Waste *Bombyx Mori* Silk Textiles as Reuseable Bio-Adsorbents for Methylene Blue Dye Removal and Oil-Water Separation

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Tables

Table S1: FTIR adsorption bands for silk fibroin conformations [1]

Fibroin Conformation	Amide I /cm^{-1}	Amide II /cm^{-1}	Amide III /cm^{-1}
β-sheet	1622–1637	1515–1525	1265
α-helix	1656–1662	1545	1240
Random coil	1638–1655	1535–1545	1235

Table S2: Adsorption capacities of silk textiles and yarns measured experimentally at 150 ppm methylene blue(MB) concentration by measuring the weight gain by the fabric.

Sample	*PW*	**PW-T**	*PWY*	*NW*	**NW-T**	**NWY**
Q_e (mg/g)	75.9 (±10.7)	93.1 (±38.4)	99.0 (±50.1)	60.2 (±10.8)	66.7 (±7.3)	67.5 (±28.7)

Table S3: MB removal data for unraveled plain-woven fabrics (PWY) at 10 ppm MB concentration.

Time	Removal efficiency (%)	Qe (mg/g)
1	93.2	2.93
3	95.9	3.02
10	96.8	3.05
30	96.9	3.05

Table S4: Separation factor (R_L) calculated for different initial MB concentrations.

ppm(mg/L)	R_L
5	0.99999999
10	0.99999998
25	0.999999951
50	0.999999901
100	0.999999803
150	0.999999704
300	0.999999408
600	0.999998815
800	0.999998421

Table S5: Removal efficiencies of PWY for MB concentrations from 5- 800 ppm.

C_e (mg/L)	Removal efficiency (%)
5	98.5
10	98.0
25	98.6
50	98.9
100	97.8
150	98.1
300	96.6
600	76.6
800	71.4

Table S6: Literature on silk adsorption capacities

Material description	Removal capacity	Reference
Silk fibroin(SF)/Graphene oxide blend films	382 mg g^{-1}	[2]
Keratin/SF aerogel	164 mg/g	[3]
Regenerated SF/cellulose composite	172 mg/g	[4]
SF/ Gold nanoparticle composite	313 mg g^{-1}	[5]
SF powder	20.6 mg/g	[6]
SF/Chitosan composite	89.0 mg/g	[7]
SF/orange peel foam	114 mg/g	[8]
Silk yarns unraveled from fabrics	145 mg/g	*This study

Table S7: Adsorption and desorption efficiency of PWY for each cycle of the reusability test.

Cycle	1	2	3	4	5	6	7	8	9	10	Average
Desorption efficiency (%)	38.6	31.3	30.2	44.1	32.0	24.6	21.8	35.4	24.4	17.9	30.0
Adsorption efficiency (%)	95.2	96.7	97.5	96.0	95.4	93.3	92.1	92.8	92.5	93.5	94.5

Figures

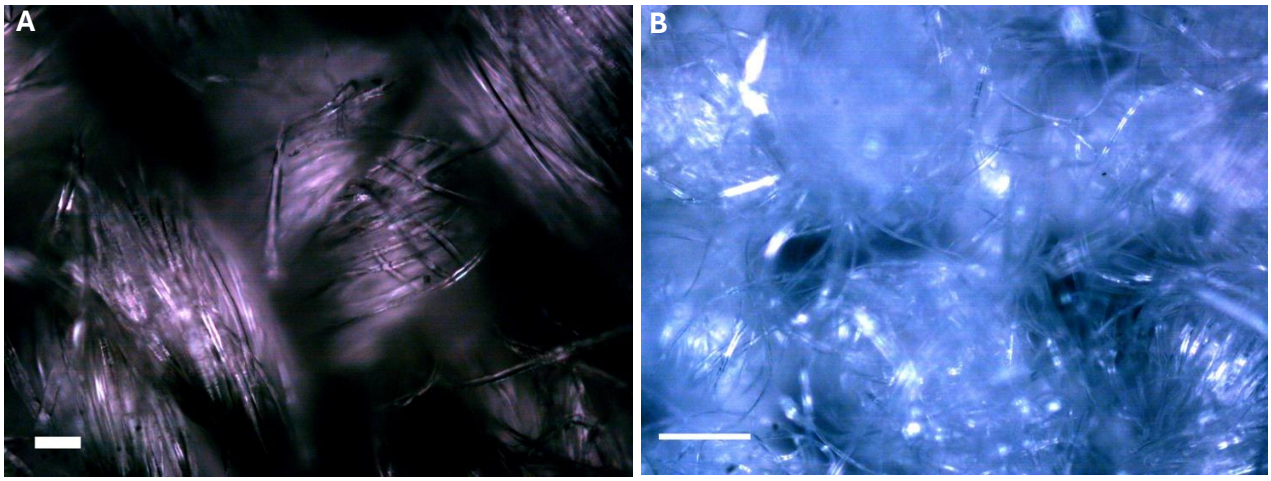


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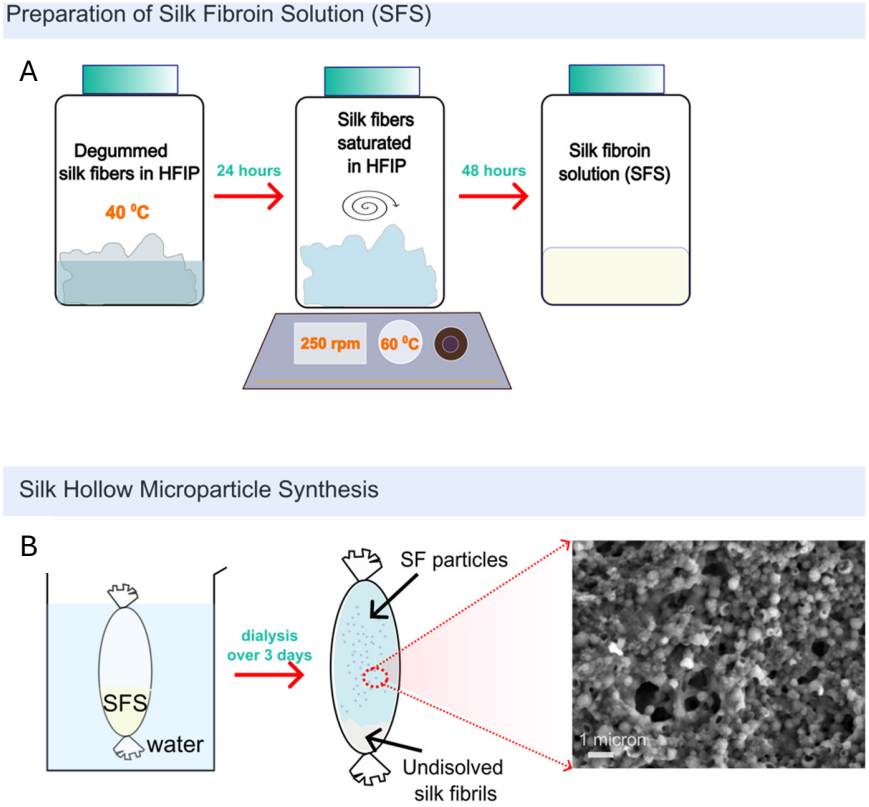


Figure S2: Graphical representation of A. preparation of silk fibroin solution and B. synthesis steps for silk hollow microparticle formation and SEM image of the silk particles.

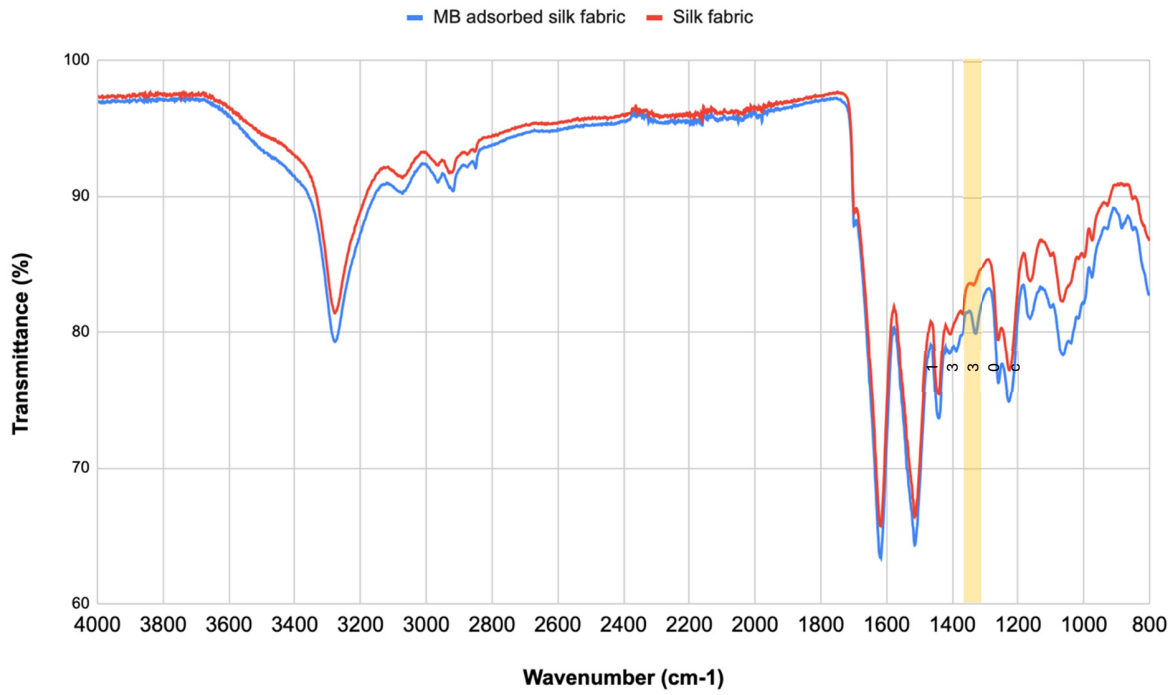


Figure S3: FTIR spectra of silk fabrics before and after MB dye adsorption

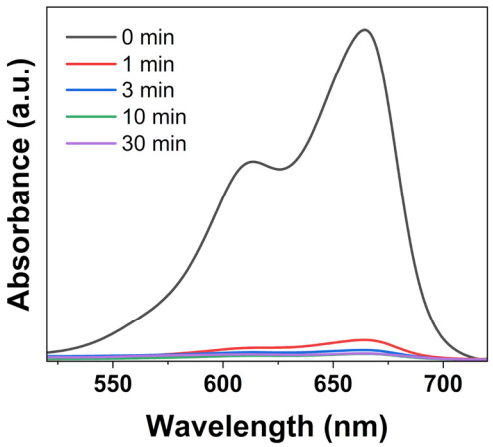
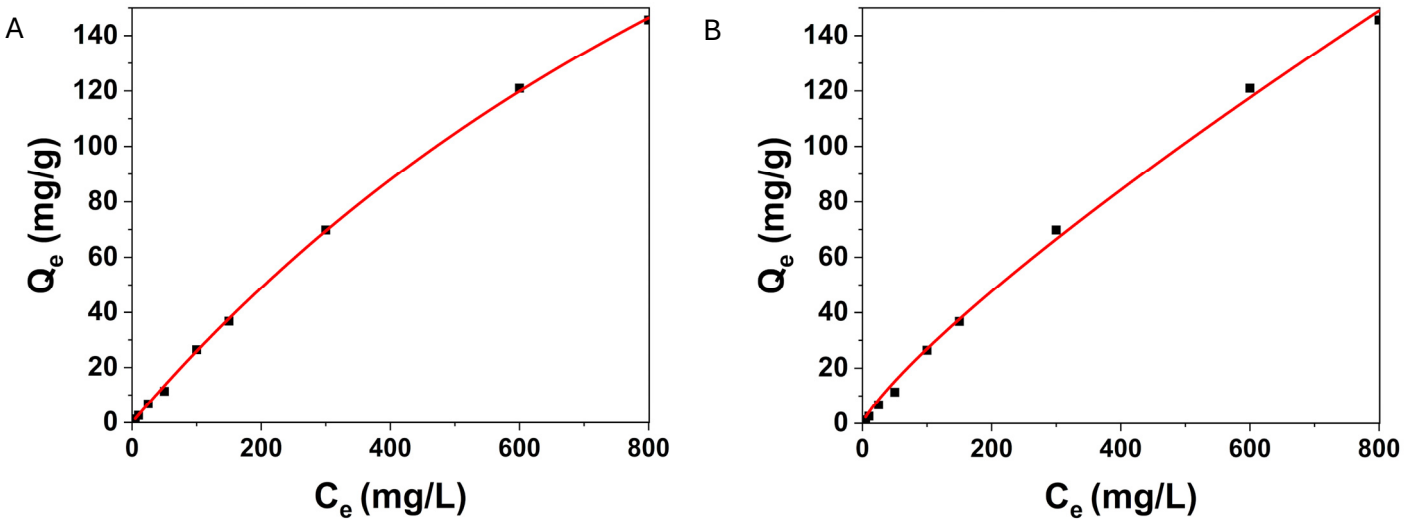


Figure S4: UV-visible spectrum depicting MB removal data for unraveled PWY fabrics at 10 ppm methylene blue concentration.



$Q_e = (Q_m \times K \times C_e)/(1+K \times C_e)$

$\log Q_e = \log K_F + (1 / n) \log C_e$

Figure S5: Nonlinear fitting for Q_e vs. C_e data points using, A. Langmuir and B. Freundlich model (respective equations are provided underneath the plots).

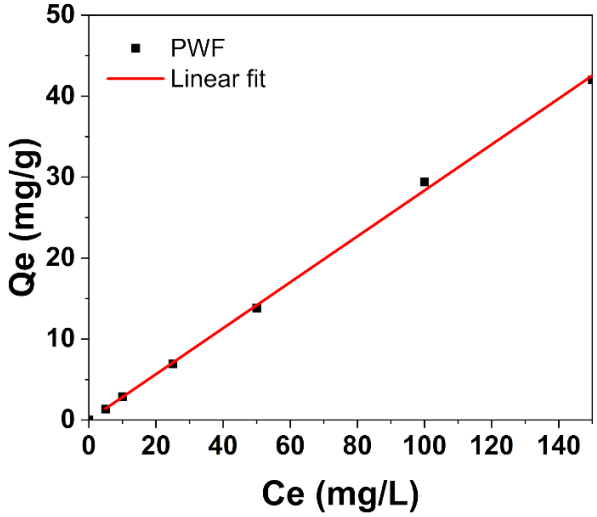


Figure S6: Linear fitting for Q_e vs. C_e data points for concentrations up to 150 ppm suggesting infinite adsorption capacity.

References

1. Zhang, X.; Pan, Z. Microstructure Transitions and Dry-Wet Spinnability of Silk Fibroin Protein from Waste Silk Quilt. *Polymers (Basel)* **2019**, *11*, doi:10.3390/polym11101622.
2. Martis, L. J., Parushuram, N., & Sangappa, Y. (2022). Preparation, characterization, and methylene blue dye adsorption study of silk fibroin–graphene oxide nanocomposites. *Environmental Science: Advances*, *1*(3), 285-296.

3. Xing, X., Zhang, X., Feng, Y., & Yang, X. (2023). Adsorption of methylene blue dye on feather keratin/silk fibroin porous aerogels. *Journal of the Taiwan Institute of Chemical Engineers*, 105298.
4. Salama, A. (2020). Cellulose/silk fibroin assisted calcium phosphate growth: Novel biocomposite for dye adsorption. *International Journal of Biological Macromolecules*, *165*, 1970-1977.
5. Parushuram, N., Ranjana, R., Harisha, K. S., Shilpa, M., Narayana, B., Neelakandan, R., & Sangappa, Y. (2022). Silk fibroin and silk fibroin-gold nanoparticles nanocomposite films: sustainable adsorbents for methylene blue dye. *Journal of Dispersion Science and Technology*, *43*(8), 1161-1176.
6. Xiao, S., Wang, Z., Ma, H., Yang, H., & Xu, W. (2014). Effective removal of dyes from aqueous solution using ultrafine silk fibroin powder. *Advanced powder technology*, *25*(2), 574-581.
7. Salama, A., Shoueir, K. R., & Aljohani, H. A. (2017). Preparation of sustainable nanocomposite as new adsorbent for dyes removal. *Fibers and Polymers*, *18*, 1825-1830.
8. Campagnolo, L., Morselli, D., Magrì, D., Scarpellini, A., Demirci, C., Colombo, M., ... & Fragouli, D. (2019). Silk fibroin/orange peel foam: an efficient biocomposite for water remediation. *Advanced Sustainable Systems*, *3*(1), 1800097.