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SUPPORTING INFORMATION

Sustainably closed loop recycling of hierarchically porous polymer microbeads for efficient removal of cationic dyes

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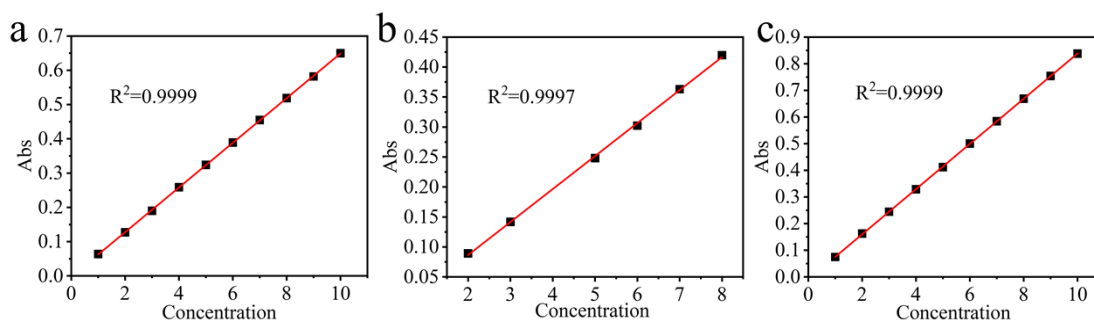
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23 **Section S.1** Standard curves of three cationic dyes



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25 Fig. S1 Standard curve of cationic dyes: (a) Methylene blue; (b) Malachite green; (c) Methyl violet 2B.

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27 The equation obtained by curve fitting.

28 Methylene blue: $A=0.0652c-0.0027$ (1)

29 Malachite green: $A=0.055c-0.0234$ (2)

30 Methyl violet 2B: $A=0.0848c-0.0094$ (3)

31 A is the absorbance of the solution, and C(mg/L) is the concentration of the solution.

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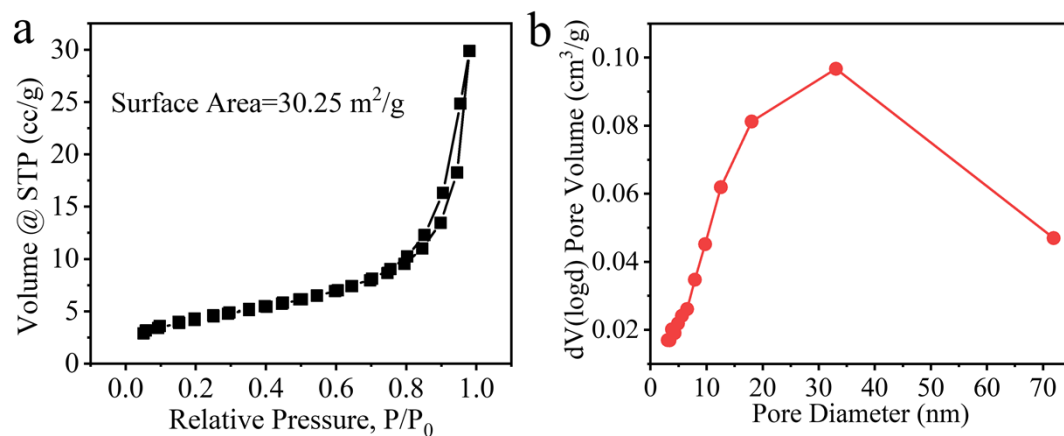
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61 **Fig. S2.** (a) Nitrogen adsorption–desorption isotherm obtained for PCP-IDA adsorbents; (b) Pore

62 size Distribution of PCP-IDA.

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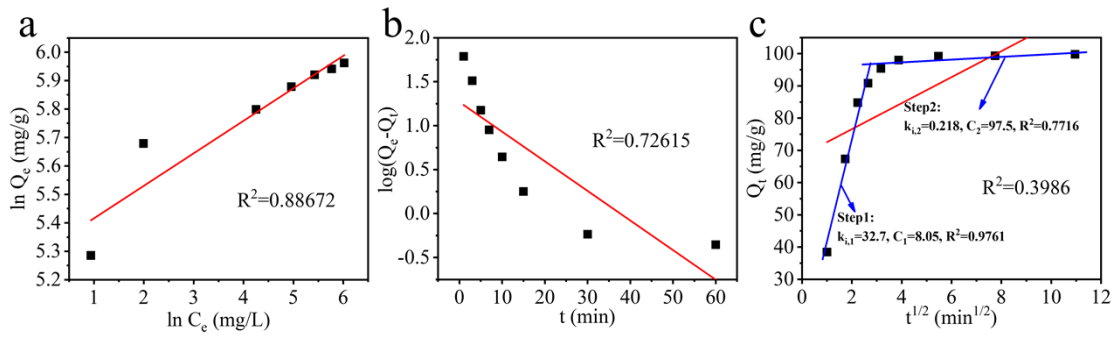
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86 **Fig. S3.** (a) Freundlich fitting curve in isothermal adsorption model. (b) The fitting result of the pseudo-
 87 first-order model; (c) The fitting result of the intra-particle diffusion kinetic model.

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111 **Table S1.** Estimated values of parameters for different kinetic models.

| Kinetic model | Parameters | | |
|--------------------------|------------------------------------|--|---------|
| Pseudo-first-order | $Q_e(\text{mg}\cdot\text{g}^{-1})$ | $k_1(\text{mg}\cdot\text{g}^{-1})$ | R^2 |
| | 99.8 | 0.776 | 0.72615 |
| Pseudo-second-order | $Q_e(\text{mg}\cdot\text{g}^{-1})$ | $k_2(\text{g}\cdot\text{mg}^{-1}\cdot\text{min}^{-1})$ | R^2 |
| | 101.01 | 0.1077 | 0.99988 |
| Intra-particle diffusion | $C(\text{mg}\cdot\text{g}^{-1})$ | $k_i(\text{mg}\cdot\text{g}^{-1}\cdot\text{min}^{-1/2})$ | R^2 |
| | 68.514 | 4.0339 | 0.39864 |

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144 **Table S2.** Isothermal adsorption fitting curve data.

| Langmuir | | | Freundlich | | |
|-----------------------|-----------------------|----------------|------------|-----------------------|----------------|
| Q _m (mg/g) | K _L (L/mg) | R ² | n | K _F (L/mg) | R ² |
| 384.62 | 0.1354 | 0.99897 | 8.7489 | 200.68 | 0.88672 |

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183 **Table S3.** The comparison of the adsorption capacity of PCP-IDA adsorbent with other reported
 184 adsorbents with similar structure or size.

| Adsorbent/mass | MB concentration/ volume | pH | Temperature | Adsorption capacity (mg/g) | Kinetic (min) | References |
|--|-----------------------------|-----|------------------|--|---------------|--------------|
| Nanocomposites poly(GDMA)/MCM-41/20 mg | 40 mg/L 40 mL | 6 | Room temperature | MB: 111.11 | 20 | 1 |
| MAA/GMA-g-PET fibers/100 mg | 40 mg/L 25mL | 10 | 298K | MB: 52.1 | 60 | 2 |
| Cell-g-AASO3H-co-GMA/50 mg | 20 mg/L 50 mL | 7 | 298K | MG: 46.23 MV: 53.53 | 120 | 3 |
| Poly GMA/DVB/200 mg | 5 mg/L 25 mL | 7 | Room temperature | MG: 13.6 | 50 | 4 |
| MCTSms-PMAA/4.5 mg | 200 mg/L 40 mL | 12 | Room temperature | MB: 211.11 | 100 | 5 |
| ATP@CCS/7.5 mg | 200 mg/L 20 mL | 10 | 298.15K | MB: 215.73 | 120 | 6 |
| PES/GO porous particles/5 mg | 150 μ mol/L 20mL | 7 | 303K | MB: 62.5 | 3600 | 7 |
| UiO-66/MIL-101(Fe)/10 mg | 50 mg/L 20mL | 9 | 298K | MB: 448.71 | 30 | 8 |
| A/ γ -Fe ₂ O ₃ /f-CNT composite beads/50 mg | 230 mg/L 50 mL | 5.2 | 298K | MB: 396.7 | 2880 | 9 |
| MPGB biosorbent/50 mg | 50 mg/L 100mL | 7 | 293K | MB: 231.5 | 60 | 10 |
| P(G-E)@IDA/10 mg | 100 mg/L 10 mL | 7 | 298K | MB: 384.62 MG: 333.33 MV: 322.58 | 15 | In this work |

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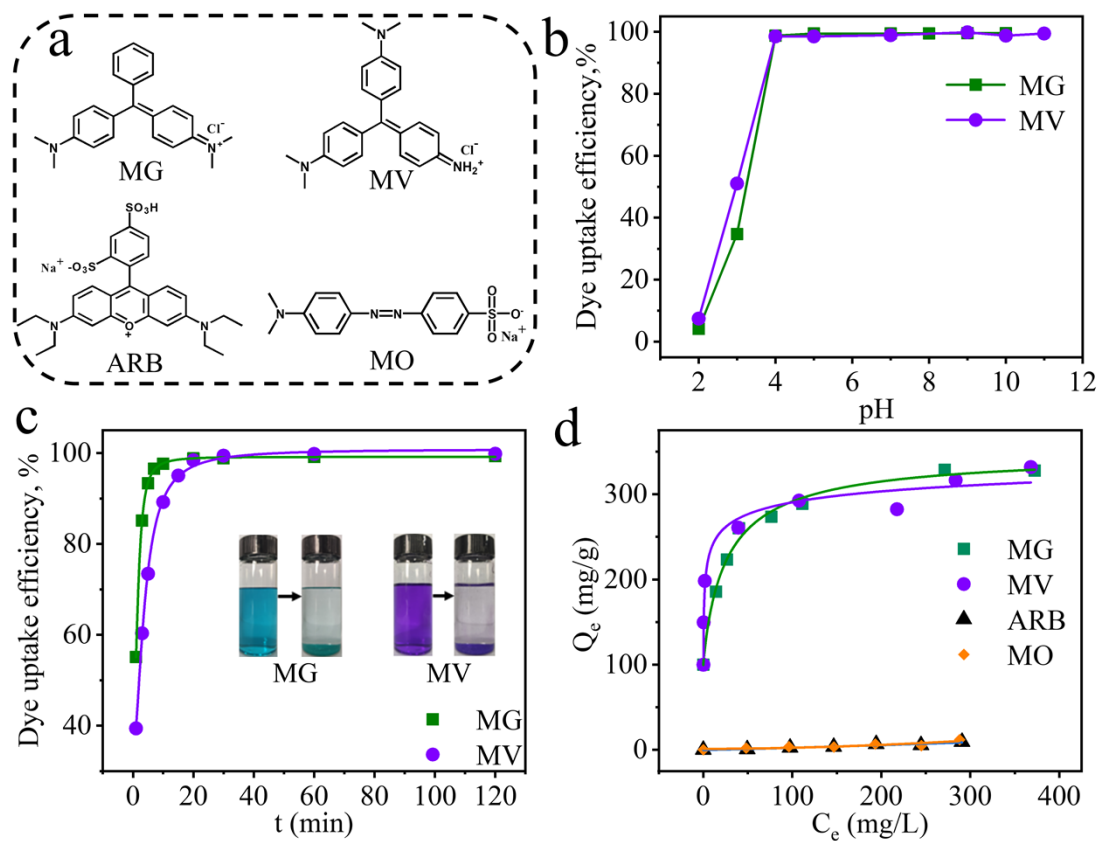
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192 **Fig. S4.** (a) The chemical structure of MG, MV, ARB and MO dyes; (b) Effect of initial PH of dye

193 solution on adsorption; (c) Rapid adsorption of MG and MV dyes; (d) The equilibrium adsorption

194 capacity of four dyes by PCP-IDA adsorbent.

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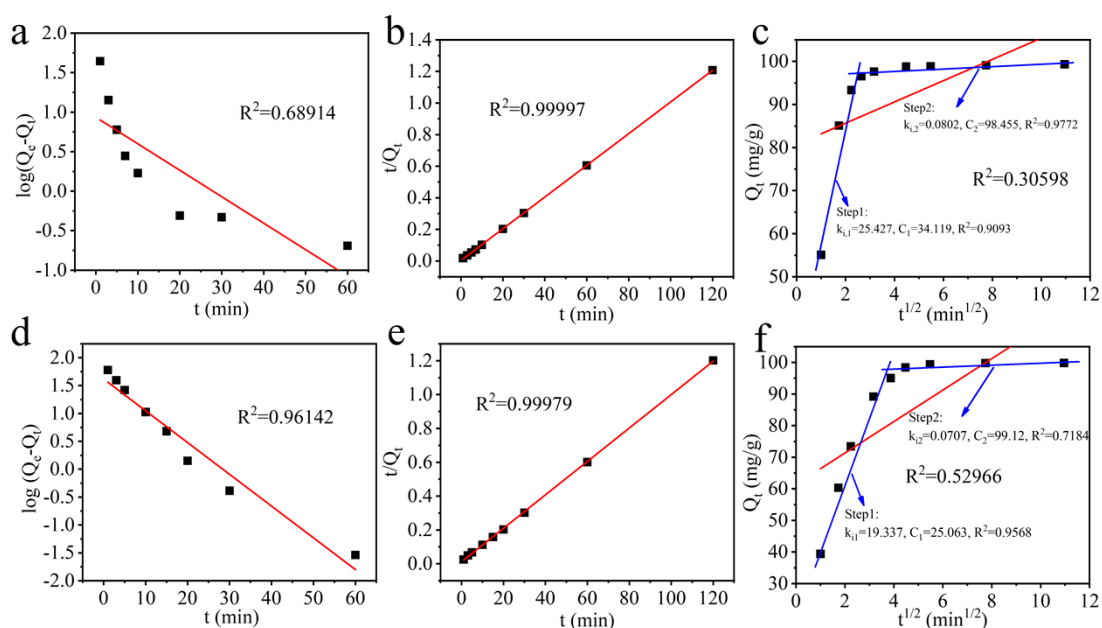
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213 **Section S.2** The related models and parameters of PCP-IDA adsorbent for MG and MV dye adsorption
214 were fitted.



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216 **Fig. S5.** (a) Pseudo-first-order model of MG; (b) Pseudo-second-order model of MG; (c) Intra-particle
217 diffusion model of MG; (d) Pseudo-first-order model of MV; (e) Pseudo-second-order model of MV; (f)
218 Intra-particle diffusion model of MV.

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241 **Table S4.** Estimated values of parameters for different kinetic models.

| Dye | Kinetic model | Parameters | | |
|-------|-----------------------------|------------------------------------|--|---------|
| MG | Pseudo-first-order | $Q_c(\text{mg}\cdot\text{g}^{-1})$ | $k_1(\text{mg}\cdot\text{g}^{-1})$ | R^2 |
| | | 99.32 | 0.0772 | 0.68914 |
| | Pseudo-second-order | $Q_c(\text{mg}\cdot\text{g}^{-1})$ | $k_2(\text{g}\cdot\text{mg}^{-1}\cdot\text{min}^{-1})$ | R^2 |
| | | 100 | 0.025 | 0.99997 |
| | Intra-particle diffusion | $C(\text{mg}\cdot\text{g}^{-1})$ | $k_i(\text{mg}\cdot\text{g}^{-1}\cdot\text{min}^{-1/2})$ | R^2 |
| | | 78.891 | 2.6216 | 0.30598 |
| MV 2B | Pseudo-first-order | $Q_c(\text{mg}\cdot\text{g}^{-1})$ | $k_1(\text{mg}\cdot\text{g}^{-1})$ | R^2 |
| | | 99.8361 | 0.1313 | 0.96142 |
| | Pseudo-second-order | $Q_c(\text{mg}\cdot\text{g}^{-1})$ | $k_2(\text{g}\cdot\text{mg}^{-1}\cdot\text{min}^{-1})$ | R^2 |
| | | 101.01 | 0.0076 | 0.99979 |
| | Intra-particle diffusion | $C(\text{mg}\cdot\text{g}^{-1})$ | $k_i(\text{mg}\cdot\text{g}^{-1}\cdot\text{min}^{-1/2})$ | R^2 |
| | | 61.408 | 4.7948 | 0.52966 |

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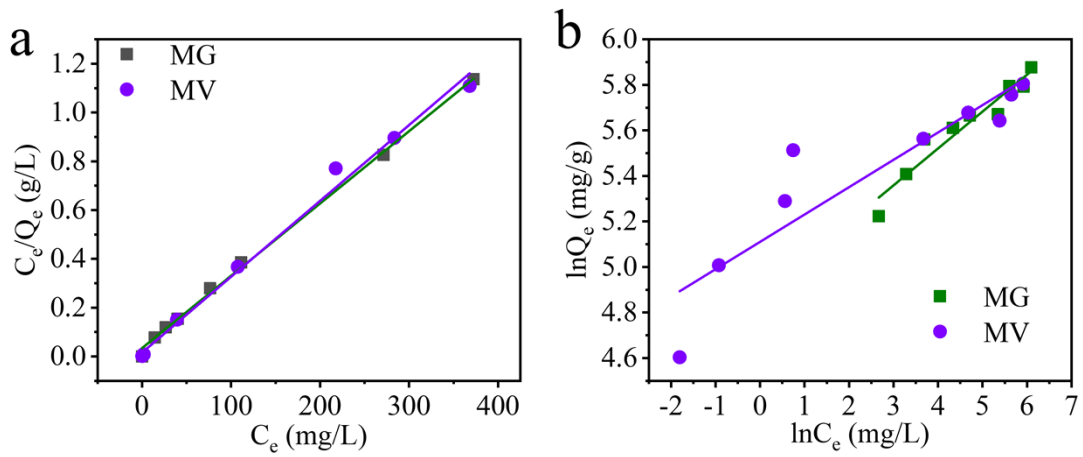
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258 **Fig. S6.** The fitting model of isothermal adsorption of MG and MV dyes: (a) Langmuir model; (b)

259 Freundlich models.

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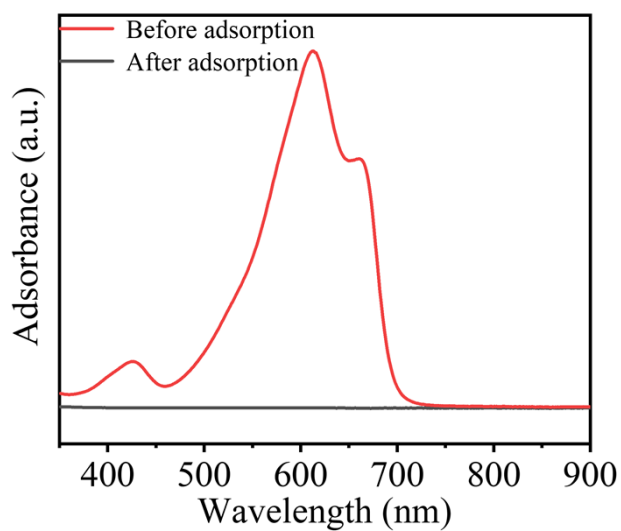
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290 **Fig. S7.** Adsorption of mixed dye solution of MB, MG and MV.

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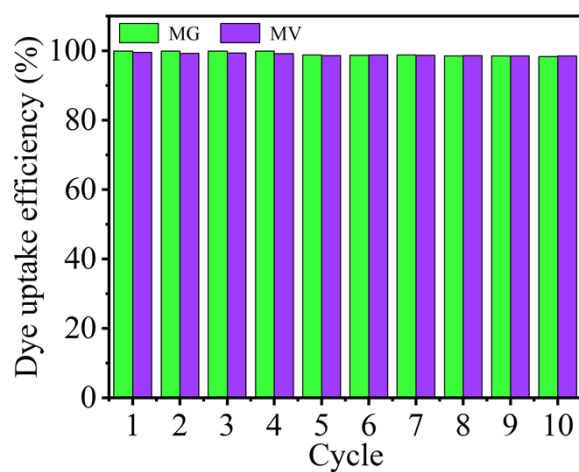
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321 **Fig. S8.** The cyclic adsorption performance of PCP-IDA adsorbent for MG and MV dyes.

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352 Reference

- 353 1. Z. Cherifi, B. Boukoussa, A. Mokhtar, M. Hachemaoui, F. Z. Zeggai, A. Zaoui, K. Bachari
354 and R. Meghabar, Preparation of new nanocomposite poly(GDMA)/mesoporous silica and its
355 adsorption behavior towards cationic dye, *React. Funct. Polym.*, 2020, **153**, 104611.
- 356 2. M. Arslan, Preparation and Application of Glycidyl Methacrylate and Methacrylic Acid
357 Monomer Mixture-Grafted Poly(ethylene terephthalate) Fibers for Removal of Methylene
358 Blue from Aqueous Solution, *J. Appl. Polym. Sci.*, 2011, **119**, 3034-3042.
- 359 3. R. K. Sharma and R. Kumar, Functionalized cellulose with hydroxyethyl methacrylate and
360 glycidyl methacrylate for metal ions and dye adsorption applications, *Int. J. Biol. Macromol.*,
361 2019, **134**, 704-721.
- 362 4. D. Husaain, M. Najam-ul-Haq, A. Saeed, F. Jabeen, M. Athar and M. N. Ashiq, Synthesis of
363 poly GMA/DVB and its application for the removal of Malachite Green from aqueous
364 medium by adsorption process, *Desalin. Water Treat.*, 2015, **53**, 2518-2528.
- 365 5. S. Y. Yu, J. L. Cui, H. Jiang, C. S. Zhong and J. Meng, Facile fabrication of functional
366 chitosan microspheres and study on their effective cationic/anionic dyes removal from
367 aqueous solution, *Int. J. Biol. Macromol.*, 2019, **134**, 830-837.
- 368 6. Q. Zhou, Q. Gao, W. J. Luo, C. J. Yan, Z. N. Ji and P. Duan, One-step synthesis of amino-
369 functionalized attapulgite clay nanoparticles adsorbent by hydrothermal carbonization of
370 chitosan for removal of methylene blue from wastewater, *Colloid Surf. A*, 2015, **470**, 248-257.
- 371 7. X. Zhang, C. Cheng, J. Zhao, L. Ma, S. D. Sun and C. S. Zhao, Polyethersulfone enwrapped
372 graphene oxide porous particles for water treatment, *Chem. Eng. J.*, 2013, **215**, 72-81.
- 373 8. A. S. Eltaweil, E. M. Abd El-Monaem, G. M. El-Subriti, M. M. Abd El-Latif and A. M.
374 Omer, Fabrication of UiO-66/MIL-101(Fe) binary MOF/carboxylated-GO composite for
375 adsorptive removal of methylene blue dye from aqueous solutions, *RSC Adv.*, 2020, **10**,
376 19008-19019.
- 377 9. S. Alvarez-Torrellas, M. Boutahala, N. Boukhalfa and M. Munoz, Effective Adsorption of
378 Methylene Blue dye onto Magnetic Nanocomposites. Modeling and Reuse Studies, *Appl. Sci-
379 Basel.*, 2019, **9**, 4563.
- 380 10. C. Li, X. J. Wang, D. Y. Meng and L. Zhou, Facile synthesis of low-cost magnetic biosorbent
381 from peach gum polysaccharide for selective and efficient removal of cationic dyes, *Int. J.
382 Biol. Macromol.*, 2018, **107**, 1871-1878.
- 383