Removal of methylene blue from aqueous solution by mesoporous silicalite-1 synthetized using carboxymethyl cellulose as template

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Removal of methylene blue from aqueous solution by mesoporous silicalite-1 synthetized using carboxymethyl cellulose as template

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Abstract

In the present work, we have developed a mesoporous silicalite-1 using CMC as a template for the removal of MB from aqueous solution. The synthesized silicalite-1 were characterised using scanning electron microscopy (SEM), transmission electron microscopy (TEM), Fourier transform infrared spectroscopy (FT-IR), X-ray diffraction (XRD), thermogravimetric analysis (TGA), Energy-dispersive X-ray spectroscopy (EDX) and N_2 adsorption-desorption isotherm (BET). XRD and FTIR analysis confirmed the formation of crystallinity and development of MFI structure in the mesoporous silicalite-1. The adsorption of MB dye on mesoporous silicalite-1 was conducted by batch adsorption method. The effect of various parameters such as adsorbent dosage, initial dye concentration, contact time and temperature on the dye uptake ability of silicalite-1 was investigated. The operating parameters for the maximum adsorption are silicalite-1 dosage (0.1 wt%), contact time (240 min), initial dye concentration (10 ppm) and temperature (30°C). The MB dye removal onto mesoporous silicalite-1 followed pseudo-second-order kinetic and Freundlich isotherm. The silicalite-1 exhibits 86% removal efficiency

even after six adsorption-desorption cycle. Therefore, the developed mesoporous silicalite-1 is an effective eco-friendly adsorbent for MB dye removal from aqueous environment.

Keywords: *Mesoporous; Silicalite-1; Methylene blue; Adsorption; Kinetics*

Introduction

In the last few decades, the quality of water is declining in an alarming rate. Dyes are one of the major pollutants which deteriorate our water resources such as river, lake etc.[1-3] Synthetic dyes constitute about 90% of the total dye used in various industries such as paints, textiles, paper, printing, plastics and cosmetics [4-6]. Azo dye, anthraquinones dyes, phthalocyanines and polymethines are the common examples of synthetic dyes which is frequently employed for different application [7, 8]. Dye dissemination into water bodies is not only aesthetically unpleasant but also causes some major issues such as low dissolved oxygen (DO) level, high BOD level and obstruction for sunlight transmission through water[8-11]. Thus, it adversely affects the aquatic organism[11]. Dyes effluent also cause serious health risk to humans such as hypersensitivity, allergy, asthma, kidney dysfunction, liver and brain disorder [12-14]. Methylene blue (MB) are recalcitrant pollutant with cariogenic and mutagenic nature. It is generally used for dying cotton, silk and wool. The structural formula of MB is shown in figure 1. It causes several health issues such as eye burns, skin irritation, vomiting and nausea to humans [15-17].

The excessive use of dye raises concern and hence its removal from aqueous effluent has become a challenging topic for researchers and scientists. Researchers have designed several novel materials for dye removal by adopting technologies such as adsorption, coagulation, oxidation, filtration, ultrafiltration etc.[18-20]. Due to simplicity, low-cost and high efficiency, adsorption is considered as one of the most effective method for dye removal [21, 22]. Activated carbon and nanoparticle are effective adsorbent for dye removal applications, however due to high cost and low efficiency its large-scale usage is limited [23]. Recently,

various low-cost and eco-friendly adsorbent were reported for removing dyes from wastewater including zeolites, kaolin, clay, montmorillonite, activated carbon, agricultural waste etc.[23-28] Silicalite-1 belongs to family of zeolite and possesses aluminium free MFI type framework. High surface area and porous nature enable it to use in different application such as adsorption, catalyst and fine chemical industrial applications [29, 30].

Jing et al. [31] fabricated TiO_2 loaded silicalite-1 for degradation of rhodamine B. They reported that adsorbent dosage plays a key role in dye removal efficiency as they observed a significant increment in dye removal efficiency with adsorbent dosage. Cheng et al. [32] reported that GO/silicalite-1 composite is as an effective adsorbent for rhodamine B. The maximum adsorption capacity (56.55 mg/g) was observed at pH=3 within 30 min of contact time. Recently, our team fabricated sodium alginate modified silicalite-1 for rhodamine B removal. The modified silicalite-1 displayed high removal efficiency than conventional silicalite-1 probably due to the high surface area and pore diameter. The reusability test was also conducted to check the industrial advantage. The result proved that sodium alginate modified silicalite-1 exhibits good regeneration capacity [33].

There is only limited research available for dye adsorption studies on silicalite-1 from wastewater. So, in this work, we made an attempt to evaluate the dye (MB) adsorption capacity of s carboxymethyl cellulose (CMC) modified silicalite-1. The effect of different parameters such as initial dye concentration, contact time, temperature, dosage on the adsorption process was also discussed in details. The adsorption kinetics and isotherm on adsorption process is also presented.

Materials and methods

Chemicals

Tetrapropylammonium hydroxide ($(CH_3CH_2CH_2)_4NOH$; TPAOH) and tetraethylorthosilicate ($C_8H_{20}O_4Si$; TEOS) were purchased from Sigma Aldrich Co. Ltd

(India). Carboxymethyl cellulose ($[C_6H_7O_2(OH)_x(OCH_2COONa)_y]_n$, CMC), and methylene blue ($C_{16}H_{18}Cl\,N_3S$, MB) were purchased from Merck Co. Ltd (India). All the chemicals were used as such without further purification.

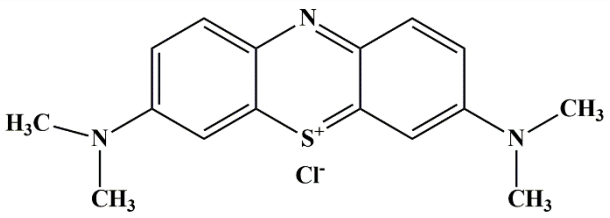
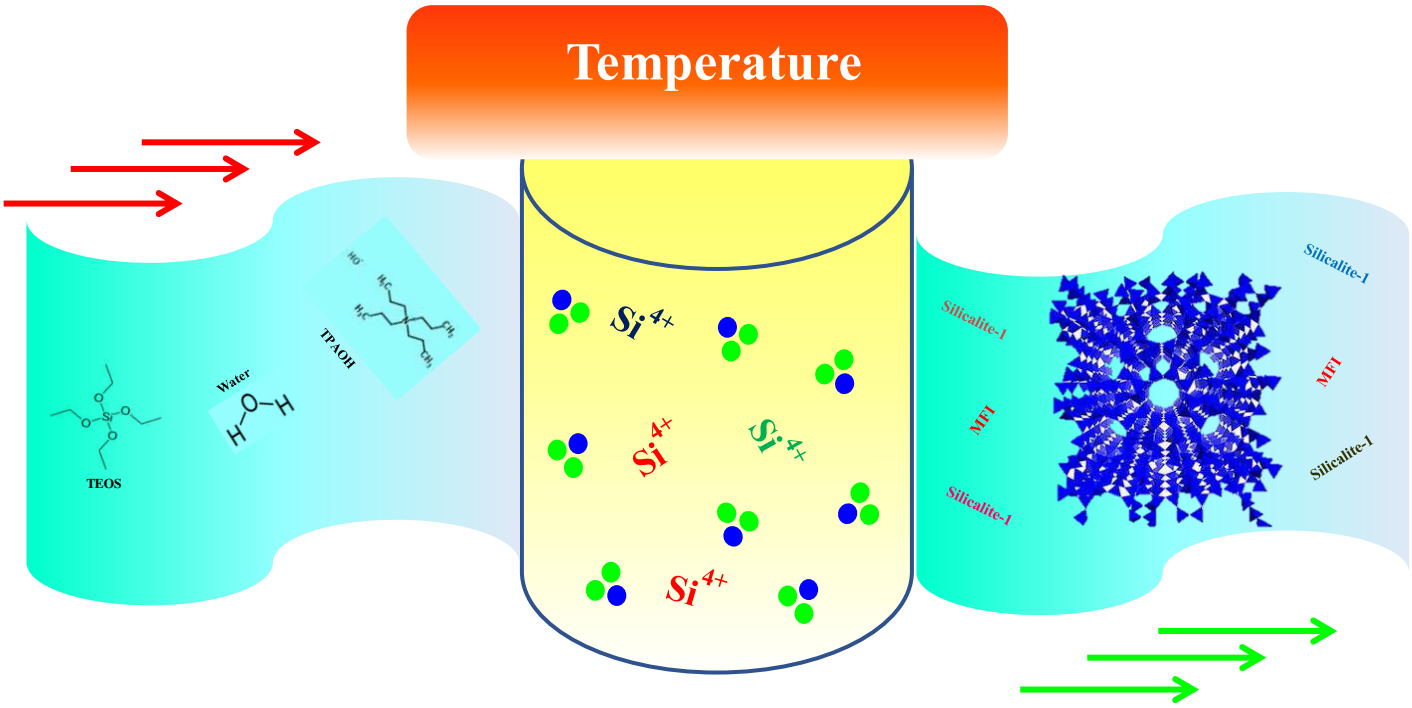


Figure 1. Structural formula of methylene blue

Synthesis of mesoporous silicalite-1

The mesoporous silicalite-1 was synthesized by adopting hydrothermal method. In a typical synthesis, 3.49 g of tetra propylammonium hydroxide (TPAOH) and 3.1 g of tetraethyl orthosilicate (TEOS) were mixed in distilled water under constant stirring for 5 h. After wards, 0.2 g of CMC is mixed to the above solution with constant stirring. The solution was concentrated in a rotavapor at 80 ^oC for 30 min to form a viscous solution. The resulting sol was introduced into a stainless autoclave for crystallisation at 80 ^oC for 24 h followed by high temperature treatment at 175 ^oC for 8 h. The obtained product was filtered, washed with water, dried and finally calcined at 550 ^oC for 5 h to remove the organic components. For comparison purpose, conventional silicalite-1 was prepared with similar procedure without adding template, CMC [34, 35].



Scheme 1. Schematic diagram of silicalite-1

Characterization

Powder X-ray diffraction (XRD) patterns in the range of 5-50º were obtained with a Rigaku Miniflex 600 diffractometer using CuKα radiation at a scanning speed of 4º/min. The characteristic vibration bands were identified by Jasco 4700 FTIR spectrometer with KBr pellet. Scanning electron microscopic (SEM) images were recorded by using a Hitachi SU6600 Variable Pressure Field Emission Scanning Electron Microscope (SEM). Transmission electron microscopic images (TEM) were taken with a JEOL JEM-2100 transmission electron microscope operated at an accelerating voltage of 200 kV. The thermal stability of the uncalcined sample was analysed by thermogravimetric (TG) analysis with Q50 instrument at a heating rate of 10ºC/min in nitrogen atmosphere. N_2 adsorption/desorption isotherm were employed to find out the surface area and pore size distributions at 77K with Micromeritics Gemini V-2380 surface area analyser. Before measurement the sample was outgassed at 573K for 3h. The Si/Al ratio of the samples was determined by electron dispersive spectroscopy (EDX) using JEOL JED-2300 instrument.

Adsorption experiment

For adsorption studies, stock solution of MB (100 ppm) was prepared in distilled water. 10, 20, 30, 40 and 50 ppm of dye solution was prepared by diluting the stock solution. The pH of the dye solution was adjusted using HCl and NaOH. The effect of the environmental parameters on dye removal was studied by varying adsorbent loading (0.02-0.10 wt%), dye concentration (10-50 ppm), contact time (20-330 min) and temperature (30-60°C). Batch adsorption experiment was employed to find out the adsorption capacity of MB onto the mesoporous silicalite-1 sample. The dye uptake performance was assessed by soaking 0.10 wt% of silicalite-1 in 50 mL of MB dye at different temperature. After predetermined time intervals the sample was withdrawn, filtered and the solution was measured using UV-Vis spectrophotometer at $\lambda_{max} \sim 668$ nm. From the calibration curve, the concentration of dye in the sample is noted. The experiments were done in triplicate and the average value is reported. The percentage removal and equilibrium concentration were calculated as follows

$$q_e = \frac{(C_0 - C_e)V}{W} \qquad (1)$$

$$R = \frac{C_0 - C_e}{C_0} \times 100(\%) \qquad (2)$$

Where C_0 and C_e are the initial and equilibrium dye concentration (mg L^{-1}), V is the volume of dye solution (L); and W is weight (g) of the adsorbent.

Desorption experiment

Desorption experiment of MB on silicalite-1 was performed by immersing 0.10 wt% of mesoporous silicalite-1 sample in 50 mL of MB solution (10 ppm, temperature- 30°C). After complete equilibrium time, the silicalite-1 was separated and eluted with 0.1 mol/L HCl to desorb the dye. The silicalite-1 was later washed with distilled water, dried and used for next cycle [36].

Result and discussion

Figure 2 shows the XRD patterns of conventional and mesoporous silicalite-1. Both silicalite-1 exhibits typical MFI framework structure with five characteristic peaks at 7.98º, 8.82º, 23.18º, 24.02º and 24.46º corresponds to (101), (020), (503), (151) and (303) refection respectively. However, with the addition of template the peak intensity of silicalite-1 slightly reduces, probably due to the development of mesopores. Thus, CMC template is efficient to generate mesoporosity in silicalite-1 without compromising its crystallinity [35, 37]. To understand the chemical composition of membrane, we performed FTIR spectroscopy. The FTIR spectra of conventional and mesoporous silicalite-1 are displayed in Figure 3. The strong absorption band at 550 cm^{-1} is attributed to double five membraned ring which is characteristic of MFI framework. The band at 796 cm^{-1} and 1105 cm^{-1} are assigned to Si-O-Si external symmetric stretching (SiO_4 tetrahedra) and Si-O-Si internal asymmetric stretching (SiO_4 tetrahedra). The band at 1230 cm^{-1} is attributed to external asymmetric stretching of Si-O bond. A weak band observed at 1640 cm^{-1} could be due to the adsorbed water present in the sample. Finally, the absorption band at 462cm^{-1} is due to Si-O-Si rocking. FTIR studies were thus complimentary to XRD study and confirmed the successful synthesis of silicalite-1 [33, 38].

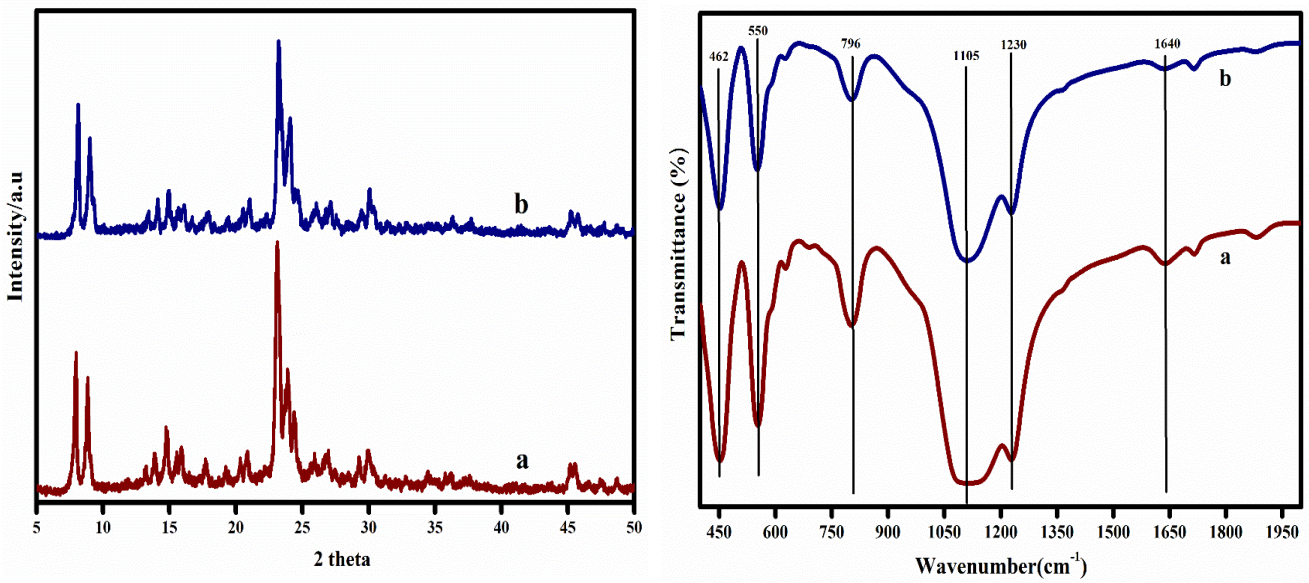


Figure 2. Powder XRD patterns and FTIR spectra of conventional and mesoporous silicalite-1

The morphology of conventional and mesoporous silicalite-1 was evaluated by SEM and TEM analysis (Figure 3 and 4). The SEM image of mesoporous silicalite-1 (Figure 3) shows a rough surface with sphere like particle in the range of 10-30 nm. The result thus suggests the formation of mesopore in the system, which is attributed to the removal of templates. TEM images further confirmed that mesoporous silicalite-1 have pore diameter in the range of 10-30 nm. The prominent lattice fringes seen in the TEM images imply a high crystallinity for the sample and are complementary to the previous reported work [39] (Figure. 4). The elemental composition of mesoporous silicalite-1 is detected using EDAX measurement. The analysis indicates that the synthesized sample contains only Si and O, thus confirming the product is silicalite-1. N_2 adsorption/desorption isotherm were employed to characterize the surface area and porosity of the sample. Figure 5(I) illustrates the N_2 adsorption/desorption isotherm of conventional and mesoporous silicalite-1. As shown in Figure 5a, the conventional silicalite-1 belongs to type 1 isotherm with major uptake of gas at a relative low pressure ($P/Po<0.2$). This indicates the presence of only micropores in the system. However, mesoporous silicalite-1 exhibits a prominent hysteresis loop at $P/Po = 0.25$ to 0.9 due to the capillary condensation of nitrogen. The result thus indicates the co-existence of micro and mesopores in the modified sample. BJH (Barrett-Joyner-Halenda) pore size distribution is shown in Figure 5(II). It is evident from figure that conventional silicalite-1 has only micropores (< 2 nm) whereas the mesoporous silicalite-1 have pore diameter in the range of 10-30 nm, thus confirming the presence of mesopores. This study further confirms that mesoporosity is generated in CMC templated sample.

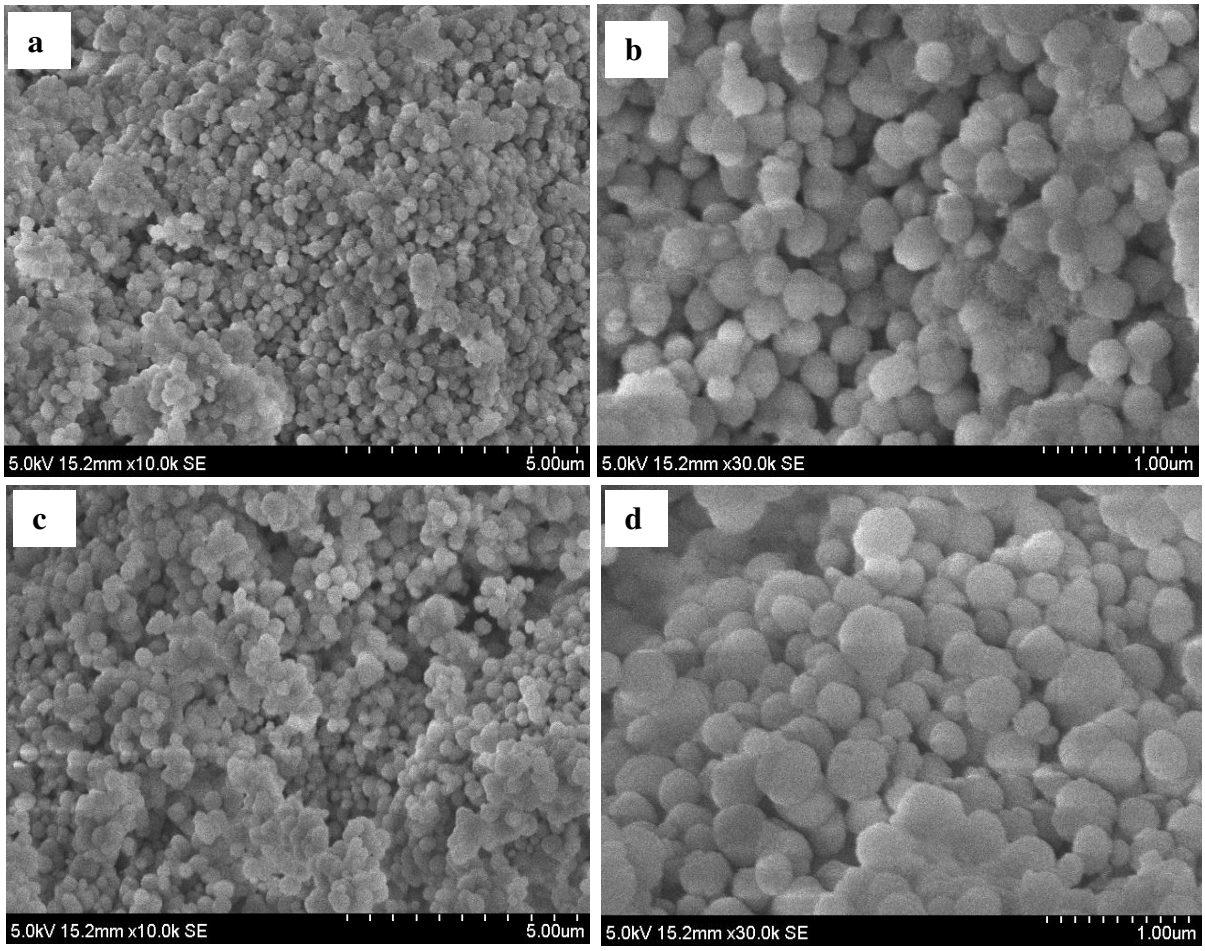


Figure 3. SEM micrographs of (a & b) conventional and (c & d) mesoporous silicalite-1

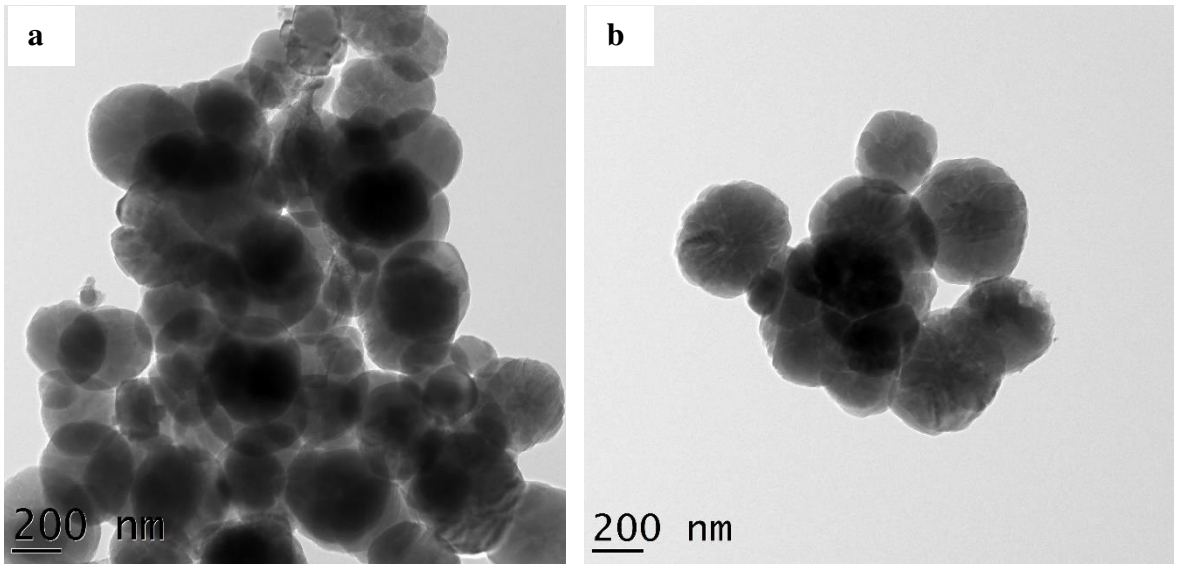


Figure 4. TEM images of mesoporous silicalite-1

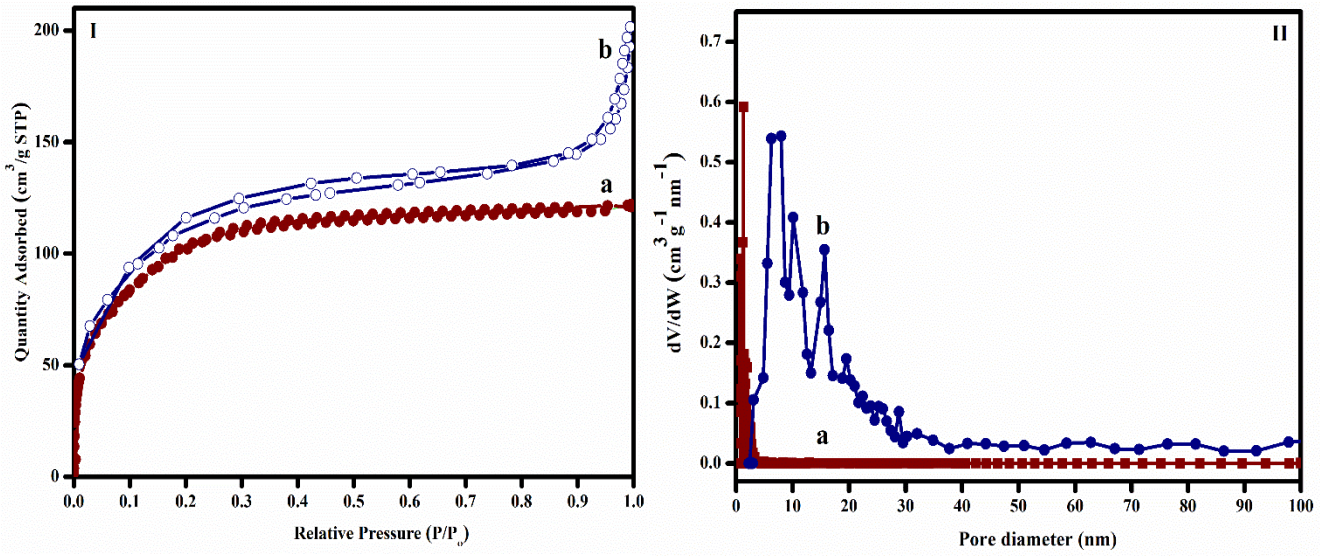


Figure 5. (I) N_2 adsorption isotherms of: a) conventional b) mesoporous silicalite-1; (II) pore size distribution of: a) conventional b) mesoporous silicalite-1

The thermal stability of conventional and mesoporous silicalite-1 is compared by thermogravimetric analysis (Figure 6). The conventional silicalite-1 showed 16% of total weight loss at 100-250°C and 300-500°C. This is attributed to the loss of water and TPAOH respectively from the silicalite-1 framework. In the case of mesopores silicalite-1, a third weight loss appears at 500°C which is due to the decomposition of template. As modified silicate possess higher weight loss (22%) than conventional silicalite-1, it is thermally more stable than conventional silicalite-1.

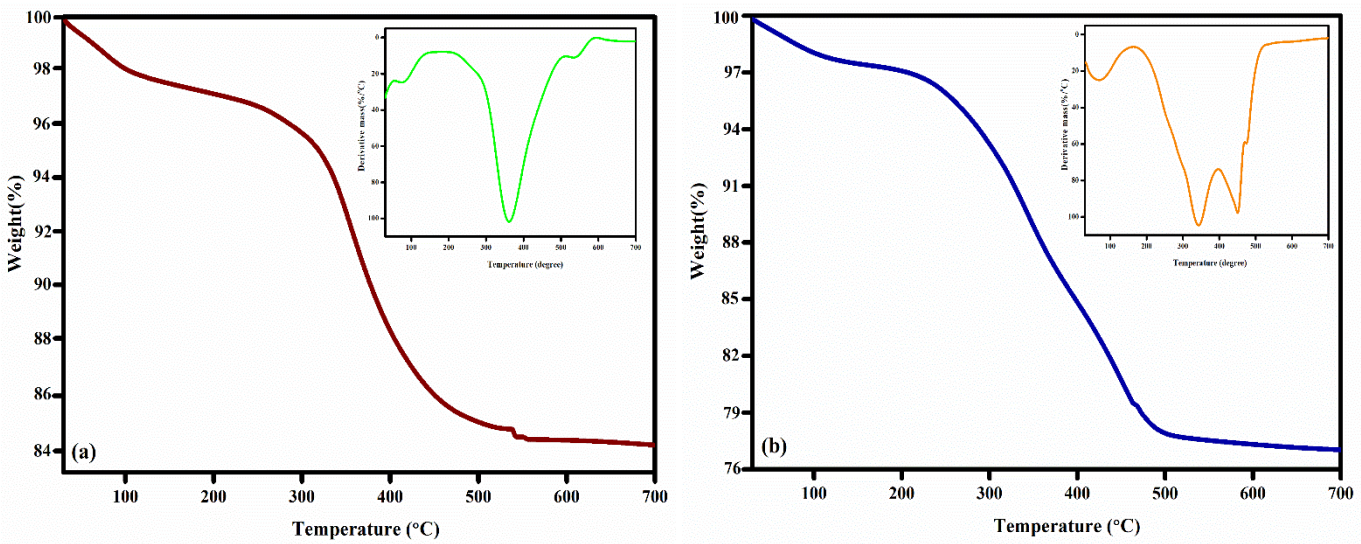


Figure 6. TGA curves of conventional and mesoporous silicalite-1 and the corresponding DTG curves are given at the inset

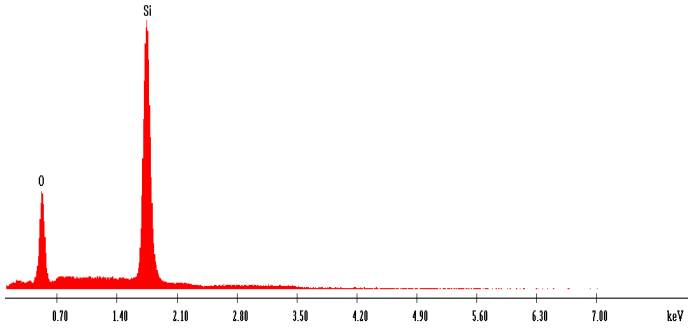


Figure 7. EDS image of mesoporous silicalite-1

Adsorption experiment of mesoporous silicalite-1 on methylene blue

Due to the presence of micro and mesoporous structure, modified silicalite-1 is expected to have good adsorption capacity for dye molecule. This prompted us to monitored the dye (MB) uptake ability of modified silicate. The influence of environmental parameters such as silicalite-1 dosage, temperature, initial dye concentration and contact time was also investigated in detail. The influence of silicate dosage on the MB removal was carried out by varying the silicalite-1 from 0.02 to 0.10 wt%. From Figure 8(a), it can clearly see that the dye adsorption capacity increases linearly with adsorbent dosage. High surface area along with the

247 presence of micro/mesoporous structure on silicalite-1 could have increases the affinity of dye
248 molecule to the adsorbent. Consequently, adsorption capacity increases and a maximum
249 adsorption capacity (4 mg/g) was achieved at 0.10 wt% of adsorption dosage. The relationship
250 between initial dye concentration and adsorption capacity of MB is shown in Figure 8(b). It is
251 evident from figure that when the initial dye concentration increases from 10 to 50 ppm, the
252 adsorption capacity increases from 4 to 16.45 mg/g. Similar results have also been reported by
253 Duman et al [40]. They observed that on increasing the initial dye concentration from 20 to 40
254 mg/L the adsorption capacity increases from 8 to 15 mg/g. Dong et al. [41] reported that
255 mesoporous SBA-15 exhibits high adsorption capacity when the initial concentration exceeds
256 230 mg/g. According to the authors high initial concentration provides a necessary driving
257 force for the diffusion of dye from aqueous to the solid phase. We have varied the immersing
258 time from 10-330 min and evaluate its effect on the MB adsorption was in the following
259 operational conditions initial dye concentration -10ppm, temperature-30°C and dosage-0.1
260 wt%. In the initial contact time, due to availability of large number of unoccupied active sites
261 on the surface of adsorbent, the rate of adsorption increases, Therefore, adsorption capacity
262 steeply rises in the initial phase of contact time. However, the rate of adsorption becomes
263 gradual 120 min and eventually at 240 min it reaches constant. This implies that the adsorption
264 site on the surface of silicalite-1 get saturated with contact time and after 240 min complete
265 occupation of adsorption site happens. Similar curve was noted by Mouni et al. they obtained
266 equilibrium at 120 min [42]. Figure 8(d) show the adsorption capacity at different temperature
267 (30 to 60°C). On increasing the temperature from 30 to 60°C, the adsorption capacity increases
268 from 4 to 7 mg/g, thus suggesting that the MB adsorption on silicalite-1 is endothermic. High
269 temperature tends to enhances the mobility of dye molecule. As a result, the dye molecule can
270 easily diffuse from the solution phase to the adsorbent phase. Therefore, high temperature

271 favours the MB adsorption on silicalite-1. Scheme 2 show the schematic representation of

272 mesoporous silicalite-1 will interact with MB dye molecule.

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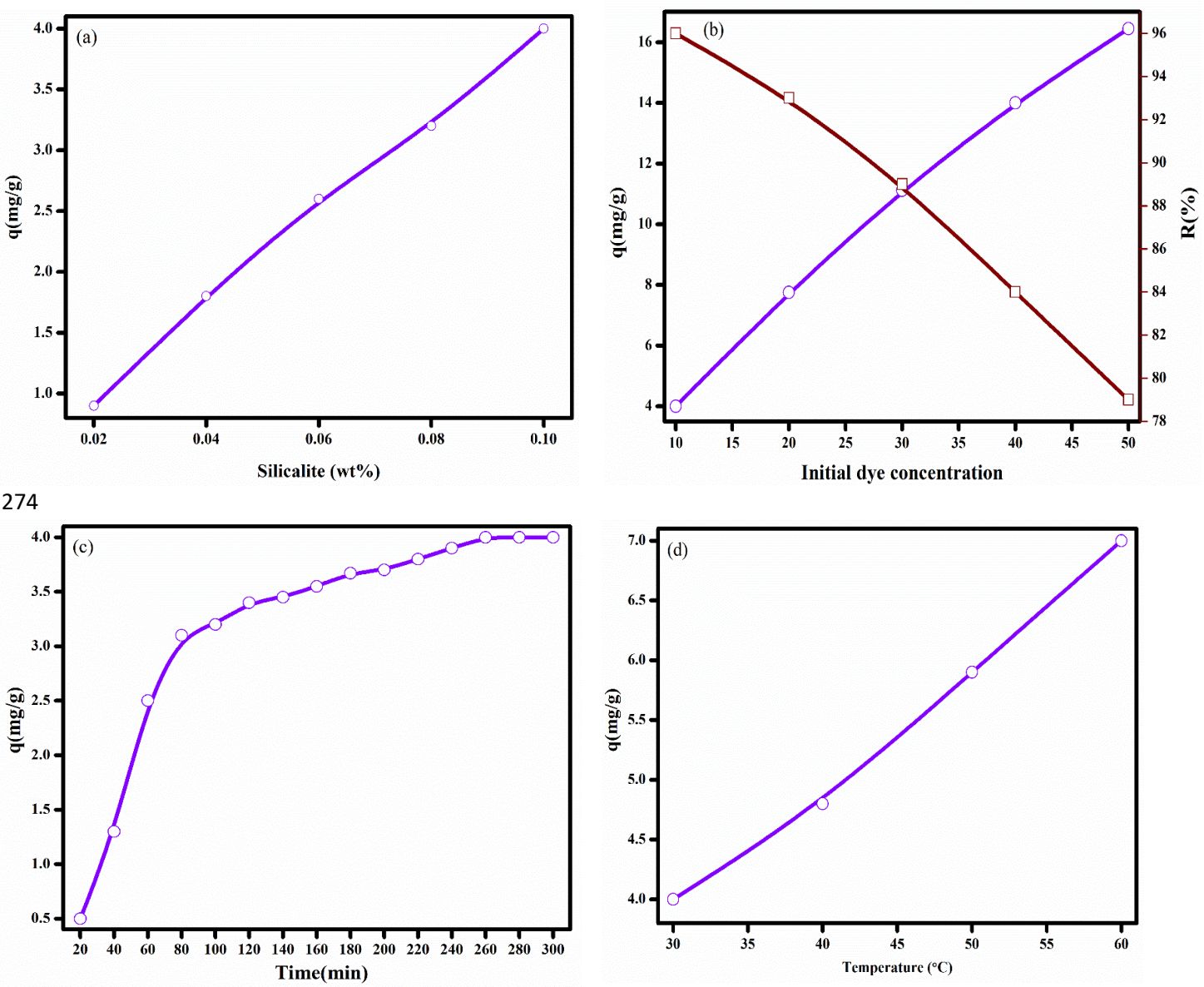

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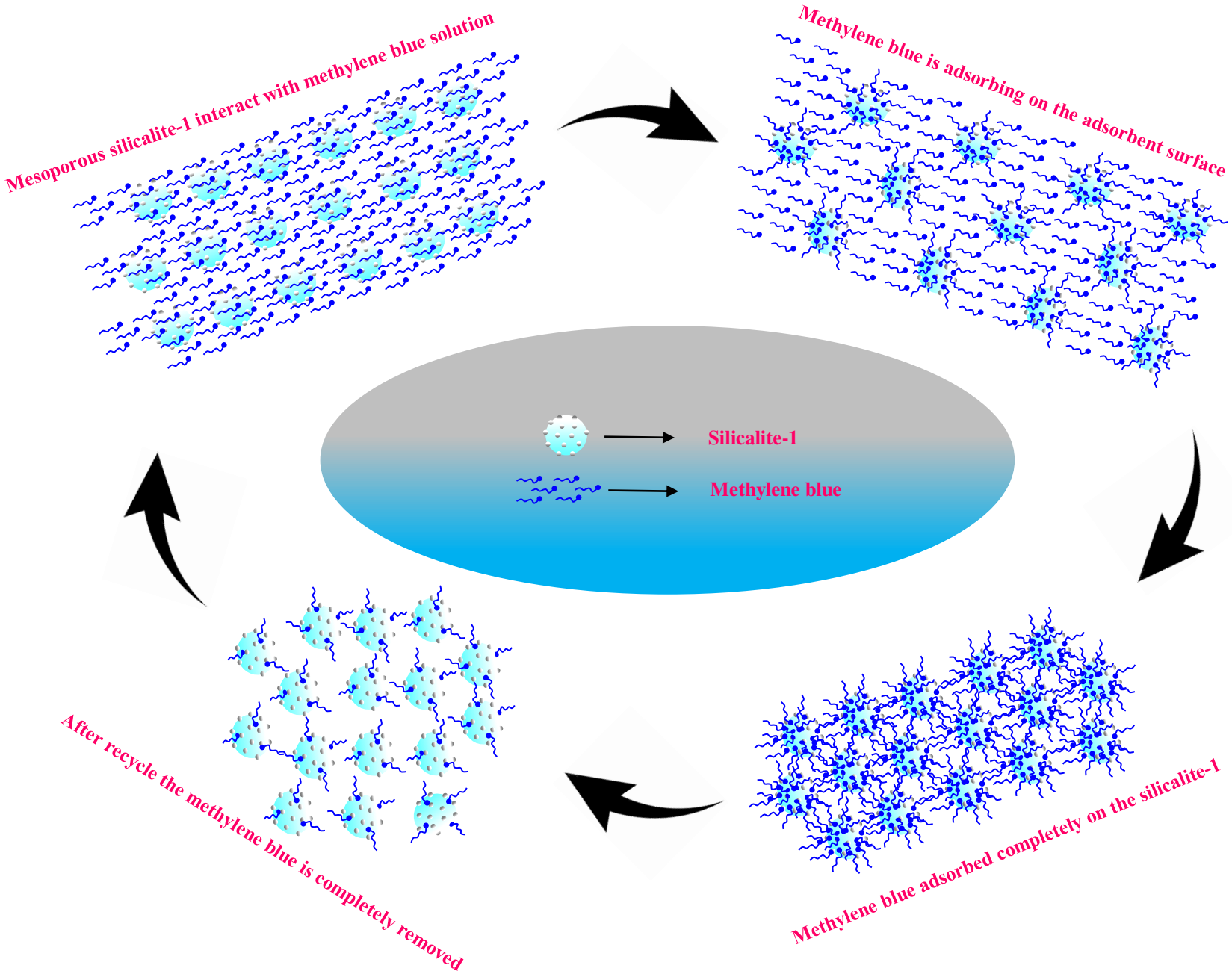
275 **Figure 8.** Effect of various parameters on the adsorption of MB onto mesoporous silicalite-1

276 a) silicalite-1 dosage b) initial dye concentration c) contact time d) temperature on the

277 adsorption process (adsorbent dosage = 0.10 wt%, initial MO concentration = 10 ppm, contact

278 time = 240 min and temperature= 30ºC



Scheme 2. Mechanism for MB adsorption on mesoporous silicalite-1

Adsorption kinetics

Adsorption kinetics was investigated to understand the adsorption mechanism and rate of adsorption of MB dye onto adsorbent. The experimental data was evaluated using two well-known models; pseudo-first-order and pseudo-second-order model. The linearized form of pseudo-first-order kinetic equation is expressed as follows [43]

$$\log(q_e - q_t) = \log q_e - \frac{K_1 t}{2.303} \quad (3)$$

where q_t (mg/g) and q_e (mg/g) are the amount of dye adsorbed at time 't' and equilibrium. K_1 are pseudo-first-order rate constant in 1/min. The graph of log ($q_e - q_t$) vs. t gives a straight with k_1 as slope and q_e as intercept.

The pseudo-second-order model is as follow[44]

$$\frac{t}{q_e} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e} \quad (4)$$

where K_2 is pseudo-second-order rate constant (g/mg min) and q_e is equilibrium adsorption capacity. The value of q_e and K_2 were obtained from the slope and intercept of plot t/q_t vs t.

The experimental data fitting on the kinetic model is illustrated in Figure 9 and the corresponding kinetic data are presented in Table 1. the validity of these models was checked by noting the correlation coefficient value (R^2). As can be seen, the experimental value fitted well in pseudo-second-order (0.99) model than pseudo-first-order (0.95). In addition to this, the correlation coefficient of pseudo-second-order model is higher than pseudo-first-order model. Therefore, pseudo-second-order is more suitable than pseudo-first order to explain kinetics of MB uptake on silicalite-1.

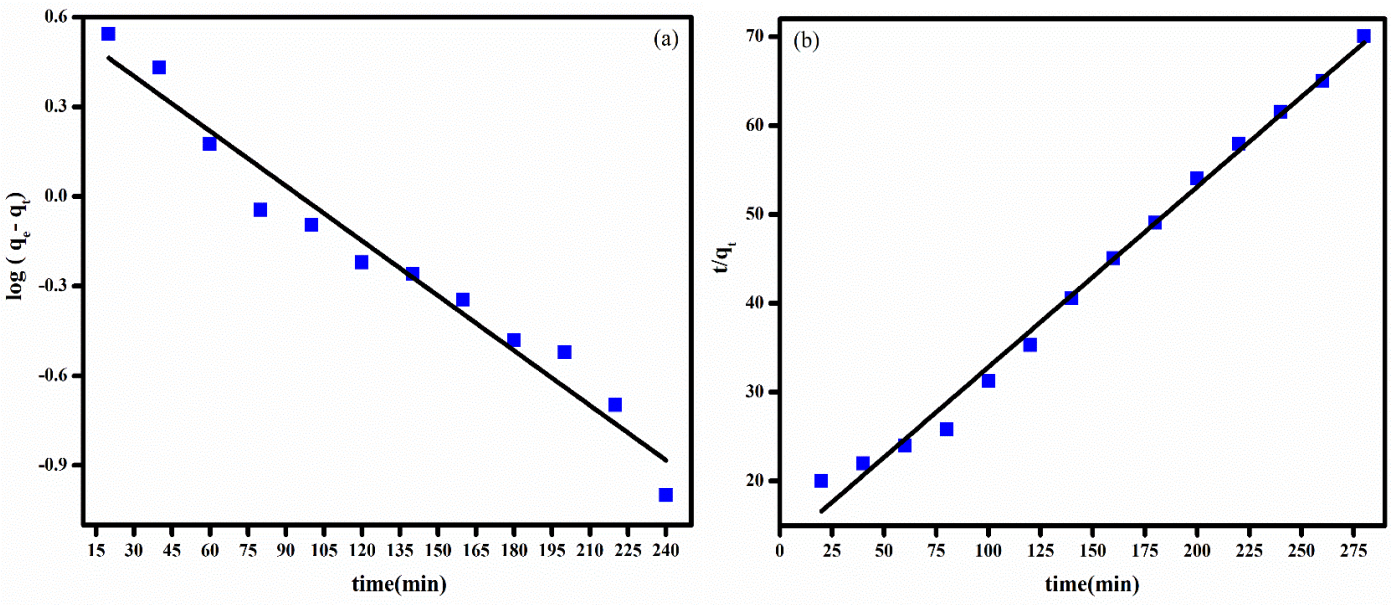


Figure 9. a) pseudo-first-order b) pseudo-second-order models for the adsorption of MB dye onto mesoporous silicalite-1

Table 1. Kinetic parameter values of adsorption kinetics for MB onto mesoporous silicalite-1

Experimental		**PFO**			**PSO**	
q_e, exp (mg g^{-1})	K_1(min^{-1})	q_e (mg g^{-1})	R^2	K_2 (g mg^{-1} min^{-1})	q_e(mg g^{-1})	R^2
4	-0.0140	3.2	0.95	0.0031	3.95	0.99

Adsorption isotherm

Adsorption isotherm is essential to understand the adsorption behaviour and interaction between the adsorbate and adsorbent. The linear form of Langmuir model is expressed as follows[45, 46]

$$\frac{C_e}{q_e} = \frac{C_e}{q_{max}} + \frac{1}{K_L q_{max}} \quad (5)$$

where q_e is the amount of MB dye adsorbed at equilibrium, C_e is the equilibrium concentration of MB dye (mg/L), q_{max} is maximum adsorption capacity, K_L and R_L are characteristic Langmuir parameters which is termed as Langmuir adsorption constant and separation factor respectively. R_L is expressed as

$$\mathbf{R_L} = \frac{\mathbf{1}}{\mathbf{1 + K_L C_0}} \quad (6)$$

Here, C_o is initial MB concentration (mg/L). The value of RL is significant and is related to the feasibility of adsorption. The adsorption process is favorable, if RL is between 0 and 1, unfavorable if R_L >1. R_L=0 and R_L=1 indicates reversible and linear adsorption respectively [52].

The Langmuir isotherm model assumes a homogeneous surface with equivalent site. According to this model, there is no interaction between the adsorbed molecules and hence multilayer adsorption will not occur. From the linear plot of C_e/q_e vs. C_e, the value of q_e and K_L is obtained. The Freundlich isotherm on the other hand assume heterogeneous surface and there is no restriction of multilayer formation. Therefore, in this model both monolayer and multilayer adsorption can take place. The linearized form of Freundlich model is represented as [47]

$$\mathbf{In\ q_e} = \frac{\mathbf{1}}{\mathbf{n}}\ \mathbf{In\ C_e} + \mathbf{In} \quad (7)$$

Where K_F and n are Freundlich constants related to adsorption capacity and degree of adsorption respectively. The value of K_F and n can be determined from the plot of ln q_e vs. ln C_e.

The data of adsorption process were applied to Langmuir and Freundlich isotherm model and the adsorption parameters are shown in Table 2. Since Freundlich isotherm gives a better fit with high correlation coefficient (R^2) it could be more suitable for MB adsorption. The value of 1/n is between 0 and 1 indicating that the MB adsorption on mesoporous silicalite-1 is a favourable process.

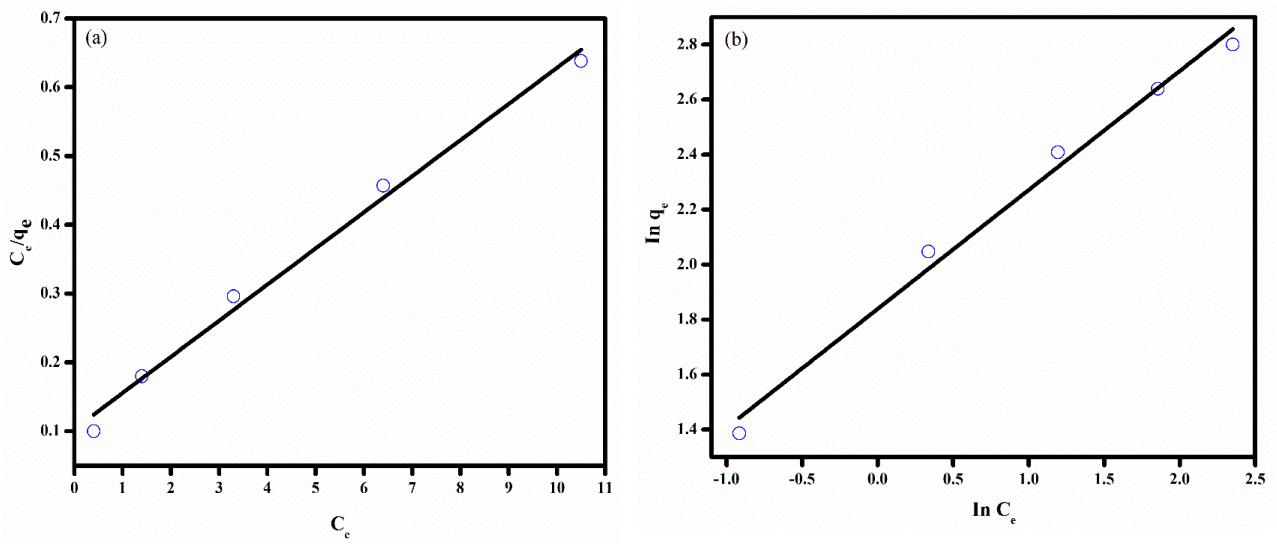


Figure 10. Isotherm model plots for the adsorption of MB a) Langmuir isotherm b) Freundlich isotherm

Table 2. Langmuir isotherm and Freundlich isotherm parameter values for MB adsorption onto mesoporous silicalite-1

Experimental	**Langmuir isotherm**			**Freundlich isotherm**		
q_e, exp (mg g^{-1})	R^2	q_{max}(mg g^{-1})	K_L	R^2	K_F (mg g^{-1})	n
4	0.977	19.04	0.509	0.995	4.3	2.31

Recyclability

The regeneration performance of the modified silicalite-1 was studied by six adsorption−desorption cycles at fixed adsorbent dose of 0.10 wt% and initial dye concentration 10 ppm. The result is presented in Figure 11. The result shows that the removal percentage of membrane at the first adsorption−desorption cycle is 95% and after six adsorption−desorption cycles, the adsorption efficiency was reduced to 86.3%. The results thus demonstrate that the silicalite-1 is effective even after multiple reuses.

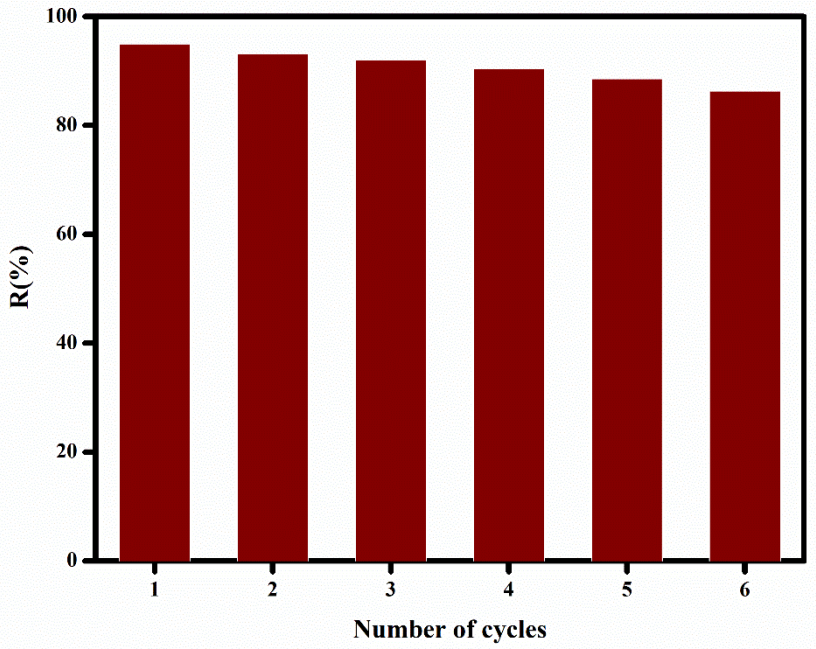


Figure 11. Reusability performance of adsorption of MB onto mesoporous silicalite-1

Conclusion

In the present study, a mesoporous silicalite-1 was developed using eco-friendly template, CMC. The successful synthesis of modified silicate was confirmed by techniques such as SEM, TEM, N_2 isotherm, XRD, FTIR. The synthesized mesoporous silicalite-1was effective to remove the cationic dye methylene blue from water. The effect of various operational parameter on the dye adsorption was studied and the result indicate that adsorption is favored at high initial concentration, high immersing time and high temperature. The dye removal was found to be governed by Freundlich isotherm model and the rate of adsorption was found to obey pseudo-second-order kinetics. The mesoporous silicalite-1 sample also show excellent recyclability. Therefore, we can conclude that modified silicalite-1 is a highly attractive candidate for the removal of MB from water.

Consent for publication

The authors confirm the consent for publication

Availability of data and material

All data generated or analysed during this study are included in this published article

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Authors contribution

Sabarish Radoor- Conceptualization, Methodology, Investigation, Validation, Writing - original draft, Software, Writing - review &editing, Formal analysis; Jasila Karayil - Investigation, Analysis, Interpretation of results, Software, Writing – review & editing, Formal analysis.; Aswathy Jayakumar - Data collection, Validation, Writing - review & editing, Formal analysis; Jyothi Mannekote Shivanna - Interpretation of results, Software, Writing – review & editing, Jyotishkumar Parameswaranpillai –Validation, Software, Writing - review & editing and Investigation; Suchart Siengchin – Editing, Funding and Supervision

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Compliance with ethical standards

Conflict of interest: The authors have declared no conflict of interest

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Figures

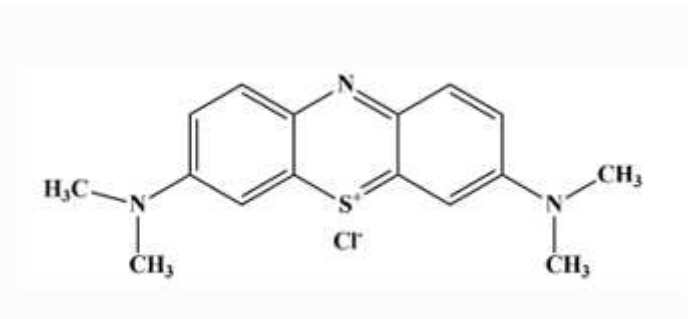

Figure 1

Structural formula of methylene blue

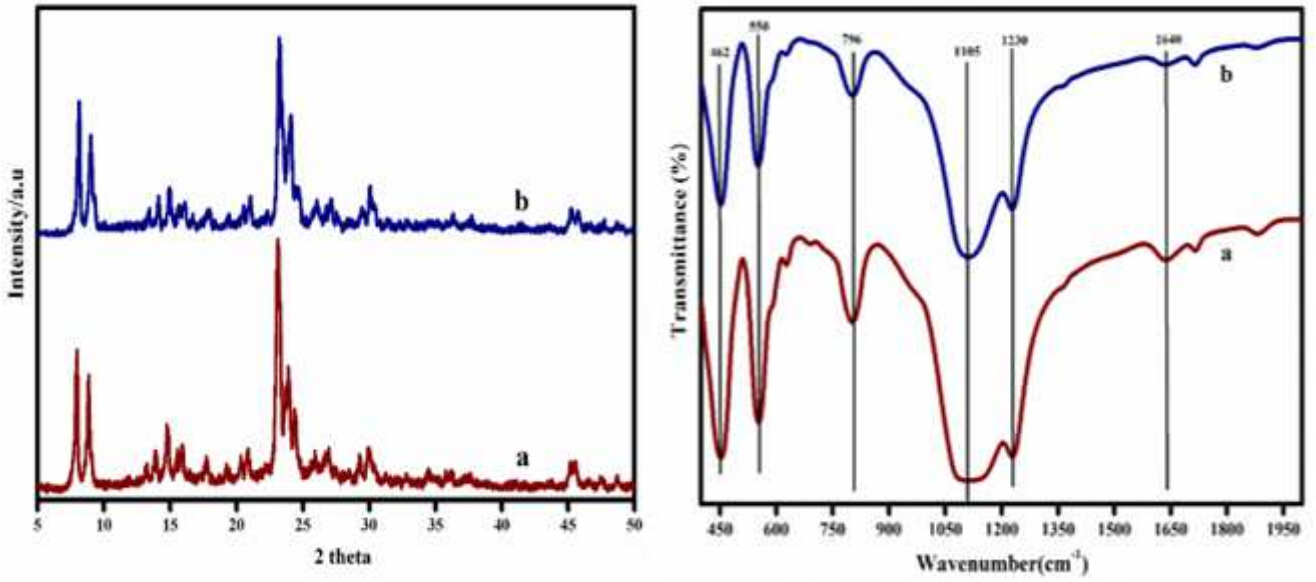

Figure 2

Powder XRD patterns and FTIR spectra of conventional and mesoporous silicalite-1

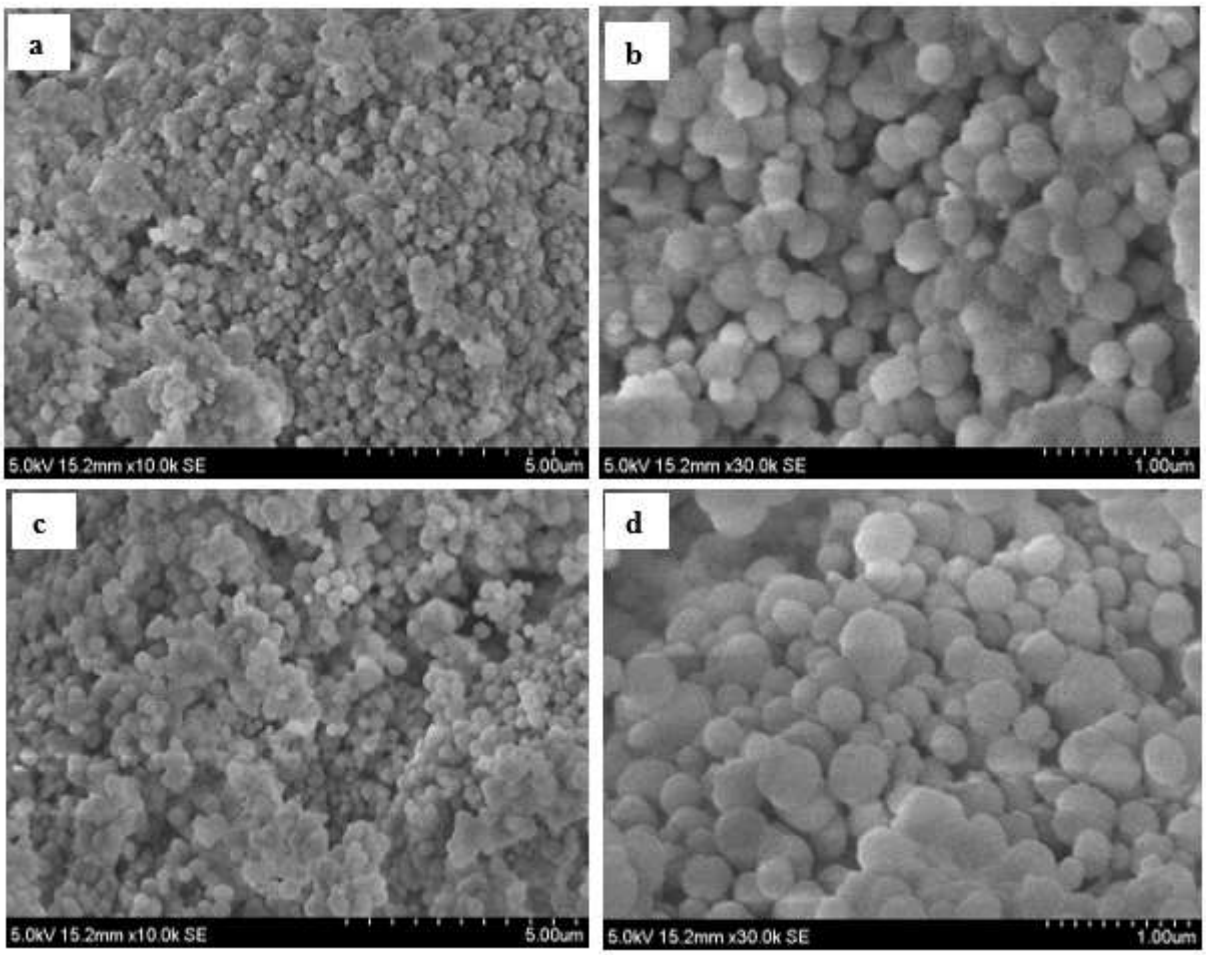

Figure 3

SEM micrographs of (a & b) conventional and (c & d) mesoporous silicalite-1

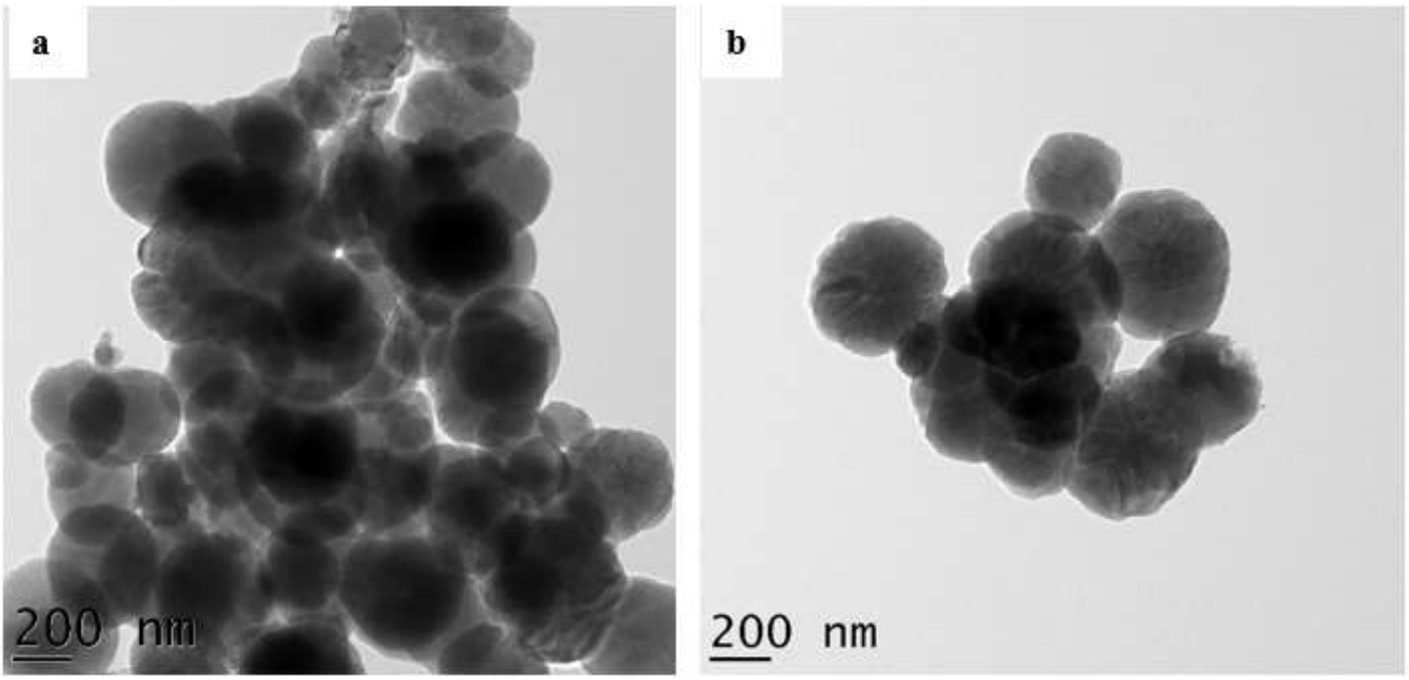

Figure 4

TEM images of mesoporous silicalite-1

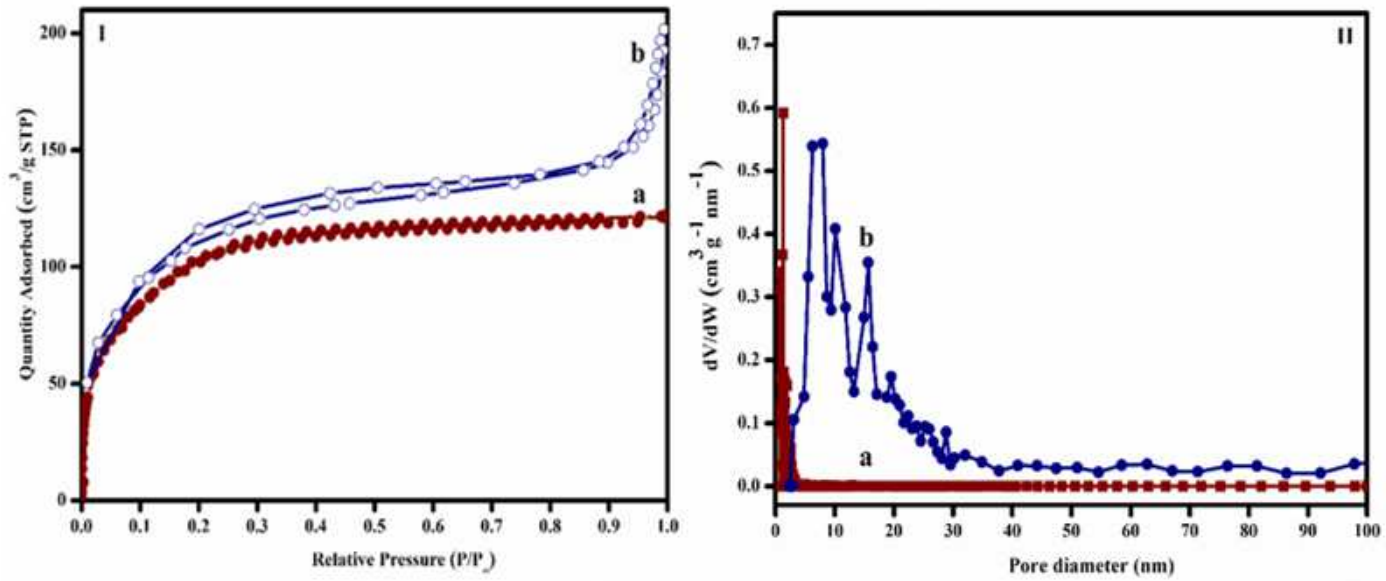

Figure 5

(I) N2 adsorption isotherms of: a) conventional b) mesoporous silicalite-1; (II) pore size distribution of: a) conventional b) mesoporous silicalite-1

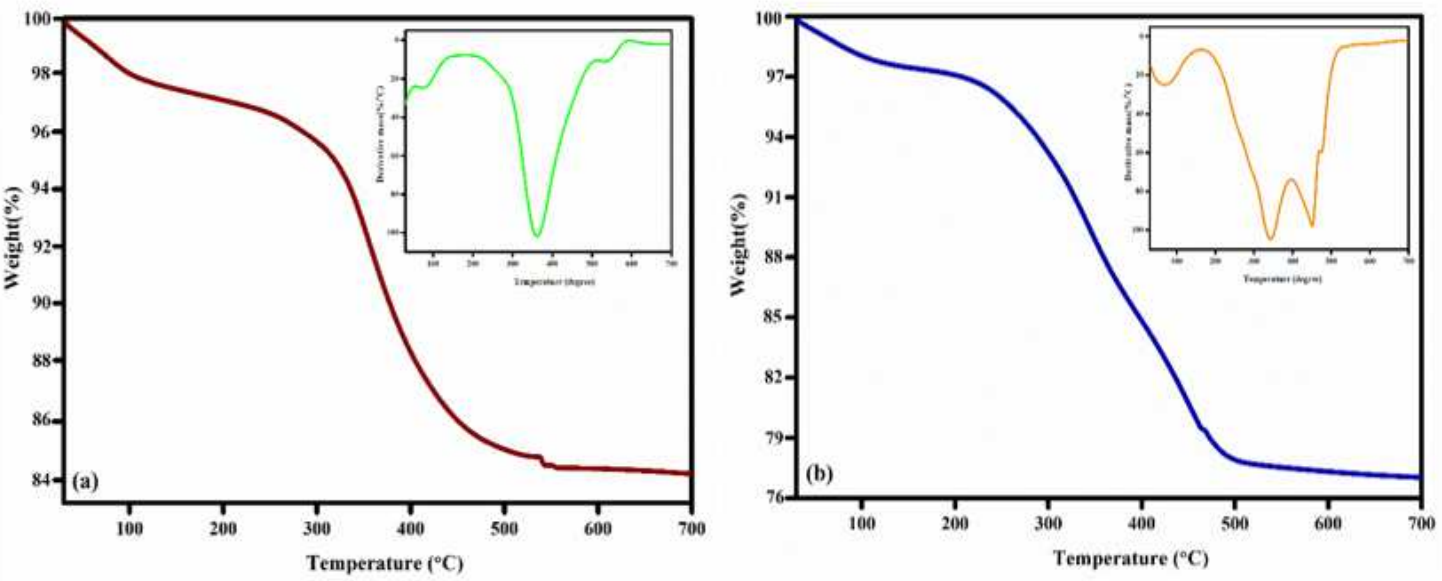

Figure 6

TGA curves of conventional and mesoporous silicalite-1 and the corresponding DTG curves are given at the inset

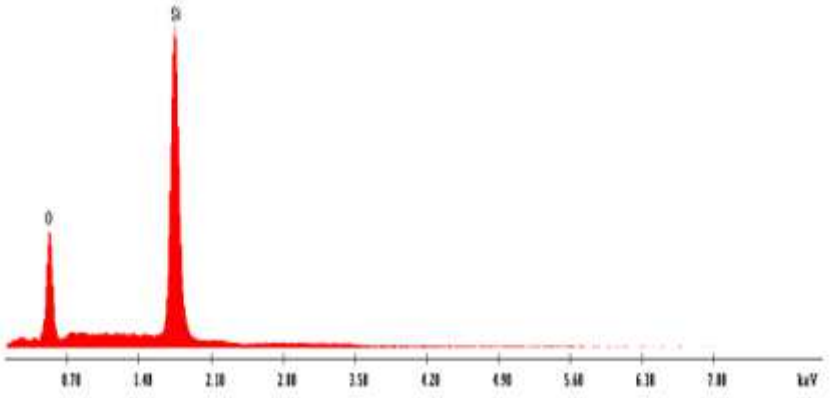

Figure 7

EDS image of mesoporous silicalite-1

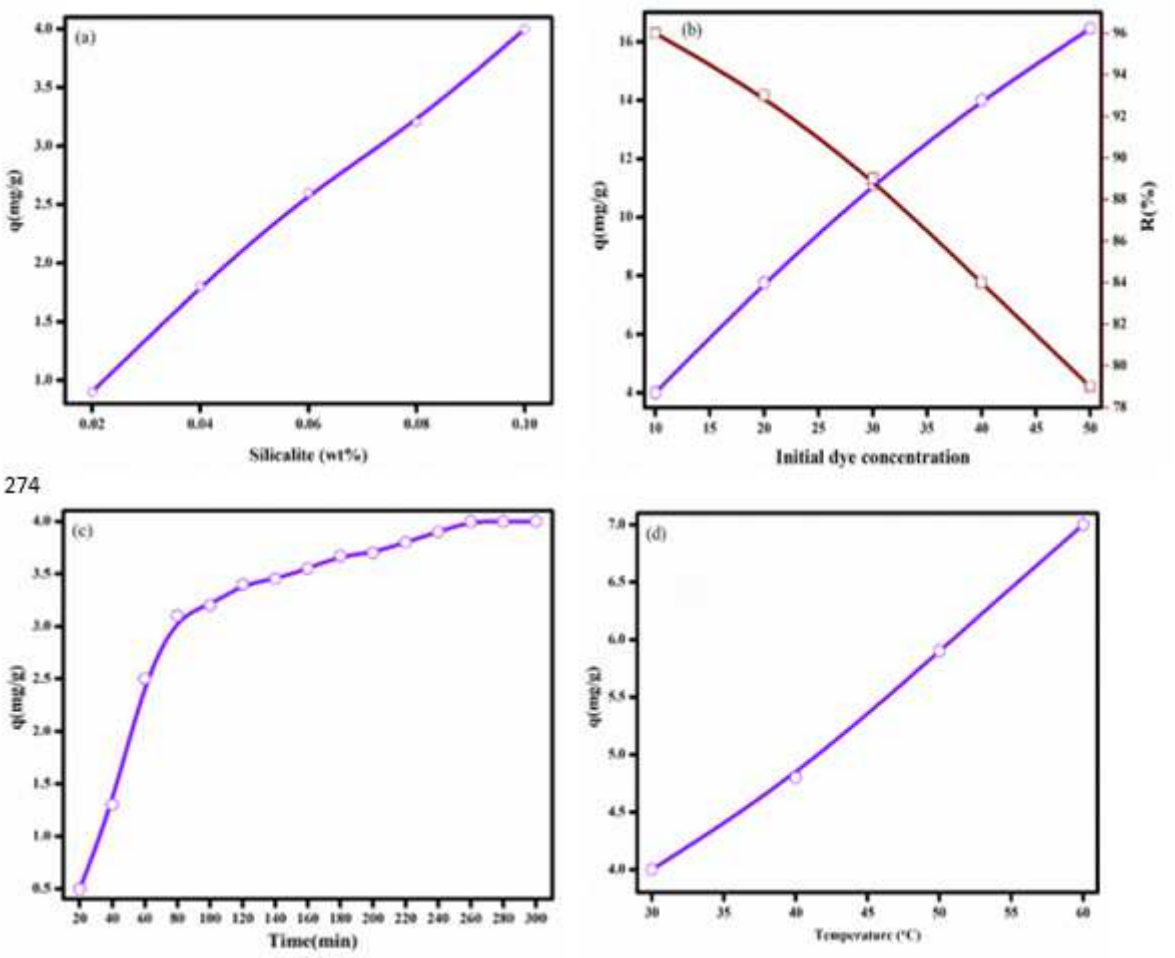

Figure 8

Effect of various parameters on the adsorption of MB onto mesoporous silicalite-1 a) silicalite-1 dosage b) initial dye concentration c) contact time d) temperature on the adsorption process (adsorbent dosage = 0.10 wt%, initial MO concentration = 10 ppm, contact time = 240 min and temperature= 30°C

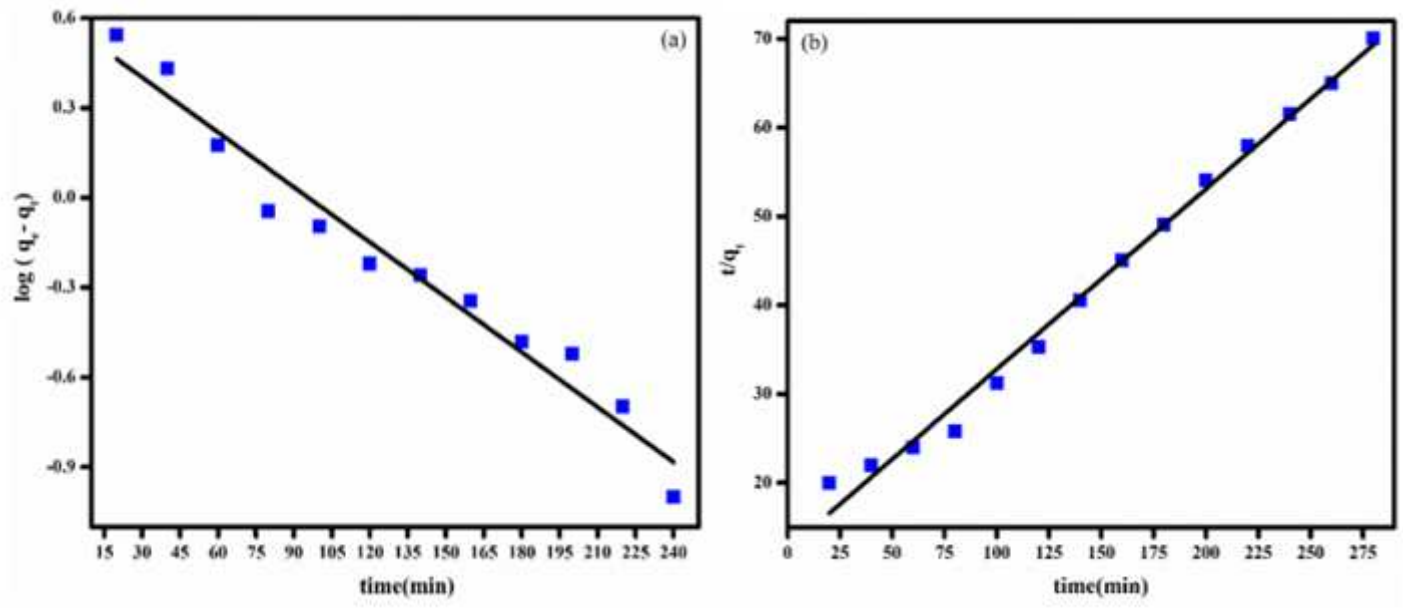

Figure 9

a) pseudo-first-order b) pseudo-second-order models for the adsorption of MB dye onto mesoporous silicalite-1

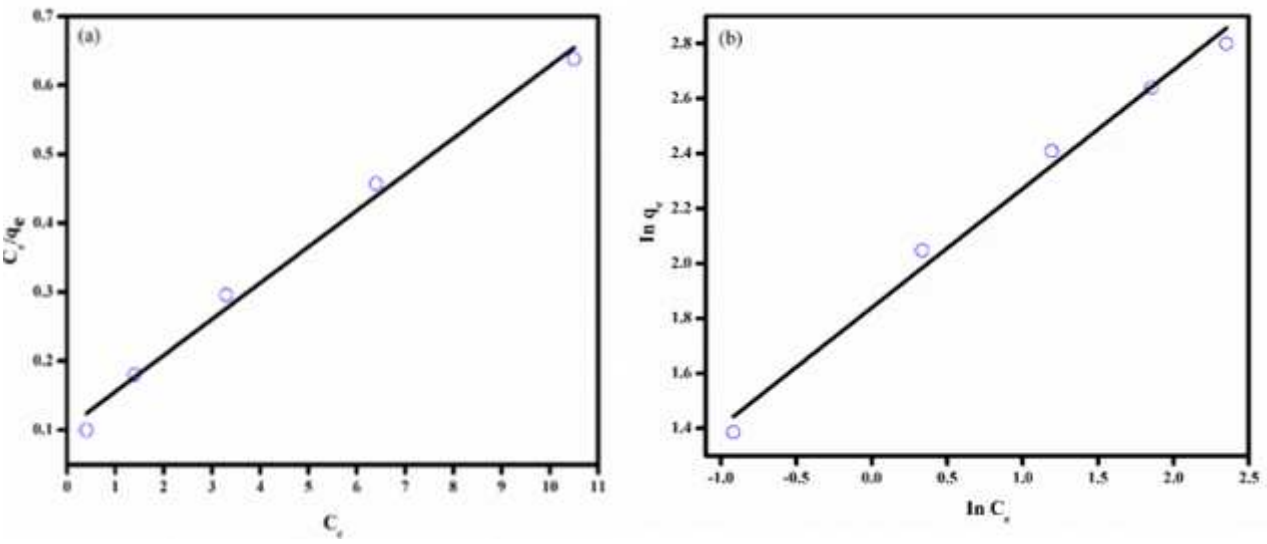

Figure 10

Isotherm model plots for the adsorption of MB a) Langmuir isotherm b) Freundlich isotherm

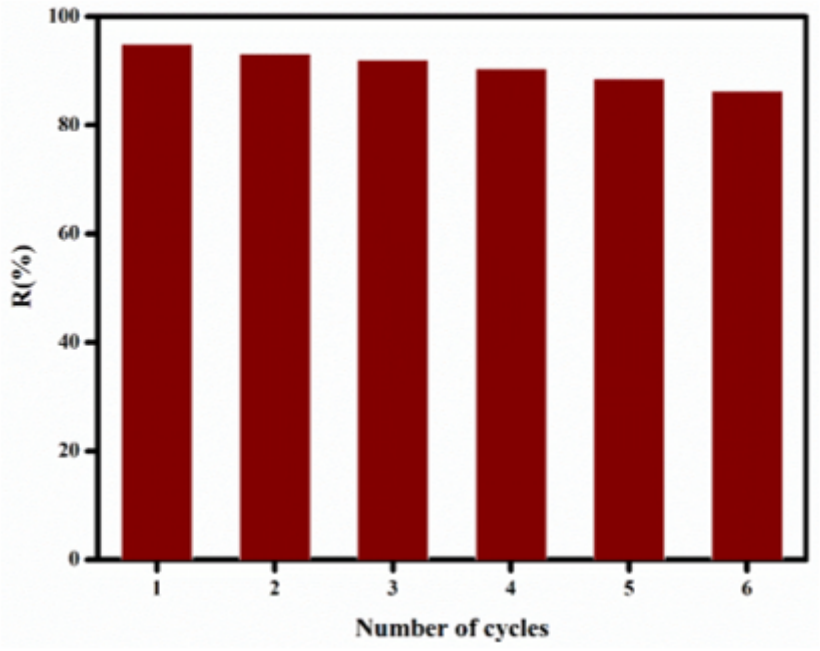

Figure 11

Reusability performance of adsorption of MB onto mesoporous silicalite-1

Supplementary Files

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