved in the removal of organic contaminants. However, considerable attention needs to be devoted to the rational designing and development of new experimental-based MXene nanomaterials with adjustable surface chemistry, numerous transition metal ions and robust features to trap pollutants. Modification of the interlayer spacing, crystallinity, and other surfaces/bulk attributes are ongoing research work for expanding the scope of MXene for wastewater remediation. Further research is vital to utilize different MXene lightweight MXene nanoarchitectures than heavier MXene to remove contaminants from wastewater. Apart from reusability and stability issues, the degradation mechanism of environmentally hazardous pollutants should be experimentally elucidated in actual reaction conditions and compared with other commercial adsorbents.

#### Credit author statement

Shuangshuang Zhang: Data curation, Writing-Original Draft, Reviewing and Editing. Muhammad Bilal: Conceptualization, Methodology, Figures, Tables, Writing-Original Draft, Resources, Reviewing and Editing. Muhammad Adeel: Data curation, Writing-Original Draft, Reviewing and Editing. Damià Barceló: Reviewing and Editing. Hafiz M.N. Iqbal: Conceptualization, Resources, Writing-Original Draft, Reviewing and Editing.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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