

Mechanochemically Synthesized Transition Metal Carbide Fillers for Coating Metallic Bipolar Plates in Proton Exchange Membrane Fuel Cells

Jil Meyers, Özgül Agbaba, Henning Buchholz, and Ferdi Schüth*

Metal bipolar plates, especially stainless steel 316 L bipolar plates, are of commercial interest due to their mechanical stability and low manufacturing cost, but they require a protective coating on their surface to improve their electrical conductivity as well as their corrosion performance. Herein, transition metal carbides (TiC, Cr₃C₂, NbC, VC, Mo₂C, and TaC) are synthesized by a mechanochemical synthesis route, incorporated as electrically conductive fillers into a polyurethane-based binder matrix, and are applied to stainless steel plates by the wet paint technique. For comparison, a commercially available graphite, the standard filler for coated metal bipolar plates, and a commercially available Cr₃C₂ are used as fillers in coatings. All protective films show good adhesion to the metal substrate and a homogeneous filler-binder distribution. Contact resistance measurements indicate that, among all the coatings, those with mechanochemically synthesized Cr₃C₂ and Mo₂C have the best conductivity performance, well below 10 mOhm cm². From potentiodynamic and potentiostatic polarization tests, the Cr₃C₂-containing coating has superior corrosion behavior, while the Mo₂C coating is unable to protect the metal from corrosion. Based on these results, Cr₃C₂-coated stainless steel is an attractive candidate for bipolar plates in proton exchange membrane fuel cells.

1. Introduction

The global increase in energy consumption and the environmental pollution caused by the use of fossil fuels are driving the demand for clean and sustainable energy applications. In recent years, fuel cell technology has attracted a lot of attention as a promising solution. Among various types of fuel cells, the proton exchange membrane fuel cell (PEMFC), which converts hydrogen and oxygen into electricity with only water as a byproduct, has been identified as an alternative, environmentally friendly

power source for the transportation sector due to its high power density, efficiency, near-zero emissions, and low operating temperature.^[1–4]

Bipolar plates are one of the key components of a PEMFC, accounting for a high fraction of the total weight and manufacturing cost. As key structural and functional components, bipolar plates account for a significant portion of both the weight and the manufacturing cost of PEMFC stacks. They are designed to perform a wide range of functions, including uniform distribution of the reactant gases to the electrodes through the flow channels, separating and electrically connecting the individual cells in a stack, and removing heat and water from the active areas. As a result, bipolar plate materials must be highly electrically and thermally conductive, chemically and mechanically stable, have low gas permeability, and be lightweight and cost effective to manufacture.^[5–7] Metallic bipolar plates have been identified as a suitable solution.

In particular, stainless steel bipolar plates have attracted commercial interest due to their high mechanical strength, good chemical stability, and low manufacturing cost. However, a potential disadvantage of stainless steel is its chemical instability when in direct contact with the acidic PEMFC environment. The corroded bipolar plates can lead to dissolution of metal ions, resulting in potential degradation of membrane performance. In addition, a corrosion layer formed on the surface of the bipolar plates can increase the electrical resistance and thus reduce the output of the cell. To improve the efficiency of the PEMFC, stainless steel plates with a protective coating on the surface have been the subject of extensive research.^[8–11]

The most common coatings for metal bipolar plates are made of noble metals such as gold and platinum, while transition metal carbides and nitrides, carbon materials, and conductive polymers provide more cost-effective alternatives.^[12–14] In the group of metal carbides, titanium carbide, and chromium carbide are among the most widely used and studied coating materials that improve the overall performance of PEMFCs by increasing the chemical stability while reducing the interfacial resistance of the metal bipolar plates.^[15–17] Conventional coating methods are mainly physical vapor deposition, including methods such as multi-arc ion plating and magnetron sputtering, which form thin, hard, adherent, and dense coatings on the metal surfaces.

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However, these methods are time consuming, costly, and have high temperature and energy requirements, making wet paint coating processes using polymer composite-based paints an attractive solution.^[18–20]

Polymer-based coatings provide excellent corrosion protection on metal bipolar plates, but their electrically insulating nature is prohibitive for practical application. Therefore, polymer composites are typically used, in which conductive fillers (e.g., metallic, carbonaceous, and ceramic particles) are dispersed in the insulating polymer matrix to improve the electrical conductivity of the protective film.^[21,22] Ceramic fillers, in which metal atoms are bonded to nonmetal atoms, are known for their excellent electrical and thermal conductivity, high chemical and mechanical stability, good corrosion resistance, and high hardness.^[23] Among various types of ceramic materials, early transition metal carbides, a class of interstitial alloys, are formed by the interstitial dissolution of carbon atoms into the lattices of the parent metal atoms. Known for their exceptional robustness, remarkable durability and wear resistance, they are used in many technological applications such as cutting tools, drills and numerous other wear-resistant components essential in the machining, mining, and construction industries. They are as well used as wear and corrosion resistant coatings on stainless steel bipolar plates.^[15–17,24–27] However, a disadvantage of metal carbides is that they are difficult to produce. Conventional methods require high temperatures in the range of 1500–2300 °C, are energy intensive, and are associated with undesirable by-products such as CO and CO₂.^[28] Another difficulty is that the gas atmosphere for the synthesis is reductive, containing hydrogen and sometimes methane, which carries a risk of explosion. The starting materials can be pure metals or oxide forms,^[27] and these high-temperature production processes include the conventional direct elemental combination of metal powders and carbon,^[29] carbothermal reduction of metal oxides,^[30,31] and direct carburization of metal powders with a gaseous source of a suitable hydrocarbon.^[32]

A novel synthesis approach that utilizes the effects of mechanical forces to induce chemical reactions offers a simple and effective method for the synthesis of metal carbides. In this alternative synthesis route, elemental metal powders and a carbon source, such as graphite, carbon nanotubes, or activated carbon, are mechanochemically reacted at room temperature, typically by ball milling.^[33–35] The use of milder reaction conditions, eliminating high reaction temperatures, hazardous gas mixtures, and solvents, makes it an alternative to conventional industrial processes.^[36] In addition, this production process can be used commercially on an industrial scale (possible vessel sizes of several hundred liters), making it suitable for a wide range of applications.^[37]

The main objective of the study is to understand the electrical conductivity and corrosion properties of composite coatings deposited on stainless steel bipolar plates when transition metal carbides are incorporated as electrically conductive filler materials, in order to create coatings with suitable properties for technical applications. We have used a mechanochemical method for the synthesis of conductive fillers that were then embedded in a polymeric binder matrix and applied to stainless steel plates using a wet paint technique. For comparison, a commercially available graphite, the standard filler for coated metal bipolar

plates, and a commercially available Cr₃C₂ were used as fillers in the coatings. Cross-cut tests and scanning electron microscope (SEM) elemental mapping analysis were performed to analyze the adhesion properties of the coating to the substrate and the distribution of the filler in the binder matrix. Interfacial contact resistance measurements as well as potentiodynamic and potentiostatic polarization methods were used to evaluate both the surface conductivity and corrosion properties of the coated metal plates.

2. Results and Discussion

2.1. Properties of Mechanochemically Synthesized Transition Metal Carbides

The initial focus of the work was the development of high-energy ball milling mechanochemical synthesis methods to produce pure single-phase carbide fillers, such as titanium carbide (TiC), chromium carbide (Cr₃C₂), niobium carbide (NbC), vanadium carbide (VC), molybdenum carbide (Mo₂C), and tantalum carbide (TaC). As starting materials, elemental metal powders and graphite were used in a Ti:C, Nb:C, V:C, and Ta:C molar ratio of 1:1, Mo:C molar ratio of 2:1 and Cr:C molar ratio of 3:2. The starting materials were milled for 1.75–11 h in a zirconia jar with zirconia balls in a Retsch high-energy ball mill *E*_{max}. The vibration frequency was 1000 rpm and the ball-to-powder weight ratio ranged from 19:1 to 11:1. A detailed description of the experimental procedure is provided in the Experimental Section of the Supporting Information, and the exact milling times and powder loadings are listed in Table S1, Supporting Information. To prevent the formation of oxides, the milling jars were filled with the appropriate amount of the metal-graphite powder mixture in a glovebox. The product from milling chromium and graphite was a mixture of different phases. Thus, this mixture was annealed for 2 h at 800 °C under argon atmosphere to form a single-phase Cr₃C₂. The structural evolution, level of contamination from the milling media, particle size, morphology and specific surface area of the synthesized carbides were investigated by powder X-ray diffraction (XRD), energy-dispersive X-ray (EDX) bulk analysis, SEM, and nitrogen physisorption analysis.

The formation processes of various metal-graphite powder mixtures milled under argon atmosphere for different periods of time are shown in Figure 1a–g. After milling the titanium-graphite powder mixture for 20 min, weak TiC reflections were observed in addition to the titanium reflections. As the milling time increased, the titanium reflections became broader, indicating decreasing particle size and/or formation of more defective domains, and the TiC reflections increased in intensity. After 105 min of milling, TiC formation was complete (Figure 1a). Due to the short milling time, the contamination by the zirconia milling media (jar and balls) could be kept below 0.5 wt% Zr. Diffraction patterns for the conversion of the chromium powder to Cr₃C₂ are given in Figure 1b. After 2 h of milling, only reflections of the chromium powder were observed. Further milling reduces the reflection intensities and increases the width of the reflections. A milling time of 6 h results in a predominantly amorphous structure with very broad background features,

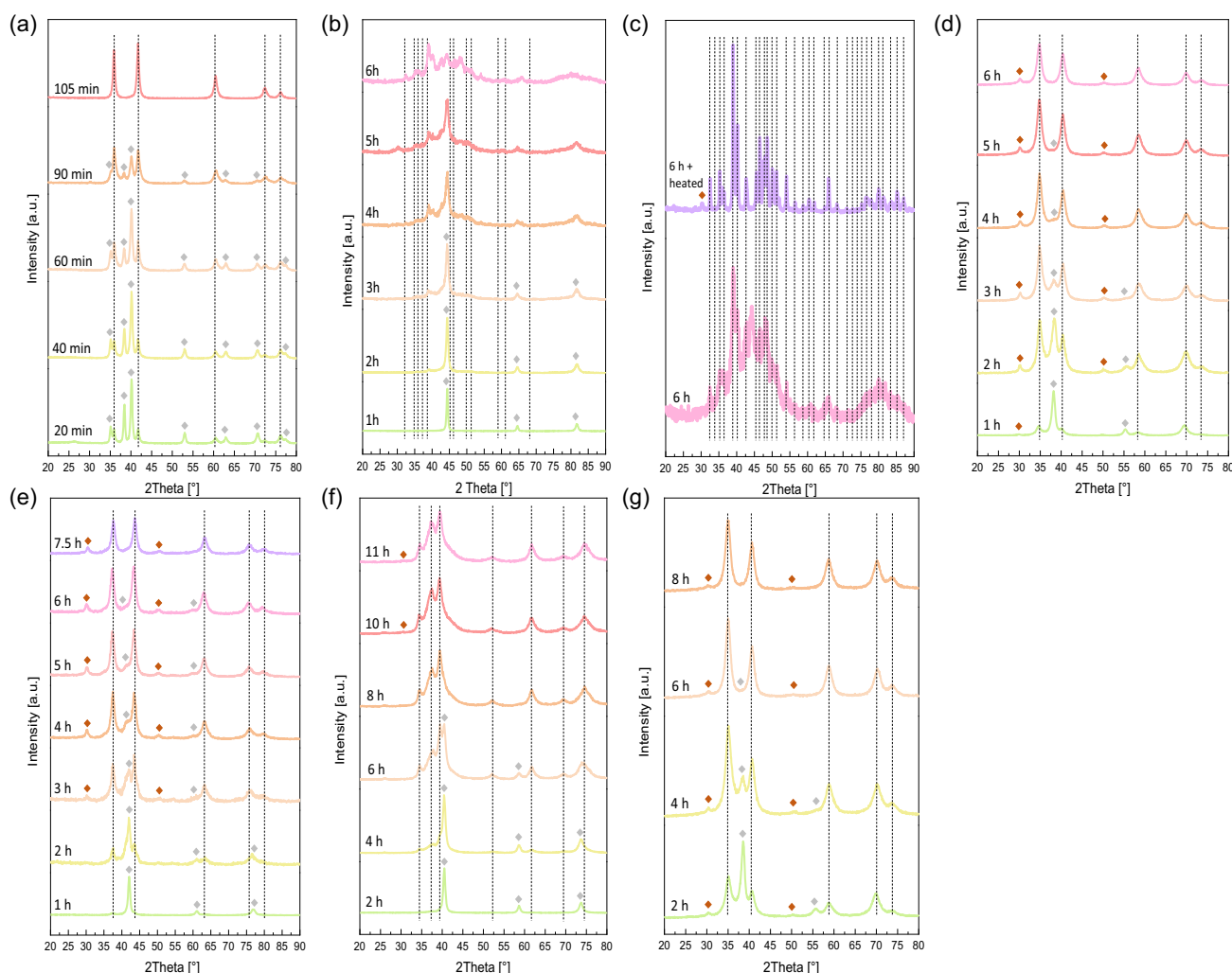


Figure 1. a–g) X-ray diffraction patterns of the reaction products of the different transition metal/graphite mixtures after different milling times. The dashed vertical lines indicate the position of the reflections of each carbide. The gray and dark orange diamonds show the reflections of the pure metal and of zirconia, respectively. (a) TiC; (b) mixture of three different chromium carbides: Cr_3C_2 , Cr_7C_3 , and Cr_{23}C_6 ; (c) mixture of chromium carbides milled for 6 h and annealed for 2 h at 800 °C under argon atmosphere, resulting in a single-phase Cr_3C_2 ; (d) NbC; (e) VC; (f) Mo_2C ; and (g) TaC.

indicating a high proportion of amorphous material; crystalline phases can be indexed to three different chromium carbides: Cr_3C_2 , Cr_7C_3 , and Cr_{23}C_6 . After milling the chromium-graphite powder mixture for 6 h and subsequently annealing it for 2 h at 800 °C under argon atmosphere, a complete set of reflections corresponding to a pure, single-phase, crystalline Cr_3C_2 was obtained (Figure 1c). It appears that the predominantly amorphous phase, also showing reflections of three types of chromium carbides, is the precursor of the Cr_3C_2 phase.^[35,38] Another feature in the XRD pattern is a small reflection corresponding to zirconia, which is an impurity from the milling media caused by the relatively long milling time. According to the SEM-EDX bulk elemental analysis, the contamination level of Cr_3C_2 was kept below 1.6 wt% Zr.

The XRD patterns of the milled niobium-graphite, vanadium-graphite, molybdenum-graphite, and tantalum-graphite powder mixtures with different milling times under argon atmosphere are shown in Figure 1d–g, respectively. It was found that

after 1 h of milling, only reflections corresponding to the elemental metals were present, except for the niobium-graphite powder mixture, where small NbC reflections were detected. After 2 h of milling, the tantalum-graphite powder mixture had small TaC reflections in addition to the Ta reflections. As the milling time increased, the reflections of the individual metals decreased and the reflections of the carbides increased. After a final milling time of 6 h for NbC, 7.5 h for VC, 11 h for Mo_2C , and 8 h for TaC, the corresponding metal reflections had disappeared completely, and only the carbide reflections remained. The results of the elemental analysis showed higher contamination levels of the carbides compared to TiC and Cr_3C_2 , in the range of 11–14 wt% Zr, while Mo_2C had a contamination level of 2 wt% Zr. The comparatively longer milling time, the hardness of the metal used and the low content of graphite in the milling jar, which acts as a lubricant, could be responsible for the higher Zr content in the carbides (see Table S2, Supporting Information).

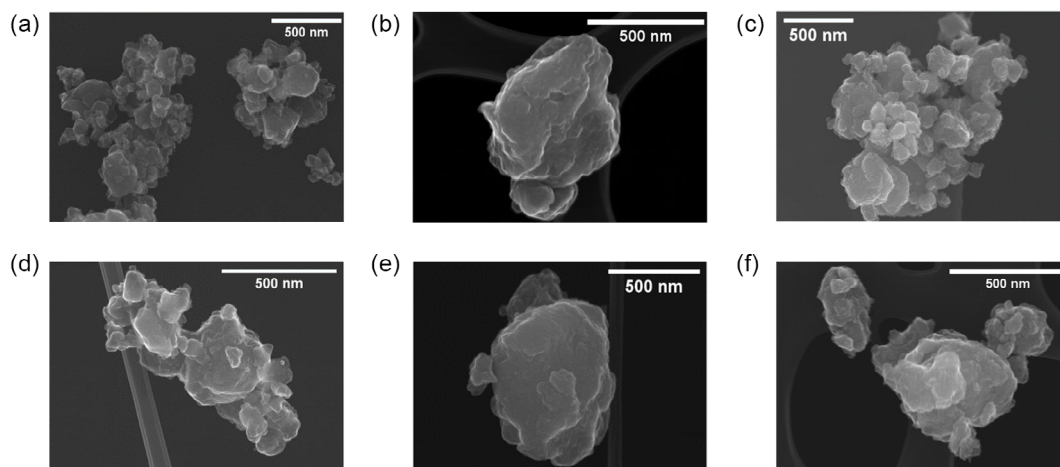


Figure 2. a–f) High resolution SEM images of the synthesized transition metal carbides. (a) TiC; (b) Cr_3C_2 ; (c) NbC; (d) VC; (e) Mo_2C ; and (f) TaC.

The particle size distribution and morphology are given in **Figure 2a–f**. All carbides were found to have an agglomerated structure with an inhomogeneous size distribution and irregular particle shape. The agglomerated particles consist of primary particles, ranging in size from a few nanometers, less than 100 nanometers, to several hundred nanometers. This broad size distribution and agglomeration of the particles can be explained by the mechanism of high-energy milling. In the early stages, the impact force deforms the particles, causing them to work harden and fracture. New surfaces are created and the particles are welded together, resulting in an increase in particle size. A wide range of particle sizes is developed. As the deformation continues, the particles work harden and begin to fracture by a fatigue failure mechanism and/or by fragmentation of loosely structured particles. The fragments produced can lead to a reduction in particle size. At this point, particle fracture predominates over cold welding. The continued impact of the milling balls refines the particle structure.^[39] The agglomeration of the carbide particles, which is also reflected in the low specific surface area of $2\text{--}14\text{ m}^2\text{ g}^{-1}$, is enhanced by the high impact energy of the high-energy ball milling process. The detailed primary average particle size (d_{average}) and the specific particle surface area (S_{BET}) of the carbides are shown in **Table 1**. Among the mechanochemically synthesized metal carbides, TiC has the smallest primary particle

size and the highest specific surface area. NbC and VC follow with similar average particle sizes and comparable surface areas. In contrast, Cr_3C_2 , Mo_2C , and TaC have the largest particle sizes and the lowest surface areas, respectively. These results confirm the general principle that the specific surface area of a material is inversely proportional to its particle size.

2.2. Preparation of the Coatings and Coating Properties

Protective coatings were formulated using a polyurethane-based binder matrix into which the mechanochemically synthesized metal carbides were incorporated as electrically conductive fillers. A commercially available graphite, referred to as the standard filler, and a commercially available Cr_3C_2 were used for performance comparison. All the formulations were prepared at the critical pigment volume concentration (CPVC), the volume ratio of filler to binder in the cured film at which the filler is completely covered by the binder. This characteristic value was determined using the oil adsorption number by placing 1 g of filler material on a glass plate and gradually adding refined linseed oil until a homogeneous paste was formed. The filler-to-binder ratio was calculated using the measured oil adsorption number and the densities of the analyzed filler and linseed oil (see Equation (S2) and (S3), as well as Table S3 and S4 in the Supporting Information for formulation details). Prior to coating, the stainless steel 316 L plates were acid-pretreated in a 10% solution of the commercially available Polinox UB A (composed of sulfuric acid, phosphoric acid, and etidronic acid) at 60°C . The plates were immersed in the heated solution for 10 min, rinsed with distilled water, and air-dried. This pickling process removes surface oxides and metal impurities (e.g., oils, greases, or rust), thereby improving the adhesion of the coating to the substrate. As reported in the literature, pretreating stainless steel substrates is essential to achieve the desired surface roughness, hydrophobicity, and oxide layer removal. These properties are necessary for strong and durable adhesion of coatings to metallic plates.^[40,41] Stainless steel plates were coated using a doctor blade in a wet paint technique process and cured in an oven at 150°C for 15 min. Coating thickness was measured using

Table 1. Overview of number-weighted average particle size, obtained by counting several hundred particles from different SEM images, and specific surface area of synthesized carbides.

Transition metal carbide	d_{average} [nm]	S_{BET} [$\text{m}^2\text{ g}^{-1}$]
TiC	85	14
Cr_3C_2	550	3
NbC	92	7
VC	100	7
Mo_2C	225	2
TaC	202	2

a metal substrate contact thickness gauge. The coating formulations, application process, and characterization methods are described in detail in the Experimental Section of the Supporting Information. All of the coatings showed dense films, with no visible defects on the metal surfaces and ranged between 15–20 μm in thickness (Table 2).

Adhesion of the coatings to the metal substrate was characterized by performing a cross-cut test with a multiblade knife, cutting a grid in the coated surface, applying an adhesive tape, and removing the tape within 5 min.^[42] The test is visually evaluated using characteristic values ranging from 0, no peeling of the coating, to 5, almost complete delamination of the coated surface (Table S5, Supporting Information). All the scratch tests performed showed high adhesion properties of the coated layer to the stainless steel plate, ranging from 0 to 1 (Table 2, see Figure S3a–k, Supporting Information for images of the cut grids of all the coatings). As an example, Figure 3 shows a stainless steel plate with a coating containing Cr_3C_2 as a filler. Since no delamination of the coated surface was observed at the cut lines, this coating was scored as 0.

Table 2. Thickness, cross-cut test characteristics, and in-plane conductivity of coated stainless steel plates.

Coating formulation	Coating Thickness [μm]	Cross-cut test	In-plane conductivity [S m^{-1}]
TiC formulated at CPVC	17.0 ± 1.6	1	$4.3 \pm 1.04 \cdot 10^6$
Cr_3C_2 formulated at CPVC	18.1 ± 1	0	$6.7 \pm 0.6 \cdot 10^6$
Cr_3C_2 exceeded CPVC by 10 wt% filler	17.9 ± 2.1	1	$8.7 \pm 0.9 \cdot 10^6$
Cr_3C_2 exceeded CPVC by 20 wt% filler	18.0 ± 1.5	0	$10.1 \pm 1.04 \cdot 10^6$
NbC formulated at CPVC	17.5 ± 1.9	0	$2.7 \pm 0.5 \cdot 10^6$
VC formulated at CPVC	15.4 ± 1.8	1	$5.0 \pm 1.7 \cdot 10^6$
Mo_2C formulated at CPVC	16.4 ± 1.3	1	$7.7 \pm 1.0 \cdot 10^6$
Mo_2C exceeded CPVC by 10 wt% filler	15.7 ± 1.4	1	$9.0 \pm 6.1 \cdot 10^6$
Mo_2C exceeded CPVC by 20 wt% filler	15.4 ± 1.3	0	$10.0 \pm 1.1 \cdot 10^6$
TaC formulated at CPVC	17.1 ± 0.7	0	$5.4 \pm 2.8 \cdot 10^6$
Commercial graphite formulated at CPVC	15.2 ± 0.5	0	74.4 ± 0.01
Commercial Cr_3C_2 formulated at CPVC	16.8 ± 0.9	1	$2.3 \pm 5.5 \cdot 10^4$

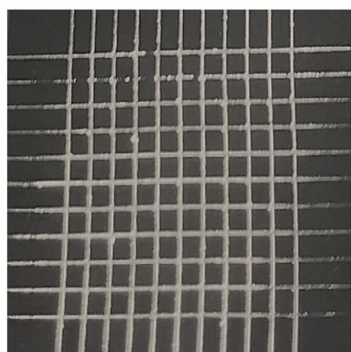


Figure 3. Cross-cut test of the coating using Cr_3C_2 as filler and formulated at CPVC.

The filler distribution in the polymer binder matrix was analyzed using SEM-EDX elemental mapping. All coatings showed a homogeneous filler loading in the polyurethane-based binder (Figure S4a–j, Supporting Information). As an example, the Cr_3C_2 coating is shown in Figure 4. Fe, Ni, and Mo, the main elements of stainless steel 316L with 68 wt%, 11 wt%, and 2 wt%, respectively,^[43] represent the metal sheet, while Cr and C represent the coating layer. The element Cr, which is found in both the coating as well as in the stainless steel, is detected in both the metal plate and the protective film, as expected. Small pieces of coating, seen at the edge of the metal plates, result from cutting the coated steel plate. The apparent presence of Mo in the coating layer does not originate from the coating material itself; rather, it is considered an artifact. Depending on the placement of the sample within the measurement device, the Mo may also be a reflection of the stainless steel plate.

The in-plane electrical conductivity of the coated metal plates showed a wide variation (Table 2). Coatings formulated with a commercial graphite or a commercial Cr_3C_2 as fillers had the lowest conductivities, while those prepared with the mechanochemically synthesized carbide fillers had much higher conductivities ranging from 4.0 to $8.0 \cdot 10^6 \text{ S m}^{-1}$. The protective films with Cr_3C_2 and Mo_2C showed the highest values. To further improve the conductivity, Cr_3C_2 and Mo_2C were incorporated at loadings into the coating, which exceeded the CPVC by 10 wt% and 20 wt%, respectively. The desired increase in conductivity was achieved by adding more electrically conductive material. Stainless steel plates with 20 wt% more filler content had a higher conductivity than those with 10 wt% more filler. In addition, the Cr_3C_2 -coated plates had overall lower values than the Mo_2C -coated sheets. A comparison of the commercially available Cr_3C_2 and the mechanochemically synthesized Cr_3C_2 showed significant differences in the in-plane conductivity. The metal plates coated with the mechanochemically synthesized Cr_3C_2 had by far the highest in-plane conductivity with a value of $6.7 \cdot 10^6 \text{ S m}^{-1}$. Additionally, it should be noted that the conductivity values of the mechanochemically synthesized metal carbides even underestimate their actual performance, as the coatings formulated with these carbide fillers contain up to 14 wt% of ZrO_2 impurities from the milling media. To maximize the performance of the conductive materials, higher filler loadings could be incorporated into the coating formulations to compensate for the insulating ZrO_2 content. However, this adjustment could negatively impact processability and the quality of the final coating.

2.3. Interfacial Contact Resistance Measurement

While the in-plane conductivities give a first indication of the conductivity properties of the coating, the practically relevant parameter is the through-plane contact resistance, since this governs the overall performance of the fuel cell stack. Thus, it is one of the most important characteristics in the fuel cell field. It was measured as a function of the compaction pressure in the range of $100\text{--}200 \text{ N cm}^{-2}$ for all coatings, with the results for the coatings formulated at CPVC shown in Figure 5a,b. The resistance values of the graphite-coated samples were in the range of $4500\text{--}6000 \text{ m}\Omega\text{cm}^2$, the plates with the commercial Cr_3C_2

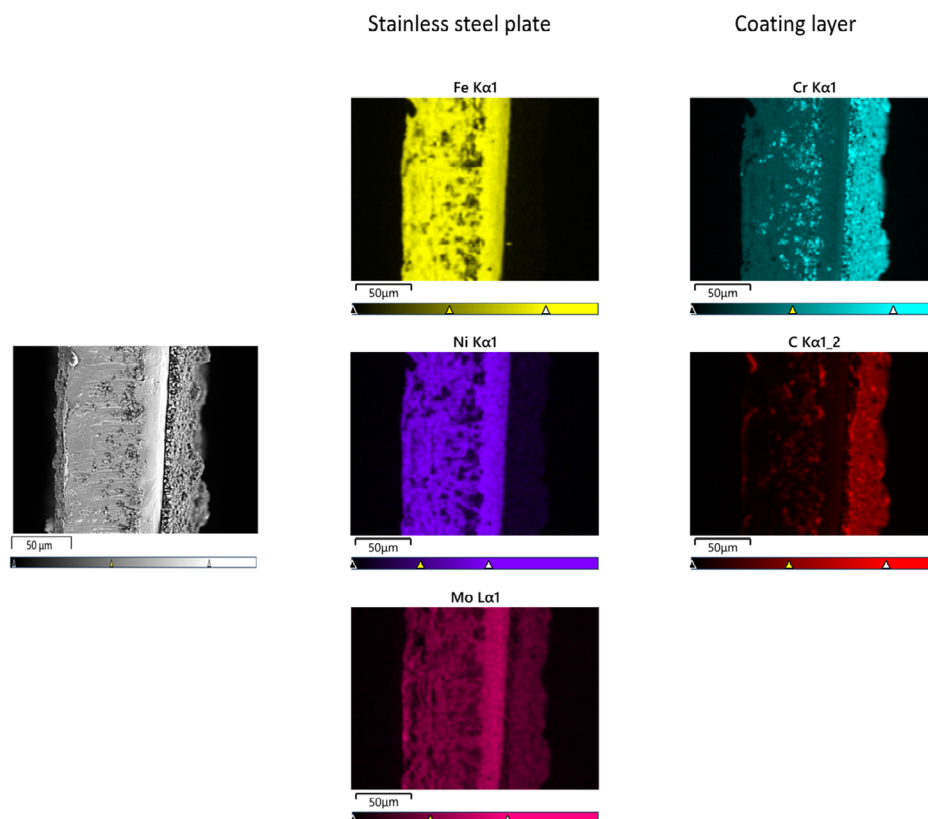


Figure 4. SEM-EDX elemental mapping micrographs of the coated plate containing Cr_3C_2 as a filler, formulated at the CPVC ratio, showing a uniform distribution of Cr_3C_2 in the polyurethane-based binder. The elements Fe, in yellow; Ni, in purple; and Mo, in violet, represent the steel plate, while Cr, in blue and C, in red, indicate the protective coating film.

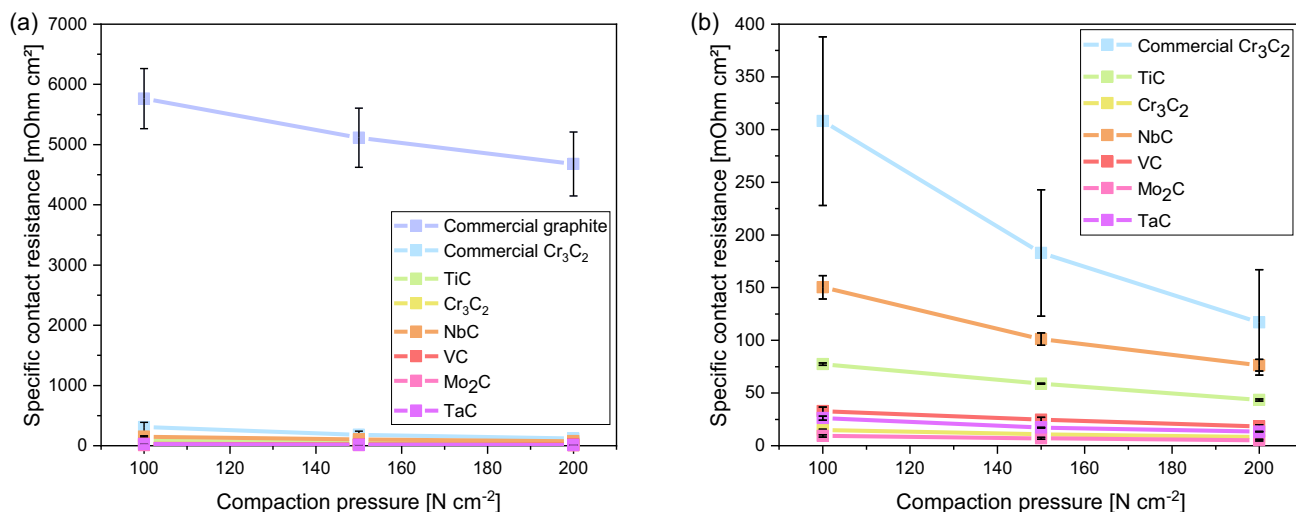


Figure 5. a,b) Contact resistance values for stainless steel plates coated with commercial graphite, commercial Cr_3C_2 , and synthesized metal carbide coating slurries formulated at CPVC. (a) Overview of all coatings and (b) enlarged view of (a).

had values between 120–310 mOhm cm^2 , while the plates coated with the mechanochemically synthesized carbides had values between 5–150 mOhm cm^2 , showing consistently lower values than the commercial fillers. Among the mechanochemically

synthesized metal carbide fillers used in coatings, the steel plates coated with Mo_2C and Cr_3C_2 had the highest through-plane conductivity (exact contact resistance values are given in Table S6, Supporting Information).

A possible explanation for the consistently lower conductivity values of the commercial Cr_3C_2 filler could be a significantly thicker, nonconducting oxide layer on the surface of the particles. The XPS spectra of both Cr_3C_2 are shown in Figure 6a,b. The carbide signal for the commercial sample has a substantially lower intensity than for the mechanochemically synthesized material, while the oxide signal exhibits the opposite trend, clearly suggesting a thicker oxide layer on the commercial material. The conventional methods of carbide synthesis mentioned in the introduction are not performed under a strictly inert atmosphere (methane–hydrogen–argon gas mixtures are used at high temperatures, sometimes carbon monoxide and water vapor are added). While the atmosphere is typically net reducing, oxygen is not completely excluded; some syntheses also start from oxide.^[27–31] This could lead to oxide formation/retention, which would affect the electrical properties. In contrast, the entire synthesis preparation of the ball-milled Cr_3C_2 (and of the other carbides), the loading of the metal–graphite powder mixture and the sealing of the milling jars, as well as the milling process itself, were performed under an argon atmosphere to avoid the formation of an oxide layer on the particles. However, a small oxide signal was also detected on the mechanochemically synthesized Cr_3C_2 particles, which may result from exposure of the precursors to oxygen prior to synthesis, from scratching out the milling jars in air, and/or handling in air until the coating was applied. Traces of Ga and Pd were also found on the surface of the particles, which are contaminations caused by working in the glovebox.

The two additional coatings, the carbide fillers Cr_3C_2 and Mo_2C , at loading exceeding the CPVC result in even lower contact resistance values, which meet the U.S. Department of Energy (DOE) technical target value of $<10 \text{ m}\Omega \text{ cm}^2$ for bipolar plates for transportation applications (Table S6, Supporting Information).^[44] Similar to the in-plane conductivity, the stainless steel plates with 20 wt% higher filler content were found to have higher through-plane conductivity than those with 10 wt% more filler, and the Mo_2C -coated plates had overall lower resistance values compared to Cr_3C_2 . The contact resistance values of Cr_3C_2 - and Mo_2C -based coated plates with increasing filler

content (exceeding the CPVC level) are presented in Figure 7a,b, respectively. The consistently lower contact resistance of Mo_2C compared to Cr_3C_2 can be attributed to Mo_2C 's intrinsically higher conductivity, as reflected by its lower electrical resistivity (the reciprocal of the electrical conductivity) at room temperature: $71 \mu\Omega \text{ cm}$ for Mo_2C compared to $75 \mu\Omega \text{ cm}$ for Cr_3C_2 .^[27] As mentioned above, the same considerations apply to the zirconia impurities in the mechanochemically synthesized carbide fillers, which reduce the material's effective conductivity. Although the “real” conductivity values could not be fully achieved, higher performance could be obtained by incorporating more filler into the polymeric binder matrix. In addition, it was observed that there is a correlation between the in-plane conductivity and the contact resistance of the coated plates, such that the higher the conductivity, the lower the contact resistance of the coatings. However, it should be mentioned that an exact quantitative correlation could not be established.

2.4. Corrosion Measurements

The corrosion performance of the coatings was analyzed according to the DOE-specified measurement protocol.^[44] To simulate the anode side of the PEM fuel cell, the electrolyte, 0.5 M H_2SO_4 containing 0.1 ppm fluoride solution, was purged with argon and heated up to the PEMFC operating temperature of 80°C , while potentiodynamic polarization tests were performed starting from an initial potential of -0.25 V versus open circuit potential (OCP) to a final potential of 0.7 V versus OCP at a scan rate of 0.5 mV s^{-1} . On the simulated cathode side, the electrolyte was flushed with oxygen, heated to operating temperature, and potentiostatic polarization tests were carried out. A potential of 0.75 V versus reversible hydrogen electrode (RHE) was applied to the system and the corrosion current density was measured for 10 h. To obtain an almost fully O_2 -saturated electrolyte, the solution was flushed with O_2 for 1 h prior to the start of the measurement. More detailed descriptions of the corrosion measurements can be found in the Experimental Section of the Supporting Information.

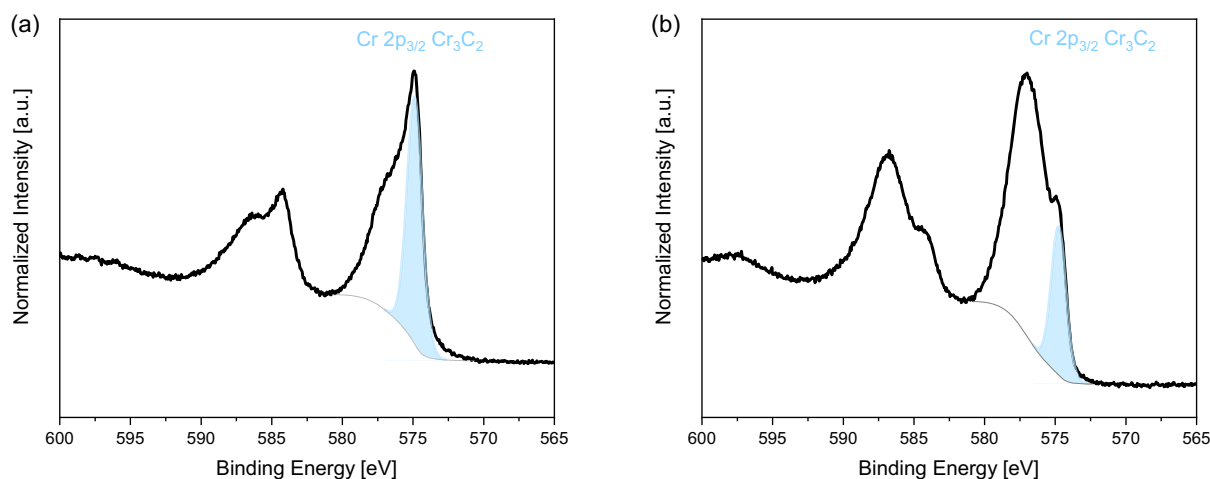


Figure 6. a,b) XPS spectra of Cr 2p region of both Cr_3C_2 samples. (a) Mechanochemically synthesized Cr_3C_2 and (b) commercial Cr_3C_2 . The signal at about 577 eV can be attributed to oxidized chromium species.

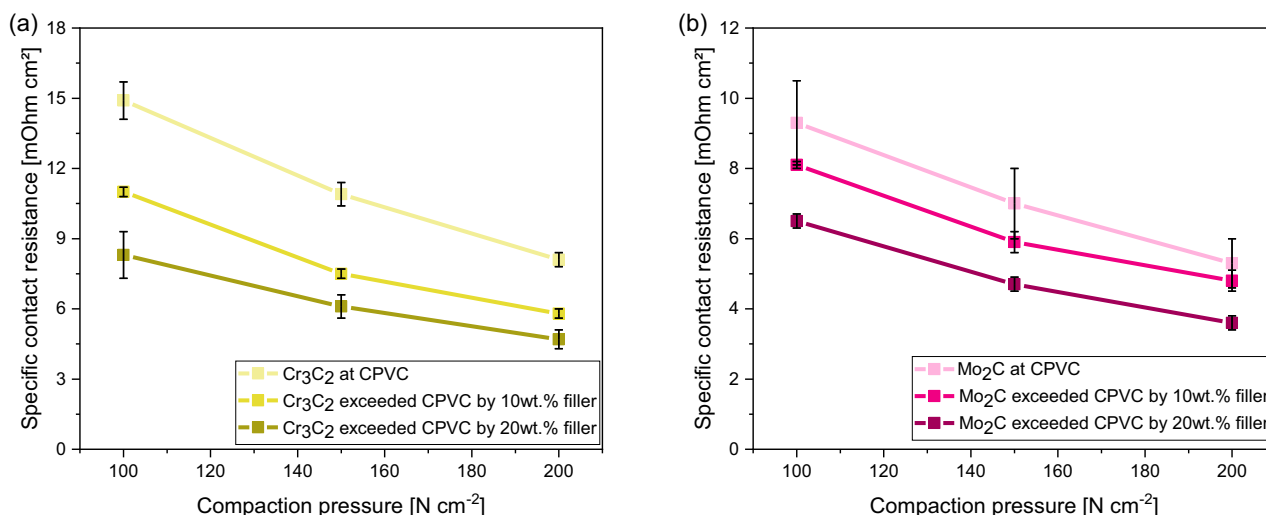


Figure 7. a,b) Contact resistance values for metallic plates coated with a higher carbide filler loading that exceeds the CPVC by 10 wt% and 20 wt%, respectively. (a) Cr_3C_2 and (b) Mo_2C .

2.4.1. Potentiodynamic Polarization Tests

The potentiodynamic curves of the protective films, formulated at CPVC, which are shown in **Figure 8**, indicate that all the coatings protect the metallic substrate from corrosion. The corresponding parameters, such as the self-corrosion potential E_{corr} , the corrosion current density i_{corr} , the polarization resistance to corrosion R_{pol} , and the corrosion rate CR , were calculated using the Tafel extrapolation method (see Equation (S4), (S5), Supporting Information) and are presented in **Table 3**. It can be seen that the lower the i_{corr} , the higher the R_{pol} , and the slower the corrosion progress. The bare stainless steel, which shows the characteristic ranges of active, passive, and transpassive regions, has the lowest corrosion resistance, while there is a wide

Table 3. Corrosion parameters of the bare and coated metal substrates at 80 °C in a 0.5 M H_2SO_4 solution bubbled with argon.

Coating formulation	E_{corr} [V vs. RHE]	i_{corr} [$\mu\text{A cm}^{-2}$]	R_{pol} [Ohm cm^2]	CR [mm per year]
Bare stainless steel 316 L	−0.07	181	64	2
TiC formulated at CPVC	0.22	0.08	$3.7 \cdot 10^5$	0.001
Cr_3C_2 formulated at CPVC	0.43	0.14	$1.8 \cdot 10^5$	0.002
NbC formulated at CPVC	0.38	0.14	$2.2 \cdot 10^5$	0.002
VC formulated at CPVC	−0.10	38	259	0.4
Mo_2C formulated at CPVC	0.23	0.40	$5.5 \cdot 10^4$	0.004
TaC formulated at CPVC	0.41	0.38	$6.5 \cdot 10^4$	0.004
Commercial graphite formulated at CPVC	0.42	0.20	$9.9 \cdot 10^4$	0.003
Commercial Cr_3C_2 formulated at CPVC	0.46	0.12	$1.7 \cdot 10^5$	0.002

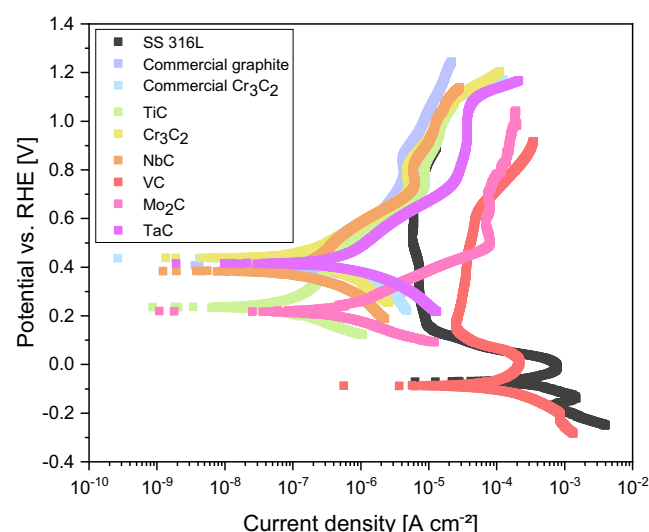


Figure 8. Potentiodynamic curves for the bare stainless steel and the CPVC formulated coatings at 80 °C in 0.5 M H_2SO_4 in a simulated anodic environment.

variation in the performance of the protective films. The VC-containing coating, with the highest i_{corr} , the lowest R_{pol} , and the fastest corrosion expansion, showed the least protection among the coatings, although E_{corr} is slightly lower than that of the bare stainless steel plate. This is followed by Mo_2C with the lowest E_{corr} among the remaining coatings. TaC, commercial graphite, both Cr_3C_2 s, the commercial and the mechanochemically synthesized, and the NbC coating all have rather similar E_{corr} , but only the last three coatings showed a similar corrosion behavior, while the TiC coating showed by far the best corrosion properties of the coated plates, even though E_{corr} is in a similar range as that of the Mo_2C coating. One possible explanation for the superior corrosion resistance of the TiC coating is its lower conductivity compared to the Cr_3C_2 and Mo_2C coatings, which implies a reduced electron transport and thus, lower current flow within its filler particle network, thereby slowing down corrosion processes. In contrast, the Cr_3C_2 and Mo_2C coatings exhibit

higher conductivity, resulting in a more efficient electron transport through their filler networks, which can facilitate electrochemical reactions and thereby accelerate corrosion. It is important to note that the i_{corr} of bipolar plates should be less than $1 \mu\text{A cm}^{-2}$ according to the DOE specifications. All coatings formulated with the commercial fillers, as well as those with the synthesized carbide fillers, meet this requirement. The only exception is the protective film containing VC, which is out of the range with an i_{corr} of almost $40 \mu\text{A cm}^{-2}$.

For the coatings with filler contents exceeding the CPVC with Cr_3C_2 and Mo_2C , the potentiodynamic curves presented in Figure 9a,b show differences in the protection of the metal plate as more filler was added to the coating. In the case of Cr_3C_2 , there was almost no difference in corrosion behavior, and none of these coatings showed active corrosion, while for the Mo_2C coated steel plates, the protection became progressively worse with increasing filler content. Even active corrosion of the substrate, indicated by an oxidation peak, occurs when the coating is formulated with a filler content exceeding CPVC by 20 wt%.

The corrosion parameters of the coatings are shown in Table 4. For the Cr_3C_2 coatings, no significant change in the characteristic values was observed. The two coatings, base Cr_3C_2 and Cr_3C_2 exceeding CPVC by 10 wt%, have almost the same level of corrosion protection, while the metal substrate covered with the coating exceeding CPVC by 20 wt% has a slightly lower protection. On the other hand, the parameters of the Mo_2C coated plates show large variations. E_{corr} decreased as more filler was added. At a final level of 20 wt% above the CPVC, E_{corr} was even slightly lower than that of the bare metal plate. Increasing the filler content by 10 wt% resulted in a 4 times higher i_{corr} , and correspondingly in a 4 times lower R_{pol} compared to the base Mo_2C sample. In fact, the effect of adding 20 wt% filler even resulted in almost 300 times higher i_{corr} and 300 times lower R_{pol} . The higher filler content can lead to higher current flow, which can cause active corrosion. In contrast, a coating consisting only of an organic polymer layer, i.e., a binder layer, would provide the best corrosion protection since no current flow can be established. One possible explanation for the minimal change in

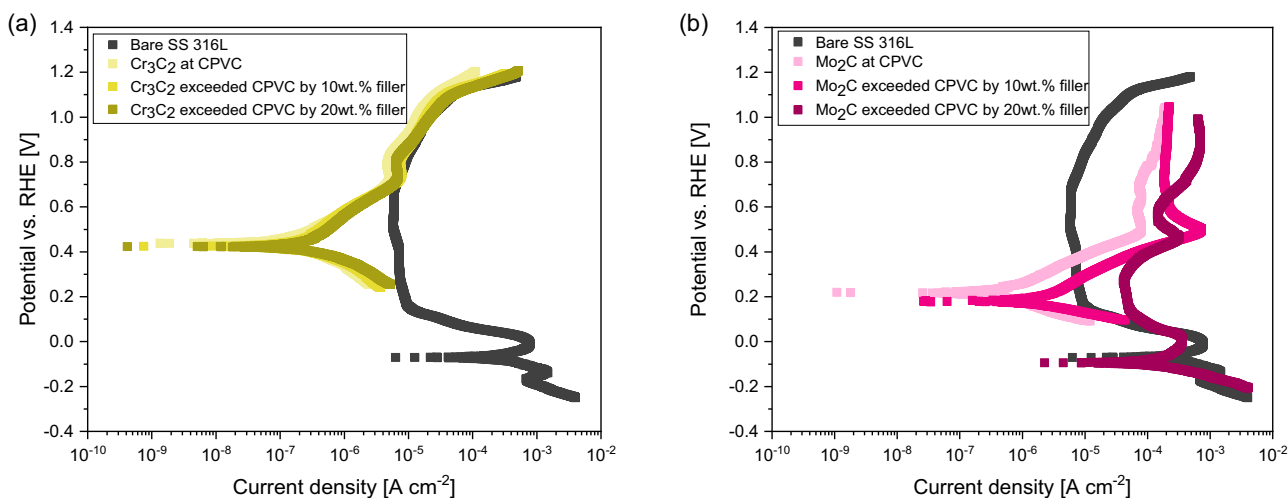


Figure 9. a, b) Potentiodynamic curves for uncoated and coated metal substrates with filler contents exceeding CPVC by 10 wt% and 20 wt%, respectively, at 80 °C in 0.5 M H_2SO_4 in simulated anodic environment. (a) Cr_3C_2 and (b) Mo_2C .

Table 4. a, b) Corrosion parameters of uncoated and coated stainless steel substrates with increased filler content at 80 °C in 0.5 M H_2SO_4 purged with argon. a) Cr_3C_2 and b) Mo_2C .

a)	E_{corr} [V vs. RHE]	i_{corr} [$\mu\text{A cm}^{-2}$]	R_{pol} [Ωcm^2]	CR [mm per year]
Bare stainless steel 316 L	−0.07	181	64	2
Cr_3C_2 formulated at CPVC	0.43	0.15	$1.8 \cdot 10^5$	0.002
Cr_3C_2 exceeded CPVC by 10 wt% filler	0.42	0.15	$1.6 \cdot 10^5$	0.002
Cr_3C_2 exceeded CPVC by 20 wt% filler	0.42	0.24	$1.2 \cdot 10^5$	0.003
b)	E_{corr} [V vs. RHE]	i_{corr} [$\mu\text{A cm}^{-2}$]	R_{pol} [Ωcm^2]	CR [mm per year]
Bare stainless steel 316 L	−0.07	181	64	2
Mo_2C formulated at CPVC	0.23	0.40	$5.5 \cdot 10^4$	0.004
Mo_2C exceeded CPVC by 10 wt% filler	0.15	1.66	$1.3 \cdot 10^4$	0.02
Mo_2C exceeded CPVC by 20 wt% filler	−0.09	116	146	1.3

the corrosion behavior of the Cr_3C_2 coating when the filler content exceeds the CPVC is the role of the filler in passivating stainless steel 316 L. A higher Cr_3C_2 content promotes the formation of a more stable and protective chromium oxide passivation layer on the metal surface, which protects the stainless steel from degradation and inhibits corrosion. In contrast, a higher Mo_2C content does not contribute to passivation; instead, it leads to pronounced active corrosion.

Regarding the DOE specifications, both Cr_3C_2 additive coatings meet the requirements with an i_{corr} below $<1 \mu\text{A cm}^{-2}$. On the other hand, both Mo_2C coatings fall far short of the corrosion performance standard, so that for practical application the Cr_3C_2 based coating appears to be the preferred option.

Following the corrosion tests, the samples were visually analyzed to evaluate the state of the coatings (Figure 10a–g, the remaining coatings can be seen in Figure S8a–e, Supporting Information). The electrochemical corrosion parameters of the coated plates is consistent with the observed appearance of the protective coating. The VC-containing coating, which had the lowest corrosion resistance among the CPVC formulated coatings, showed the greatest degradation of the film, with even a large portion of the film having been delaminated. As the corrosion performance improved, the specimens showed only a minor coating delamination. The TiC coating with the lowest corrosion current density and the highest corrosion resistance had no visible coating defects. (Figure S8, Supporting Information) For the coatings with increased amounts of Cr_3C_2 and Mo_2C filler, the corrosion performance deteriorated and more damage to the protective films was observed for the more highly filled coatings. A possible explanation could be the penetration of the electrolyte through the protective film due to the incomplete inclusion of filler particles in the binder matrix at levels above the CPVC formulation. When the coating is prepared at CPVC, all

filler particles can principally be covered with binder. As more filler particles are added to the coating, the probability increases that individual particles will not be completely encapsulated by the polymer material. As a result, electrolyte can penetrate through the protective layer, causing the coating to swell and separate from the metal plate (Figure S6, Supporting Information).

Since in a PEM fuel cell, metal ions generated by active corrosion can migrate through the membrane, and a level of 5–10 ppm can cause performance degradation in the ionic conductivity of the membrane,^[45] the metal ion concentration of the electrolyte, especially the metal ion concentration of Fe, Cr, Ni, and Mo, which are the main elements of 316 L stainless steel, was analyzed after the corrosion test (Table 5). The metal ion concentration of the other coatings can be seen in Table S9, Supporting Information. All of the coatings, except Mo_2C , were found to protect the stainless steel from corrosion by reducing the concentration of metal ions in the electrolyte compared to the bare stainless steel. Another observation is that the Cr concentration in the electrolyte is approximately ten times higher for the commercial Cr_3C_2 -based coating than for the synthesized Cr_3C_2 -based coating. This difference can be attributed to the lower adhesion strength of the commercial carbide coating (adhesion rating of 1) compared to the superior adhesion achieved with the synthesized carbide coating (adhesion rating of 0).

In the case of the two higher filled Cr_3C_2 and Mo_2C coatings, it was seen that the concentration level of both metal ions, Cr and Mo, increased as more filler material was added to the coating formulation. However, they were still lower than those of the pure steel in the electrolyte. In terms of Mo ion concentration, Mo_2C coatings with 10 and 20 wt% more filler showed lower corrosion protection and higher contamination levels in the electrolyte than the pure metal plate. The combination of a high Mo concentration and the presence of 5 ppm of Fe in the electrolyte,

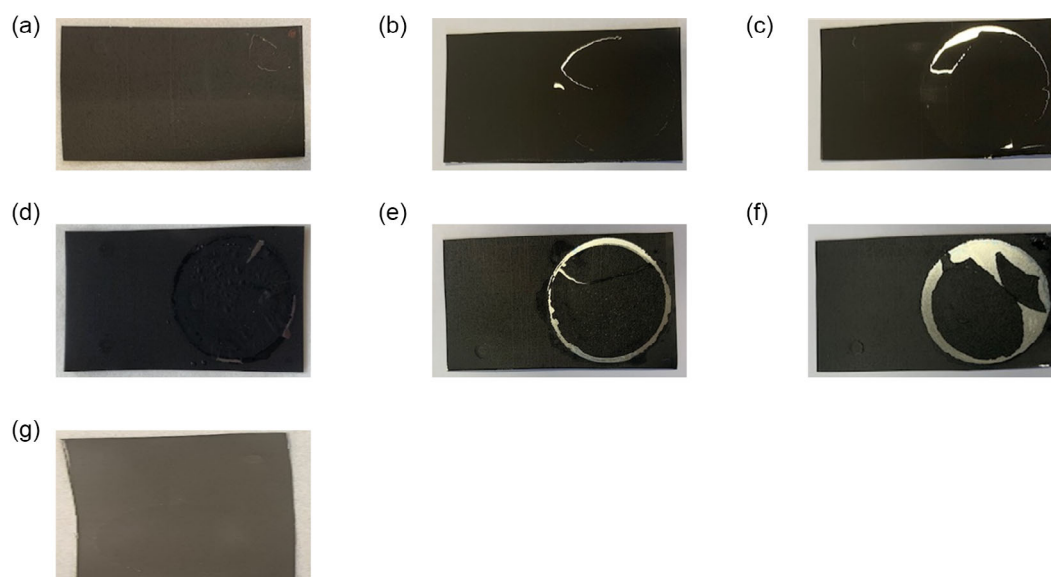


Figure 10. a–g) Photographic images of the coated stainless steel plates after potentiodynamic polarization tests. (a) Cr_3C_2 coating formulated at CPVC; (b) Cr_3C_2 coating exceeding CPVC by 10 wt%; (c) Cr_3C_2 coating exceeding CPVC by 20 wt%; (d) Mo_2C coating formulated at CPVC; (e) Mo_2C coating exceeding CPVC by 10 wt%; (f) Mo_2C coating exceeding CPVC by 20 wt%; and (g) commercial Cr_3C_2 coating formulated at CPVC. The round feature indicates the area where the electrolyte contacts the plates, sealed by an O-ring.

Table 5. Metal ion concentration in the electrolyte after potentiodynamic polarization tests performed at 80 °C in 0.5 M H₂SO₄ bubbled with argon.

	Fe [ppm]	Cr [ppm]	Ni [ppm]	Mo [ppm]
Bare stainless steel 316 L	11.9	3.5	2.3	0.4
Cr ₃ C ₂ formulated at CPVC	0.07	0.01	0.9	–
Cr ₃ C ₂ exceeded the CPVC by 10 wt% filler	0.1	0.06	–	–
Cr ₃ C ₂ exceeded the CPVC by 20 wt% filler	0.1	0.2	–	–
Mo ₂ C formulated at CPVC	0.02	–	0.01	2.1
Mo ₂ C exceeded the CPVC by 10 wt% filler	0.2	–	0.5	4.5
Mo ₂ C exceeded the CPVC by 20 wt% filler	5.0	1.1	1.3	11.8
Commercial Cr ₃ C ₂ formulated at CPVC	0.06	0.1	0.02	–

particularly in the coating with 20 wt% higher Mo₂C filler content, can accelerate the chemical degradation of the Nafion membrane in a PEMFC system. This occurs via Fenton reactions, in which iron ions react with hydrogen peroxide produced at the PEMFC's anode and cathode due to oxygen crossover or as an intermediate of the oxygen reduction reaction. These reactions generate hydroxyl and hydroperoxyl radicals that attack the polymer backbone of the membrane, resulting in structural damage and membrane thinning. Consequently, the membrane's ionic conductivity decreases, ultimately reducing the overall performance of PEMFCs.^[45–47]

2.4.2. Potentiostatic Polarization Tests

The potentiostatic curves representing the long-term corrosion stability of the protective films formulated at CPVC, shown in Figure 11a,b, indicate that almost all the coatings are effective in protecting stainless steel from corrosion (detailed i_{corr} values are given in Table S7, Supporting Information). The corrosion performance of the bare stainless steel with an i_{corr} in the range of 5 $\mu\text{A cm}^{-2}$ was improved by most of the coatings; only the

VC-containing coating failed in terms of protection, showing an i_{corr} around 6 $\mu\text{A cm}^{-2}$. This was followed by the Mo₂C coating with only slightly better corrosion properties compared to the bare substrate, TaC and TiC coatings with similar levels of corrosion resistance, NbC coating with an i_{corr} of less than 1 $\mu\text{A cm}^{-2}$, and commercial graphite together with both Cr₃C₂ coatings provided the best protection against the metal degradation with an i_{corr} below 0.5 $\mu\text{A cm}^{-2}$. According to the DOE guidelines, which specify that bipolar plates in a PEMFC cathode side should have an i_{corr} of less than 1 $\mu\text{A cm}^{-2}$, coatings with both commercial fillers as well as with the mechanochemically synthesized Cr₃C₂ and NbC fillers are able to meet the technical requirements, while the i_{corr} of the remaining coatings ranged from 1.5 to 6 $\mu\text{A cm}^{-2}$.

For the two higher filled coatings of Cr₃C₂ and Mo₂C, the potentiostatic curves show differences in the corrosion performance of the coated metal plates as more filler is added to the coating formulation (Figure 12a–d, detailed i_{corr} values can be found in Table S8, Supporting Information). In the case of Cr₃C₂, there was almost no change in the corrosion behavior, while for the Mo₂C coated steel plate, the corrosion protection deteriorated with increasing filler content. No significant increase in i_{corr} was observed for either Cr₃C₂ coating. Compared to the Cr₃C₂ coating formulated at CPVC, the two samples with higher filler content provided slightly less metal protection at the same current density range. However, the Mo₂C-coated plates showed a decrease in corrosion protection as more filler was added to the coating. Increasing the filler content by 10 wt% resulted in an i_{corr} that was twice that of the CPVC formulated coating. At a final level of 20 wt% more filler, the increase was lower, ≈ 1.5 times higher after testing. The use of higher filler levels in coatings can result in higher current densities. As mentioned above, the same principle applies: increasing the Cr₃C₂ filler content enhances the formation of the stainless steel passivation layer, resulting in minimal changes in corrosion behavior. In contrast, the Mo₂C content does not protect the metal surface, leading to more pronounced corrosion as the filler loading increases. A pure polymer binder layer on the metal plate

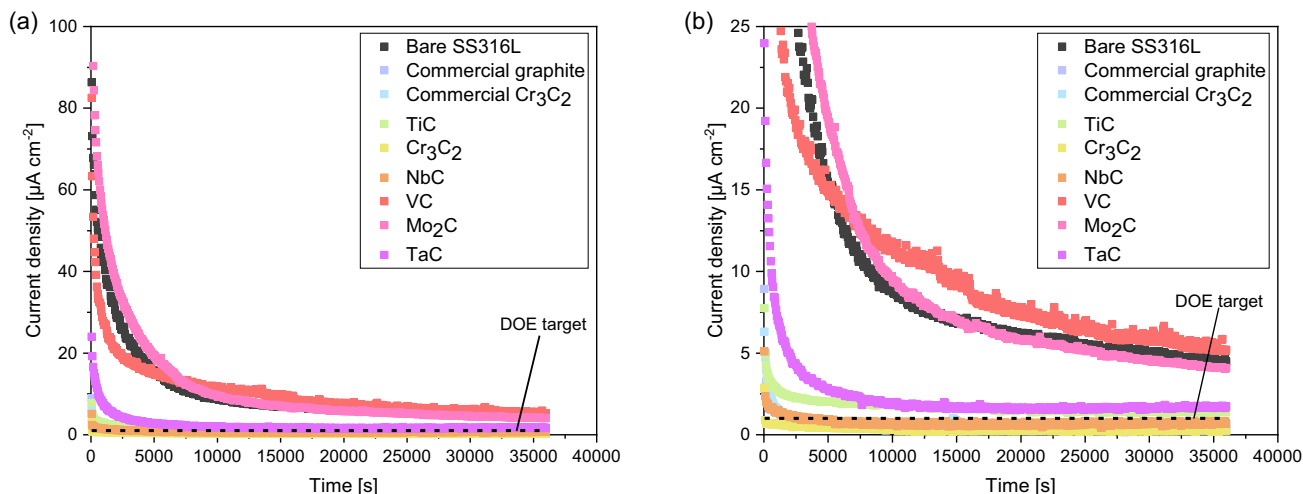


Figure 11. a,b) Potentiostatic curves for the bare stainless steel and CPVC formulated coatings at 80 °C in 0.5 M H₂SO₄ in a simulated cathodic environment. (a) Overview of all samples and (b) magnified image of (a).

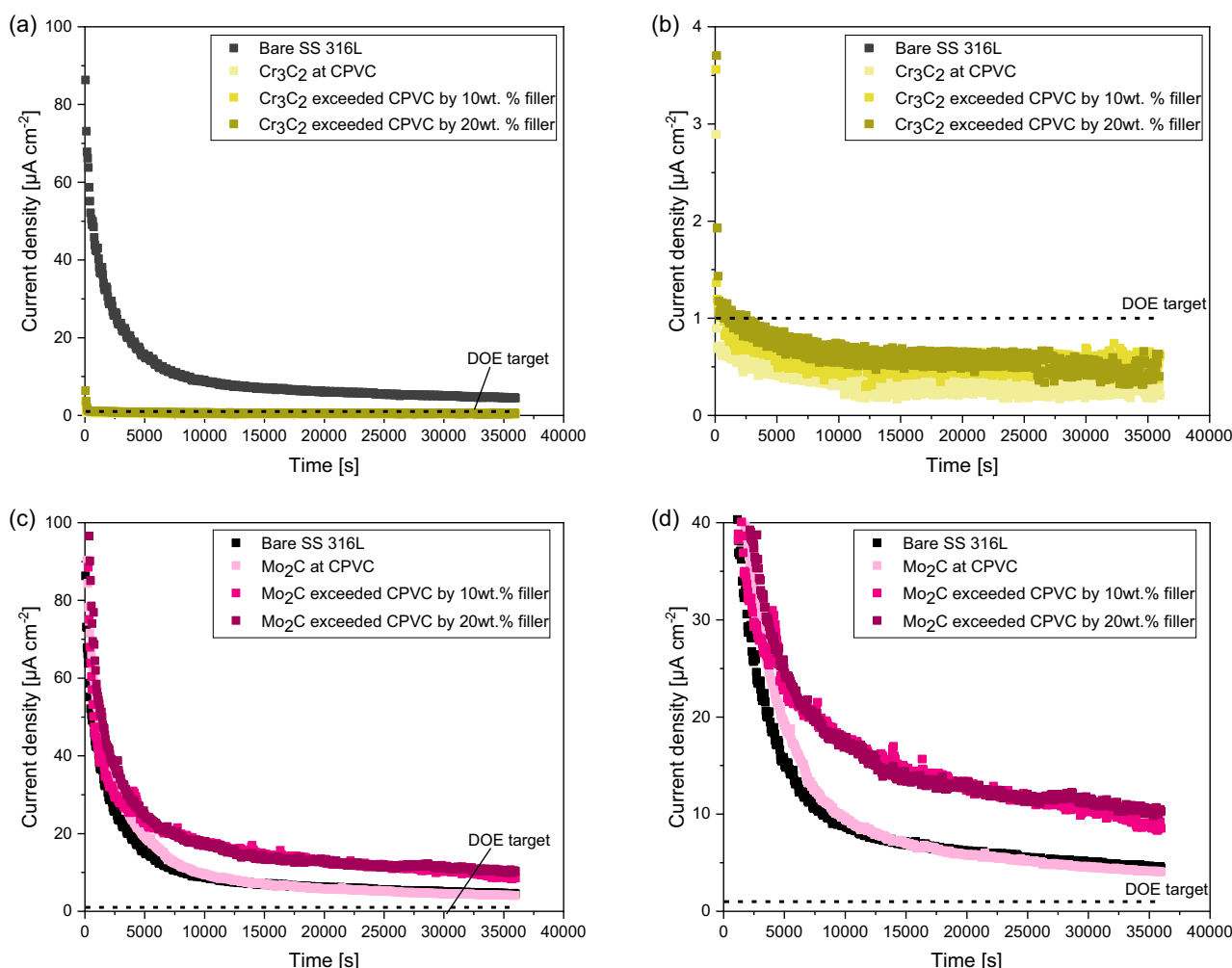


Figure 12. a–d) Potentiostatic curves for uncoated and coated metal substrates with filler contents exceeding CPVC by 10 wt% and 20 wt%, respectively, at 80 °C in 0.5 M H₂SO₄ in simulated cathodic environment. (a) Cr₃C₂; (b) magnified graph of (a); (c) Mo₂C; and (d) magnified graph of (c).

would provide the best corrosion protection. In terms of DOE specifications, both of the Cr₃C₂ additive coatings meet the requirements with an i_{corr} of less than $<1 \mu\text{A cm}^{-2}$, while both Mo₂C coatings fail to meet the corrosion performance standard.

Following the potentiostatic tests, the samples were visually inspected to analyze the state of the coatings (Figure 13a–g). The electrochemical corrosion parameters of the coated samples correlate closely with the observed appearance of the protective films. Coatings with high metal protection showed little or only partial delamination. As the protective performance deteriorated, increasing delamination of the coating was observed. In comparison to the potentiodynamic polarization tests, the protective films showed poorer properties in terms of adhesion to the metal substrate. Even after only several hours of exposure to the electrolyte, swelling of the coated film was observed in some of the low corroding samples (Figure S9a–e, Supporting Information). Partial or complete delamination of the coating occurred after the O-ring mask seal was removed. One explanation could be that swelling of the coatings causes electrolyte penetration into the polymer matrix, resulting in wetting of the metal substrate

and delamination of the film. For the higher-loaded Cr₃C₂ and Mo₂C coatings, as more electrically conductive material was added to the formulation, the corrosion performance decreased and more damage to the protective films was observed. As above, this could be attributed to incomplete coverage of the filler with polymer as the CPVC level was exceeded. This would allow the electrolyte to penetrate through the protective film, and peel from the metal substrate.

The metal ion concentration of the most common elements found in stainless steel 316 L, Fe, Cr, Ni, and Mo, was analyzed in the electrolyte, as described above for the other electrode (Table 6). The metal ion concentration of the remaining coatings can be seen in Table S10, Supporting Information. Most of the coatings were found to protect the stainless steel from corrosion and reduce the concentration of metal ions in the electrolyte compared to the bare metal. Even the VC-coated plate showed lower concentrations of the metals, although the corrosion performance of the sample was inferior to the bare steel. An increase in transition metal concentration was observed for Cr₃C₂ coatings with higher filler content. When the final filler content

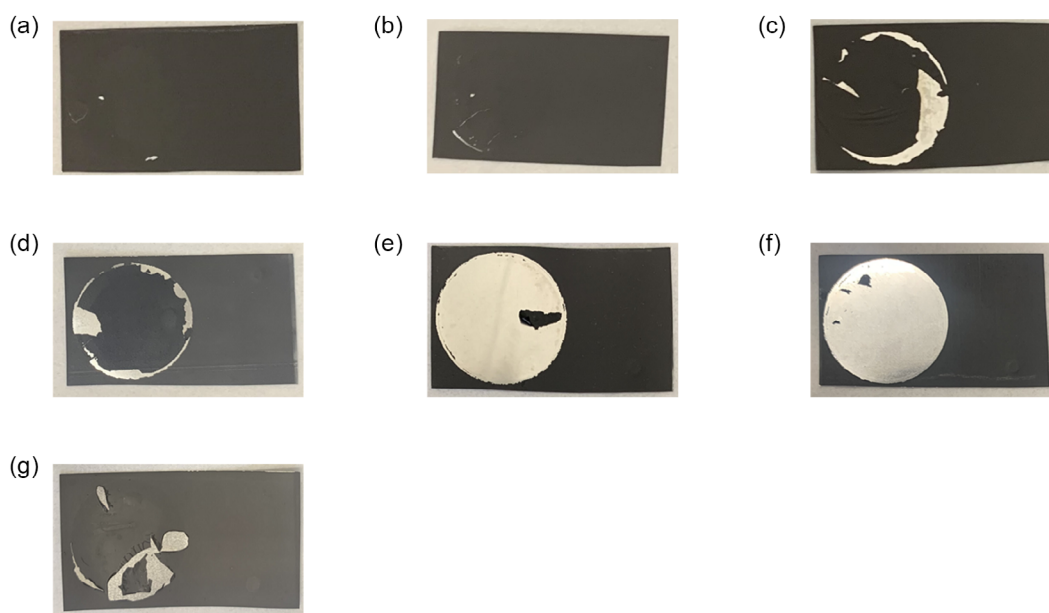


Figure 13. a–g) Photographic images of the coated stainless steel plates after potentiostatic polarization measurements. (a) Cr_3C_2 coating formulated at CPVC; (b) Cr_3C_2 coating exceeding CPVC by 10 wt%; (c) Cr_3C_2 coating exceeding CPVC by 20 wt%; (d) Mo_2C coating formulated at CPVC; (e) Mo_2C coating exceeding CPVC by 10 wt%; (f) Mo_2C coating exceeding CPVC by 20 wt%; and (g) commercial Cr_3C_2 coating formulated at CPVC. The round feature indicates the area where the electrolyte contacts the plates, sealed by an O-ring.

Table 6. Metal ion concentration in the electrolyte after performing potentiostatic polarization tests performed at 80 °C in 0.5 M H_2SO_4 bubbled with oxygen.

	Fe [ppm]	Cr [ppm]	Ni [ppm]	Mo [ppm]
Bare stainless steel 316 L	48.1	9.7	7.8	0.7
Cr_3C_2 formulated at CPVC	0.1	0.1	–	–
Cr_3C_2 exceeded the CPVC by 10 wt% filler	1.8	22.0	0.3	0.06
Cr_3C_2 exceeded the CPVC by 20 wt% filler	32.0	94.0	14.9	2.2
Mo_2C formulated at CPVC	0.4	0.08	0.6	82
Mo_2C exceeded the CPVC by 10 wt% filler	0.3	0.06	0.06	95
Mo_2C exceeded the CPVC by 20 wt% filler	28	5	4.1	84
Commercial Cr_3C_2 formulated at CPVC	0.2	0.06	–	0.04

exceeded the CPVC by 20 wt%, the concentrations of Cr, Ni, and Mo far exceeded those of pure stainless steel. For Mo_2C coatings, a filler loading exceeding the CPVC level by 10 wt% resulted in higher Mo release than a 20 wt% excess. For both higher filled Cr_3C_2 and Mo_2C coatings, however, the Cr and Mo concentrations reached values well above those observed for the bare stainless steel plate.

3. Conclusion

In this work, a mechanochemical synthesis method for electrically conducting early transition metal carbides for use as fillers in conductive polymer-based coatings was developed. The

electrical conductivity and corrosion performance of composite coatings based on these carbides on stainless steel plates for use as bipolar plates in PEM fuel cells were investigated. The carbides were incorporated as electrically conductive fillers into a polyurethane-based binder matrix and applied to stainless steel plates using a doctor blade in a wet paint technique. All coatings exhibited good adhesion to the metal substrates and a homogeneous filler-binder distribution. Through-plane conductivity measurements showed that the synthesized Cr_3C_2 and Mo_2C samples had the lowest contact resistance of all the coatings. Increasing their filler content leads to coatings meeting the DOE technical target value of $<10 \text{ m}\Omega \text{ cm}^2$ for bipolar plates for transport applications, with protective films containing Mo_2C showing slightly better conductivity performance than Cr_3C_2 in both cases. The consistently higher conductivity values of the coatings with mechanochemically synthesized fillers could be attributed to a thinner insulating oxide layer on the filler particle surface. In the synthesis of commercial carbides by conventional methods, air is not systematically excluded, which can lead to the formation of a surface/subsurface oxide layer on the particles that can affect the electrical properties. On the other hand, the preparation of the starting materials and the milling process of the ball-milled carbides were performed under an argon atmosphere to prevent oxide formation.

Potentiodynamic polarization tests, which simulate rapid corrosion measurements, indicated that high conductivity coatings do not necessarily provide good corrosion resistance. While the Mo_2C coating provided limited protection, the Cr_3C_2 coating offered excellent corrosion resistance even at high filler loading. Similar corrosion behavior was observed in long-term corrosion measurements. For both coatings with excess filler, the Cr_3C_2 protective films didn't show a significant difference in corrosion

protection, while both Mo₂C coatings demonstrated a clear deterioration. Visual inspection of the coatings and inductively coupled plasma optical emission spectroscopy (ICP-OES) results were consistent with the electrochemical corrosion parameters of the coated metal plates, with metal corrosion being more severe in a simulated cathode environment. These findings position mechanochemically synthesized Cr₃C₂ as an attractive filler for protective, conductive composite coatings on PEMFC metal bipolar plates, which meet the US DOE technical targets for commercial applications. This opens a promising pathway for cost-effective and durable coating technologies. Scale-up of the materials to kilogram quantities is in progress to prepare larger-scale testing.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

A patent application on the subject of this study has been filed. Creation of a company MechSyn to commercialize the development described here is in progress.

Author Contributions

Jil Meyers: conceptualization (lead); formal analysis (lead); investigation (lead); methodology (lead); visualization (lead); writing—original draft (lead). **Özgül Agbaba:** supervision (supporting); validation (equal); writing—review and editing (equal). **Henning Buchholz:** investigation (supporting); methodology (supporting); validation (supporting); writing—review and editing (supporting). **Ferdi Schüth:** funding acquisition (lead); project administration (lead); resources (lead); supervision (lead); validation (equal); writing—review and editing (lead).

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

bipolar plates, composite coatings, conductive fillers, mechanochemical reactions, proton exchange membrane fuel cells, stainless steel

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