## **Electronic Supporting Information**

# Eliminating Waste with Waste: Transforming Spent Coffee Grounds into Microrobots for Water Treatment

Amit Kumar Singh,  ${}^{\ddagger a*}$  Tarini Basireddy ${}^{\ddagger b}$  and Jeffrey L. Moran ${}^{*a, c}$ 

<sup>a</sup> Department of Mechanical Engineering, George Mason University, 10920 George Mason Circle, Manassas, VA 20110, USA.

<sup>b</sup> Thomas Jefferson High School for Science and Technology, Alexandria, VA 22312, USA. <sup>c</sup> Department of Bioengineering, George Mason University, 10920 George Mason Circle, Manassas, VA 20110, USA.

\*E-mail: asingh91@gmu.edu (A.K.S.), jmoran23@gmu.edu (J.L.M.)

<sup>‡</sup>These authors contributed equally to this work.

## 1. Materials

The espresso coffee grounds were purchased from Cafe Bustelo (brick pack) and brewed once using a Bialetti Moka Cup 6 coffee maker. Iron (II, III) oxide (magnetite) nanoparticles (IONPs, Cat. No: 637106-25G), L-ascorbic acid (AA), molecular biology-grade ethanol (200 proof, Cat. No: E7023-1L), Oil red O dye, and acetone were purchased from Sigma Aldrich (USA). Neodymium (NdFeB) magnets (N52-grade) were purchased from K & J Magnetics (USA). Disposable 100  $\mu$ m Nylon mesh cell strainer and ultrapure distilled (DI) water were purchased from Fisherbrand (USA) and Invitrogen (USA), respectively. Natural seawater was procured from Carolina Biological supply company (USA). Methylene blue (MB) was purchased from HiMedia Laboratories (USA). Engine oil was procured from Valvoline Inc. (USA). Spherical polystyrene (PS) microbeads (mean particle size: 60  $\mu$ m) were purchased from Amazon (USA). All the glasswares and Whatman filter papers (Grade 1) were purchased from VWR International (USA). The aforementioned chemicals were used without further purification.

## 2. Characterization techniques

Field emission scanning electron microscopy (JEOL-IT500HR, USA) was used to examine the samples. The samples were vacuum-dried and placed on carbon tape adhered to a stub and gold-sputtered for FESEM analysis. Gold sputtering was performed at 20 mA ion current for 3 min (1 cycle) using a Denton Vacuum Desk V sputter coater (USA). The sample elemental analysis was measured by energy dispersive X-ray spectroscopy (EDX) in FESEM. The FTIR analyses were performed on a Nicolet iS10 FTIR Spectrometer (Thermo Scientific, USA). Before testing, samples were dried and formed into KBr pellets and the measurements were obtained in the 400-4000 cm<sup>-1</sup> spectral range in transmission mode with 64 scans at a resolution of 4 cm<sup>-1</sup>. The zeta potential (surface charge) and dynamic light scattering (DLS) measurements were performed in Malvern Zetasizer Nano ZS (USA) using DTS-1070 folded capillary zeta cells. The measurements for zeta potential were taken by suspending the samples in 0.1X PBS at pH 7.4. For dye removal experiments, the liquid samples were scanned at 665 nm using a Tecan Safire 2 Microplate Reader (USA), and full range scan was performed using Cary 60 UV-Vis spectrophotometer (Agilent, USA). The experimental videos were recorded with a Redmi Note 7 Pro smartphone (Xiaomi, India). Microrobot motion was captured under the Leica DMi8 inverted microscope with Leica application software (LAS-X) software. Various experiments were conducted by applying a neodymium bar magnet (N-52 grade) with a magnetic field gradient of 109 mT mm<sup>-1</sup>, and the applied magnetic field was measured with a Tunkia TD8620 Handheld Digital Tesla Meter High Precision Gaussmeter (Amazon, USA). To edit the videos, Xilisoft and Movavi software were used, along with ImageJ software<sup>1</sup> for tracking motions within the clips. Free Studio software package was used to extract the images from the video file. ChemDraw was used for drawing chemical structures.

## 3. Elemental mapping and IONP Size distribution



**Figure S1.** (A) The image shows the area selected (rectangle in red) on the surface of uncoated SCGs for EDX analysis. The scale bar is 20  $\mu$ m. (B) EDX spectrum of the SCGs and the percentage weight (Wt%) distribution of the elements. (C) The image shows the area selected (rectangle in red) on the CoffeeBot's surface for EDX analysis. The scale bar is 20  $\mu$ m. (D) EDX spectrum of the CoffeeBots and the percentage weight (Wt%) distribution of the elements. (E) FESEM image shows deposition of magnetic IONPs in the form of aggregates on the surface of the CoffeeBots. The scale bar is 1  $\mu$ m. (F) Size distribution of commercial IONPs (Sigma).

#### **Description of Figure S1**

**Figures S1A** and **S1C** depict the area designated (shown as a red rectangle) for the elemental mapping of uncoated SCGs and CoffeeBots, respectively. In addition, **Figures S1B and S1D** illustrate the EDX spectra and elemental weight percentage of the uncoated SCG and CoffeeBot surfaces, respectively. The EDX analysis of an uncoated SCG particle (**Figures S1B**) reveals that it is composed of 54.59% carbon (C), 34.59% oxygen (O), 6.9% nitrogen (N), and trace amounts of sulfur (S) about 0.13%. The EDX analysis of CoffeeBots (**Figures S1D**) suggests that the composition of CoffeeBots includes 38.72% carbon (C), 31.87% oxygen (O), and 20.45% iron sourced from IONPs, along with smaller percentages of nitrogen (N) at 3.47%, and sulfur (S) traces at 0.37%. In both specimens, the presence of elemental gold (Au) was identified, which can be attributed to the utilization of Au sputtering during FESEM sample preparation. The surface of CoffeeBots is depicted in **Figure S1E**, showcasing the attachment of aggregates consisting of discrete IONPs with sizes varying from 200 nm to 700 nm. The DLS technique was utilized to measure the size of commercial IONPs, which further verified the nanoparticles' size distribution. The average diameter of these particles was approximately 570 nm, with sizes ranging from 200 – 800 nm (**Figure S1F**).

### 4. Porosity and pore size distribution of the CoffeeBots

The density of the CoffeeBots must first be determined in order to determine their porosity. According to Shi *et al.*,<sup>2</sup> the density of the CoffeeBots was determined using pycnometer method. An excess of ethanol and 1 g of adsorbent were added to the pycnometer and sealed with a glass stopper. The capillary holes in the stopper were then used to remove any excess ethanol. The density of the CoffeeBots were calculated using the following equation:

where  $D_c$  is the density of the CoffeeBots;  $M_1$  is the weight of the dry CoffeeBots,  $M_2$  is the total weight of the pycnometer and the filled ethanol (without adsorbent);  $M_3$  is the is the weight of the adsorbed ethanol (calculated from the difference between the weight of the ethanol and dried CoffeeBots);  $M_4$  is the total weight of the pycnometer together with the inserted CoffeeBots and ethanol; and  $D_e$  is the density of ethanol. All the measurements were repeated thrice and the average has been reported. The average density of the CoffeeBots ( $D_c$ ) were estimated to be ~ 464 kg/m<sup>3</sup> using the measured data reported below in **Table S1**.

Parameters	Recorded data		
$M_1$	1 g		
$M_2$	24.2 g		
M <sub>3</sub>	2.3 g		
M4	25.8 g		
$D_e$	0.789 g/ml at 20°C		
D <sub>w</sub>	1570 kg/m <sup>3</sup>		
Calculated <b>D</b> <sub>c</sub>	0.464 g/ml or, 464 kg/m <sup>3</sup>		
Calculated Porosity	70.44%		

 Table S1. Measured and calculated data for porosity calculations of the CoffeeBots.

The porosity was estimated from the dry CoffeeBots using the following equation reported<sup>10</sup> in previous literature:

$$Porosity = \left(1 - \frac{\rho_c}{\rho_w}\right) \times 100\%....(2)$$

where  $\rho_c$  is the average density of the CoffeeBots estimated from equation (1) and  $\rho_w$  is the density of solid wood (1570 kg/m<sup>3</sup>),<sup>2-5</sup> as standard. The porosity of the CoffeeBots were calculated to be 70.44%. The pore size distribution on the surface of the CoffeeBots were determined using ImageJ analysis of the SEM images.



**Figure S2.** (A) Representative SEM image showing pores of varying dimensions on the surface of CoffeeBots. The scale bar is 20  $\mu$ m. (B) Representative post-processed image of (A) showing pores distribution as black spots after ImageJ analysis. The scale bar is 20  $\mu$ m. (C) Histogram showing average distribution of pore sizes over 3 separate SEM images.

## **Description of Figure S2**

SEM images of the surface of the CoffeeBots before and after image processing using the "thresholding" approach in ImageJ software<sup>6</sup> are shown in **Figures S2A** and **S2B**, respectively. The pore size distribution studies (**Figure S2C**) from SEM images captured from three

different sites of a CoffeeBot's surface confirmed the presence of micropores of varying size, ranging from  $\sim 0.5$  to 70 µm, thereby indicating a hierarchical microporous structure.



## **5.** Hydrophobicity and Wettability of CoffeeBots

**Figure S3.** (A) Resting water droplet on a cluster of hydrophobic CoffeeBots at the indicated times. (B) Relationship between contact angle of the sessile water droplet and corresponding time. (C) Negligible contact angle between oil droplet and CoffeeBots indicates oleophilicity of the CoffeeBots.

## **Description of Figure S3**

The water contact angle (WCA) method was used to assess the wettability and hydrophobicity of the CoffeeBots. **Figure S3** illustrates a sessile water droplet (50  $\mu$ L) resting on the cluster of dry CoffeeBots on top of a glass slide. To quantify the contact angle, the shape of the water droplet was fitted using the "contact angle" plugin of ImageJ software (**Figure S3A**). The measured contact angle between water droplet and CoffeeBots is ~ 139°, providing evidence of their hydrophobic nature. It is evident from **Figure S3B** that the contact line remains stable over ~50 minutes, evincing the stability of the CoffeeBots' hydrophobicity. Additionally, an assessment was conducted to determine the affinity of CoffeeBots towards motor oil by measuring the contact angle between them. It was observed that oil droplets were quickly absorbed by CoffeeBots, leading to a negligible contact angle within 1 sec (**Figure S3C**). This indicates that CoffeeBots have a strong affinity for oils and possess high oleophilicity.



## 6. Zeta Potential (surface charge) measurements

**Figure S4.** Zeta potential measurements of various samples. Numerical values below each bar give the mean (in mV) of the recorded data for the indicated sample.

## **Description of Figure S4**

The plot depicts the zeta potential at 25°C for SCGs, IONPs, CoffeeBots and polystyrene (PS) microbeads in a 0.1X PBS buffer solution of pH 7.4. The average zeta potential (surface charge) values for SCGs, IONPs, CoffeeBots and PS beads were recorded to be -21.7 mV, -62.3 mV, -42.7 mV and -42.1 mV, respectively.

## 7. Chemical durability of the CoffeeBots



**Figure S5.** Contact angle of water resting on a bed of CoffeeBots that have been exposed to a strong acid, strong base, salt solution, and pure water respectively.

#### **Description of Figure S5**

The chemical durability of the hydrophobic CoffeeBots was evaluated by exposing them to a strong acid, strong base, and concentrated salt solution, using pure water as a control. We immersed 60 mg of CoffeeBots in 10 mL of 1M HCl, 1M NaOH, 1M NaCl, and water for 12 hr each. As a figure of merit for chemical durability, we conducted contact angle measurements on the CoffeeBots (after drying at 100°C for 8 h) to confirm that they retained their hydrophobicity (and thus kept their ability to capture microplastics and oil droplets). In each case, a sessile water droplet (50  $\mu$ L) was placed on 60 mg of dry CoffeeBots placed onto a glass slide. To measure the contact angle, the spherical water droplet was fitted using the "contact angle" plugin in ImageJ. The water contact angles (in an air medium) were recorded as ~129°, ~98°, and ~134° for CoffeeBots treated with 1M HCl, 1M NaOH, and 1M NaCl, respectively. The contact angle of the control (pure water) droplet was measured to be ~139°. These results are summarized in **Figure S5**. From these data, it is evident that the CoffeeBots exhibited chemical resistance as they maintained their hydrophobic nature even when exposed to corrosive liquids and salt solutions.

## 8. Velocity profile measurements



Figure S6. Speed variation of ~450  $\mu$ m CoffeeBots with change in magnetic field strength. Description of Figure S6

**Figure S6** shows the variation in speed ( $V_m$ ) of ~450 µm CoffeeBots with changing external magnetic fields from 95 mT to 15 mT while immersed in seawater at a distance of 2 cm away from the magnetic pole. The microrobots moved linearly toward the nearest magnetic pole under magnetic guidance. Under a lower magnetic field strength (15 mT), the CoffeeBots averaged ~920 µm s<sup>-1</sup>, which corresponds to 2 body lengths per second. With a magnetic field

strength of 95 mT, the CoffeeBots could move at an average speed of ~5500  $\mu$ m s<sup>-1</sup>, which corresponds to ~12 body lengths per second. In addition, the experiments indicated that the motor could be accelerated by further increases in the external magnetic field.



## 9. The calibration plot for MB dye experiments

Figure S7. Calibration curve of Methylene Blue (MB) dye at wavelength of 665 nm.

## **Description of Figure S7**

**Figure S7** shows the calibration curve of absorbance (*A*) at a wavelength of 665 nm versus varying MB dye concentrations (*C*, in mg/L) in seawater, measured using a microplate reader. The linear correlation between the MB dye concentration and the absorbance peak intensity at 665 nm can be expressed as a linear mathematical equation: A = 0.03102 C + 0.04861. In order to determine an unknown concentration of MB in seawater, this mathematical expression was used for all the experiments.<sup>11</sup>

#### 10. Reusability of AA@CoffeeBots

For reusability tests, AA@CoffeeBots were retrieved at the end of the reaction and washed twice 50% with ethanol for removal of surface-absorbed MB dye molecules.<sup>12</sup> The samples were dried at 65°C for 5 h. The dried samples were placed on a watch glass and were immersed in 400  $\mu$ L of ascorbic acid solution to reload the sample with ascorbic acid. Following this, the watch glass was then placed in an oven at 65°C for at least 12 h to evaporate the solvent. The aforesaid method for AA@CoffeeBots-mediated water treatment was repeated each time followed by reloading of ascorbic acid for five consecutive reusability tests.



11. Rate constant measurements for MB dye experiments

**Figure S8.** The MB dye removal rate kinetics plots for (A) Uncoated SCGs (B) Motile CoffeeBots (C) Ascorbic Acid (D) Stationary AA@CoffeeBots and (E) Motile AA@CoffeeBots. The experiments were carried out in triplicate and the findings were presented as mean values with corresponding standard deviations.

## **Description of Figure S8**

**Figure S8** depicts the time-dependent MB dye removal by 50 mg of microrobots immersed in 8 mL of seawater with 10 mg/L MB dye contamination. The logarithmic of MB concentrations plotted as a function of the reaction time, where 'C' denotes the concentrations of MB dye in the solution at any time, and 'C<sub>0</sub>' denotes the initial dye concentration of 10 mg/L. The recorded MB concentration in the solution fits to a straight indicates that MB removal follows first-order kinetic rate law, where slope (k) represents observed first-order rate constant (in min<sup>-1</sup>).

## 12. Ascorbic acid and Methylene Blue reaction



**Figure S9.** Schematic representation showing the reduction of Methylene Blue (MB) by Ascorbic acid (AA).

#### **Description of Figure S9**

Previous studies by El-Aila<sup>13</sup> suggests that under alkaline conditions, the blue-coloured MB dye is reduced to the colorless reduced form, known as leucomethylene blue (LMB) by AA (in excess). The AA undergoes oxidation to form dehydroascorbic acid (DHA) in the process of MB reduction.<sup>14,15</sup>

#### 13. Oil separation efficiency (OSE) calculations and reusability of CoffeeBots

The OSE was determined using following equation stated below:

$$OSE \ (\%) = \left(\frac{W_0 - W_1}{W_0}\right) \times 100.$$

where,  $W_1$  is the weight of oil spill (in mg) left on glass slide after clean-up operation, and  $W_0$  is the initial weight (in mg) of the oil droplet.<sup>11,16</sup> The average data along with standard error bars was recorded using a set of three experiments.

For reusability tests, 30 mg of CoffeeBots were introduced onto a droplet of engine oil (25 mg) drop-cast onto a glass slide, followed by a stay for 3 min. After this, the CoffeeBots were magnetically collected and subsequently by acetone wash.<sup>2</sup> The exhausted motors were suspended in 10 ml of acetone and vortexed for 2 min. Once again, the motors were magnetically retrieved and washed twice with acetone, followed by oven drying at 65°C for 2

h. For testing of its reusability, CoffeeBots were again introduced into oil samples and the process of oil sorption was carried out for five consecutive cycles. The percentage of recovered oil (OSE) after each cycle was calculated as stated previously. Three experiments were used to record the average data and standard error bars.

### 14. Microplastics removal efficiency calculations and CoffeeBots reusability

The removal efficiency of PS beads from seawater was calculated using the following equation:

Removal efficiency (%) = 
$$\left(\frac{M_0 - M_1}{M_0}\right) \times 100$$

where,  $M_1$  is final mass (in mg) of residual PS beads after filtration, and  $M_0$  is the initial mass (in mg) of the PS beads before treatment.<sup>3</sup> The average data along with standard error bars was recorded using a set of three experiments.

For reusability tests, 10 mg of spherical PS microbeads (20  $\mu$ m -140  $\mu$ m) were uniformly dispersed in 8 ml of seawater sample by ultrasonication. 50 mg of CoffeeBots were added to the contaminated water, followed by 1 h standby period. After the treatment, a neodymium bar magnet was used to separate microrobots from the solution. The retrieved CoffeeBots were suspended in 10 ml of ethanol and vortexed for 10 min and subsequently the treated solution was filtered using a Whatman filter paper. The retrieved CoffeeBots and filter paper with remnant PS beads were oven-dried at 80°C overnight to remove the ethanol and water, respectively. The remnant PS beads were again introduced into microplastic contaminated seawater solution and the process of removal was carried out for five consecutive cycles. The percentage of microplastic removal after each cycle was calculated as stated previously. Three experiments were used to record the average data and standard error bars.

Pollutant	Materials	Magnetic	Physical/Chemical	Removal	Reference
Туре		Propulsion	<b>Pre-treatment</b>	Efficiency	From
			of SCGs for	(in %)	ESI
			coating magnetic		
			particles		
Methylene	Fe <sub>3</sub> O <sub>4</sub> /SCGs	No	Yes	85%	17
Blue	Fe <sub>3</sub> O <sub>4</sub> /SCGs	No	Yes	90%	18
Dye	Activated carbon	No	Yes	Not	19
Removal	from SCGs/ Fe <sub>3</sub> O <sub>4</sub>			defined	
	SCG biochar/ Fe <sub>3</sub> O <sub>4</sub>	No	Yes	98%	20
	Activated carbon	No	Yes	Not	21
	from SCGs/			defined	
	Fe <sub>3</sub> O <sub>4</sub> /Chitosan				
	Fe3O4/ Polyvinyl	No	Yes	91%	22
	alcohol/SCGs				
	Fe <sub>3</sub> O <sub>4</sub> /SCGs	No	Yes	98%	23
	Fe <sub>3</sub> O <sub>4</sub> /SCGs	No	Yes	95%	24
	Fe <sub>3</sub> O <sub>4</sub> /SCGs	No	Yes	90%	25
	Fe <sub>3</sub> O <sub>4</sub> /SCGs	No	Yes	99%	26
	Ferrofluid/SCGs	No	No	80%	27
	Fe <sub>3</sub> O <sub>4</sub> /SCGs/Ascorbic	Yes	No	93%	This
	Acid				work
Oil Spills	Maghemite/SCGs	No	Yes	Not	28
				defined	
	Activated carbon	No	Yes	99%	29
	from SCGs/ Fe <sub>3</sub> O <sub>4</sub>				
	Fe <sub>3</sub> O <sub>4</sub> /SCGs	Yes	No	99%	This
					work
Plastics	Fe <sub>3</sub> O <sub>4</sub> /SCGs	Yes	No	64%	This
					work

## 15. Comparison of various magnetic spent coffee ground (SCGs) for water treatment

**Table S2.** Comparative study of previously published magnetic SCGs with CoffeeBots for water treatment.

## 16. Proof-of-Concept water treatment tank architecture





## **Description of Figure S10**

CoffeeBots have the potential to be utilized in a limited area by integrating a customized treatment tank onto a ship. This water-remediation solution would consist of a tank with an input pipe for contaminated water and an output pipe for clean water.<sup>30,31</sup> Furthermore, the setup may incorporate triaxial Helmholtz coils surrounding the tank that can be externally controlled by an operator. These coils can generate complex three-dimensional (3D) magnetic fields, which would drive the CoffeeBots' motion in optimal trajectories to facilitate efficient pollutant removal from the water. After treatment, the CoffeeBots could be collected (e.g., by diverting them using a DC magnetic field to a separate holding area) at the end of treatment to allow purified water to flow out. The magnetic field magnitude and direction could be optimized to improve the fluid mixing conditions, as shown previously using magnetic microplatelet suspensions, as shown previously<sup>32-34</sup>, and thus maximize convective transport enhancement of pollutant to the CoffeeBots' surface. Optimizing the magnetic field offers a promising way to take full advantage of the magnetic properties of the CoffeeBots.

## **17. Supporting Videos details**

**Video S1:** Magnetic actuation of a CoffeeBot in upward, downward, right, and left directions. **Video S2:** A CoffeeBot magnetically navigated in complex G, M, and U trajectories.

**Video S3:** Methylene Blue dye decolorization in seawater by an assembly of magnetically actuated AA@CoffeeBots.

Video S4: Magnetic guidance of a CoffeeBot to capture and transport oil droplet in seawater.

**Video S5:** An extremely fast-moving CoffeeBot captures and transports free floating oil droplet under the influence of magnetic fields.

Video S6: Oil droplet manipulation by an assembly of magnetically-propelled CoffeeBots.

Video S7: An assembly of CoffeeBots employed for removal of oil slick from seawater.

Video S8: Magnetically-guided CoffeeBot for trapping of microplastic beads.

## References

- 1. M. D. Abramoff, P. J. Magalhaes, and S. J. Ram, Biophotonics Int., 2004, 11, 36-42.
- 2. C. Shi, T. Wang, S. Roy, S. S. Chopra, G. Chen, J. Shang, J. Tian and Y. S. Ok, *ACS EST Engg.*, 2023, **3**, 1297–1307.
- 3. C. Akarsu, H. Kumbur and A. E. Kideys, Water Sci. Technol., 2021, 84, 1648–1662.
- 4. Q. Fu, F. Ansari, Q. Zhou, L. A. Berglund, ACS Nano, 2018, 12, 2222–2230.
- 5. L. J. Gibson and M. F. Ashby, *Cellular Solids: Structure and Properties*, Cambridge University Press, Cambridge, 2nd edn., 1997.
- 6. M. Haeri and M. Haeri, J. Open Res. Softw., 2015, 3, e1.
- 7. J. Baggenstoss, R. Perren and F. Escher, Eur. Food Res. Technol., 2008, 227, 1357–1365.
- 8. Y. Liu, Y. Peng, T. Zhang, F. Qiu and D. Yuan, Cellulose, 2018, 25, 3067–3078.
- 9. H. Li, Q. Zhong, Q. Sun, B. Xiang and J. Li, Langmuir, 2022, 38, 3493-3500.
- 10. G. Shi, M. Wu, Q. Zhong, P. Mu and J. Li, *Langmuir*, 2021, 37, 7843-7850.
- 11. A. K. Singh, T. Bhuyan, S. Maity, T. K. Mandal and D. Bandyopadhyay, *ACS Appl. Nano Mater.*, 2020, **3**, 3459–3470.
- 12. J. Fan, B. Zhang, B. Zhu, W. Shen, Y. Chen and F. Zeng, Water, 2023, 15, 183.
- 13. H. J. El-Aila, Tenside, Surfactants, Deterg., 2012, 49, 26-31.
- 14. P. K. Sen, P. Mukherjee and B. Pal, J. Mol. Liq., 2016, 224, 472-479.
- 15. M. Lackovičová, T. Baranyaiová and J. Bujdák, Appl. Clay Sci., 2019, 181, 105222.
- 16. Z. Chen, C. Zhou, J. Lin, Z. Zhu, J. Feng, L. Fang and J. Cheng, *J. Sol-Gel Sci. Technol.*, 2018, **85**, 23–30.
- 17. N. Besharati, N. Alizadeh and S. Shariati, J. Mex. Chem. Soc., 2018, 62, 110-124.
- 18. A. Zuorro and R. Lavecchia, Appl. Mech. Mater., 2013, 394, 3-7.

19. M. F. Rizkiana, Hidayatullah, A. Rosalina, B. A. Fachri and H. Harada, *IOP Conf. Ser. Mater. Sci. Eng.*, 2021, **1053**, 012007.

20. T. P. Krishna Murthy, B. S. Gowrishankar, R. H. Krishna, M. N. Chandraprabha and B. B. Mathew, *Environ. Chem. Ecotoxicol.*, 2020, **2**, 205–212.

21. I. K. Santosa and M. F. Rizkiana, J. Biobased Chem., 2023, 3, 66-75.

22. M. Pham Thanh and T. Le Van, BIO Web Conf., 2021, 30, 02008.

23. A. Zuorro, A. Di Battista, and R. Lavecchia, Chem. Eng. Trans., 2013, 35, 1375-1380.

24. T. Wirawan, N. Hindryawati and N. T. Widodo, AIP Conf. Proc., 2022, 2668, 030013.

25. P. Pietrzyk, E. I. Borowska, P. Hejduk, B. C. Camargo, M. Warczak, T. P. Nguyen, *et. al.*, *Environ. Sci. Pollut. Res.*, 2023, **30**, 62689–62703.

26. F. Taleb, M. Ammar, M. ben Mosbah, R. ben Salem and Y. Moussaoui, *Sci. Rep.*, 2020, **10**, 110048.

27. I. Safarik, K. Horska, B. Svobodova and M. Safarikova, *Eur. Food Res. Technol.*, 2012, 234, 345–350.

J. C. Silva, G. E. Oliveira, R. D. Toledo Filho and F. G. Souza, *Macromol. Symp.*, 2018, 380, 1–6.

29. L. Acosta, D. Galeano-Caro, O. E. Medina, F. B. Cortés and C. A. Franco, *Processes*, 2021, 9, 63.

30. M. Urso, M. Ussia and M. Pumera, Nat. Rev. Bioeng., 2023, 1, 236–251.

31. C. D. Powell, A. J. Atkinson, Y. Ma, M. Marcos-Hernandez, D. Villagran, P. Westerhoff and M. S. Wong, *J. Nanoparticle Res.*, **22**, 48.

32. J. E. Martin and K. J. Solis, Soft Matter, 2014, 10, 3993-4002.

33. J. E. Martin and K. J. Solis, Soft Matter, 2015, 11, 7130-7142.

34. K. J. Solis and J. E. Martin, Soft Matter, 2013, 9, 9182–9188.