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MXENES FOR ENVIROMENTAL APPLICATIONS

The development of new methods for obtaining clean water is one of the most important global problems of mankind. It is important to purify water from harmful impurities that can adversely affect human health and the environment. Natural renewable energy sources, such as solar radiation, can be perfectly used for cleaning, distilling and desalination of seawater. The analysis of the study showed that MXene with unique and customizable properties provide new strategies for effective water purification and desalination. Experimental results indicate that representatives of the two-dimensional materials family (Mxene) have demonstrated improved light absorption and water evaporation characteristics. Thus, MXenes demonstrate a huge potential in the production of solar energy in the form of steam to solve the problem of shortage of drinking water. And also, according to the results of research, MXene showed a high degree of adsorption of heavy metal ions, such as Pb (II), Cr (VI), Hg (II) and U (VI). The large surface area, abundance of active sites, good dispersibility and reducibility, and the fact that many cations can be spontaneously inserted between the layers of two-dimensional MXenes make them promising candidates for water purification from toxic oxidants associated with heavy metal ions, radionuclides, and organic dyes.

Key words: Mxene, coefficient of performance (COP), adsorption

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Қоршаған ортаны қорғау үшін MXenes қолдану

Таза су алудың жаңа әдістерін жасау адамзаттың ең маңызды жаһандық мәселелерінің бірі болып табылады. Суды адам денсаулығына және қоршаған ортаға теріс әсер ететін зиянды қоспалардан тазарту маңызды. Күн радиациясы сияқты табиғи жаңартылатын энергия көздерін теңіз суын тазарту, айдау (дистилляция) және тұзсыздандыру үшін пайдалануға болады. Зерттеудің талдауы бірегей және теңшелетін қасиеттері бар MXenes суды тиімді тазартудың және тұщыландырудың жаңа стратегияларын қамтамасыз ететінін көрсетті. Эксперимент нәтижелері екі өлшемді материалдар тобының (Mxenes) өкілдері жарықты сіңіру және судың булану сипаттамаларының жақсарғанын көрсетті. Осылайша, MXenes ауыз су тапшылығы мәселесін шешу үшін бу түріндегі күн энергиясын өндіруде үлкен әлеуетті көрсетеді. Сондай-ақ, зерттеу нәтижелері бойынша MXenes Pb (II), Cr (VI), Hg (II) және U (VI) сияқты ауыр металл иондарының адсорбциялануының жоғары дәрежесін көрсетті. Үлкен бетінің ауданы, белсенді учаскелердің көптігі, жақсы дисперстілігі мен тотықсыздануы және екі өлшемді MXenes қабаттарының арасына көптеген катиондардың өздігінен енуі оларды ауыр металл иондарымен, радионуклидтермен байланысты улы тотықтырғыштардан суды тазарту үшін перспективалы кандидаттар етеді.

Түйін сөздер: Mxene, өнімділік коэффициенті (COP), адсорбция

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Применение MXenes в области защиты окружающей среды

Разработка новых методов получения чистой воды является одной из важнейших глобальных проблем человечества. Важно очищать воду от вредных примесей, которые могут плохо воздействовать на здоровье человека и окружающую среду. Природные возобновляемые источники энергии, подобные как солнечная радиация, могут отлично употребляться для чистки, дистилляции и опреснения морской воды. Анализ исследования показал, что MXenes с уникальными и настраиваемыми свойствами обеспечивают новые стратегии эффективной очистки и опреснения воды. Экспериментальные результаты свидетельствуют о том, что представители семейства двумерных материалов (Mxenes) продемонстрировали улучшенные характеристики поглощения света и испарения воды. Таким образом, MXenes демонстрируют огромный потенциал в производстве солнечной энергии в виде пара для решения проблемы нехватки питьевой воды. А также, по результатам исследований MXenes показали высокую степень адсорбции ионов тяжелых металлов, таких как Pb (II), Cr (VI), Hg (II) и U (VI). Большая площадь, обилие активных сайтов, хорошая распыляемость и восстанавливаемость, а также тот факт, что многие катионы могут спонтанно вставляться между слоями двумерных MXenes, делает их перспективными кандидатами для очистки воды от токсичных окислителей, связанные с ионами тяжелых металлов, радионуклидов и органических красителей.

Ключевые слова: Mxene, коэффициент полезного действия (КПД), адсорбция.

Introduction

Ecological/Environmental applications

With the rapid growth of the world population and the rise of agriculture and industrial production, a shortage of drinking water is predicted [1]. Therefore, the development of new methods for obtaining clean, fresh water is one of humankind's most important global problems [2]. To improve water quality, it is necessary to remove harmful impurities that can negatively affect human health and the environment. For example, some of the most common pollutants are heavy metal ions, toxic gases, dyes, hormones, and organic solvents [1]. Among all water pollutants, heavy metal ions (Pb (II), Cr (VI), Hg (II), and U (VI)) are in first place in terms of toxicity to the human body. It is also important to purify water from various organic dyes, which are difficult to biodegrade due to their stable molecular structure [3]. Recently photothermal conversion, membrane separation, and adsorption have become the most popular water purification methods [4–6].

Photothermal conversion

Studies show that natural renewable energy sources such as solar radiation can be effectively used for the purification, distillation, and desalination of seawater [7]. For such purposes, solar steam generators based on the principle of photothermal conversion have demonstrated their effectiveness. In the classical solar steam generators, solar energy gets absorbed by the receiver and then used to heat up the main fluid due to thermal conductivity and thermal convection [1,8]. The coefficient of performance (COP) of such systems is less than 50% since most of the absorbed solar thermal energy is used to heat the main fluid. For efficient water evaporation due to light absorption and reduction of heat losses in the system, the evaporator must have high hydrophilicity and photothermal conversion, excellent light absorption characteristics, and a porous structure to ensure sufficient water supply to the absorbers [9]. In addition, photothermal materials should not sink the water since placed directly at the water-air interface reduces heat loss to bulk water. Recent studies have shown that MXene exhibits high efficiency in absorbing solar energy and has great potential for solar steam generator applications [10].

Literature Review

Methods for obtaining Mxene, a solar steam generator for desalination of water based on a hydrophobic membrane with Ti_3C_2

For example, in 2017, the most popular representative of the MXene family, Ti_3C_2, was studied as a solar photothermal material [10]. For its synthesis, the Ti_3AlC_2 MAX phase was used, which was treated with an aqueous solution of HF for selective etching of aluminum, followed by immersion in DMSO and sonication. After that, the aqueous suspension of MXenes was filtered through a hydrophilic membrane of polyvinylidene fluoride (PVDF) 0.22 μm thick. Thus, a thin layer of stacked MXene sheets was formed on the surface of the PVDF substrate. To test the characteristics of photothermal water evaporation, the MXene-PVDF photothermal membrane was modified by adding functional groups using a solution of polydimethylsiloxane (PDMS) in hexane. Ti_3C_2 demonstrated close to 100% light to heat conversion and excellent light absorption. In this case, the wavelength of the laser source, 473 or 785 nm, did not adversely affect the efficiency of titanium carbide in any way, which indicates the ideal characteristics of photothermal conversion. In addition, a thin MXene-PVDF membrane with a thermal barrier provided a photothermal evaporation efficiency of about 84%. The disadvantage of such a membrane was that it was wetted in water, although after modification with PDMS it could float on the surface of water on its own. The non-wetting surface naturally repels water and prevents it from entering the membrane during desalination with the help of solar radiation [11]. Such high characteristics of Ti_3C_2 suggest that MXene-based membranes can provide effective water purification from contamination and environmentally friendly long-term operation [12].

Later, in 2018, a self-floating solar steam generator for water desalination based on a hydrophobic membrane with Ti_3C_2 was presented [11]. Two-dimensional titanium carbide nanosheets were synthesized by etching the Ti_3AlC_2 MAX phase with an HCl/LiF solution. MXene membrane was prepared by filtering delaminated Ti_3C_2 nanosheets modified with a hydrolysis solution of perfluorodecyltrimethoxysilane (PFDTMS) (0.5 vol% acetic acid, 2.0 vol% PFDTMS and 97.5 vol% isopropanol) onto a mixed ester membrane cellulose with a pore size of 0.22 microns. The solar steam generator device consisted of three components: membrane with Ti_3C_2 on filter membrane (solar energy absorber, vapor generator, and salt blocker), commercial polystyrene foam (heat insulator and flotation device), and nonwoven wicks (waterway due to capillary effect). To test the effectiveness of the hydrophobic MXene membrane, a hydrophilic counterpart was also prepared. Modification of Ti_3C_2 with PFDTMS led to the formation of functional groups on the -CF_3 surface, which were responsible for the hydrophobic character. Figure 1 shows a schematic illustration of the evaporation process of hydrophilic and hydrophobic Ti_3C_2 membranes. In the case of a hydrophilic membrane, the capillary effect facilitates seawater penetration, leading to the continuous crystallization of salts on the membrane surface with water evaporation. Accumulation of large amounts of salt can cause severe damage to the membrane, as shown in Figure 1a. However, a hydrophobic membrane is non-wettable due to its ability to block water together with dissolved salts under the membrane while vapors freely leave its pores (Figure 1d). Therefore, using a hydrophobic Ti_3C_2 membrane in a steam generator device promotes long-term and stable desalination using solar energy. For example, the concentration of the four primary ions (Na^+, K^+, $Mg^{2+,}$ and Ca^{2+}) can be markedly reduced to the level of 99.5%, which indicates effective desalination of seawater. It was also found that the membrane was able to efficiently vaporize typical contaminants (filtration rate of about 100%) that may be contained in wastewaters, such as organic dyes, reactive dark blue (RhB) and methyl orange (MO), heavy metal ions, Cu^{2+} and Cr^{6+}, and other organic substances, including benzene and acetone (Figure 1f).

Ju et al. [6] presented a macroporous three-dimensional MXene architecture (3DMA) for high-efficiency solar steam production. Using a two-step immersion method, such structures were obtained by applying $Ti_3C_2T_x$ MXene layers onto a melamine foam (MF) scaffold. At the first stage, the MP was immersed in polyvinyl alcohol (PVA) solution and dried in a vacuum. At the second stage, the resulting MA/PVA composite scaffold was immersed in a suspension of MXenes, followed by vacuum drying. PVA acted as an adhesion promoter between MXene and the scaffold. Figure 1 a,b shows the difference in the morphology of melamine foam before and after modification of $Ti_3C_2T_x$. A decrease in pore size was observed as the MXene nanolayers were collected on the scaffold walls in the form of large chunks. To assess the efficiency of solar steam generation, the resulting porous structure was placed at the water-air interface while it floated freely on the water surface without any support. The efficiency of 3DMA for a solar radiation intensity of 1000 $W{\cdot}m^{-2}$ was 82.4% at an evaporation rate of 1.309 $kg{\cdot}m^{-2}{\cdot}h^{-1}$, and at an illumination of 5000 $W{\cdot}m^{-2}$, it reached 88.1% at an evaporation rate of 6.997 kg $\cdot M^{-2}{\cdot}h^{-1}$ [9]. The resulting three-dimensional structure was effective for the desalination of seawater, which was manifested by a significant decrease in the concentration of Na^+, K^+, $Mg^{2+,}$ and Ca^{2+} cations (Figure 1c). In addition, when treating wastewater from methyl orange and methylene blue (MB) dyes, 3DMA demonstrated a capture rate close to 100%, as shown in Figure 1d.

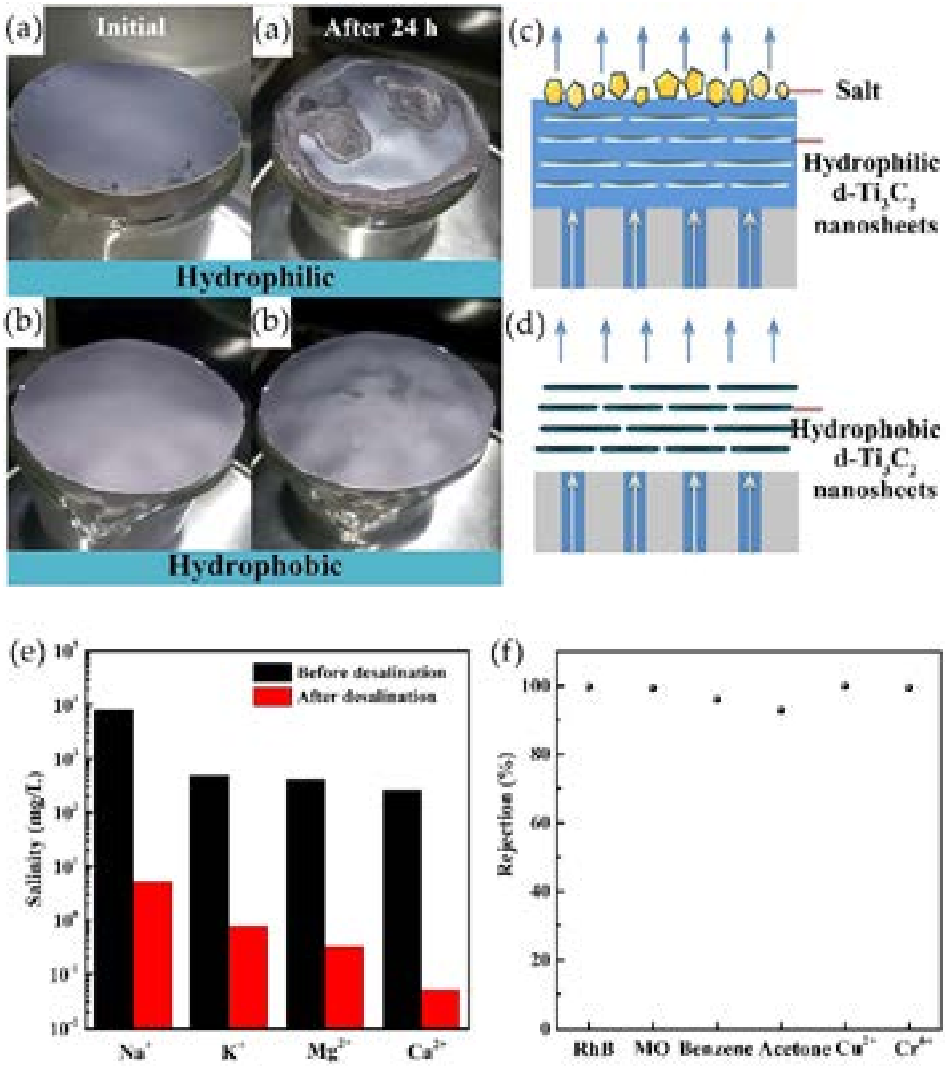

Figure 1 – Optical photographs of (a) hydrophilic and (b) hydrophobic membranes with Ti_3C_2 before and after 24 hours of desalination. Schematic representation of the desalination process by solar steam generation using (c) hydrophilic and (d) hydrophobic membranes. (e) Results of measuring the salinity of Na^+, K^+, Mg^{+} and Ca^+ primary ions before and after desalination. (f) Filtration efficiency of organic substances and heavy metal ions. Reprinted with permission from [11] 2018 Royal Society of Chemistry.

Steam generator based on $Ti_3C_2T_x$

Additionally, a steam generator based on nanocoatings of two-dimensional $Ti_3C_2T_x$ MXene was described for the first time, which imitated the hierarchical textures of black scales of the West African gaboon viper (*Bitis rhinoceros*) [13]. MXene nanolayers were obtained by etching the $Ti_3Al_2C_2$ MAX phase in a LiF-HCl fluoride solution followed by ultrasonic-assisted delamination. The aqueous MXene solution was then sprayed onto thermosensitive polystyrene substrates and air-dried. Then, the samples were warmed up to a temperature above the glass transition temperature of polystyrene while still below 140 °C to exclude degradation of MXene. This caused them to shrink to ≈50% of the original length and ≈25% of the original area. Repeating the described deformation process several times, a biomimetic hierarchical structure of MXene was obtained, where the primary crumpled structure mimicked the micro-ridges of the black scales of vipers and the secondary structure (more minor folds) resembled the texture of nano-ridges. The presence of such a branched texture of biomimetic $Ti_3C_2T_x$ nanocoatings promoted multiple scattering and reflection of light, causing broadband light absorption (84.9–86.9%) and improved equilibrium temperature at illumination of 1000 W·m^{-2} (58.2–62.6 °C) compared to conventional flat samples MXene (46.8% – 64.0% and 50.4–58.1 °C). An increase in the complexity of the structure (degree of deformation) of the hierarchical nanocoating caused a further increase in light absorption and thermal conductivity up to 93.2% and 65.4 °C, respectively. Solar steam generating devices based on such biomimetic MXene nanocoatings demonstrated a high evaporation rate of about 1.33 kg·m-2·h^{-1} at a solar radiation intensity of 1000 W·m^{-2}. It has also been shown that they can be used to implement flexible solar/electric heaters. Consequently, the MXene surface treatment technology had a significant impact on the final performance of the device.

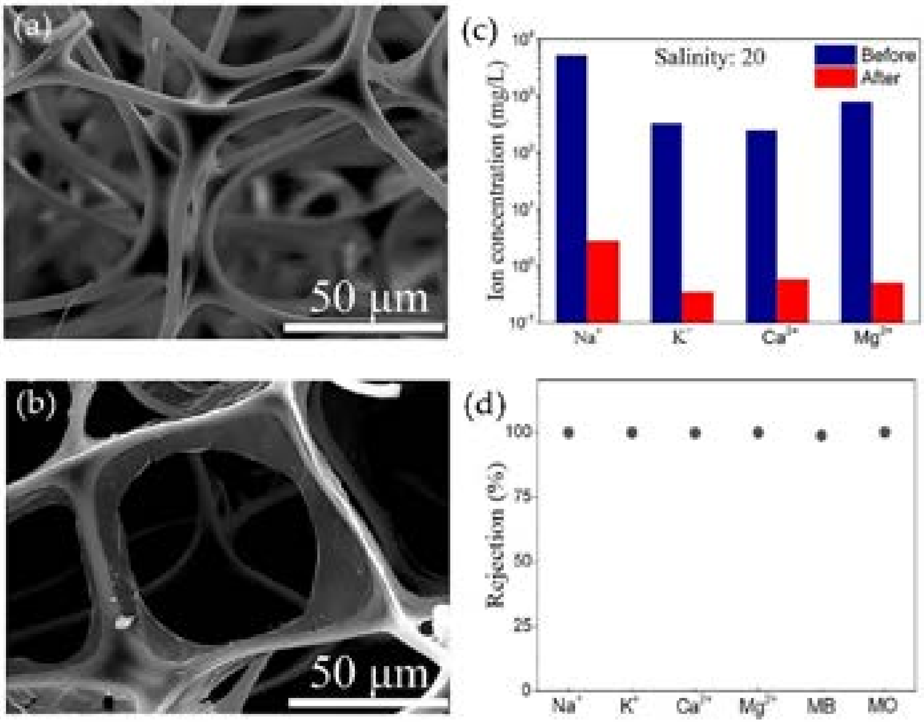

Figure 2 – SEM images of melamine foam (a) before and (b) after immersion in a solution of polyvinyl alcohol and MXene suspension, followed by drying. (c) Concentrations of Na^+, K^+, $Mg^{+,}$ and Ca^+ cations in a standard seawater sample with a salinity of 20 before and after evaporation. (d) Degree of trapping of metal ions and dyes (methylene blue, MS, and methyl orange, MO). Reprinted with permission from [9] 2019 Royal Society of Chemistry.

Highly efficient conversion of solar energy into steam using composite material PDA@MXene

In 2020, Zhao et al. reported a new strategy for highly efficient solar energy conversion to steam using composite polydopamine@$Ti_3C_2T_x$ (PDA@MXene) microspheres [14]. Mixing a colloidal solution of exfoliated MXene and an aqueous dispersion of PDA led to the formation of many aggregates, which were self-assembled PDA microspheres wrapped in $Ti_3C_2T_x$ (PDA@MXene). Numerous hydrogen bonds ensured the adhesion between nanosheets and microspheres. The photothermal layer of composite microspheres demonstrated a synergistic effect in efficiently absorbing solar radiation and converting solar radiation into steam. The steam generation efficiency at an illumination of 1000 and 4000 W·m^{-2} reached 85.2% and 93.6%, respectively. In addition, the PDA@MXene photothermal layer allowed the production of pure water from seawater with a salt capture rate of over 99%. The maximum rate of water evaporation using a PDA@MXene membrane with a loading mass of microspheres of about 0.8 mg/cm^2 was 1.276 kg·m^{-2}·h^{-1} at a radiation intensity of 1000 W·m^{-2}. The photothermal layer of the composite was mechanically stable and effectively maintained stable light absorption and water evaporation characteristics after several lighting cycles. However, experiments with seawater desalination showed that the surface of the hydrophilic membrane becomes contaminated after 10 hours of desalination due to salt crystallization. In this case, washing off these crystals did not affect the subsequent characteristics of the evaporation of membranes based on composite PDA@MXene microspheres.

Thus, it can be concluded that MXenes, with their unique and customizable properties, provide new strategies for efficient water purification and desalination. So far, only $Ti_3C_2T_x$ was used as photothermal materials. However, strong experimental results suggest that other members of this large family of two-dimensional materials will also be effective and possibly exhibit improved light absorption and water evaporation characteristics. Therefore, MXenes show tremendous potential in generating solar power in steam to solve the problem of drinking water shortages.

Water purification

Adsorption

In addition to other strategies, MXenes are actively used for another popular water purification method, namely adsorption. This is due to its simplicity, economy, and the absence of secondary pollution [15]. Adsorption is considered the most effective method for removing heavy metal ions because other strategies such as biological processing or chemical reactions cannot metabolize them or break them down. However, for the material used as an adsorbent to be effective, it has to have a large surface area and high functionality [16].

It was found that titanium exhibits a high sorption affinity for metal ions [17]. Therefore, in 2014, Peng and colleagues [18] investigated the sorption capacity of Ti-based MXene to purify water from lead ions. They suggested that since the surface of individual Ti_3C_2 nanolayers possesses terminals with functional OH or F groups, the substitution of the cation in the hydroxyl group can lead to the activation of ion exchange sites, providing sites for sorption of toxic metals. Moreover, the large area of the layered MXenes promotes the binding of metal ions that have to be removed [19]. For the sorption of Pb (II), the authors proposed $Ti_3C_2(OH/ONa)_xF_{2-x}$, which was synthesized by selective etching of the Ti_3AlC_2 MAX phase followed by alkalinization-intercalation. As a result of the treatment, some of the terminal surface F groups were partially replaced by OH groups, and OH groups were converted to ONa groups. The results of kinetic tests showed that sorption equilibrium was reached in just 120 s. In addition, $Ti_3C_2(OH/ONa)_xF_{2-x}$ made it possible to lower the Pb (II) content in wastewater to levels below 10 µg/L. Moreover, the adsorption properties of the material were reversible. Such a high sorption capacity was associated with hydroxyl groups in activated Ti sites, where the Pb (II) ion-exchange was facilitated by forming a hexagonal potential trap. However, the residual amount of F groups weakened the adsorption of lead cations [20].

Later, papers were published in which the use of MXenes for water purification by adsorption from other toxic metals was investigated. For example, $Ti_3C_2T_x$ nanolayers synthesized by selective etching of Ti_3AlC_2 in a 10% HF solution followed by ultrasonic delamination have demonstrated excellent ability to remove Cr (VI) [21]. Hexavalent chromium is a strong oxidizing agent that is highly toxic to plants, animals, and humans. Improper disposal of wastewater after various industrial processes causes severe groundwater pollution and damages the environment [21]. The maximum removal capacity for Cr (VI) was 250 mg/g. At the same time, the residual concentration of hexavalent chromium after purification (less than 5 parts per billion) was much lower than the standard for drinking water recommended by the World Health Organization (0.05 parts per million). Removal of Cr (VI) by two-dimensional $Ti_3C_2T_x$ nanolayers occurs via a reduction reaction. MXenes reduce Cr (VI) to a less toxic form of Cr (III), which then can be removed without alkaline treatment at an optimum pH of 5.0. It has also been shown that $Ti_3C_2T_x$ can be used for the reductive removal of other oxidants such as $K_3[Fe(CN)_6]$, $KMnO_4$ and $NaAuCl_4$.

In 2016, Wang and co-authors demonstrated for the first time the use of multilayer V_2CT_x MXene as a potentially effective adsorbent for U (VI) [22]. Efficient treatment of nuclear waste is becoming a serious environmental problem, as long-lived actinide contamination threatens the environment even in small quantities due to their radiological and chemical toxicity. Therefore, the use of MXene as an adsorbent for trapping radionuclides is a promising direction. The V_2CTx powder was obtained by immersing the V_2AlC MAX phase powder in a 40% concentrated HF solution, followed by stirring. V_2CT_x was generally stable in the process of capturing U (VI) from aqueous solution in the pH range from 3.0 to 5.0, but its sorption behavior showed a significant dependence on pH. The best results were obtained at pH 5.0. For example, the maximum absorbent capacity of a powder containing 32 at.%

of the unreacted V_2AlC powder was 174 mg/g. The adsorption capacity of pure two-dimensional MXene nanolayers reached about 256 mg/g. The weak binding of uranium ions at acidic pH levels (lower than 3) also suggests that multilayer V_2CT_x can be regenerated with acidic solutions. Due to functional -OH/-O and -F surface groups, which play a role in the heterogeneous adsorption sites, the capture of U (VI) can be described by a heterogeneous adsorption model. The results of experiments and theoretical calculations showed that V_2CT_x adsorbs uranium ions by forming bidentate adsorption configurations with hydroxyl groups attached to V atoms. The adsorption process proceeded according to the ion exchange mechanism, which was confirmed by deprotonation of the hydroxyl group after binding to U (VI).

In addition to Pb (II), Cr (VI), and U (VI), Fu et al. showed in 2020 that MXene could also adsorb Hg (II) [23]. Since mercury, which is one of the most toxic heavy metals, is widely used in the process of extraction of precious metals, e.g. from electronic devices and industrial catalysis, excessive environmental pollution causes irreparable harm to human health and natural ecosystems [24, 25]. For practical use, it is desirable that the adsorbent can effectively remove Hg (II) ions from water even under harsh chemical conditions. The acid-base resistance of M-Ti_3C_2 nanosheets was evaluated at various pH values ranging from 1 to 12 (Fig. 2c). M-Ti_3C_2 nanosheets (dosage 0.1 g/L) were able to remove less than 10% Hg (II) at pH≤2.0. In contrast, the removal efficiency increased significantly at higher pH, and more than 99.0% of Hg (II) was removed over a wide pH range from 4 to 10. Moreover, M-Ti_3C_2 showed high removal efficiency (>97.0%) even in highly alkaline environments (pH > 11).

In parallel with the first experiment to remove Pb (II), it was demonstrated in 2014 that $Ti_3C_2T_x$ could also be effectively used for the adsorption of the cationic dye methylene blue (MB) [26]. The reaction of the MXene with an aqueous solution of MB under ambient conditions was described by three stages: (1) active adsorption of MB molecules on the surface of $Ti_3C_2T_x$; (2) an increase in the disordered packing of layers; and (3) oxidation of the MXene.

It has also been demonstrated that multi-layer V_2CT_x MXene could be successfully used as an adsorbent for removing colorants in wastewaters. Usually, dyes in water are found in the form of ions, so they can be removed using special adsorbents. More efficient adsorption of V_2CT_x, compared to the most studied $Ti_3C_2T_x$ could be explained by its simpler unit cell and greater distance between the layers [27].

Due to unique intrinsic properties, MXenes are of greater interest as adsorbents than graphene oxide, which consists of only one element, has a simpler and insufficiently functionalized surface, and a high cost [20, 28, 29].

Conclusion

A large surface area, an abundance of active sites, good dispersibility and reducibility, as well as the fact that many cations could spontaneously intercalate between layers of two-dimensional MXenes, make them promising candidates for water purification from toxic oxidants associated with heavy metal ions, radionuclides, and organic dyes. This review shows a solution to problems with the lack of drinking water through by the use of MXenes. To date, only two representatives of a large family of these materials, $Ti_3C_2T_x$ and V_2CT_x, have been studied as adsorbents. However, the emergence of new work with other MXenes is expected to substantially enrich the fundamental understanding of adsorption mechanisms and significantly expand the range of their possible applications for environmental cleaning.

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