Scalable Manufacturing of Flexible and Highly Conductive $Ti_3C_2T_x$/PEDOT:PSS Thin Films for Electromagnetic Interference Shielding

Supplementary data

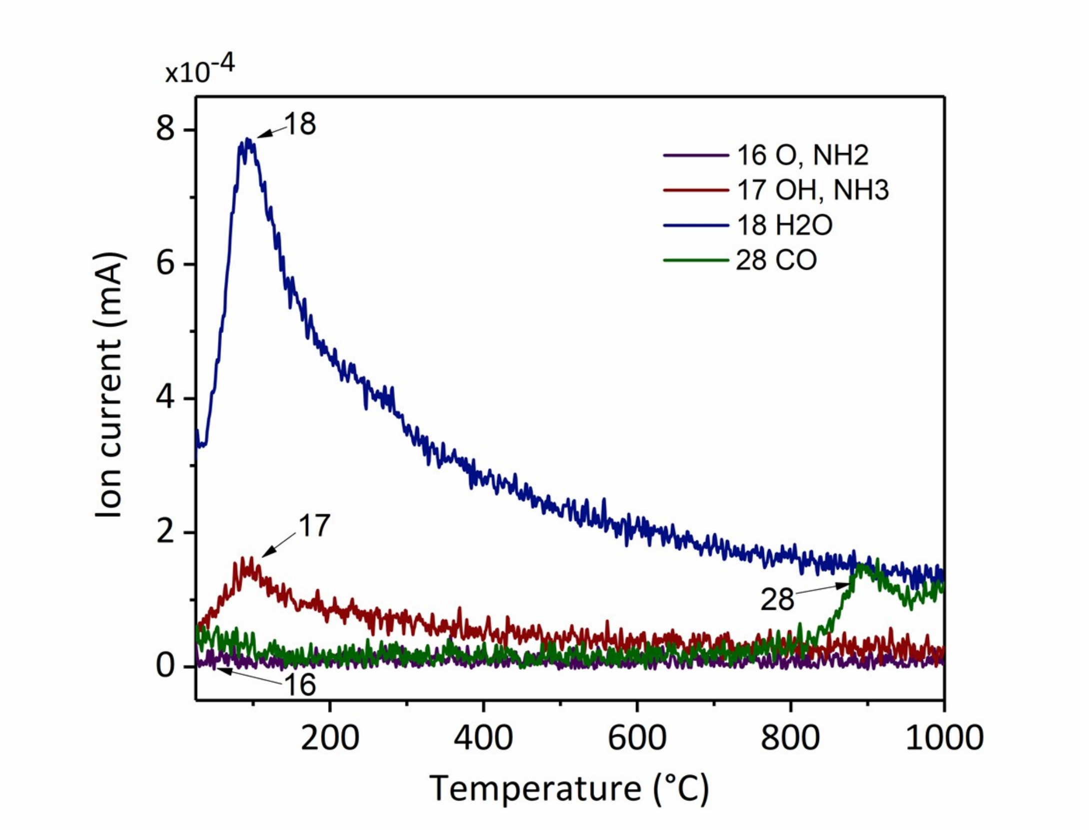

Figure S1. Sample mass spectrometry analysis of $Ti_3C_2T_x$.

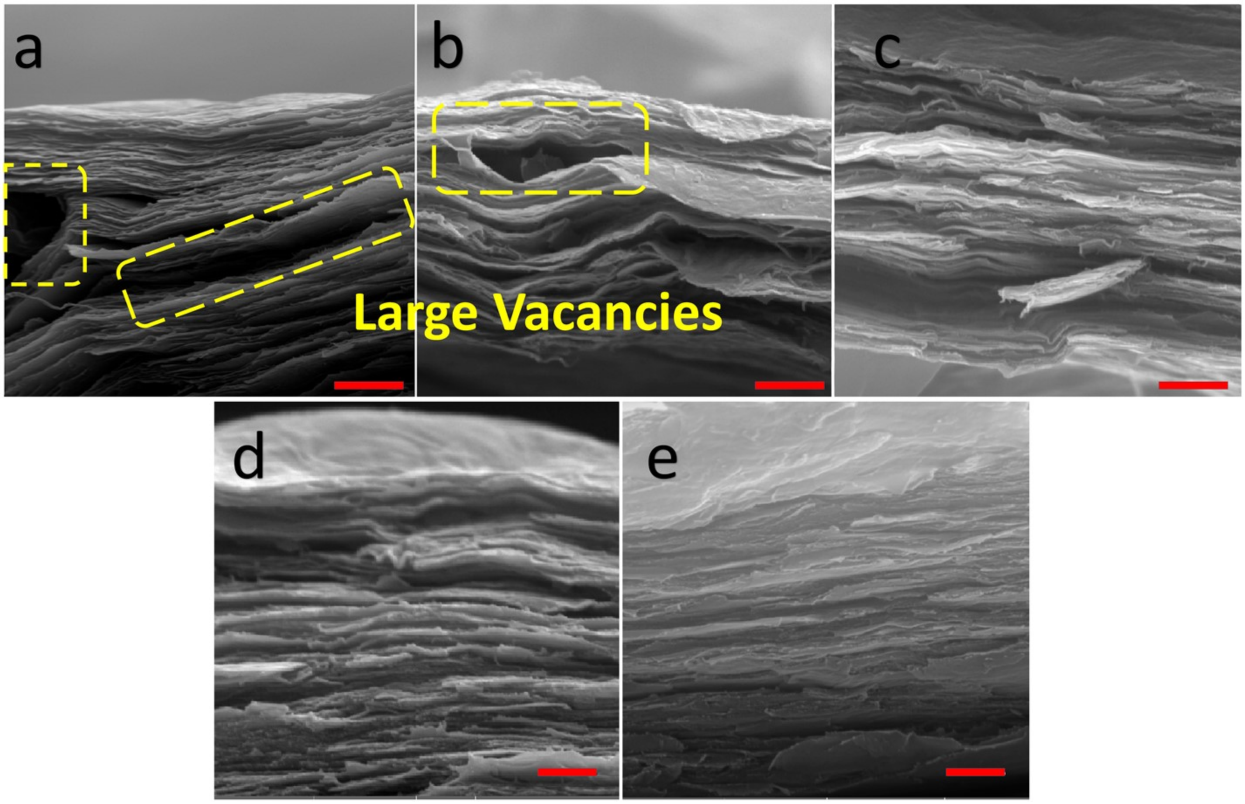

Figure S2. Cross-section SEM images of (a) the pristine $Ti_3C_2T_x$ and $Ti_3C_2T_x$/PEDOT:PSS composite films containing (b) 10, (c) 25, (d) 35, and (e) 50 wt% of the polymer (scale is 2 μm). The large vacancies shown in Figures S3a-b were observed everywhere in the dried film.

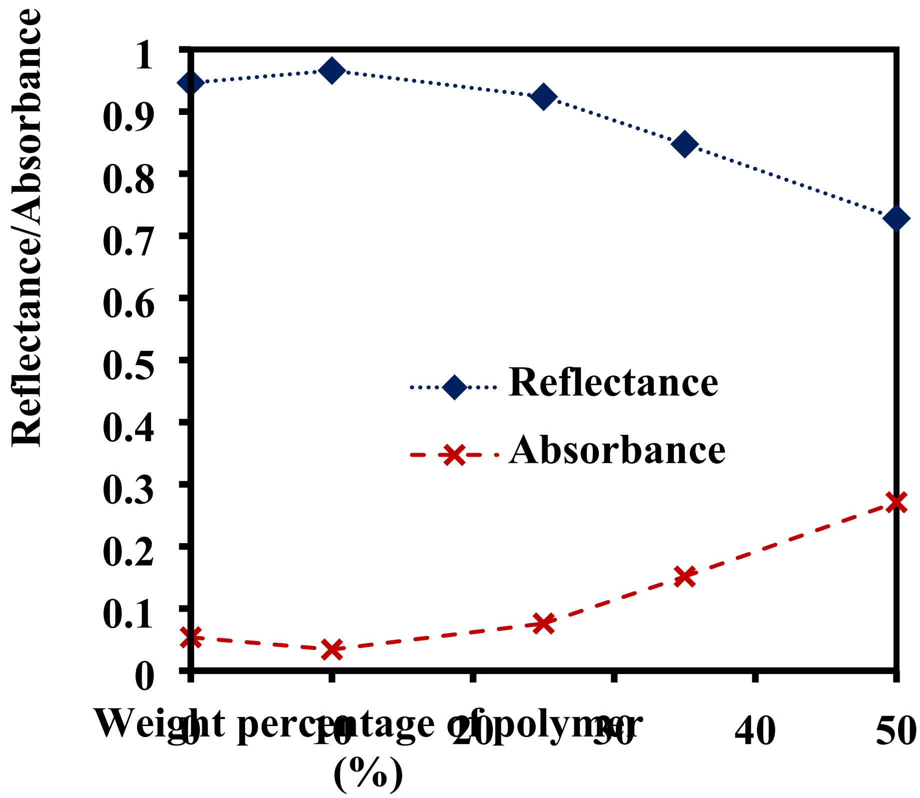

Figure S3. Reflectance and absorbance of pristine $Ti_3C_2T_x$ and the composite films that contained various concentrations of polymer.

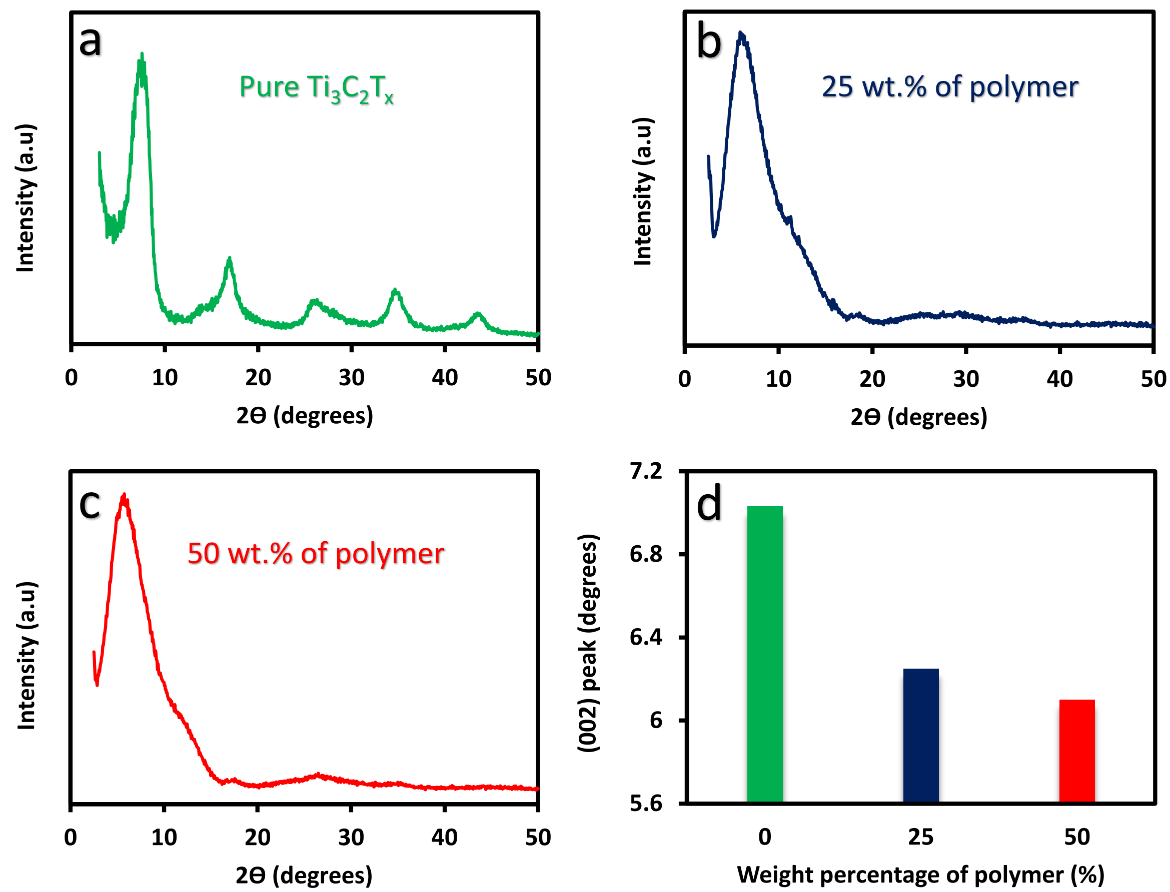

Figure S4. XRD patterns of (a) pure $Ti_3C_2T_x$ and the composite films that contained (b) 25 and (c) 50 wt.% of PEDOT:PSS. (d) Compression between the 002 peaks of the pristine $Ti_3C_2T_x$ and the composite films that contained various concentrations of polymer.

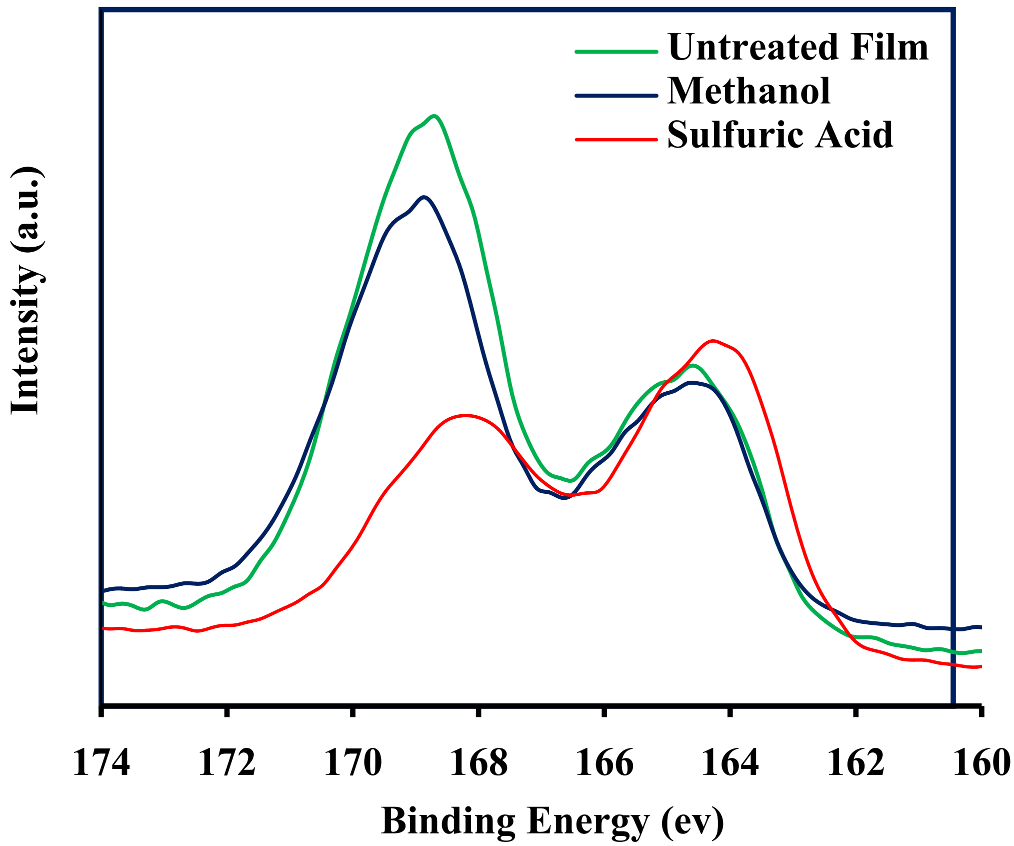

Figure S5. XPS results for the composite film containing 25 wt.% of PEDOT:PSS before and after post-treatment with sulfuric acid and methanol.

Table S1. Previous studies that used drop-casting or inkjet printing approach for PEDOT:PSS and its composites.

Material	Substrate	Method	Reference
Cu-$Bi_{0.5}Sb_{1.5}Te_3$ /PEDOT:PSS	Silicon dioxide (Activated by plasma)	Drop-casting	1
PEDOT:PSS	Glass (Activated by UV/ozone)	Drop-casting	2
PEDOT:PSS/ethanol, and Graphene dots (GDs)/ PEDOT:PSS/ethanol	Fluorine-doped SnO_2	Drop-casting	3
SnSe/PEDOT:PSS/ dimethyl sulfoxide (DMSO)	Glass	Drop-casting	4
PEDOT:PSS coated Te nanorod/ PEDOT:PSS	Glass	Drop-casting	5
Bi_2Te_3 nanosheet/PEDOT: PSS	Glass	Drop-casting	6
PEDOT: PSS	Silicon	Drop-casting	7
PEDOT: PSS	Glass wafer	Drop-casting	8
PEDOT: PSS	Silicon dioxide (Activated by plasma)	Drop-casting	9
PEDOT: PSS	Glass	Drop-casting	10
DMSO/PEDOT:PSS	Polydimethylsiloxane (PDMS)	Drop-casting	11
PEDOT:PSS)/waterborne polyurethane (WPU)/ Carbon nanocoils (CNCs)	Glass or elastomeric substrates (Activated by plasma)	Drop-casting	12
PEDOT:PSS/ single-walled carbon nanotubes (SWCNTs)	Polyimide	Drop-casting	13
DMSO/PEDOT:PSS and ethylene glycol (EG)/ PEDOT:PSS	Polypropylene, glass, and poly(ethyleneterephthalate) (PET)	Drop-casting	14
PEDOT:PSS/SWCNTs/ DMSO	Glass	Drop-casting	15
Bi_2Te_3 nanosheet/PEDOT: PSS	Glass	Drop-casting	16
Cellulose/PEDOT:PSS/DMSO/glycerol	Glass	Drop-casting	17
PEDOT: PSS	Glass	Drop-casting	18
PEDOT: PSS/DMSO	Glass (Activated by aqua regia)	Drop-casting	19
Graphene oxide (GO)/PEDOT:PSS/DMSO	Glass (Activated with a mixture of concentrated sulfuric acid and hydrogen peroxide)	Drop-casting	20
PEDOT:PSS/ CNTs	Polyimide	Drop-casting	21
PEDOT: PSS/DMSO	Glass	Drop-casting	22
PEDOT: PSS	Glass	Drop-casting	23
Ag_2Se NW/PEDOT:PSS	Glass	Drop-casting	24
PEDOT:PSS/ GDs	Fluorine-doped SnO_2	Drop-casting	25
PEDOT-PSS/SWCNTs	PET	Inkjet printing	26
PEDOT-PSS/vegetal glycerol	Polyimide	Inkjet printing	27
PEDOT:PSS/DMSO	Pre-treated polyethylene naphthalate	Inkjet printing	28
PEDOT:PSS	Polyimide (Activated by plasma)	Inkjet printing	29
PEDOT:PSS	PET	Inkjet printing	30
PEDOT-PSS/ ethanol/ diethylene glycol	Polyimide (Activated by plasma)	Inkjet printing	31

Table S2. Average static, advancing, and receding contact angles of water on the polycarbonate sheet.

	Average static contact angle	Average advancing contact angle	Average receding contact angle
Before treatment	81.6	89.2	69.6
After treatment for 30 min	24.2	28.6	19.9

Table S3. Average static, advancing, and receding contact angles of the aqueous solution of $Ti_3C_2T_x$ or $Ti_3C_2T_x$/PEDOT:PSS (25 wt. %) on activated polycarbonate sheet.

	Average static contact angle	Average advancing contact angle	Average receding contact angle
$Ti_3C_2T_x$	17.3	21.1	14.2
$Ti_3C_2T_x$ /PEDOT:PSS (25 wt.%)	15.9	19.3	0

Table S4. Previous studies of thickness, conductivity, SE_T, and specific EMI shielding effectiveness of various nanocomposites.

Material	Thickness (mm)	Conductivity (σ cm^{-1})	SE_T (dB)	SE_T/Density/thickness (dB cm^2 g^{-1})	Reference
$Ti_3C_2T_x$/PEDOT:PSS	0.011	340.5	42.1	19498	32
Ti_2CT_x/Polyvinyl alcohol (PVA)	0.1	2.0×10^{-4}	26	5136	33
$Ti_3C_2T_x$/Cellulose nanofiber (CNF)	0.035	1.43	39.6	7029	34
$Ti_3C_2T_x$/Silver nanowires (AgNW)/ Nanocellulose (NC)	0.0169	300	42.74	16724	35
$Ti_3C_2T_x$/Aramid nanofibers (ANF)	0.012	-	34.71	21971	36
$Ti_3C_2T_x$/PVA	0.027	7.16	44.4	9343	37
Polystyrene/Graphene	2.5	1.25	29	322	38
Poly(3-hydrobutyrate-co-3-hydroxyvalerate) (PHBV)/ AgNW	0.018	-	45.9	19678	39
$Ti_3C_2T_x$/Polyacrylonitrile (PAN)/TiO_2	0.045	92.68	32	4085	40
$Ti_3C_2T_x$/rGO/Epoxy	2	6.95	56.4	9400	41
Graphene/PEDOT:PSS	0.8	6.84	70	841	42
V_2O_5/Polyaniline (PANI)	6	0.016	34.7	2770.3	43
$Ti_3C_2T_x$/AgNW/CNF	0.035	373.78	59.7	10647	44
$Ti_3C_2T_x$/Xanthan	0.00684	115.29	34.1	14,490	45
Poly vinylidene fluoride (PVDF)/MWCNT	0.9	0.074	32	259	46
Graphene/ PMMA	0.24	3.11	19	1042	47

$Ti_3C_2T_x$/PANI	0.376	3.25	35.3	1700	[48]
Poly(oxymethylene) (POM)/ MWCNT	2	3.33	58.6	344.4	[49]
$Ti_3C_2T_x$/CNF	0.047	7.39	24	2647	[50]
MWCNT/Polycarbonate	2.16	3	39	164	[51]
$Ti_3C_2T_x$/PEDOT:PSS	0.007	2900 ± 400	55.42	38079	This work

Table S5. Compositions of the $Ti_3C_2T_x$ MXene and $Ti_3C_2T_x$ MXene/PEDOT:PSS solutions used for drop-casting. One mL of each of the solutions was used for covering 9 cm^2 of polycarbonate sheet.

Sample name	PEDOT:PSS content (mg)	$Ti_3C_2T_x$ MXene content (mg)	Water content (mL)
Pure $Ti_3C_2T_x$	-	15	1
10 wt.%	1.5	13.5	
25 wt.%	3.75	11.25	
35 wt.%	5.25	9.75	
50 wt.%	7.5	7.5	

Equations

1. The EMI reflection loss is calculated using:

$$SE_R = 168 + 10log\left(\frac{\sigma}{\mu f}\right) = 10\ log\left(\frac{1}{1-R}\right) \quad (S1)$$

where σ is the conductivity, μ is the magnetic permeability of the shield relative to air, and f is the frequency in Hz and R is the reflectance.

2. The EMI absorption loss:

$$SE_A = 8.68t(\sqrt{\frac{\sigma\omega\mu}{2}}) = 8.68\frac{t}{\delta} = 10\ log\ (\frac{1-R}{T}) \quad (S2)$$

where t is the thickness of the shield, ω is the angular frequency in rad/s, δ is the skin depth of the shield, and T is the transmittance.

3. Reflectance (R):

$$R = {S_{11}}^2 \quad (S3)$$

where S_{11} is the reflected voltage magnitude divided by the incident voltage magnitude in port 1.

4. Transmittance (T):

$$T = {S_{21}}^2 \quad (S4)$$

where S_{21} is the transmitted voltage magnitude from port 1 to port 2 divided by the incident voltage magnitude in port 1.

5. Absorbance (A):

$$A = 1 - R - T \quad (S5)$$

References

1. Y. Wang, M. Hong, W.-D. Liu, X.-L. Shi, S.-D. Xu, Q. Sun, H. Gao, S. Lu, J. Zou and Z.-G. Chen, *Chem. Eng. J.*, 2020, **397**, 125360.
2. T. A. Yemata, A. K. K. Kyaw, Y. Zheng, X. Wang, Q. Zhu, W. S. Chin and J. Xu, *Polym. Int.*, 2020, **69**, 84-92.
3. C.-P. Lee, K.-Y. Lai, C.-A. Lin, C.-T. Li, K.-C. Ho, C.-I. Wu, S.-P. Lau and J.-H. He, *Nano Energy*, 2017, **36**, 260-267.
4. H. Ju and J. Kim, *Chem. Eng. J.*, 2016, **297**, 66-73.
5. Q. Meng, Q. Jiang, K. Cai and L. Chen, *Org. Electron.*, 2019, **64**, 79-85.
6. Y. Du, K. Cai, S. Chen, P. Cizek and T. Lin, *ACS Appl. Mater. Interfaces.*, 2014, **6**, 5735-5743.
7. Y. Honma, K. Itoh, H. Masunaga, A. Fujiwara, T. Nishizaki, S. Iguchi and T. Sasaki, *Adv. Electron. Mater.*, 2018, **4**, 1700490.
8. P. Stadler, D. Farka, H. Coskun, E. D. Głowacki, C. Yumusak, L. M. Uiberlacker, S. Hild, L. N. Leonat, M. C. Scharber and P. Klapetek, *J. Mater. Chem. C.*, 2016, **4**, 6982-6987.
9. S. Xu, M. Hong, X.-L. Shi, Y. Wang, L. Ge, Y. Bai, L. Wang, M. Dargusch, J. Zou and Z.-G. Chen, *Chem. Mater.*, 2019, **31**, 5238-5244.
10. N. Massonnet, A. Carella, O. Jaudouin, P. Rannou, G. Laval, C. Celle and J.-P. Simonato, *J. Mater. Chem. C.*, 2014, **2**, 1278-1283.
11. J. Liu, X. Wang, D. Li, N. E. Coates, R. A. Segalman and D. G. Cahill, *Macromolecules*, 2015, **48**, 585-591.
12. S. Xu, Z. Fan, S. Yang, Y. Zhao and L. Pan, *Chem. Eng. J.*, 2021, **404**, 126064.
13. S.-H. Chung, D. H. Kim, H. Kim, H. Kim and S. W. Jeong, *Mater. Today Commun.*, 2020, **23**, 100867.
14. C. Liu, B. Lu, J. Yan, J. Xu, R. Yue, Z. Zhu, S. Zhou, X. Hu, Z. Zhang and P. Chen, *Synth. Met.*, 2010, **160**, 2481-2485.
15. S. Liu, H. Li and C. He, *Carbon*, 2019, **149**, 25-32.
16. H. Song, C. Liu, H. Zhu, F. Kong, B. Lu, J. Xu, J. Wang and F. Zhao, *Journal of electronic materials*, 2013, **42**, 1268-1274.
17. D. Belaineh, J. W. Andreasen, J. Palisaitis, A. Malti, K. Håkansson, L. Wagberg, X. Crispin, I. Engquist and M. Berggren, *ACS Applied Polymer Materials*, 2019, **1**, 2342-2351.
18. T. A. Yemata, Y. Zheng, A. K. K. Kyaw, X. Wang, J. Song, W. S. Chin and J. Xu, *Org. Electron.*, 2020, **81**, 105682.
19. Q. Zhu, E. Yildirim, X. Wang, X. Y. D. Soo, Y. Zheng, T. L. Tan, G. Wu, S.-W. Yang and J. Xu, *Frontiers in chemistry*, 2019, **7**, 783.
20. F. Li, K. Cai, S. Shen and S. Chen, *Synth. Met.*, 2014, **197**, 58-61.
21. W. Lee, Y. H. Kang, J. Y. Lee, K.-S. Jang and S. Y. Cho, *RSC Adv.*, 2016, **6**, 53339-53344.
22. L. Ouyang, C. Musumeci, M. J. Jafari, T. Ederth and O. Inganäs, *ACS Appl. Mater. Interfaces.*, 2015, **7**, 19764-19773.
23. S. Zhang and F. Cicoira, *Adv. Mater.*, 2017, **29**, 1703098.
24. D. Park, M. Kim and J. Kim, *Polymers*, 2021, **13**, 210.
25. C.-P. Lee, C.-A. Lin, T.-C. Wei, M.-L. Tsai, Y. Meng, C.-T. Li, K.-C. Ho, C.-I. Wu, S.-P. Lau and J.-H. He, *Nano Energy*, 2015, **18**, 109-117.
26. A. Denneulin, J. Bras, F. Carcone, C. Neuman and A. Blayo, *Carbon*, 2011, **49**, 2603-2614.
27. L. D. Garma, L. M. Ferrari, P. Scognamiglio, F. Greco and F. Santoro, *Lab on a Chip*, 2019, **19**, 3776-3786.

28. C. Bali, A. Brandlmaier, A. Ganster, O. Raab, J. Zapf and A. Hübler, *Materials Today: Proceedings*, 2016, **3**, 739-745.
29. M. Borghetti, M. Serpelloni, E. Sardini and S. Pandini, *Sensors and Actuators A: Physical*, 2016, **243**, 71-80.
30. A. Rivadeneyra, M. Bobinger, A. Albrecht, M. Becherer, P. Lugli, A. Falco and J. F. Salmerón, *Polymers*, 2019, **11**, 824.
31. M. Borghetti, M. Ghittorelli, E. Sardini, M. Serpelloni and F. Torricelli, *IEEE Transactions on Instrumentation and measurement*, 2016, **65**, 2137-2144.
32. R. Liu, M. Miao, Y. Li, J. Zhang, S. Cao and X. Feng, *ACS Appl. Mater. Interfaces.*, 2018, **10**, 44787-44795.
33. H. Xu, X. Yin, X. Li, M. Li, S. Liang, L. Zhang and L. Cheng, *ACS Appl. Mater. Interfaces.*, 2019, **11**, 10198-10207.
34. B. Zhou, Z. Zhang, Y. Li, G. Han, Y. Feng, B. Wang, D. Zhang, J. Ma and C. Liu, *ACS Appl. Mater. Interfaces.*, 2020, **12**, 4895-4905.
35. M. Miao, R. Liu, S. Thaiboonrod, L. Shi, S. Cao, J. Zhang, J. Fang and X. Feng, *J. Mater. Chem. C.*, 2020, **8**, 3120-3126.
36. H. Wei, M. Wang, W. Zheng, Z. Jiang and Y. Huang, *Ceram. Int.*, 2020, **46**, 6199-6204.
37. X. Jin, J. Wang, L. Dai, X. Liu, L. Li, Y. Yang, Y. Cao, W. Wang, H. Wu and S. Guo, *Chem. Eng. J.*, 2020, **380**, 122475.
38. D.-X. Yan, P.-G. Ren, H. Pang, Q. Fu, M.-B. Yang and Z.-M. Li, *J. Mater. Chem.*, 2012, **22**, 18772-18774.
39. S. Yang, Y.-Y. Wang, Y. Song, L.-C. Jia, G.-J. Zhong, L. Xu, D.-X. Yan, J. Lei and Z.-M. Li, *J. Mater. Chem. C.*, 2021.
40. Y. Wang, H.-K. Peng, T.-T. Li, B.-C. Shiu, H.-T. Ren, X. Zhang, C.-W. Lou and J.-H. Lin, *Chem. Eng. J.*, 2021, 128681.
41. S. Zhao, H.-B. Zhang, J.-Q. Luo, Q.-W. Wang, B. Xu, S. Hong and Z.-Z. Yu, *ACS nano*, 2018, **12**, 11193-11202.
42. N. Agnihotri, K. Chakrabarti and A. De, *RSC Adv.*, 2015, **5**, 43765-43771.
43. A. P. Narayanan, K. N. Unni and K. P. Surendran, *Chem. Eng. J.*, 2021, **408**, 127239.
44. B. Zhou, Q. Li, P. Xu, Y. Feng, J. Ma, C. Liu and C. Shen, *Nanoscale*, 2021, **13**, 2378-2388.
45. Y. Sun, R. Ding, S. Y. Hong, J. Lee, Y.-K. Seo, J.-D. Nam and J. Suhr, *Chem. Eng. J.*, 2021, **410**, 128348.
46. B. Bhaskara Rao, N. Kale, B. Kothavale and S. Kale, *AIP Advances*, 2016, **6**, 065107.
47. H.-B. Zhang, Q. Yan, W.-G. Zheng, Z. He and Z.-Z. Yu, *ACS Appl. Mater. Interfaces.*, 2011, **3**, 918-924.
48. G. Yin, Y. Wang, W. Wang, Z. Qu and D. Yu, *Adv. Mater. Interfaces.*, 2021, 2001893.
49. J. Li, J.-L. Chen, X.-H. Tang, J.-H. Cai, J.-H. Liu and M. Wang, *J. Colloid Interface Sci.*, 2020, **565**, 536-545.
50. W.-T. Cao, F.-F. Chen, Y.-J. Zhu, Y.-G. Zhang, Y.-Y. Jiang, M.-G. Ma and F. Chen, *Acs Nano*, 2018, **12**, 4583-4593.
51. S. Pande, A. Chaudhary, D. Patel, B. P. Singh and R. B. Mathur, *RSC Adv.*, 2014, **4**, 13839-13849.