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Two-dimensional $\text{Ti}_3\text{C}_2\text{T}_x$ MXene nanosheets for CO_2 electroreduction in aqueous electrolytes†

Sarathkumar Krishnan, , Senthilkumaran Marimuthu, , Mayank K. Singh and
Dhirendra K. Rai *

Electroreduction of carbon dioxide (CO_2) to valuable synthons is a sustainable and viable technique to reduce the carbon footprint in the atmosphere. In this work, two-dimensional $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheets have been investigated for selective CO_2 adsorption and its subsequent electroreduction to valuable products in an aqueous electrolyte medium. Various physical and electrochemical characterizations have been performed to ascertain the structural, morphological, and electrocatalytic characteristics of $\text{Ti}_3\text{C}_2\text{T}_x$. $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheets show a CO_2 adsorption capacity of 0.16 mmol g^{-1} under ambient conditions and exhibit a maximum faradaic efficiency of 96% towards CO_2 electroreduction to C_1 and C_{2+} products, mainly CO (42.2%), methanol (23.6%), ethanol (20.1%), and acetone (10.1%). Besides, it also shows high catalytic stability for product formation for a tested period of 72 h. This report sheds new light on the design of titanium-based advanced catalysts for efficient CO_2 conversion in a benign aqueous medium.

1. Introduction

In this era, energy production and transportation are mainly dependent on fossil fuels which increases the emission of greenhouse gases into the atmosphere. Particularly, carbon dioxide (CO_2) is one of the primary greenhouse gases, increasing dramatically in the earth's atmosphere. Rising CO_2 levels ($>417 \text{ ppm}$, as per NOAA-Mauna Lab Observatory, 2022) adversely affect the earth's atmosphere.^{1,2} This mandates an urgent need to mitigate CO_2 emissions in the atmosphere. Besides its antagonistic environmental impact, CO_2 is also an abundant C_1 source for several organic products. Therefore, efforts involving CO_2 mitigation by transforming it into valuable products have currently drawn huge attention.^{3,4} With the aim of CO_2 mitigation efforts, among the different methods reported earlier, electrochemical conversion of CO_2 into value-added products (C_1 and C_{2+} carbon products) is considered an efficient and viable route for the sustainable ecosystem due to its ambient operating conditions.^{4,5}

In the process of CO_2 electroreduction, various C_1 products (carbon monoxide, methane, formate, methanol), C_2 products (ethanol, ethylene), and C_{2+} products (propanol, acetone, acetate) can be formed using different homo/heterogeneous

catalysts. Among them, methanol, ethanol, ethylene, and acetone are highly promising chemical synthons in industry as well as synthetic fuels to meet fuel scarcity. However, the chemically inert CO_2 molecule significantly hinders the formation of various carbon products.⁶ Additionally, the process of CO_2 electroreduction confronts two major challenges: (i) the inferior binding capability of the CO_2 reduction intermediate on electrode materials and (ii) the competitive hydrogen evolution reaction (HER) occurring during the electroreduction process.^{7,8} To address these issues, the design of an electrocatalyst is of substantial interest. Recently, various transition metal-based oxides/hydroxides⁹ and metal-free carbonaceous materials¹⁰ have been reported for CO_2 electroreduction. Among them, copper (Cu) and copper-containing electrocatalysts are well-recognized for the formation of the above-mentioned C_2 and C_{2+} products due to their effective binding ability with $^*\text{CO}$ and $^*\text{CHO}$ intermediates.^{11,12} The quest for relatively low-cost and earth-abundant highly active catalysts with enhanced charge transfer, which is highly desired to steer the enhancement in the fundamental catalytic activity towards value-added C_2 and C_{2+} products, still continues.

In the last decade, new families of two-dimensional materials, namely, metal carbides, nitrides, and carbonitrides (MXenes), discovered by Gogotsi *et al.*, have aroused much attention. The applications of MXenes are extensively explored in the field of supercapacitors,¹³ batteries,¹⁴ electrochemical sensors,¹⁵ and water splitting¹⁶ applications due to their excellent electronic conductivity, inherent surface functional groups, and tunable interlayer spacing.^{17,18} MXenes ($\text{M}_{n+1}\text{X}_n\text{T}_x$, where M is the transition element, X is carbon or nitrogen, and T_x is the surface

Sustainable Energy & Environmental Materials (SEEM) Lab, Department of Metallurgical Engineering and Materials Science (MEMS), Indian Institute of Technology Indore, Simrol, Indore 453552, India. E-mail: dkrain@iiti.ac.in;
Tel: +91 731 660 3278

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* These authors contributed equally to this work.

functional group such as $-O$, $-F$ and $-OH$) are obtained by selective etching of A elements (A represents group 13 and 14 elements) from their parent MAX phases which have a general formula of $M_{n+1}AX_n$.¹⁹

Apart from their wide range of applications, recent experimental and theoretical investigations proved the efficacy of layered MXenes for CO_2 capture and its electrocatalytic conversion.^{20–22} Reports have suggested that the interlayer spacing between MXene lamellar structures and its high electrical conductivity are conducive to CO_2 adsorption and its efficient conversion.^{23–25} It has been identified that the inherent surface functional groups ($T_x = -F$, $-O$, and $-OH$) in MXenes can change the MXene work function that effectively favors CO_2 reduction intermediate adsorption. Also, the metal centers in MXenes can increase CO_2 adsorption through non-covalent ($O=C=O_{lp} \cdots M$) or covalent (surface-bound CO_2) interactions.^{24,26,27} For instance, Wang *et al.* reported two kinds of MXenes ($Ti_3C_2T_x$ and V_2CT_x) for CO_2 capture and obtained a maximum CO_2 adsorption capacity of 5.79 mmol g^{-1} at 40 bar pressure.²⁵ In recent years, a few reports investigated MXenes for electrocatalytic conversion of CO_2 . For example, Qu *et al.* investigated that the use of N-doping and titanium vacancies in the Ti_3C_2 MXene shows 92% faradaic efficiency towards CO production.²⁸ Hondoko *et al.* reported the use of Ti_2CT_x and Mo_2CT_x for the electrocatalytic conversion of CO_2 into formic acid as a primary product with a 56% faradaic efficiency. They suggested that the inherent functional groups (T_x) present in the materials are the reason for such high performance.²² Ti_3C_2 and Mo_2C were investigated by Attanayake *et al.* for CO_2 reduction and showed a faradaic efficiency of 90% towards CO production by using ionic liquid-based electrolytes.²⁰ Also, a copper immobilized $Ti_3C_2Cl_x$ MXene derived from a quaternary MAX phase ($Ti_3(Al_{1-x}Cu_x)C_2$) was reported for electrochemical CO_2 reduction, where methanol and ethanol were the major products due to the unsaturated electronic structure of Cu.²⁸ Bao *et al.* synthesized a copper single atom decorated $Ti_3C_2T_x$ MXene for effective carbon monoxide reduction and achieved 98% of multi-carbon products. This is due to the formation of a favorable C–C coupling to generate the key intermediates ($*CO-CHO$) for multi-carbon product formation.²⁹

Of all MXene-based CO_2 electroreduction processes reported so far, most use either complex organic electrolytes or high temperature or demonstrate electroreduction of the CO intermediate instead of directly using CO_2 . There is enormous room available to explore the fundamental catalytic activity of MXenes using aqueous electrolytes for electrocatalytic CO_2 conversion. In continuation of this exciting research field, herein, we have prepared the $Ti_3C_2T_x$ MXene by an *in situ* etching method and used it as an active electrode material for electrocatalytic CO_2 conversion using simple aqueous $KHCO_3$ electrolyte. Laursen *et al.* and Hondoko *et al.* suggested that the use of Ti-based MXenes is beneficial in CO_2 electroreduction due to their hydrogen-opposing behavior, which is beneficial for promising CO_2 reduction.^{22,30} The as-prepared material has been investigated using various physical and electrochemical characterization techniques. Unlike the product

formation obtained in organic electrolyte systems, we achieved the highest faradaic efficiency of 96% towards mono- as well as multi-carbon products such as CO, methanol, ethanol, and acetone in 0.5 M $KHCO_3$ aqueous electrolyte. Moreover, the $Ti_3C_2T_x$ MXene electrocatalyst exhibits excellent long-term stability. The use of an earth-abundant electrocatalyst for the transformation of CO_2 in a benign aqueous medium into value-added products highlights the sustainable character of this methodology.

2. Experimental section

2.1. Chemicals

Potassium bicarbonate ($KHCO_3$), methanol, ethanol, acetone, dimethyl sulfoxide (DMSO), lithium fluoride (LiF), and hydrochloric acid (HCl) were purchased from SRL Chemicals, India. Ti_3AlC_2 was procured from Y-Carbon Ltd, Ukraine. All the chemicals were used without any further purification. A Nafion N-117 membrane was procured from Alfa Aaser, India. Deionized (DI) water with a resistivity of $18.2 \text{ M}\Omega \text{ cm}$ was obtained from a Millipore ultrapure water purification system.

2.2. Preparation of the MXene ($Ti_3C_2T_x$)

$Ti_3C_2T_x$ was prepared by a top-down approach based on previous literature.¹⁹ In a typical synthesis, 1.6 g of LiF was added to a solution containing 5 mL of DI water and 15 mL of 12 M HCl in a Teflon container (HF-safe container). The as-obtained mixture was stirred at 100 rpm at 35°C for 5 min to obtain a homogeneous solution. Afterward, Ti_3AlC_2 (1 g) was added slowly and stirred for 24 h at 35°C . Then, the resultant mixture was repeatedly washed in a centrifuge at 4000 rpm until the pH of the suspension reached a value more than 6. Finally, the sedimented $Ti_3C_2T_x$ was dried at 60°C in a vacuum oven for 8 h.

2.3. Modification of the glassy carbon electrode (GCE) by $Ti_3C_2T_x$ coating

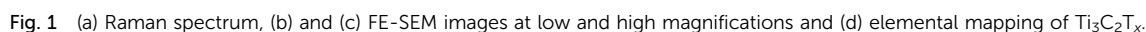
5 mg of $Ti_3C_2T_x$ and 5 wt% of Nafion solution (10 μL) were taken in 1 mL of ethanol and sonicated for 15 min. The obtained $Ti_3C_2T_x$ slurry was applied to the freshly polished glassy carbon electrode (5 mm diameter) by repeatedly coating the electrode with 10 μL drops of the suspension. The deposition of the suspension was repeated five times. Each time the coated suspension was allowed to dry before applying the next drop of suspension.

2.4. Physical characterization

Powder X-ray diffraction (PXRD) was performed using a Rigaku Ultima IV X-ray diffractometer with Cu K α radiation (1.54 Å) in the range of 5° to 60° (step size of 0.02°). Field emission-scanning electron microscopy (FE-SEM, JEOL-7610F Plus) was used to investigate the surface morphology of the as-prepared material. The elemental composition and their distribution were examined using energy-dispersive X-ray spectroscopy (EDX) and elemental mapping, respectively. The Raman spectrum was recorded using a LabRAM HR Evolution spectrometer



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3.1. CO₂ adsorption studies

From the two adsorption isotherms, it is evident that decreased adsorption capacity is observed at elevated temperature, which implies that physical adsorption is the main force for the interaction between CO_2 and $\text{Ti}_3\text{C}_2\text{T}_x$ at low CO_2 pressure.³⁸ Also, a linearly increasing adsorption isotherm is observed with increasing relative CO_2 pressure until the measured range, implying that, owing to its mesoporous nature, the surface of $\text{Ti}_3\text{C}_2\text{T}_x$ is still not saturated with CO_2 , and even more

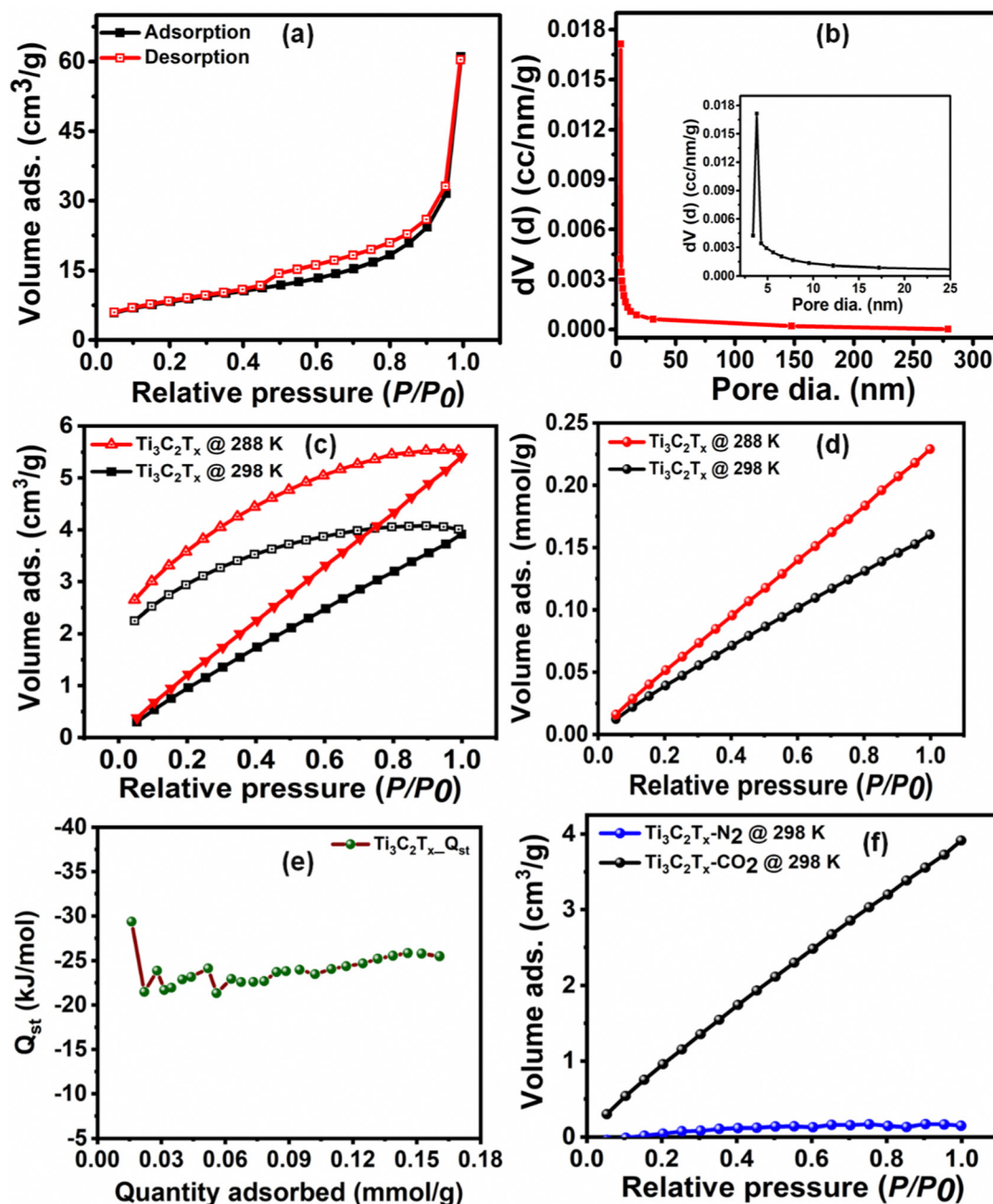
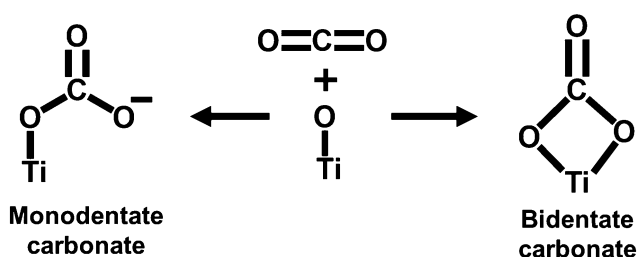


Fig. 2 (a) and (b) BET N_2 adsorption-desorption isotherm and pore size distribution, (c) and (d) CO_2 adsorption-desorption isotherm in $\text{cm}^3 \text{g}^{-1}$ and CO_2 adsorption isotherm in mmol g^{-1} at 288 and 298 K, (e) isosteric heat of adsorption and (f) CO_2 selectivity studies over N_2 at 298 K.



Scheme 1 Possible CO_2 interaction on the MXene surface.

CO_2 adsorption can be achieved upon increasing the relative pressure.³¹

The adsorption affinity of CO_2 on the $\text{Ti}_3\text{C}_2\text{T}_x$ adsorbent has been calculated (Fig. 2(e)) from the CO_2 adsorption isotherms at 288 and 298 K using the enthalpy of adsorption ($-Q_{st}$) obtained from the Clausius-Clapeyron equation (eqn (1)). The average enthalpy of adsorption is found to be $-23.6 \text{ kJ mol}^{-1}$, which lies in the physisorption range (-10 to -50 kJ mol^{-1}).³⁹ An almost constant value of the enthalpy of adsorption at all surface coverage suggests the homogeneous surface of the $\text{Ti}_3\text{C}_2\text{T}_x$ adsorbent involving a similar type of interaction all

over its surface.^{38,40,41} To understand the practicability of the adsorbent, the CO₂/N₂ selectivity on Ti₃C₂T_x has been investigated using the Henry law equation (Fig. 2(f)). The calculations indicate that the Ti₃C₂T_x adsorbent exhibits almost 22 times selectivity for CO₂ over N₂ at room temperature, suggesting its remarkable potential as a catalyst in CO₂ conversion reactions. Although CO₂ and N₂ have almost similar kinetic diameters (CO₂: 330 pm; N₂: 364 pm), the higher electric quadrupole moment of CO₂ facilitates its interactions with the highly polar surface functional groups on the Ti₃C₂T_x surface, which renders enhanced selectivity.

3.2. Electrochemical analysis

To evaluate the electrocatalytic activity of the Ti₃C₂T_x modified glassy carbon electrode (GCE) (Ti₃C₂T_x-GCE), linear sweep voltammetry (LSV) has been carried out using a standard three-electrode system in 0.5 M KHCO₃ aqueous electrolyte in the potential range between −1.6 and 0 V (*vs.* Ag/AgCl) at a scan rate of 10 mV s^{−1} (Fig. 3(a)). In the N₂ saturated electrolyte, the obtained higher current density is attributed to the H₂ evolution reaction. In the case of CO₂-saturated electrolyte, the current density is higher than that in the case of N₂-saturated electrolyte, which is due to the simultaneous reduction of both H⁺ and CO₂. Additionally, an anodically shifted electrochemical potential has been observed for the CO₂-saturated electrolyte (−1.37 V *vs.* Ag/AgCl) compared to the N₂-saturated electrolyte (−1.51 V *vs.* Ag/AgCl) corresponding to a constant current density of −5 mA cm^{−2}. These observations reveal that the electrode material active sites efficiently facilitate the electron transfer across the electrode interface and eventually help to enhance the CO₂ reduction process. To assess if the coated Ti₃C₂T_x is the sole cause for this excellent electrocatalytic performance, we also investigated the catalytic activity of the bare GCE under similar conditions (Fig. 3(a)). The observed rise in the current densities for both N₂ and CO₂ saturated electrolytes is insignificant, which confirms that the overall product formation is exclusively due to the catalytic activity of Ti₃C₂T_x.

The effect of electrolyte concentration is also an important factor for effective product formation. Therefore, we also

investigated and compared the performance of the Ti₃C₂T_x-GCE at two different concentrations of electrolyte, namely 0.1 M and 0.5 M KHCO₃ (Fig. S5, ESI†). The results suggest that with increasing concentration of KHCO₃ electrolyte from 0.1 M to 0.5 M the current density also increases. In general, the local environment around the active materials during electrolysis is known to influence the CO₂ reduction reaction (CO₂RR). Nakona *et al.* also observed high current density at higher KHCO₃ concentrations, however with decreased faradaic efficiency for CO₂RR products. They suggested that, after a certain concentration of KHCO₃, the K⁺ ions generated from the electrolyte also determine the product selectivity. At higher electrolyte concentrations, the K⁺ ions start accumulating around the negatively charged electrode materials, which hinders the CO₂RR by preventing CO₂ from accessing the electrode, resulting in the dominance of the HER.^{42,43} Moreover, being a basic salt (formed from a strong base and a weak acid), the higher concentration of KHCO₃ increases the pH of the solution. It has been suggested that > 8 pH of electrolyte causes deprotonation of the CO₂ carrier HCO₃[−] into CO₃^{2−} and, thus, decreases the CO₂ carrier concentration.^{43,44} This also favors the HER and decreases the faradaic efficiency of CO₂RR products, despite the increase in the overall current density. Considering these facts, we chose a 0.5 M KHCO₃ concentration (causing pH ~ 7.47, just below 8; Fig. S6, ESI†) for further studies.

To examine the types of products formed during the CO₂ electroreduction, constant potential electrolysis, *i.e.*, chronoamperometry (CA), has been carried out in an H-type cell using a Nafion membrane as the proton exchanger and separator (Fig. S1, ESI†). Prior to the extensive investigation, we also investigated the effect of electrolyte concentration using the CA experiment at −1.1 V (*vs.* Ag/AgCl) at two different electrolyte concentrations (0.1 M and 0.5 M KHCO₃). The evaluation of CO₂RR products formed at two different electrolyte concentrations was done by ¹H NMR (Fig. S7, ESI†). Fig. S7 (ESI†) clearly suggests that the product formation in 0.1 M KHCO₃ is negligible. In contrast, in the case of 0.5 M KHCO₃ electrolyte, well-distinguished peaks for ethanol (δ (ppm): 3.55 (q), 1.08 (t)),

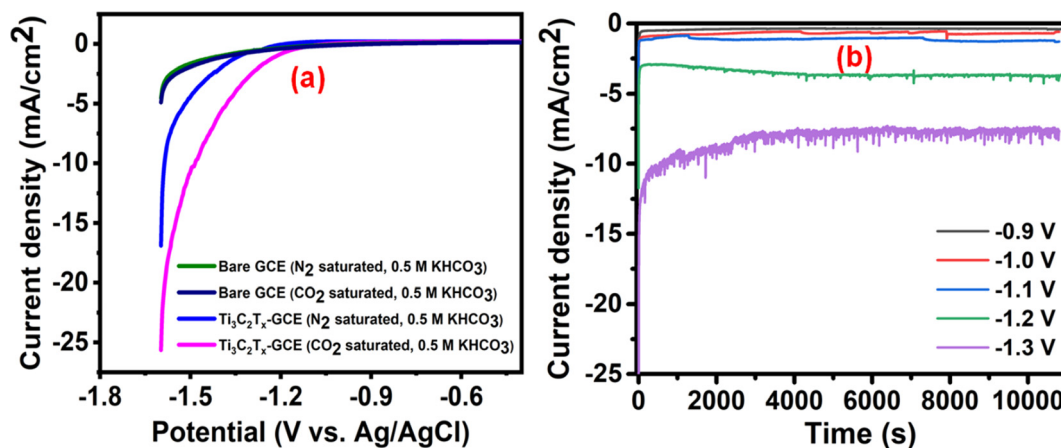


Fig. 3 (a) LSV curves in the presence of 0.5 M KHCO₃ at 10 mV s^{−1} and (b) CA profiles of the Ti₃C₂T_x-modified GCE at different potentials (*vs.* Ag/AgCl).

methanol (δ (ppm): 3.26 (s)), and acetone (δ (ppm): 2.13 (s)) are observable.⁴⁵ Therefore, choosing a 0.5 M KHCO_3 electrolyte concentration not only enhances the current density, as observed in the LSV experiment, but also facilitates the CO_2 electroreduction.

To assess the effect of potential on the CO_2RR , the CA experiment was performed at different potentials (-0.9 to -1.3 V) (Fig. 3(b)). The potential choice was based on the onset potential (-1.1 V) obtained from the CO_2 -saturated $\text{Ti}_3\text{C}_2\text{T}_x$ -GCE LSV curve. The CA experiments were carried out at -0.9 V, -1.0 V, -1.1 V, -1.2 V, and -1.3 V (vs. Ag/AgCl) for 3 h, and the corresponding ^1H NMR spectra are shown in Fig. S8 (ESI[†]). The time vs. current density profiles with different potentials are shown in Fig. 3(b), which confirms the stable current densities obtained at all selected onset potentials. In the case of the CA profile at -1.3 V, some disturbance was observed, which was due to vigorous H_2 evolution causing bubble formation on the electrode.

Faradaic efficiency (FE) is an important measure of the catalytic activity of an electrocatalyst. The variation of the percentage FEs of the total liquid product along with the current density at different potentials is shown in Fig. 4(a). The concentrations of various products formed during the electrolysis were quantified from the standard plot obtained by plotting their relative ^1H NMR peak with respect to known

concentrations (Fig. S9, ESI[†]). From Fig. 4(a), it can be observed that, upon increasing the potential, the %FE of liquid products as well as current density gradually increases until -1.1 V (vs. Ag/AgCl) (maximum FE obtained: 53.8%), whereas a further increase in the potential causes a drastic reduction in %FE along with a sudden increase in the current density. This observation corroborates that, beyond -1.1 V, the HER is strongly favored over the CO_2RR . The breakup of faradaic efficiency for individual liquid products at different voltages is shown in Fig. 4(b). It was observed that, for the highest obtained FE of 53.8%, at -1.1 V (vs. Ag/AgCl), the highest contribution came from the formation of methanol, which is comparable to most of the reported catalysts (Table S1, ESI[†]). In addition to liquid products, gaseous products such as CO , CH_4 , C_2H_6 , and C_2H_4 are also known to be formed in the CO_2RR . Therefore, to calculate the overall %FE, we also quantified the gaseous products at -1.1 V vs. Ag/AgCl using gas chromatography (Fig. S10, ESI[†]), which suggested the formation of only CO (42.2%). Based on the NMR and gas chromatography studies, the overall FE of the $\text{Ti}_3\text{C}_2\text{T}_x$ electrocatalyst, at -1.1 V vs. Ag/AgCl, was calculated to be 96%.

In general, the $\text{Ti}_3\text{C}_2\text{T}_x$ electrode was observed to favor the formation of all C_{1-3} products, suggesting a strong interaction of its surface with the CO_2RR intermediates.^{46–48} This is mainly due to the presence of $-\text{OH}$, $-\text{O}$ and $-\text{F}$ surface functional

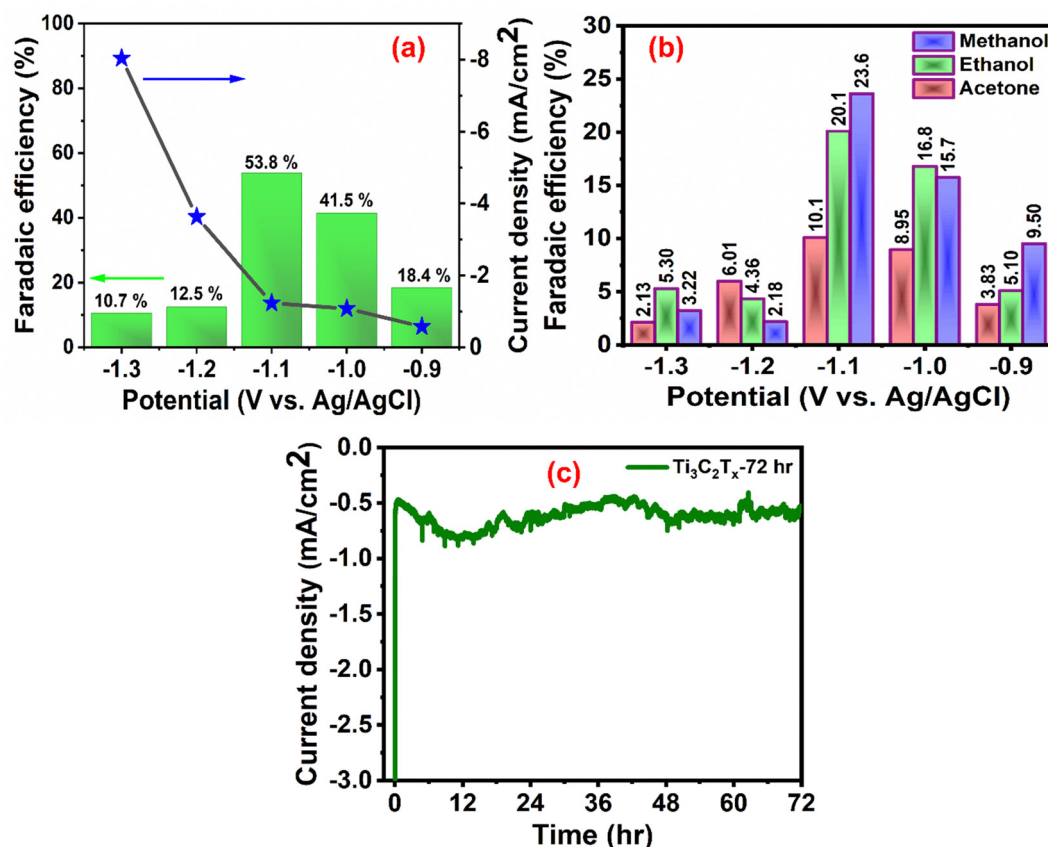


Fig. 4 (a) The total liquid product %FE and the corresponding current density, (b) individual liquid product %FE, and (c) long-term stability test at -1.1 V vs. Ag/AgCl for 72 h.



groups, which activate the electrode surface for CO₂ reduction.^{22,46} It was further supplemented by calculating the electrochemical active surface area (ECSA) of Ti₃C₂T_x in terms of double layer capacitance (*C_{dl}*) from its cyclic voltammetry curves at different scan rates (10–50 mV s^{−1}).⁴⁹ Fig. S11 (ESI†) shows the ECSA plot of Ti₃C₂T_x, which has the maximum *C_{dl}* value of 3.19 mF cm^{−2}. This value is comparable to most of the recently reported electrocatalysts, confirming the presence of a higher number of electrocatalytic active sites for the excellent CO₂RR performance.^{22,49,50}

The stability of the electrocatalyst for a long-time operation is one of the important concerns for finding the best catalyst for CO₂ electroreduction. We investigated the long-term stability of the Ti₃C₂T_x-GCE for 72 h at a constant potential of −1.1 V (Fig. 4(c)). After 72 h of electroreduction, no discernible change in the current density was observed, which signifies the excellent stability of the Ti₃C₂T_x-GCE. The slight fluctuation in the chronoamperometry curve is due to bubble formation and explosion during continuous electrolysis. The same phenomena have also been observed in a previously reported investigation.²⁹ To further investigate any alteration in the structural features of the catalyst after 72 h of electrolysis, FE-SEM, EDX, elemental mapping, and PXRD studies were also carried out (Fig. S12–S14, ESI†). FE-SEM images reveal the preservation of its two-dimensional morphology with slight agglomeration. EDX and elemental mapping confirm the uniform presence of all the elements pertaining to Ti₃C₂T_x along with electrolyte ions, confirming the overall compositional integrity of the sample after long-time electrolysis. Moreover, the PXRD pattern of the used catalysts shows all the related peaks of Ti₃C₂T_x with reduction and broadening of some peaks, indicating partial oxidation of some Ti sites. To investigate the changes in charge transfer dynamics, electrochemical impedance spectroscopy (EIS) of the Ti₃C₂T_x catalyst before and after the extended electrolysis was carried out (Fig. S15, ESI†). EIS measurements were performed in the frequency range of 0.1 Hz to 100 kHz with an alternating current amplitude of 10 mV. It was observed that, after prolonged electrolysis, the bulk resistance of the catalyst shows slight enhancement (*R_b*: 17.2 Ω (for fresh) and 24.3 Ω (for used)), as can be observed at high frequencies. However, the appearance of a semicircle was observed for the used catalyst, which corresponds to charge transfer resistance (*R_{ct}*) due to the presence of CO₂-converted or adsorbed products (CO₂^{*}, HCO₃[−] or any liquid product) on the electrode surface which increases the impedance.^{51,52}

4. Conclusion

The Ti₃C₂T_x MXene was synthesized by a less hazardous *in situ* etching process, which was used as a selective CO₂ adsorbent and an electrocatalyst for CO₂ electroreduction. The presence of inherent functional groups, high conductivity, and metal sites in the MXene facilitates enhanced gas adsorption and endows the MXene with efficient electrocatalytic properties. Detailed volumetric CO₂ gas adsorption studies reveal that the MXene material exhibits a maximum adsorption capacity of 0.16 mmol g^{−1}

at 298 K with good CO₂/N₂ selectivity. In addition, the thermodynamic calculation suggests that the physical adsorption behavior is the main mechanism for CO₂ adsorption on the MXene material at low pressure. The as-prepared MXene exhibits excellent electrocatalytic properties towards CO₂ electroreduction in an aqueous electrolyte with an overall faradaic efficiency of 96% towards various C₁ (CO, CH₃OH) and C₂₊ (CH₃CH₂OH, CH₃COCH₃) products. The electrocatalyst exhibits a remarkably stable electroreduction performance for a duration of 72 hours as investigated by chronoamperometry using a laboratory-made H-type cell. These striking catalytic features of the MXene demonstrate its use as an attractive and abundantly available electrocatalyst for the conversion of CO₂ into multi-carbon products in a benign medium.

Conflicts of interest

There are no conflicts to declare.

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