Multifunctional Nanocomposites with High Strength and Capacitance Using 2D MXene and 1D Nanocellulose

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The family of two-dimensional (2D) metal carbides and nitrides, known as MXenes, are among the most promising electrode materials for supercapacitors thanks to their high metal-like electrical conductivity and surface-functional-group-enabled pseudocapacitance. A major drawback of these materials is, however, the low mechanical strength, which prevents their applications in lightweight, flexible electronics. A strategy of assembling freestanding and mechanically robust MXene (Ti$_3$C$_2$Tx) nanocomposites with one-dimensional (1D) cellulose nanofibrils (CNFs) from their stable colloidal dispersions is reported. The high aspect ratio of CNF (width of ≈3.5 nm and length reaching tens of micrometers) and their special interactions with MXene enable nanocomposites with high mechanical strength without sacrificing electrochemical performance. CNF loading up to 20%, for example, shows a remarkably high mechanical strength of 341 MPa (an order of magnitude higher than pristine MXene films of 29 MPa) while still maintaining a high capacitance of 298 F g$^{-1}$ and a high conductivity of 295 S cm$^{-1}$. It is also demonstrated that MXene/CNF hybrid dispersions can be used as inks to print flexible micro-supercapacitors with precise dimensions. This work paves the way for fabrication of robust multifunctional MXene nanocomposites for printed and lightweight structural devices.
and properties to be considered for practical structural materials for flexible electronics.

We here use carboxymethylated cellulose nanofibrils (CNFs) as a functional additive to assemble with MXenes. CNFs as freestanding films have shown a high toughness and a large strain (larger than 6%) at break.27 CNFs also show interesting abilities to disperse and stabilize low-dimension electronic materials such as carbon nanotubes (CNTs),28,29 PEDOT,30 and graphene,31 and can be used as the scaffold/binders for the fabrication of composites.32–35 The thin dimension of CNFs we used, with a width of ≈3.5 nm and length reaching several micrometers, offer the possibility of scaffolding/binding individual MXene flakes into high-strength materials without introducing a high amount of insulating phases between the conductive layers. We show that CNFs and MXene Ti3C2Tx colloidal dispersions form indeed stable, homogeneous hybrid at different weight ratios. The freestanding hybrid film made from this hybrid dispersion can simultaneously integrate a high electric conductivity (690 S cm−1), high mechanical strength (154 MPa), high Young’s modulus (41.9 GPa), and high specific capacitance (325 F g−1) with 10% CNF loading. To the best of our knowledge, the combination of these values is among the highest reported to date for structural supercapacitor electrodes. These properties stem from the strong interfacial interactions between Ti3C2Tx flakes and CNF. We further demonstrate the feasibility of using Ti3C2Tx/CNF hybrid for patterning highly flexible functional electronics with the minimum resolution (≈120 μm) of the gaps between interdigital electrodes.

We prepared the MXene/CNF hybrid dispersions by mixing the colloidal dispersion of delaminated Ti3C2Tx (d-Ti3C2Tx) MXene flakes and carboxymethylated CNFs at different mass ratios (Figure 1a). These two materials are both negatively charged when dispersed in water: the as-prepared d-Ti3C2Tx MXene had a zeta potential of −51.4 mV at a pH of 7, and CNF had a zeta potential of −71.8 mV at a pH of 7 (Figure 1b). The individual Ti3C2Tx flakes are around 2 nm thick and several hundred nm in lateral size (Figure 1c,d). We prepared carboxymethylated CNFs from a fully bleached sulfite pulp from Scandinavian softwood that was subsequently carboxymethylated to a charge around 600 μmol g−1.36 The individual nanofibrils have a high aspect ratio with a uniformly square cross-section of around 3.5 nm and length reaching several micrometers (Figure 1e,f). Figure 1b shows the zeta potential change of Ti3C2Tx/CNF hybrid dispersions with the different mass concentration of CNF as a function of the time. The hybrid dispersions showed high stability with insignificant changes in the zeta potential, such as Ti3C2Tx−20% CNF with a zeta potential of −52.9 mV after one month. Over this period, the hybrid dispersions did not have any precipitates or phase separations, suggesting a stable blend of the two colloids.

To study the interaction between the two phases, we used colloidal probe atomic force microscopy (AFM)39 in liquid to measure the interfacial interaction between MXene flakes and nanocellulose fibrils. We selected a model system consisting of a planar surface coated with Ti3C2Tx flakes and a probe coated with CNFs (for details see the Experimental Section and Figure S1a of the Supporting Information). Figure 1g,h shows the force–distance measurements between Ti3C2Tx flake’s surface and the CNF-coated probe upon approach and on separation in liquid (Milli-Q water and NaCl solution). The approach data (Figure 1g; Figure S1b, Supporting Information) show long-range electrostatic repulsion for all solutions containing NaCl (10−6 to 10−3 M) whereas for pure water there is also an attractive force at separations less than ≈50 nm. We propose that electrostatic forces between CNFs and Ti3C2Tx flakes are responsible for the long-range repulsion, which in turn favor the stabilization of the dispersion. The separation data (Figure 1h) show the force required to separate the two surfaces. We calculated the adhesion between Ti3C2Tx surface and the CNF-coated probe as shown in Figure 1i. The adhesion decreased from 1.0 mJ m−2 in Milli-Q water to 0.4 mJ m−2 in 10−3 M NaCl. This result is consistent with our previous reports for graphene–CNF system.39 We attribute the adhesion between the Ti3C2Tx flakes and CNFs to the functional groups (−OH, −O−, −F) present on the outer transition metal layer surface of Ti3C2Tx flakes. Additionally, these results indicate that the highest adhesion between the surfaces can be achieved at the low ionic strength regime, i.e., Milli-Q water. We, therefore, conducted all fabrication of Ti3C2Tx/CNF composites in water.

The attraction force between CNFs and Ti3C2Tx flakes offers an excellent opportunity of binding MXene flakes with a strong CNF scaffold. We fabricated freestanding composite nanopapers made from Ti3C2Tx/CNF dispersions using vacuum filtration (Figure 2a). Figure 2, and Figures S2 and S3 (Supporting Information) show the morphologies of the freestanding nanopapers of pristine Ti3C2Tx, MXene and CNF as well as their composites. We found that the composites have similar layered structures as the pristine MXene films. With the increase of CNF loading, nanofibril structures appeared at the cross-sectional view of layers and even between adjacent layers. We note that the freestanding nanopapers possess smooth surfaces even at high CNF loading, with a roughness Ra of around 10 nm from a 3 × 3 μm2 area (Figure S3, Supporting Information).

XRD pattern (Figure 3a) of the pristine Ti3C2Tx MXene shows clear diffraction peaks related to MXene basal planes. The peak at 6.9° (d-spacing of 12.8 Å) corresponds to (0002) planes of MXenes. This peak moved from 6.1° to 5.3° as CNF loading increased from 5% to 40%, corresponding to a shift in the (0002) planes’ distance from 14.4 to 16.7 Å. These results suggest a substantial amount of CNF incorporated into nanocomposites as a function of CNF content, which is in agreement with the data of FTIR and contact angles (Figure S4, Supporting Information).

Loading of CNF to the MXene films significantly enhanced the mechanical strength of the MXene Ti3C2Tx/CNF nanocomposites. The freestanding films of Ti3C2Tx/CNF can be folded or wrapped into various shapes without any cracking (Figure 2b). The tensile strength, i.e., stress at break, of Ti3C2Tx−5% CNF increased significantly to 68.0 from 25.8 MPa for pristine MXene Ti3C2Tx film (Figure 3d, Figure S5 (Supporting Information), and Table 1). The tensile strength increased up to 271.5 MPa for Ti3C2Tx−60% CNF, which is around an order of magnitude higher than pristine MXene films, and is more than two-times higher than that of previously reported Ti3C2Tx/CNF composites (with the highest tensile strength of 135 MPa).47

To demonstrate the effect of the geometry of nanocellulose on the composite, we also used cellulose nanocrystals (CNCs) which are shorter than CNF to build MXene/CNF composites.
As shown in Figure S6 of the Supporting Information, CNCs are slightly wider than CNF with a width of \( \approx 10 \) nm. Yet these nanocrystals are several orders of magnitude shorter than the nanofibrils. We found that CNCs also enable to form nanocomposite with MXene \( \text{Ti}_3\text{C}_2\text{T}_x \) (Figure S7, Supporting Information), but the \( \text{Ti}_3\text{C}_2\text{T}_x/\text{CNC} \) nanocomposite films became brittle and shattered into pieces when CNC loading exceeded 20% (Figure S9, Supporting Information). XRD data of \( \text{Ti}_3\text{C}_2\text{T}_x/\text{CNC} \) show, almost, an unchanged position of (0002) peak and rapidly decreasing diffraction intensity with increasing CNC loading compared to that of the pristine MXene film (Figure 3b), suggesting that the CNC does not effectively incorporate into between adjacent MXene layers. This is consistent with the observation from AFM data of \( \text{Ti}_3\text{C}_2\text{T}_x/\text{CNC} \) films that show a lot of CNC clusters on the surface of films (Figure S10, Supporting Information). As expected, the \( \text{Ti}_3\text{C}_2\text{T}_x/\text{CNC} \) nanocomposites showed no significant enhancement in the mechanical properties, probably due to a combination of the short length of CNC with an aspect ratio of 10–30 and its inability to effectively entangle MXene flakes (Figure S11, Supporting Information).

These results suggest that the improvement in MXene/CNF mechanical strength originates from two factors. (1) Strong interfacial interactions, originating mostly from van der Waals interactions between the materials with possible contributions from hydrogen bonding between the hydroxyl and carboxylic terminations of CNFs and the hydroxyl termination groups of MXene.
MXene flake. (18,37) (2) The geometrical proportions between the 1D and 2D materials, where the CNF, which is thin, yet longer than the MXene flakes, serve as bridges to bind MXenes on the same plane and locks the adjacent layers. This bridging does not occur for shorter CNCs (Figure S11, Supporting Information).

We were able to further improve the mechanical strength of the fabricated Ti₃C₂Tₓ/CNF nanopapers by using a vacuum pressing process. After vacuum pressing, the nanopapers with 5% CNF, 10% CNF, 20% CNF, 40% CNF, and 60% CNF showed an enhanced tensile strength of 139.1, 181.3, 340.6, 416.1, 339.4, and 330.1 MPa, respectively (Figure 3c,d). These values are higher than the initial tensile strength of the Ti₃C₂Tₓ/CNF (Figure S5 and Table S1, Supporting Information) because vacuum-pressed nanopapers form stronger percolating CNF networks, and a more compact structure with stronger interactions between CNF and MXene. The XRD data of Ti₃C₂Tₓ/CNF (Figure S12, Supporting Information) shows a decrease in the d-spacing after vacuum-pressing, because some intercalated water between Ti₃C₂Tₓ/CNF dries out of the structure. We hypothesize that the use of other CNFs with more functional groups, such as TEMPO (2,2,6,6-tetramethyl 1-piperidinyloxy) modified CNF, could further enhance the structural properties of the Ti₃C₂Tₓ/CNF nanocomposites. (38)

The electrical conductivity of the Ti₃C₂Tₓ/CNF films can also be tailored by varying the CNF content. Pristine Ti₃C₂Tₓ films fabricated using vacuum filtration had a conductivity of 1.85 × 10⁵ S m⁻¹. Figure 3e. 5% CNF loading slightly decreased

**Figure 2.** Morphology characterization of Ti₃C₂Tₓ MXene/CNF nanopapers. a) Schematics for the fabrication of Ti₃C₂Tₓ/CNF nanopapers and their digital photographs. b) Photography of a Ti₃C₂Tₓ/CNF nanopaper folded into a shape of airplane to show the mechanical flexibility. c–h) AFM peak-force error images (top) and SEM cross-sectional images (bottom) of Ti₃C₂Tₓ MXene (c), Ti₃C₂Tₓ–10% CNF (d), Ti₃C₂Tₓ–20% CNF (e), Ti₃C₂Tₓ–40% CNF (f), Ti₃C₂Tₓ–80% CNF (g), and pristine CNF papers (h). For AFM images the corresponding height images are presented in Figure S3 of the Supporting Information.
the conductivity to $1.12 \times 10^5$ S m$^{-1}$, and after increasing the CNF loading to 20%, a high conductivity of $2.95 \times 10^4$ S m$^{-1}$ was still retained (Table 1). As shown in Figure S13 of the Supporting Information, Ti$_3$C$_2$T$_x$/CNF nanopapers have higher electronic conductivity than those reported for Ti$_3$C$_2$T$_x$/PVA composites at similar loading ratios of the organic materials.[17] We hypothesize that the higher conductivity is attributed to the thinner CNF dimension ($\approx 3.5$ nm), which may reduce the insulating gaps between Ti$_3$C$_2$T$_x$ flakes. Ashby plot (Figure 3f) of conductivities versus tensile strengths, highlights that our Ti$_3$C$_2$T$_x$/CNF nanopapers have better integration of mechanical properties and electronic conductivities, than other reported MXene-based structural composites. This improvement is required for high-performance structural energy storage.

To evaluate the electrochemical properties of the Ti$_3$C$_2$T$_x$/CNF nanocomposite and understand the effect of CNF contents on their charge storage properties, we conducted electrochemical measurements in a three-electrode setup using 3 m sulfuric acid as the electrolyte. Figure 4a schematically shows the three-electrode setup used in our experiment. Cyclic voltammogram (CV) curves of Ti$_3$C$_2$T$_x$ MXene and different Ti$_3$C$_2$T$_x$/CNF nanocomposite electrodes are shown in Figure 4b and Figure S14 (Supporting Information). The addition of CNF to Ti$_3$C$_2$T$_x$ MXene does not significantly lower the capacitance of the nanocomposites compared to the pristine MXene paper, and the vacuum pressing has negligible effect on the response and capacitances...
of the Ti3C2Tx/CNF nanopaper electrodes (Figure S15, Supporting Information). When the CNF loading is below 20 wt%, the composite electrodes showed mostly capacitive behavior with significant redox peaks associated with the protonation and change in the oxidation status of Ti atoms[49,50] as shown in Figure 4c for Ti3C2Tx–5% CNF electrode (and Figure S14 of the Supporting Information for other compositions). We measured gravimetric capacitances of 369, 324, and 298 F g⁻¹ for Ti3C2Tx/CNF nanocomposite electrodes with 5%, 10%, and 20% CNF loadings, respectively, at a scan rate of 2 mV s⁻¹ (Figure 4d). The capacitance decreased with CNF loading because CNF is electrochemically inactive in the potential window of Ti3C2Tx[18] Figure 4d,e, shows the normalized capacitances of different Ti3C2Tx/CNF nanocomposites. Figure S16 of the Supporting Information shows the gravimetric capacitances of Ti3C2Tx/CNF nanocomposites with a linear decrease by increasing CNF loading from 5 to 20 wt%. For example, the capacitances of the Ti3C2Tx–5% CNF (332 F g⁻¹) or Ti3C2Tx–20% CNF (285 F g⁻¹) is only ~5% or ~20% less than that of the pristine Ti3C2Tx film (348 F g⁻¹) at a scan rate of 5 mV s⁻¹. The inset of Figure 4d shows that the normalized gravimetric capacitances, calculated solely based on the weight of Ti3C2Tx using the composition ratios of the initial hybrid dispersions, remain almost the same for all electrodes, demonstrating the identical amount of Ti3C2Tx used in the preparation of the films. This suggests that the composition of the Ti3C2Tx/CNF nanocomposite films is close to the composition of the initial hybrid dispersions. This happens because the length of CNF and the lateral size of MXene particles are both larger than the pore size of PVDF membrane, and because the extremely large aspect ratio of CNFs (~300) promotes entanglement of the fibers,[10] which create a scaffold structure to support the MXene flakes.

The addition of only 5% CNF to Ti3C2Tx resulted in a negligible decrease (around 5%) in the gravimetric capacitance of the nanocomposite electrode with a very similar rate-capability to that of pristine MXene paper electrode. The mechanical strength of the Ti3C2Tx–5% CNF electrode, however, increased by 260% from 25.8 to 68.0 MPa. The increase in mechanical strength is more significant for Ti3C2Tx–20% CNF nanocomposite paper which showed an order of magnitude higher mechanical strength (~217 MPa) with only 20% lower gravimetric capacitance. The significant improvement in the mechanical strength of the fabricated paper electrodes thus outweighs the slight decrease in their electrochemical properties and renders them especially useful in applications where structural multifunctional electrodes with high mechanical robustness as well as high capacitances are required. The semilog plot of the gravimetric capacitance versus mechanical strength in Figure 4f, shows that the Ti3C2Tx/CNF nanopaper electrodes display the best integration of the capacitance and mechanical strength among some of the previously reported notable multifunctional supercapacitor electrodes such as MXene/rGO,[43] and MXene/PVA,[18] rGO/Aramid nanofiber,[11] rGO/cellulose,[12] rGO and rGO/PANI,[8] rGO/MnO2,[7] and CNT/PPy.[42] This indicates that in Ti3C2Tx/CNF composites, an optimized trade-off between mechanical properties and energy storage is obtained, where the mechanical strength is significantly improved compared to pristine Ti3C2Tx while the electrochemical properties of Ti3C2Tx MXene are mostly retained.

Figure 4g shows the electrochemical impedance spectroscopy results for Ti3C2Tx/CNF electrodes. As shown, addition of CNF resulted in a small increase in the ion transport resistance of the hybrid electrodes as evident from the Nyquist plots and in a good agreement with CV experiments. The electrodes, however, showed almost a vertical rise in the imaginary impedance at low frequencies indicating the facile accessibility of electrolyte’s ions to charge storage sites on the surface of titanium carbide layers. CNFs act as spacers in between MXene layers and improve the proton transport in the nanocomposite electrode.[25,43,44] We compared the capacitive response of the nanocomposite electrodes to that of pristine MXene electrodes by considering a power-law relationship between the peak current (iₚ) and scan rate (ν), iₚ = aνᵇ. In this equation “a” and “b” are variables, where b = 0.5 corresponds to diffusion-limited reactions and b = 1 corresponds to surface-controlled (capacitive) reactions. All Ti3C2Tx/CNF nanopaper electrodes displayed a similar capacitive response to that of the pristine MXene film (Figure 4h), with a highly capacitive charge storage properties up to scan rates of 200 mV s⁻¹. These results further show that the addition of CNF does not significantly impact the rate-capability and capacitive charge storage mechanism in Ti3C2Tx/CNF nanocomposite electrodes but does significantly

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Table 1. Physical properties of Ti3C2Tx, MXene, Ti3C2Tx/CNF, and CNF papers.

| Samples       | 2θ [°] | d-spacing [Å] | Conductivity [S m⁻¹] | Contact angle [°] | Tensile strength [MPa] | Strain at fracture [%] | Young’s modulus [GPa] |
|---------------|-------|---------------|----------------------|------------------|------------------------|------------------------|------------------------|
| MXene Ti3C2Tx | 6.90  | 12.80         | 1.85 × 10⁻¹          | 68               | 25.8 ± 1.9             | 0.10 ± 0.08            | 22.3 ± 5.9             |
| Ti3C2Tx–5% CNF| 6.14  | 14.38         | 1.12 × 10⁻¹          | 63               | 68.0 ± 4.8             | 0.16 ± 0.11            | 41.4 ± 0.2             |
| Ti3C2Tx–10% CNF| 5.99  | 14.74         | 6.90 × 10⁻⁴         | 57               | 153.6 ± 9.1            | 0.39 ± 0.04            | 41.9 ± 2.6             |
| Ti3C2Tx–20% CNF| 5.80  | 15.23         | 2.95 × 10⁻⁴         | 48               | 217.4 ± 7.7            | 0.57 ± 0.03            | 41.0 ± 1.8             |
| Ti3C2Tx–40% CNF| 5.28  | 16.73         | 1.96 × 10⁻³         | 35               | 269.6 ± 15.7           | 1.92 ± 0.38            | 26.6 ± 2.5             |
| Ti3C2Tx–60% CNF| –     | –             | 1.02 × 10⁻²         | 30               | 271.5 ± 17.7           | 2.18 ± 0.11            | 18.5 ± 1.8             |
| Ti3C2Tx–80% CNF| –     | –             | 1.17 × 10⁻²         | 24               | 52.5 ± 3.9             | 1.32 ± 0.83            | 6.58 ± 2.7             |
| Ti3C2Tx–5% CNC | 6.84  | 12.91         | 4.46 × 10⁻⁴         | 62               | 85.7 ± 16.4            | 0.87 ± 0.24            | 16.8 ± 1.9             |
| Ti3C2Tx–20% CNC| –     | –             | 2.63 × 10⁻⁴         | 32               | 34.1 ± 2.6             | 0.17 ± 0.09            | 20.2 ± 2.9             |
| CNF           | –     | –             | –                    | 22               | 145.7 ± 14.3           | 3.15 ± 0.33            | 8.25 ± 0.4             |
increase their mechanical strength. We also investigated the cyclic performance of the Ti$_3$C$_2$Tx paper electrodes in 3 m sulfuric acid electrolyte, as shown in Figure 4i where a Ti$_3$C$_2$T$_x$–5% CNF showed almost a 100% capacitance retention after 10 000 cycles at a charging/discharging rate of 10 A g$^{-1}$.

We fabricated micro-supercapacitors by employing a combined process of wax printing and vacuum-assisted filtration as shown in Figure 5a. The micro-supercapacitor structures (Figure 5b) had a resolution of ≈900 µm width and 1 cm length for interdigitated electrodes, and ≈130 µm gaps between the individual electrodes (Figure 5c), which is smaller than those previously reported for laser patterning method (450 µm gaps). Top-view SEM images of micro-supercapacitor exhibit some wrinkles resulting from the porous structure of the filter membrane, while the cross-sectional SEM image of the electrodes show stacked layers similar to Ti$_3$C$_2$T$_x$/CNF nanofibers (Figure 5d,e). We could control the thickness of the printed electrodes to around 1.7 µm for Ti$_3$C$_2$T$_x$–10% CNF electrodes and fabricate interdigitated electrodes with well-defined edges and good separation between the fingers. We
transferred the patterned electrodes onto flexible substrates using scotch tape. The successful fabrication of Ti$_3$C$_2$Tx/CNF-based micro-supercapacitors through our approach was attributed to the functionality of CNFs, which increase the contact angle and the patternability of the Ti$_3$C$_2$Tx/CNF dispersions and improves the mechanical properties of the Ti$_3$C$_2$Tx/CNF electrodes, for flexible printed electronics. Without using any metal current collectors, the Ti$_3$C$_2$Tx/CNF electrodes could act as both the current collector and active material in the fabricated micro-supercapacitors to store...
electrochemical energy. The symmetric Ti$_3$C$_2$T$_x$/CNF-based interdigitated micro-supercapacitors displayed ideal capacitive behaviors as shown in Figure S5h. The Ti$_3$C$_2$T$_x$-5% CNF-based micro-supercapacitor displayed a specific capacitance of 25.3 mF cm$^{-2}$ at a scan rate of 2 mV s$^{-1}$, similar to the value of 28.6 mF cm$^{-2}$ (2 mV s$^{-1}$) for pristine Ti$_3$C$_2$T$_x$ MXene-based micro-supercapacitor (Figure 5g). The micro-supercapacitors fabricated with CNF loadings of up to 10 wt% only showed a slightly lower capacitance (Figure 5g), at higher rates while maintaining a comparable performance with that of pristine Ti$_3$C$_2$T$_x$ MXene-based micro-supercapacitor. Among the different fabricated micro-supercapacitors, the Ti$_3$C$_2$T$_x$-10% CNF-based device showed the best rate performance (Figure 5f; Figures S17 and S18). In addition, the Ti$_3$C$_2$T$_x$/CNF-based micro-supercapacitors could deliver a high integrated output of energy and power densities, with an energy density of 0.08 µWh cm$^{-2}$ at the power density of 145 µW cm$^{-2}$ (Figure 5i). The fabricated Ti$_3$C$_2$T$_x$/CNF micro-supercapacitors also showed excellent life cycle. For example, the Ti$_3$C$_2$T$_x$-10% CNF micro-supercapacitor exhibited a capacitance retention of 86.8% after 10 000 cycles at a current density of 0.57 mA cm$^{-2}$ (Figure 5j). In addition, because of the enhanced mechanical properties of the Ti$_3$C$_2$T$_x$/CNF electrodes, the fabricated micro-supercapacitor devices could be bent and rolled without noticeable degradation in their charge storage performance as shown in Figure 5k.

The combination of these properties renders the Ti$_3$C$_2$T$_x$/CNF hybrid and their printed structures as promising electrode materials for flexible energy storage devices and printed electronics. For example, Ti$_3$C$_2$T$_x$/CNF hybrid can be used to fabricate paper-based microfluidic and electronic structures on a wax-defined paper, with a minimum feature size of 800 µm for conductive wires (Figures S19–S22, Supporting Information). This is attributed to the strong interaction between Ti$_3$C$_2$T$_x$ flakes and cellulose fibers from the hybrid dispersion and also from the paper-based substrates, as we showed in our AFM force measurements earlier.

In summary, we have demonstrated that a blend of aqueous dispersions of the 2D material, Ti$_3$C$_2$T$_x$ MXene, and the 1D material, cellulose nanofibrils CNFs can be used to rationally design and fabricate multifunctional nanocomposites with three notable properties. (1) A high mechanical strength of 340 MPa, more than an order of magnitude higher than the pristine MXene film with 29 MPa, originating from the attractive force between the two particles in water (which we analyzed with AFM) and further from the geometrical entanglement between them. (2) A high electronic conductivity of 2.95 × 10$^4$ S m$^{-1}$, probably resulting from the small width of the CNF (around 3.5 nm) allowing better conductivity between the 2D flakes. (3) A high gravimetric capacitance of ~298 F g$^{-1}$, since the addition of CNF does not limit the accessible sites for ion transport and pseudocapacitive charge storage. These combined metrics are higher than those previously reported for structural supercapacitor electrodes.[1,5–7,11,29,30] We further showed that the MXene/CNF dispersion can be easily printed into various shapes for applications in electronics and micro-supercapacitors with high power and energy densities. Further fundamental studies on systems combining 1D and 2D materials should address questions about the effect of geometry and surface properties on the self-assembly process that defines the properties of the nanocomposite. These systems can find more applications in areas such as structural energy storage,[1] printed electronics and paper-based diagnostics.[48]

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

Acknowledgements
W.T. and A.V.M. contributed equally to this work. M.M.H. acknowledges the European Research Council (Grant No. 715268). W.T., Z.W., and M.H.H. acknowledge the Wallenberg Wood Science Centre at KTH for funding through the Knut and Alice Wallenberg Foundation. Z.W. acknowledges the Wenner-Gren Foundation for funding. A.V.M. acknowledges the support from Alabama EPSCoR Graduate Research Scholar Program (GRSP Round 12 and 13). M.B. acknowledges the support from Auburn University’s Inramural Grants Program (IGP). The authors thank Invenntia (RISE Sweden) for providing nanocellulose. W.T. and M.H.H. led the project and developed the idea. W.T. prepared and characterized the MXene/nanocellulose composites, conducted the electrochemical tests of microsupercapacitors, and analyzed the data with supervision by M.M.H. A.V.M. prepared and characterized MXene dispersions with supervision by M.B. A.V.M. conducted the electrochemical tests in three-electrode system, and analyzed the data. L.O. contributed to experiment design, designed the patterning on Whatman paper, prepared the micro-supercapacitor devices, and analyzed the data. M.R. prepared and characterized the CNC, and conducted the AFM force measurements. Z.W. and J.E. prepared the CNF dispersion, T.P. assisted for the AFM measurement and analyzed the data. All authors contributed to writing the manuscript.

Conflict of Interest
The authors declare no conflict of interest.

Keywords
2D titanium carbide, MXenes, nanocellulose, nanocomposites, supercapacitors

Received: May 9, 2019
Revised: June 20, 2019
Published online: August 13, 2019

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