



MXenes: The two-dimensional influencers

A B S T R A C T

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MXenes have significantly impacted materials science and nanotechnology since their discovery in 2011. Theoretical calculations have predicted more than 100 possible compositions of MXenes and lab-scale fabrication of more than 40 MXene structures has been reported to date. The unique characteristics of MXenes have made them an ideal fit for a wide variety of applications, including energy storage, environmental, electronics, communications, gas and liquid separations and adsorption, biomedical, and optoelectronics. MXene attracted many researchers, and as a result, publication trends on MXene have grown exponentially in recent years. By 2021, MXenes have already shown promise in several research areas, including energy storage devices, electromagnetic interference shielding, nanocomposites, and hybrid materials. In parallel, new applications are emerging where MXenes outperform other nanomaterials, such as in tribology. MXene compositions are also being expanded rapidly. Here, we briefly overview the history, properties, trends, and application of MXenes to better understand their potentials and familiarize new audiences with this 2D material family.

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MXenes are a novel family of 2D early transition metal carbides and nitrides that were discovered in 2011 [1]. $M_{n+1}X_nT_x$ formula describes the 2D structure of MXene, generally constructed by $n+1$ layers of early transition metals—labeled as M, interleaved by n layers of carbon or nitrogen—labeled as X, and surface terminations such as F, O, OH, or Cl — labeled as T_x [2,3]. The fabrication of MXenes starts with topochemical selective etching of the precursor material, usually a MAX phase. MAX phases are layered carbides and nitrides with a hexagonal closed packed structure (A in MAX stands for A-group elements of the periodic table, such as Al, Ga, Si) [4] and they play an essential role in the quality of MXene [5]. The selective etching is done by removing the A-layer atoms from the MAX phase and continues with the delamination and exfoliation of loosely stacked 2D MXene flakes [5]. MXene can be prepared in multilayered (ML) powder forms or single-flake colloidal solutions [6]. ML-MXenes are made of stacks of loosely attached MXene flakes, while colloidal MXenes are obtained by exfoliation of the ML-MXenes to single-flake MXenes [7]. Fig. 1a represents some schematic examples of common and newly discovered MXene structures.

To date, four different composition formulas of MXenes have been synthesized as M_2XT_x , $M_3X_2T_x$, $M_4X_3T_x$, and $M_5X_4T_x$. $Ti_3C_2T_x$, which is an $M_3X_2T_x$ structure, is the first reported MXene [1] and is the most studied MXene [9]. A schematic of $M_3X_2T_x$ is shown at the top left in Fig. 1a. Soon after, M_2XT_x and $M_4X_3T_x$ (Fig. 1a) were discovered in 2012 by synthesizing Ti_2CT_x and $Ti_4C_3T_x$ as well as solid solutions in M and X sites [10]. The number of layers (n in $M_{n+1}X_nT_x$) is controlled by the precursor structure, such as MAX phases [4]. For example, Ti_2CT_x and $Ti_3C_2T_x$ are made by etching the Al layers from two different MAX phases of Ti_2AlC

and Ti_3AlC_2 , respectively. The MXene dependence on the precursor means that novel MAX phase precursors are required to make thicker MXenes ($M_{n+1}X_nT_x$ with larger n) or new chemistry of M and X. In 2019, a MAX phase with five layers of M (M_5AlC_4) was discovered as $(Mo_4V)AlC_4$. By selective etching of the Al layers from this phase, the first $M_5X_4T_x$ as $(Mo_4V)C_4T_x$ was synthesized (Fig. 1a) [11].

Theoretical calculations on the MAX phases and MXenes have predicted more than 100 possible compositions of MXenes [12]. By synthesizing MAX phases with different elements, such as solid solutions on the M and X sites, solid solution MXenes can be made; this creates a vast compositional space to fabricate an infinite number of MXene compositions and tune physio-chemical properties of MXene by mixing different transition metals or synthesizing carbonitrides. While the first solid-solution MXene was synthesized in 2012 as $(Ti,Nb)_2CT_x$, Ti_3CNT_x , and $(Cr,V)_3C_2T_x$ [10], several MXene solid solutions were reported in 2020 [13,14]. In addition to solid solutions with random elemental compositions, combining certain transition metals in proper stoichiometric ratios, enables fabrication of ordered double-transition metals, such as in-plane ordered MXenes when $n = 1$ (e.g. $(Mo_{2/3}Ti_{1/3})CT_x$) [3] or out-of-plane ordered MXenes when $n = 2$ and 3 (e.g., $Mo_2TiC_2T_x$ and $Mo_2Ti_2C_3T_x$) [15]. In general, electron microscopy has been essential in understanding novel MXene structures, MXenes compositions, and defect distributions, as it has been reviewed by Alnoor et al. [16]. Additional tuning of the MXenes' precursor chemistry (M and X elements) can yield novel compositions and structures of MXenes. An example of these new compositions is high-entropy MXenes. Nemani et al. [17] reported the successful fabrication of two high-entropy $M_4X_3T_x$ MXenes, $TiVCrMoC_3T_x$ and $TiVNbMoC_3T_x$ by

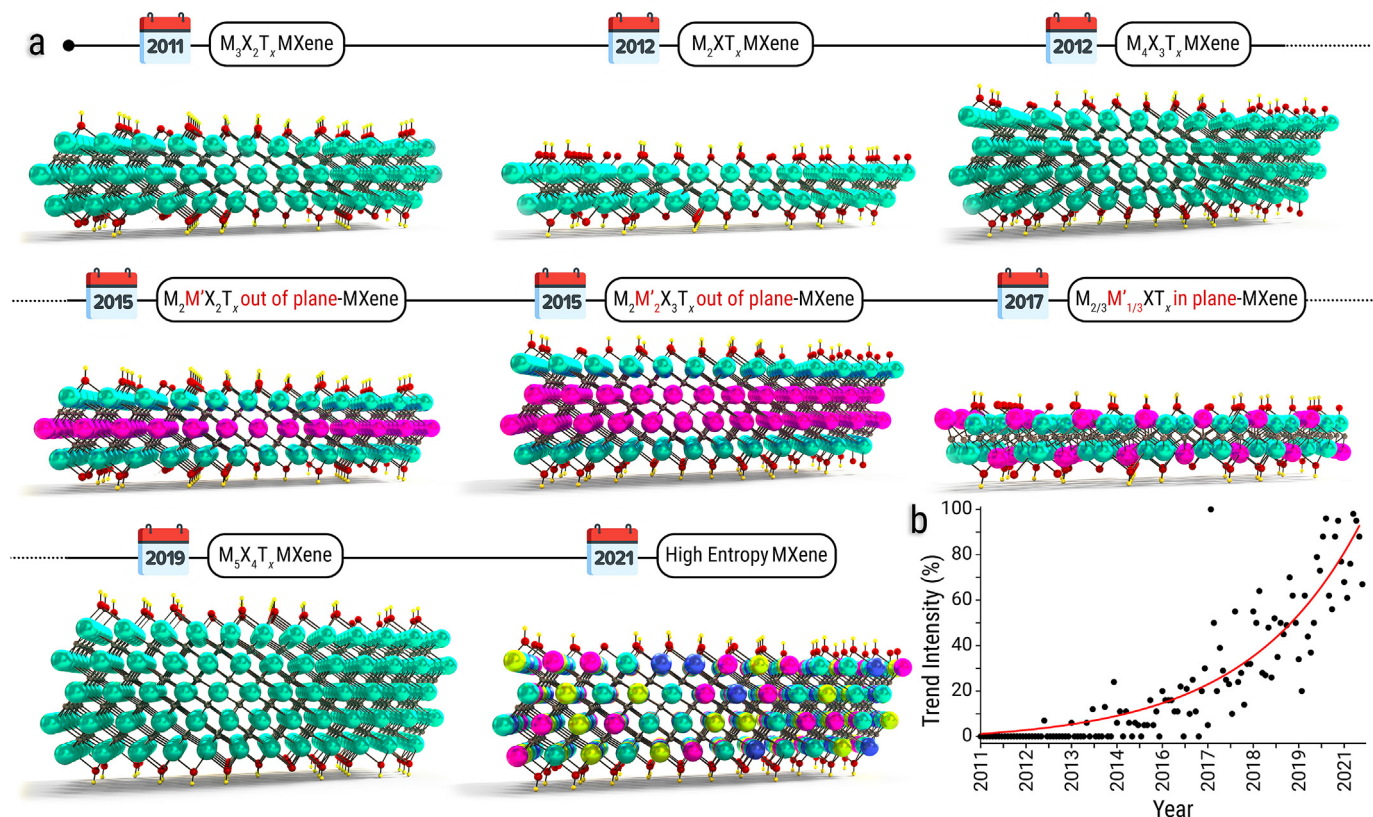


Fig. 1. (a) The schematic illustration of different MXene structures. $Ti_3C_2T_x$, as an example of $M_3X_2T_x$ MXene (top left), was the first discovered MXene in 2011, followed by the discovery of M_2XT_x and $M_4X_3T_x$ in 2012. The family of MXenes was expanded by the addition of ordered double-transition metal MXenes, out-of-plane ordered in 2015 followed by in-plane ordered in 2017. $M_5X_4T_x$ solid solution MXene and high-entropy $M_4X_3T_x$ MXenes are among the latest additions to the MXenes family. The green, yellow, pink, and blue color spheres represent early transition metals in MXenes. Surface terminations of MXenes are randomly scattered and attached to the outer atomic layer of the MXene structure, which are mainly O, F, or OH groups. (b) The monthly Google Trends data, collected over the decade from users' engagement with the word "MXene." These data are proportional, and the highest value is normalized to 100. Since 2015, there has been a considerable increase in trend intensity. Google Trends data in this work were mined using the "pytrends" Python library [8].

fabricating precursors of high-entropy MAX carbides; $TiVCr-MoAlC_3$, and $TiVNbMoAlC_3$, respectively. These are two high-entropy phases in the potentially large family of high-entropy MXenes (Fig. 1a bottom). This discovery was followed by the report of a M_2XT_x high-entropy MXenes in the same year [18]. The discovery of high-entropy MXenes adds another level of tunability to this fast-growing field.

The functional groups (T_x) in MXene structures are mainly governed by fluorine groups (F), hydroxyl (OH), and oxygen (O). Density functional theory (DFT) studies have predicted that the choice of MXene surface terminations can affect the electronic and magnetic properties of MXenes [19,20]. Bae et al. have shown that the choice of DFT methods is also critical in predicting MXenes' behavior and it can affect the predicted characteristics. For example, by studying 22 different M_2CT_x MXenes ($M = Sc, Y, La, Ti, Zr, Hf, V, Nb, Ta, Mo, W$, and $T = O, F$) with DFT and four different methods of Perdew–Burke–Ernzerhof (PBE), SCAN, HSE06, and PBE + U, they identified new magnetic states in V_2CO_2 , V_2CF_2 , and Mo_2CF_2 [19]. Almost all the experimental studies on MXenes have a mixture of different surface terminations ($-F, -O, -OH$), which make the realization of these unique properties challenging. However, a study in 2020 showed successful modification of MXenes with uniform surface terminations [21]. This implies that many DFT predictions on MXenes electronic and magnetic properties in the past ten years can be realized experimentally. In general, control of MXene properties via surface terminations and

modifications is an area that needs further investigation using both experimental and computational approaches.

Since the discovery of MXenes, more than 8300 authors have published their studies on MXenes or MXene-based materials in more than 400 journals. Over 1450 institutions from 62 countries were involved in this rapid expansion of MXene research [22]. The number of published articles on MXene exceeded 3000 counts in 2020. In 2020 and 2019, more than 1493 and 903 papers were published on MXene, respectively, while for years 2018 and 2017, this number was only 469 and 226, respectively [22]. Beyond the available data on the Web of Science, we investigated the monthly Google Trends data collected over the past decade, as shown in Fig. 1b. This graph represents users' search interest in the term "MXene" worldwide over the past decade. The data value is proportional, and a value of 100 is the peak engagement for the term "MXene." An exponential curve fitted the trend intensity data to determine the intensive growth of "MXene" engagement with users on the internet. This graph clearly shows a considerable increase in engagement since 2015 with continuous development. The growth in the number of publications and the exponential trend in the users' online search on MXenes combined with MXenes' wide variety of compositions demonstrate the rapid exploration of MXenes in various applications.

One of the applications in which MXenes outperform other materials is electromagnetic interference (EMI) shielding. Ti-containing MXenes, such as $Ti_3C_2T_x$ and Ti_3CNT_x , are shown to

have EMI shielding effectiveness higher than those of copper and aluminum thin films at the same thicknesses at the micrometer levels or less [23,24]. MXenes' EMI shielding can be further tuned by the use of different transition metals (M in MXenes) or by changing the number of layers (n), which typically has a direct correlation with MXenes' electrical conductivity [13]. Additionally, polymers have been used with MXenes to fabricate MXene-polymer composites to tune the EMI shielding effectiveness [21]. In general, in recent years, MXenes have been extensively used as filler nanomaterials in polymeric matrices that intensified their recognition in various research fields [25]. The hydrophilic nature of MXenes allows for interaction and reaction with hydrophilic polymers, e.g., polydiallyldimethylammoniumchloride, polyvinyl alcohol, and polyacrylic acid [25]. Compared to the other materials, MXenes offer specific advantages in polymeric composites owing to MXenes' surface functionalities, a wide range of compositions, high aspect ratios, and high surface area of the 2D flakes. When MXenes are homogeneously dispersed into the polymer chains, considerable improvements can be achieved in thermal, morphological, mechanical, and rheological properties [25]. The number of journal articles published on MXene-polymer nanocomposites (NCs) from 2018 to 2020 was more than 150, while the sum published between 2011 and 2017 was lower than 50 [25]. In recent years, conductive polymers, such as poly (aniline), polypyrrole, and polystyrene sulfonate, have been among the first choices of studies for interaction with MXene. These NCs can be used as strain sensors, electromagnetic shields, energy storage devices, and gas sensors [26,27]. Another advantage of MXene-polymer composites is the increase in the shelf-life of MXenes. In general, MXenes susceptibility to oxidation is the primary factor to be considered. Although the oxidation rate of MXene in NCs depends on their exposure to air, water, and temperature, embedding MXene flakes in solid polymer NCs can slow down the oxidation [28,29].

Electrochemical energy storage is the first and most studied application of MXenes [9]. MXenes' high electronic and ionic conductivity makes them an appealing choice for use in batteries and supercapacitors. New findings in MXenes compositions and composites have elevated their previous performance in energy storage properties and they remain an attractive target for these applications. For example, recently, Organi et al. [30] fabricated a free-standing, ultralightweight, additive and binder-free $\text{Ti}_3\text{C}_2\text{T}_x$ MXene through unidirectional freeze casting. Their study showed that $\text{Ti}_3\text{C}_2\text{T}_x$ MXene aerogel could be aligned in submillimeter domains along the temperature gradient with strain resistance up to 50%. This MXene aerogel revealed outstanding electrochemical response along with superb rate performance, high specific capacity, and high cyclic stability. This study shows that preventing MXene flakes from restacking during aerogel fabrication would rule out the need for electrochemical cycling to gain maximum capacity. The authors also showed that MXene aerogels' mechanical and electrical properties depend on how the 2D flakes are oriented in the aerogel's structure. The excellent electro-mechanical properties of these aerogels make them good candidates as high-quality strain sensors [30].

MXene flakes' outstanding mechanical strength, bending rigidity, and control over the 2D flake thicknesses make them a potential material for applications in tribology [31]. It has been reported that the addition of only 0.8 wt% of $\text{Ti}_3\text{C}_2\text{T}_x$ can improve the antifriction properties of base oil [32]. Huang et al. [31] coated the $\text{Ti}_3\text{C}_2\text{T}_x$ MXene on Si substrate to investigate the tribological properties of $\text{Ti}_3\text{C}_2\text{T}_x$ MXene. They measured the coefficient of friction of $\text{Ti}_3\text{C}_2\text{T}_x$ MXene to be 0.0067 ± 0.0017 , which is 3.3 times lower than that of Si substrate, achieving superlubricity with MXenes. This work opens opportunities for exploring the potential of MXenes coatings as novel solid lubricants for various applications. However, for the

industrial application of MXenes to become widespread and scale, it is necessary to mitigate the environmental hazards and costs of MXene fabrication.

Among the barriers to the large-scale production of many MXenes are the high cost of raw materials and environmental hazards. Reducing the fabrication cost by modification in the chemical usage or precursor feed of MXene would positively impact these two challenges. As for the raw materials used for the MAX phase synthesis, replacing the transition metal (for example, Ti) with transition metal oxide (for example, TiO_2) can considerably reduce the cost of MAX phase fabrication [33]. Further cost reduction is possible by using inexpensive A-layer atom sources, such as aluminum or silicon, as well as the source of carbon as the X-layer. Metal scraps are affordable sources of A-layer elements; for example, aluminum has 33 million tons of available scraps worldwide [33]. Recycled aluminum has lower costs and greenhouse gas emissions than bauxite ore [34]. In addition, graphitic carbon can be replaced with recycled carbon from tires by a simple sulfonation-pyrolysis process, which would decrease the production cost of carbon by up to 50% [35]. Furthermore, the pyrolysis process itself lowers the CO_2 emission compared to fossil fuels. Jolly et al. [33] synthesized $\text{Ti}_3\text{C}_2\text{T}_x$ MXene from the Ti_3AlC_2 MAX phase, produced from secondary aluminum, titanium dioxide, and tire-recycled carbon. This MXene exhibits electrical conductivity of $5857 \pm 680 \text{ S/cm}$. In addition, their $\text{Ti}_3\text{C}_2\text{T}_x$ MXene's electrochemical performance as an electrode in supercapacitors was comparable with those of conventionally fabricated $\text{Ti}_3\text{C}_2\text{T}_x$. MXenes from elemental powders. This method established a new path to optimize the environmental side-effects and costs related to MXene production. While MXenes, made by their conventional process, are already being explored for different commercial applications, their fabrications via cost-effective methods and sources with no environmental hazards can further speed up their industrial applications.

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