

# Hierarchically 3-D porous Structure of Silk Fibroin-Based Bio-Composite Adsorbent for Water Pollutant Removal

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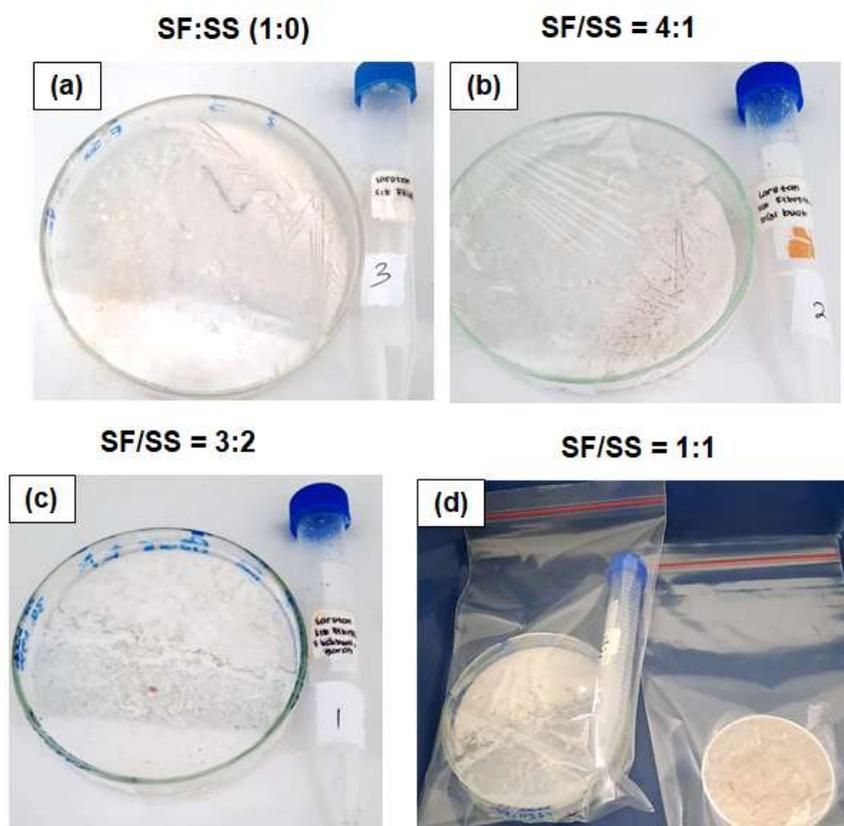
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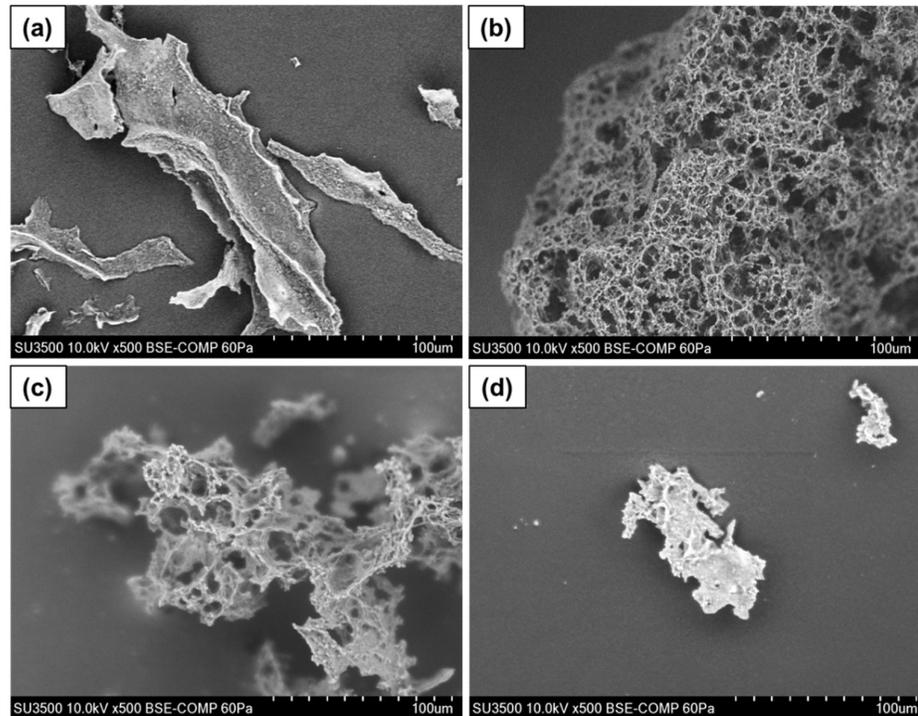
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**Figure S1.** Physical appearance of SF:SS composite powders at various composition.

**Figure S1** shows the freeze-dried product of SF:SS at various composition. The viscous solutions of SF:SS were stored in a freeze-drying flask and rotating the flask in a bath, called a shell freezer, which is cooled by mechanical refrigeration. The SF:SS product was frozen slowly, cycled up and down in temperature. This cycling process is called annealing and the freezing temperature was conducted at  $-100^{\circ}\text{C}$ . Finally, SF:SS composite powders with various composition were successfully obtained.



**Figure S2.** SEM images of SF:SS at various composition. (a) SF:SS=1:0; (b) SF:SS= 4:1; (c) SF:SS=3:2 and (d) SF:SS=1:1. (in Low magnification).

**Figure S2** shows the surfaces of 3D scaffolds of the sample at the composition (b) 4:1 and (c) 3:2 which exhibit a morphology of irregular pore structures with some regions of interconnectivities when magnified. The surfaces of the pore walls were inhomogeneous and smooth (see **Fig S2 (b)**). It can be seen that the pore formation was markedly affected by the compositions of SF:SS. When the amount of soursop seed (SS) applied to the RSF solution was lower (i.e., SF:SS =1:0 and SF:SS = 1:1), the formed 3D scaffolds showed no pore structure formation. However, when the amount of soursop seed (SS) was higher (i.e., 4:1), the obtained 3D scaffolds of SF:SS exhibited inhomogeneous and interconnected porous structure.

Examples of temporal removal mechanism of CV and  $\text{Cu}^{2+}$  monitored by UV-vis spectroscopy are shown in Figure S3 and Figure S4 as follows:

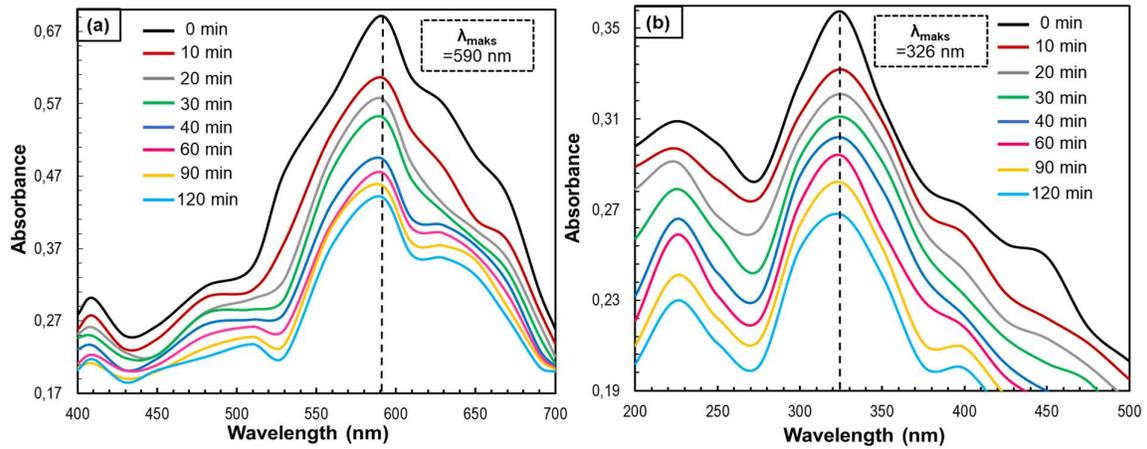


Figure S3. UV-Vis absorption spectra recorded from adsorption experiment using SF:SS (4:1) composites for (a) crystal violet (C<sub>0</sub>= 10 ppm) and (b) Cu<sup>2+</sup> metal ions (C<sub>0</sub>= 10 ppm).

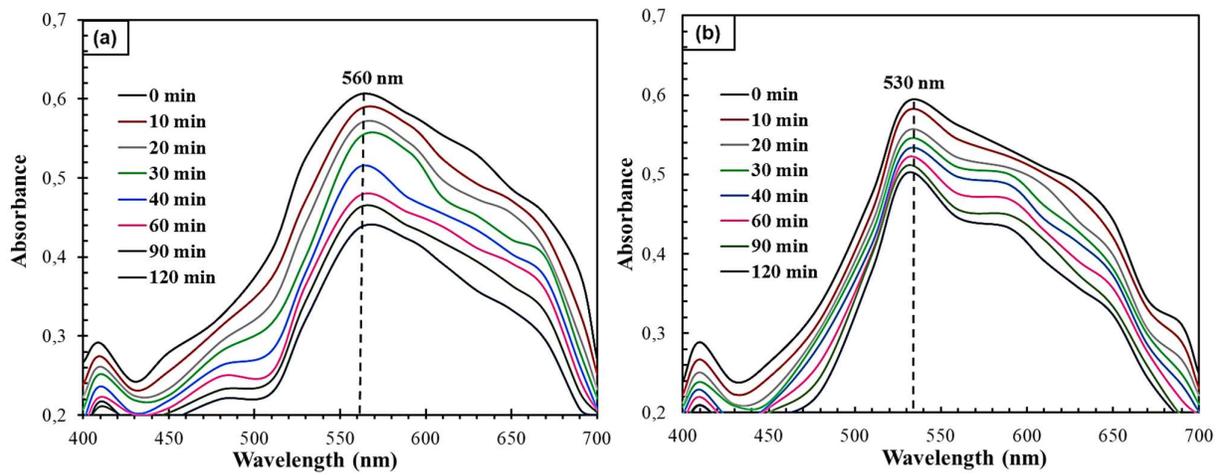
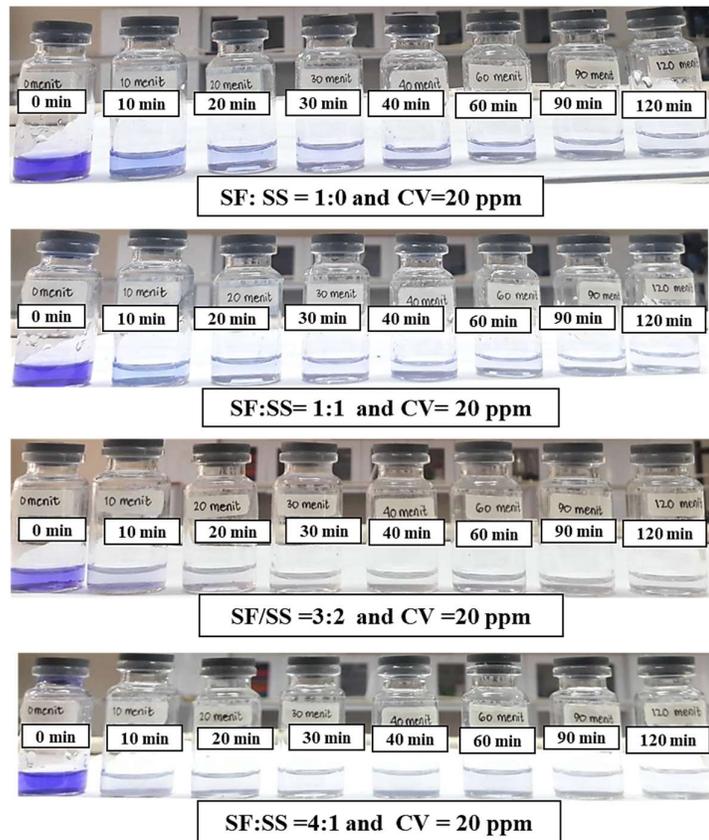


Figure S4. UV-Vis absorption spectra recorded from adsorption experiment using SF:SS (3:2) and (1:1) composites for crystal violet (C<sub>0</sub> = 10 ppm).



**Figure S5.** Photograph showing adsorption of the cationic dye (crystal violet) by the dried SF:SS at various compositions.