

## Documents

Nugraha, M.W.<sup>a</sup>, Wirzal, M.D.H.<sup>a</sup>, Ali, F.<sup>b</sup>, Roza, L.<sup>c</sup>, Sambudi, N.S.<sup>a d</sup>

**Electrospun polylactic acid/ tungsten oxide/ amino-functionalized carbon quantum dots (PLA/WO<sub>3</sub>/N-CQDs) fibers for oil/water separation and photocatalytic decolorization**

(2021) *Journal of Environmental Chemical Engineering*, 9 (5), art. no. 106033, .

DOI: 10.1016/j.jece.2021.106033

<sup>a</sup> Department of Chemical Engineering, Universiti Teknologi PETRONAS, Seri Iskandar, Perak 32610, Malaysia

<sup>b</sup> Department of Biotechnology Engineering, Faculty of Engineering, International Islamic University Malaysia, Kuala Lumpur, 53100, Malaysia

<sup>c</sup> Program Studi Pendidikan Fisika, Fakultas Keguruan dan Ilmu Pendidikan, Universitas Muhammadiyah Prof. Dr. Hamk, Jakarta Timur, Indonesia

<sup>d</sup> Centre of Urban Resource Sustainability (CUREs), Universiti Teknologi PETRONAS, Seri Iskandar, Perak 32610, Malaysia

#### Abstract

Oily wastewater generated from various industrial processes and oil spillages is a massive threat to the environment and human health. Hence, it is essential to develop novel material for practical oil/water separation and recovery. By incorporating WO<sub>3</sub> (tungsten oxide)/ N-CQDs (amino-functionalized carbon quantum dots) into the matrix of polymer, the alteration of wettability behavior of electrospun PLA fiber is reported in this present work. The modified PLA/WO<sub>3</sub>/N-CQDs fibers shows a significantly enhanced hydrophobicity of the fibers while maintaining surface super-oleophilicity. These behaviors are achieved by modifying the surface contact angle and surface morphology of the composite PLA fiber using WO<sub>3</sub>/N-CQDs as filler. The PLA/WO<sub>3</sub>/N-CQDs EDA fiber exhibits the highest water contact angle (WCA) at 132.37°, oil absorption capacity at 35.752 g/g for n-hexane, oil separation performance at 8,326.048 L m<sup>-2</sup> h<sup>-1</sup> for n-hexane and 11,961.364 L m<sup>-2</sup> h<sup>-1</sup> for n-heptane, and decolorization of methylene blue (MB) at 91.80%. Additionally, the PLA/WO<sub>3</sub>/N-CQDs EDA fiber demonstrated an excellent separation performance and durability after ten times cyclic separation performance test. This present study contributes to a potential application in the field of oily wastewater treatment. © 2021 Elsevier Ltd

#### Author Keywords

Adsorption; Contact angle; Fiber preparation; Oil/water separation; Porous fiber

#### Index Keywords

Adsorption, Aromatic compounds, Contact angle, Durability, Fibers, Health risks, Hexane, Morphology, Nanocrystals, Semiconductor quantum dots, Surface morphology, Tungsten compounds, Wastewater treatment, Water absorption; Carbon quantum dots, Electrospuns, Fiber preparation, Functionalized, N-Hexane, Oil/water separation, Photocatalytic decolorization, Porous fibers, Separation performance, Tungsten oxide; Separation

#### References

- Ismail, N.H., Salleh, W.N.W., Ismail, A.F., Hasbullah, H., Yusof, N., Aziz, F., Jaafar, J. **Hydrophilic polymer-based membrane for oily wastewater treatment: a review** (2020) *Sep. Purif. Technol.*, 233.
- Abuhasel, K., Jeng, Y.T., Munusamy, Y., Kchaou, M., Alquraish, M. **Latex-based membrane for oily wastewater filtration: study on the sulfur concentration effect** (2021) *Appl. Sci.*, 11, p. 1779.
- Monash, P., Pugazhenth, G. **Effect of TiO<sub>2</sub> addition on the fabrication of ceramic membrane supports: a study on the separation of oil droplets and bovine serum albumin (BSA) from its solution** (2011) *Desalination*, 279, pp. 104-114.

- Gore, P.M., Gawali, P., Naebe, M., Wang, X., Kandasubramanian, B.  
**Polycarbonate and activated charcoal-engineered electrospun nanofibers for selective recovery of oil/solvent from oily wastewater**  
(2020) *SN Appl. Sci.*, 2, pp. 1-13.
- Farrington, J.W.  
**Oil pollution in the marine environment i: inputs, big spills, small spills, and dribbles**  
(2013) *Environment*, 55, pp. 3-13.
- Jafarinejad, S.  
**Recent developments in the application of sequencing batch reactor (SBR) technology for the petroleum industry wastewater treatment**  
(2017) *Chem. Int.*,
- Sun, H., Xu, Z., Gao, C.  
**Multifunctional, ultra-flyweight, synergistically assembled carbon aerogels**  
(2013) *Adv. Mater.*,
- Zhang, L., Wu, J., Wang, Y., Long, Y., Zhao, N., Xu, J.  
**Combination of bioinspiration: a general route to superhydrophobic particles**  
(2012) *J. Am. Chem. Soc.*,
- Chen, Y., Wang, N., Guo, F., Hou, L., Liu, J., Liu, J., Xu, Y., Co<sub>3</sub>O<sub>4</sub>, A.  
**nano-needle mesh for highly efficient, high-flux emulsion separation**  
(2016) *J. Mater. Chem. A.*,
- Zhu, H., Chen, D., An, W., Li, N., Xu, Q., Li, H., He, J., Lu, J.  
**A robust and cost-effective superhydrophobic graphene foam for efficient oil and organic solvent recovery**  
(2015) *Small*, 11, pp. 5222-5229.
- Xue, Z., Sun, Z., Cao, Y., Chen, Y., Tao, L., Li, K., Feng, L., Wei, Y.  
**Superoleophilic and superhydrophobic biodegradable material with porous structures for oil absorption and oil-water separation**  
(2013) *RSC Adv.*, 3, p. 23432.
- Gu, J., Xiao, P., Chen, P., Zhang, L., Wang, H., Dai, L., Song, L., Chen, T.  
**Functionalization of biodegradable PLA nonwoven fabric as superoleophilic and superhydrophobic material for efficient oil absorption and oil/water separation**  
(2017) *ACS Appl. Mater. Interfaces*, 9, pp. 5968-5973.
- Shi, J., Zhang, L., Xiao, P., Huang, Y., Chen, P., Wang, X., Gu, J., Chen, T.  
**Biodegradable PLA nonwoven fabric with controllable wettability for efficient water purification and photocatalysis degradation**  
(2018) *ACS Sustain. Chem. Eng.*, 6, pp. 2445-2452.
- Ma, L., He, J., Wang, J., Zhou, Y., Zhao, Y., Li, Y., Liu, X., Qu, M.  
**Functionalized superwetable fabric with switchable wettability for efficient oily wastewater purification, in situ chemical reaction system separation, and photocatalysis degradation**  
(2019) *ACS Appl. Mater. Interfaces*, 11, pp. 43751-43765.
- Ye, B., Jia, C., Li, Z., Li, L., Zhao, Q., Wang, J., Wu, H.  
**Solution-blow spun PLA/SiO<sub>2</sub> nanofiber membranes toward high efficiency oil/water separation**

- (2020) *J. Appl. Polym. Sci.*, 137, p. 49103.
- Di Camillo, D., Ruggieri, F., Santucci, S., Lozzi, L.  
**N-doped TiO<sub>2</sub> nanofibers deposited by electrospinning**  
(2012) *J. Phys. Chem. C.*, 116, pp. 18427-18431.
  - Zhu, J., Zhang, S., Wang, L., Jia, D., Xu, M., Zhao, Z., Qiu, J., Jia, L.  
**Engineering cross-linking by coal-based graphene quantum dots toward tough, flexible, and hydrophobic electrospun carbon nanofiber fabrics**  
(2018) *Carbon N. Y.*, 129, pp. 54-62.
  - Bibi, R., Wei, L., Shen, Q., Tian, W., Oderinde, O., Li, N., Zhou, J.  
**Effect of amino functionality on the uptake of cationic dye by titanium-based metal organic frameworks**  
(2017) *J. Chem. Eng. Data*, 62, pp. 1615-1622.
  - Wang, S., Li, X., Liu, Y., Zhang, C., Tan, X., Zeng, G., Song, B., Jiang, L.  
**Nitrogen-containing amino compounds functionalized graphene oxide: Synthesis, characterization and application for the removal of pollutants from wastewater: a review**  
(2018) *J. Hazard. Mater.*, 342, pp. 177-191.
  - (Eric) Hu, X., Liu, L., Luo, X., Xiao, G., Shiko, E., Zhang, R., Fan, X., Li, C.  
**A review of N-functionalized solid adsorbents for post-combustion CO<sub>2</sub> capture**  
(2020) *Appl. Energy*, 260.
  - Lei, S., Zeng, M., Huang, D., Wang, L., Zhang, L., Xi, B., Ma, W., Cheng, Z.  
**Synergistic high-flux oil-saltwater separation and membrane desalination with carbon quantum dots functionalized membrane**  
(2019) *ACS Sustain. Chem. Eng.*, 7, pp. 13708-13716.
  - Liu, L., Chen, H., Shiko, E., Fan, X., Zhou, Y., Zhang, G., Luo, X., (Eric) Hu, X.  
**Low-cost DETA impregnation of acid-activated sepiolite for CO<sub>2</sub> capture**  
(2018) *Chem. Eng. J.*, 353, pp. 940-948.
  - Xing, H.T., Chen, J.H., Sun, X., Huang, Y.H., Su, Z.B., Hu, S.R., Weng, W., Huang, Y.  
**NH<sub>2</sub>-rich polymer/graphene oxide use as a novel adsorbent for removal of Cu(II) from aqueous solution**  
(2015) *Chem. Eng. J.*, 263, pp. 280-289.
  - Fan, L., Luo, C., Li, X., Lu, F., Qiu, H., Sun, M.  
**Fabrication of novel magnetic chitosan grafted with graphene oxide to enhance adsorption properties for methyl blue**  
(2012) *J. Hazard. Mater.*, 215-216, pp. 272-279.
  - Song, B., Wang, T., Sun, H., Shao, Q., Zhao, J., Song, K., Hao, L., Guo, Z.  
**Two-step hydrothermally synthesized carbon nanodots/WO<sub>3</sub> photocatalysts with enhanced photocatalytic performance**  
(2017) *Dalton Trans.*, 46, pp. 15769-15777.
  - Jamila, G.S., Sajjad, S., Leghari, S.A.K., Long, M.  
**Nitrogen doped carbon quantum dots and GO modified WO<sub>3</sub> nanosheets combination as an effective visible photo catalyst**

- (2020) *J. Hazard. Mater.*, 382.
- Zhang, J., Zhang, X., Dong, S., Zhou, X., Dong, S.  
**N-doped carbon quantum dots/TiO<sub>2</sub> hybrid composites with enhanced visible light driven photocatalytic activity toward dye wastewater degradation and mechanism insight**  
(2016) *J. Photochem. Photobiol. A Chem.*, 325, pp. 104-110.
  - Ahmadi, M., Guinel, M.J.F.  
**Synthesis and characterization of tungstite (WO<sub>3</sub>·H<sub>2</sub>O) nanoleaves and nanoribbons**  
(2014) *Acta Mater.*, 69, pp. 203-209.
  - Liu, X.J., Guo, M.L., Huang, J., Yin, X.Y.  
**Improved fluorescence of carbon dots prepared from bagasse under alkaline hydrothermal conditions**  
(2013) *BioResources*, 8, pp. 2537-2546.
  - Wang, Y., Li, X., Dai, X., Zhan, Y., Ding, X., Wang, M., Wang, X.  
**Anchoring ZIF-67 particles on amidoximerized polyacrylonitrile fibers for radionuclide sequestration in wastewater and seawater**  
(2020) *J. Hazard Mater.*, 395.
  - Aseri, N.S., Lau, W.J., Goh, P.S., Hasbullah, H., Othman, N.H., Ismail, A.F.  
**Preparation and characterization of polylactic acid-modified polyvinylidene fluoride hollow fiber membranes with enhanced water flux and antifouling resistance**  
(2019) *J. Water Process Eng.*, 32.
  - Arunima, S.R., Deepa, M.J., Geethanjali, C.V., Saji, V.S., Shibli, S.M.A.  
**Tuning of hydrophobicity of WO<sub>3</sub>-based hot-dip zinc coating with improved self-cleaning and anti-corrosion properties**  
(2020) *Appl. Surf. Sci.*, 527.
  - Jain, S., Sanger, A., Chauhan, S., Chandra, R.  
**Hydrogen sensing properties of nanostructured Pd/WO<sub>3</sub> thin films: role of hydrophobicity during recovery process**  
(2014) *Mater. Res. Express*, 1.
  - Yuan, Z., Wu, X., Jiang, Y., Li, Y., Huang, J., Hao, L., Zhang, J., Wang, J.  
**Carbon dots-incorporated composite membrane towards enhanced organic solvent nanofiltration performance**  
(2018) *J. Memb. Sci.*, 549, pp. 1-11.
  - Shang, W., Ye, M., Cai, T., Zhao, L., Zhang, Y., Liu, D., Liu, S.  
**Tuning of the hydrophilicity and hydrophobicity of nitrogen doped carbon dots: A facile approach towards high efficient lubricant nanoadditives**  
(2018) *J. Mol. Liq.*, 266, pp. 65-74.
  - Haase, M.F., Stebe, K.J., Lee, D.  
**Continuous fabrication of hierarchical and asymmetric bijel microparticles, fibers, and membranes by solvent transfer-induced phase separation (STRIPS)**  
(2015) *Adv. Mater.*, 27, pp. 7065-7071.

- Dai, X., Cao, Y., Shi, X., Wang, X.  
**The PLA/ZIF-8 nanocomposite membranes: the diameter and surface roughness adjustment by ZIF-8 nanoparticles, high wettability, improved mechanical property, and efficient oil/water separation**  
(2016) *Adv. Mater. Interfaces*, 3.
- Ho, C.S., Abidin, N.H.Z., Nugraha, M.W., Sambudi, N.S., Ali, F., Wirzal, M.D.H., Kasmiarno, L.D., Adli, S.A.  
**Electrospun porous polylactic acid fibers containing CdS for degradation of methylene blue**  
(2020) *Fibers Polym.*, 21, pp. 1212-1221.
- Krasian, T., Punyodom, W., Worajittiphon, P.  
**A hybrid of 2D materials (MoS<sub>2</sub> and WS<sub>2</sub>) as an effective performance enhancer for poly(lactic acid) fibrous mats in oil adsorption and oil/water separation**  
(2019) *Chem. Eng. J.*, 369, pp. 563-575.
- Zhan, Y., Liu, Y., Liu, Q., Liu, Z., Yang, H., Lei, B., Zhuang, J., Hu, C.  
**Size-controlled synthesis of fluorescent tungsten oxide quantum dots via one-pot ethanol-thermal strategy for ferric ions detection and bioimaging**  
(2018) *Sens. Actuators, B Chem.*, 255, pp. 290-298.
- Xu, J., Zhang, J., Gao, W., Liang, H., Wang, H., Li, J.  
**Preparation of chitosan/PLA blend micro/nanofibers by electrospinning**  
(2009) *Mater. Lett.*, 63, pp. 658-660.
- Luo, Y.B., Wang, X.L., Xu, D.Y., Wang, Y.Z.  
**Preparation and characterization of poly(lactic acid)-grafted TiO<sub>2</sub> nanoparticles with improved dispersions**  
(2009) *Appl. Surf. Sci.*, 255, pp. 6795-6801.
- Doma, A.S., Hassan, N., AbdEl-Hamid, A.I., Soliman, H.M.A.  
**Adsorption of methylene blue dye on hydrothermally prepared tungsten oxide nanosheets**  
(2020) *Egypt. J. Chem.*, 63, pp. 483-498.
- Nayak, A.K., Lee, S., Choi, Y.I., Yoon, H.J., Sohn, Y., Pradhan, D.  
**Crystal phase and size-controlled synthesis of tungsten trioxide hydrate nanoplates at room temperature: enhanced Cr(VI) photoreduction and methylene blue adsorption properties**  
(2017) *ACS Sustain. Chem. Eng.*, 5, pp. 2741-2750.
- Madrakian, T., Maleki, S., Gilak, S., Afkhami, A.  
**Turn-off fluorescence of amino-functionalized carbon quantum dots as effective fluorescent probes for determination of isotretinoin**  
(2017) *Sens. Actuators, B Chem.*, 247, pp. 428-435.
- Liu, X., Pang, J., Xu, F., Zhang, X.  
**Simple approach to synthesize amino-functionalized carbon dots by carbonization of chitosan**  
(2016) *Sci. Rep.*, 6, pp. 1-8.
- Sial, Q.A., Javed, M.S., Lee, Y.J., Duy, L.T., Seo, H.  
**Flexible and transparent graphene-based supercapacitors decorated with**

**nanohybrid of tungsten oxide nanoflakes and nitrogen-doped-graphene quantum dots**

(2020) *Ceram. Int.*, 46, pp. 23145-23154.

- Babu, V.J., Nair, A.S., Peining, Z., Ramakrishna, S.  
**Synthesis and characterization of rice grains like Nitrogen-doped TiO<sub>2</sub> nanostructures by electrospinning-photocatalysis**  
(2011) *Mater. Lett.*, 65, pp. 3064-3068.
- Mallick, S., Ahmad, Z., Touati, F., Bhadra, J., Shakoor, R.A., Al-Thani, N.J.  
**PLA-TiO<sub>2</sub> nanocomposites: thermal, morphological, structural, and humidity sensing properties**  
(2018) *Ceram. Int.*, 44, pp. 16507-16513.
- Zhang, J., Liu, J., Wang, X., Mai, J., Zhao, W., Ding, Z., Fang, Y.  
**Construction of Z-scheme tungsten trioxide nanosheets-nitrogen-doped carbon dots composites for the enhanced photothermal synergistic catalytic oxidation of cyclohexane**  
(2019) *Appl. Catal. B: Environ.*, 259.
- Gondal, M.A., Dastageer, M.A., Khalil, A.  
**Synthesis of nano-WO<sub>3</sub> and its catalytic activity for enhanced antimicrobial process for water purification using laser induced photo-catalysis**  
(2009) *Catal. Commun.*, 11, pp. 214-219.
- Ren, Z., Liu, X., Chu, H., Yu, H., Xu, Y., Zheng, W., Lei, W., Li, C.  
**Carbon quantum dots decorated MoSe<sub>2</sub> photocatalyst for Cr(VI) reduction in the UV-vis-NIR photon energy range**  
(2017) *J. Colloid Interface Sci.*, 488, pp. 190-195.
- Makuła, P., Pacia, M., Macyk, W.  
**How to correctly determine the band gap energy of modified semiconductor photocatalysts based on UV-vis spectra**  
(2018) *J. Phys. Chem. Lett.*, 9, pp. 6814-6817.
- Gu, J., Xiao, P., Chen, P., Zhang, L., Wang, H., Dai, L., Song, L., Chen, T.  
**Functionalization of biodegradable PLA nonwoven fabric as superoleophilic and superhydrophobic material for efficient oil absorption and oil/water separation**  
(2017) *ACS Appl. Mater. Interfaces*,
- Abdel Messih, M.F., Ahmed, M.A., Soltan, A., Anis, S.S.  
**Synthesis and characterization of novel Ag/ZnO nanoparticles for photocatalytic degradation of methylene blue under UV and solar irradiation**  
(2019) *J. Phys. Chem. Solids*, 135.
- Mahmud, R.A., Shafawi, A.N., Ahmed Ali, K., Putri, L.K., Md Rosli, N.I., Mohamed, A.R.  
**Graphene nanoplatelets with low defect density as a synergetic adsorbent and electron sink for ZnO in the photocatalytic degradation of Methylene Blue under UV-vis irradiation**  
(2020) *Mater. Res. Bull.*, 128.
- Visa, M., Bogatu, C., Duta, A.  
**Tungsten oxide – fly ash oxide composites in adsorption and photocatalysis**  
(2015) *J. Hazard. Mater.*, 289, pp. 244-256.

- Abdellah, M.H., Nosier, S.A., El-Shazly, A.H., Mubarak, A.A.  
**Photocatalytic decolorization of methylene blue using TiO<sub>2</sub>/UV system enhanced by air sparging**  
(2018) *Alex. Eng. J.*, 57, pp. 3727-3735.
- Gusain, D., Srivastava, V., Sillanpää, M., Sharma, Y.C.  
**Kinetics and isotherm study on adsorption of chromium on nano crystalline iron oxide/hydroxide: linear and nonlinear analysis of isotherm and kinetic parameters**  
(2016) *Res. Chem. Intermed.*, 42, pp. 7133-7151.
- Ernawati, L., Wahyuono, R.A., Muhammad, A.A., Nurislam Sutanto, A.R., Maharsih, I.K., Widiastuti, N., Widiyandari, H.  
**Mesoporous WO<sub>3</sub>/TiO<sub>2</sub> nanocomposites photocatalyst for rapid degradation of methylene blue in aqueous medium**  
(2019) *Int. J. Eng. Trans. A Basics*, 32, pp. 1345-1352.
- Nugraha, M.W., Zainal Abidin, N.H., Supandi, Sambudi, N.S.  
**Synthesis of tungsten oxide/ amino-functionalized sugarcane bagasse derived-carbon quantum dots (WO<sub>3</sub>/N-CQDs) composites for methylene blue removal**  
(2021) *Chemosphere*, 277.
- Batool, F., Akbar, J., Iqbal, S., Noreen, S., Bukhari, S.N.A.  
**Study of isothermal, kinetic, and thermodynamic parameters for adsorption of cadmium: an overview of linear and nonlinear approach and error analysis**  
(2018) *Bioinorg. Chem. Appl.*, 2018.
- Smrithi, S.P., Kottam, N., Arpitha, V., Narula, A., Subramanian, A.G.N., K.R.V.  
**Tungsten oxide modified with carbon nanodots: Integrating adsorptive and photocatalytic functionalities for water remediation**  
(2020) *J. Sci. Adv. Mater. Devices*, 5, pp. 73-83.
- Elsayed, E.M., Elnouby, M.S., Gouda, M.H., Elessawy, N.A., Santos, D.M.F.  
**Effect of the morphology of tungsten oxide embedded in sodium alginate/polyvinylpyrrolidone composite beads on the photocatalytic degradation of methylene blue dye solution**  
(2020) *Materials (Basel)*, 13, p. 1905.
- Yan, F., Kong, D., Fu, Y., Ye, Q., Wang, Y., Chen, L.  
**Construction of carbon nanodots/tungsten trioxide and their visible-light sensitive photocatalytic activity**  
(2016) *J. Colloid Interface Sci.*, 466, pp. 268-274.

**Correspondence Address**

Sambudi N.S.; Department of Chemical Engineering, Malaysia; email: soraya.sambudi@utp.edu.my

**Publisher:** Elsevier Ltd

**ISSN:** 22133437

**Language of Original Document:** English

**Abbreviated Source Title:** J. Environ. Chem. Eng.

2-s2.0-85110038048

**Document Type:** Article

**Publication Stage:** Final

**Source:** Scopus

---

**ELSEVIER**

Copyright © 2021 Elsevier B.V. All rights reserved. Scopus® is a registered trademark of Elsevier B.V.