









Research Article

Remediation of Methyl Red Dye from Aqueous Solutions by Using Biosorbents Developed from Floral Waste

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The disposal of biological waste into water bodies is a major global concern as it leads to water pollution resulting in the loss of plenty of revenue in the cleaning of water bodies. Here, in the present research work, sacred flowers were collected, segregated, sun-dried, and powdered. The dried floral powders (marigold and rose) were characterized by field emission scanning electron microscopy (FESEM), electron diffraction spectroscopy (EDS), Fourier transforms infrared spectroscopy (FT-IR), and X-ray diffraction (XRD). The microscopy revealed the irregular spherical shape of the sheet-like structure whose size varies in microns. The EDS revealed the elemental composition which was dominated by mainly carbon and oxygen. The XRD shows the presence of carbon (10-25th) in the amorphous form and the absence of any crystalline phase in the biosorbents. The FT-IR showed peaks that conformed to the presence of functional groups like -OH and a carbonyl group. The dried powders were used as an economical and eco-friendly biosorbent for the removal of methyl red (MR) dye from the aqueous solutions by batch adsorption study. After 60 minutes of contact time, the marigold powder (MGP) and rose petal powder (RPP) showed decolorization of 61.16% and 56.08% for 2 ppm of MR dye. The kinetic revealed that the dye removal reaction does not follow the pseudo-first-order as well as the pseudo-second-order. The utilization of such waste-based biosorbents will minimize solid waste and also will provide an economical biosorbent for the removal of environmental pollutants.

1. Introduction

Since ancient times only, India is known as the land of temples, especially in cities like Haridwar, Varanasi, Dwarka, and Mathura. [1]. Besides this, India is also having a large number of mosques, churches, and other religious places

where every day, a huge number of flowers are offered by the devotees [2]. In addition, these flowers are also used in huge amounts in various marriage events, funerals, last-rite, and other social gatherings. Every year, all these major religious places dispose of millions of tonnes of dried flowers into the river or other water bodies or are dumped as solid

municipal waste [3, 4]. The disposal of tonnes of dried or semidried flowers in important rivers, like the Ganga, and the Yamuna leads to water pollution [5, 6]. Every year, various government organizations like “Namami Gange,” national river conservation plan (NRC), Yamuna action plan (YAP), River Development and Ganga Rejuvenation (MOWR), and Atal mission for rejuvenation and urban transformation (Amrut), spend a huge amount of money for cleaning the important and sacred rivers of the country. Besides this, there are several nongovernmental organizations (NGOs) like “The ISHA foundation” and “Jeevitnadi-Living River Foundation” which are involved in cleaning rivers in India. The disposal of flowers in the rivers increases pollution by increasing the biological oxygen demand (BOD), nitrogen content, and other parameters of the water [7], hence affecting the flora and fauna of the river or the aquatic ecosystem. As per reports in 2019, every year, about 8×10^9 kilogram of flowers is dumped into river Ganga alone which leads to water pollution [8].

To date, various techniques like precipitation, adsorption, filtration, nanofiltration, and absorption are applied for the removal of both organic and inorganic pollutants from the wastewater [9]. Most of these techniques are quite simple and economical but less effective and energy intensive. So, the utilization of such biological waste as an adsorbent for the remediation of dyes, pesticides, etc. may reduce the cost of wastewater treatment. The adsorption method is easy to implement, reliable, less expensive, and has no formation of any toxic product at any step [10]. So, adsorption is the most preferred technique. Moreover, being organic in nature, the bioadsorbents easily get degraded or mineralized in the environment. There are several investigators which have reported the utilization of the abovementioned techniques for wastewater treatment and other environmental cleanups. Elgarahy et al. reported the sorption of cationic and anionic dyes from wastewater by using an untapped sepia-based composite [11]. Further, Elwakeel et al. reported the remediation of chromate ions from the wastewater by a magnetic Schiff's base sorbent which was based on the waste peels of shrimps [12]. Further, recently, Elwakeel et al. developed a product named 2-mercaptobenzimidazole which was functionalized with chitosan. The functionalized product was used for the effective remediation of methylene blue in a batch and column experiment. The authors have reported MB dye removal percentage of up to 94.5% [13]. Another approach by Tsuchiai et al., Ishizuka et al., and Shi et al. remediated the MB dye by using beach bivalve shells collected from the Port Said coastal area (Egypt) as an adsorbent. The dye removal percentage with this adsorbent reached 93.6% [14–16].

Biosorbents are biological materials used to passively remove contaminants from a solution [17, 18]. Activated carbon is widely used in sorbent matter in biosorption practice due to its great physicochemical structural properties. One major drawback of this activated carbon is its low regeneration capacity and expensive manufacturing [19]. Biological waste is one of the best substitutions for activated carbon as an adsorbent due to its low-cost, environmental friendly, high yield, biodegradable nature, high regeneration capacity, high abundance, and renewable nature [20]. The effective transformation of biological waste into a biosorbent

reduces the ecological risk imposed by dumping such waste into the natural environment. Moreover, such practices also minimize the cost of solid waste management [21]. There are several types of biosorbents [22–24] such as (1) tea-industry waste [25], (2) sugar industry waste (bagasse) [26], (3) peach and apricot stones, (4) antibiotic waste, (5) sludge, (6) waste green sands, (7) fly ash [6, 27, 28], and incense sticks ash [4, 29–31].

El-Azazy et al. utilized green tea waste (GTW) as an adsorbent for the removal of methylene blue (MB) from the wastewater where the investigators achieved a maximum adsorption capacity of 96.58% MB dye by using about 68.28 mg/g GTW [25]. Kerrou et al. used sugarcane bagasse (SB) to remove organic dyes from the wastewater. The removal rate of MB dye was 80.27% to 98.49% with the increase of the mass of adsorbent from 0.05 g to 5 g due to an increase in a specific area [26]. Abatal et al. utilized pitahaya fruit (*Hylocereus* spp.) peels for the remediation of Pb(II), Cd(II), Co(II), and Ni(II) from the wastewater where the biosorption capacity of pitahaya peel was 77%, 72%, 74%, and 74% for Pb(II), Cd(II), Co(II), and Ni(II), respectively [32]. Saranya et al. developed biosorbents from floral waste for the removal of lead ions from the aqueous solutions [33].

A large number of heavy metals, pesticides, dyes [34], etc. are pollutants that cause environmental threats all over the world [35]. Nowadays, heavy metals are the most threatening environmental pollutants which affect public health due to their nonbiodegradable nature. The increase in the use of pesticides for controlling pests to ensure food security causes environmental pollution [36]. Dye is used in large amounts in textiles, paper, leather, cosmetics, food, plastics, rubber, and the pharmaceutical industry. The discharge of dyes in water bodies adversely affects the overall ecosystem [37]. Therefore, there is an immediate requirement for the remediation of environmental pollutants including dyes from the environment by using low-cost, eco-friendly, and biodegradable biosorbents such as flowers [38].

These flowers are mainly consisting of carbon, which can be used in its natural form for the development of various value-added materials. The value-added materials developed from such floral waste will be economical and eco-friendly. Moreover, the development of such value-added products from floral waste will also help in the reduction of the solid waste arising from the disposal of sacred flowers into the rivers [21]. There are several examples where *Rosa damascena* mill sacred flowers were used for the manufacturing of value-added materials like an essential oil. Adeel et al. reported the extraction of dye pigment from such sacred flowers offered at religious places [39]. Vankar et al. utilized the residue left after dye extraction from the *Hibiscus rosa-sinensis* flower. Here, investigators have used such floral residue for the removal of heavy metals from the aqueous solutions [40]. Elangovan et al. also reported the remediation of hexavalent and trivalent chromium by using biosorbent prepared from palm flower (*Borassus aethiopum*) [41]. Waghmode et al. formulated a media for the growth of an actinomycete, i.e., *Microbispora* sps from the floral extracts of *Madhuca latifolia* L [3]. Luis et al. utilized marigold

flowers for the extraction of xanthophyll pigments under optimized conditions [42]. Pu et al. evaluated the chemical properties and other parameters of safflowers [43]; while Sato et al. developed safflower yellow B and carthamin red pigment from the same flower [44]. So, from the literature, it is evident that floral waste has been used as a source of value-added material. Davamani et al. utilized flower waste for the removal of chromium ions from the tannery effluent. Here, flower waste biomass absorbed around 70% chromium from the tannery effluent [45]. Aman et al. used rose biomass for the removal of chromium, mercury, and zinc from the contaminated waters. The absorption capacity of 100 ppm of chromium, mercury, and zinc was 5.26, 6.76, and 4.07 mg/g, respectively [46]. Mondal et al. also utilized the waste marigold flower powder for the removal of heavy metals, i.e., Cd (II) and Cr (VI) from aqueous solutions [47]. Tolcha et al. used the flower of *Typha latifolia* for the uptake of multiclass pesticide residues from contaminated water. These studies show the removal of diazinon, atrazine, chlorothalonil, ametryn, chlorpyrifos, and dimethametryn pesticides from the wastewater using a batch experiment [48]. Elango and Govindasamy used temple waste flowers for the removal of color from textile dyeing effluent. The maximum color removal efficiency of activated carbon was 98.17% at a 200 mg dose of biosorbent, and color removal was high at 95.83% at 100 min [49]. From the above piece of literature work, it is evident that floral waste could act as a potential biosorbent for the removal of both organic and inorganic pollutants from wastewater [36]. Generally, *Tegetes erecta* (TE) is used as garlands in prayer, *C. infundibuliformis* (CI) while bowing heads, and *P. tuberosa* are used at religious places to worship Gods, which is evident from the literature. So, the majority of the literature available in the scientific domain is based on these flowers only [3].

Methyl Red ($C_{15}H_{15}N_3O_2$) is an azo-dye compound having a molecular weight of 269.31 g/mol. It is chiefly used as a pH indicator in laboratories as its color changes with solution pH. The dye imparts red color in acidic solutions having a pH of 4.4, yellow color in solutions having a pH higher than 6.2, whereas orange color in the solutions having a pH range of 4.4-6.2. MR is generally used as a model dye for various experimental studies due to its intense color in aqueous systems. The dye has low biodegradability because of the presence of benzene rings in its structure. Methyl red is a toxic organic compound and is known to cause respiratory problems, digestive tract infections, skin and eye irritation, carcinogenesis, mutagenesis, and teratogenesis. Hence, wastewater containing such toxic organic contaminants has to be treated prior to its disposal or discharge into the aquatic systems [50-52]. The utilization of floral waste from religious places as an adsorbent will solve the flower-based waste disposal problems and environmental pollution [3].

Recently, various investigators have used different metallic/nonmetallic nanoparticles, biosorbents, clay, etc. for the remediation of methyl orange dyes from the wastewater. Bhowmik et al. developed a new magnetic nanoparticle having a mixed phase of $CaFe_2O_4$ and $MnFe_2O_4$ magnetic nanocomposite (CaF-MnF-MNC) by coprecipitation method.

Further, the MR dye was removed from the aqua matrix by using this nanocomposite. The authors achieved very high efficiency for the dye remediation, i.e., 99.88% [53]. In another work reported by Bhowmik et al. developed a magnetic nanocomposite (Fe_2O_3/Mn_3O_4) for the increased remediation of MO dyes from the aqueous solutions. They performed neural network modelling and response surface methodology optimization. Further, Bhowmik et al. reported the development of magnetic $CaFe_2O_4/MnFe_2O_4$ nanocomposite and utilized for the remediation of Eriochrome Black-T (an anionic dye) from the aqua matrix by simple adsorption technique. Here, the investigators reported the dye remediation by simple shaking condition, batch stirring, and sonoassisted, and out of which, the latter technique was most efficient which removed the EBT dye up to 98% [54]. In another work by Bhowmik et al. where the investigators developed a nanocomposite of calcium ferrite and zirconia and utilized them further for the remediation of MO dyes from the aqua matrix. Here, the investigators have mainly focused on the optimization parameters for the remediation. Under optimized conditions, investigators reported the removal of MO dye from the aqua matrix of up to 98.92% [54].

Yadav et al. extracted ferrous particles from incense sticks ash and modified them into iron oxide nanoparticles (IONPs). Further, the investigator utilized the synthesized IONPs for the remediation of methylene blue from the aqueous solution and the efficiency attained was about 70% [55]. Modi et al. also provided descriptive information about the utilization of zinc oxide nanoparticles for the removal of methylene blue from wastewater [56].

Several investigators have used metallic particles as an adsorbent for the removal of MR dye and other pollutants. For instance, Elwakeel et al. provided detailed information about metal/mineral-incorporating materials for water treatment. The authors have mainly focused on the remediation of Cr (VI) from wastewater using metallic particles [57]. Mashabi et al. provided detailed information about the remediation of dyes by using adsorbents made from chitosan or glycidyl methacrylate [58]. One more article by Elwakeel reported descriptive information about the detailed information for the applications of chitosan resins in wastewater treatment. The chitosan was used as a bioadsorbent for the remediation of pollutant removal from wastewater [59].

There were several objectives for performing this experiment. Firstly, to suggest the utilization of floral waste as an economical and biodegradable bioadsorbent from various religious places. Secondly, to assess the potential of the recycled flowers-based bioadsorbents for the remediation of MR dye from the aqueous solutions. Thirdly, to minimize the solid waste arising from the disposal of flowers at rivers or other water bodies and in municipality areas. One final objective of this study was to suggest an economical and effective method to maintain the cleanliness of the river water.

In the current research work, authors have emphasized the utilization of sacred flowers offered at temples, mosques, and other religious places for the development of bioadsorbents. The current methods involve several steps like a

collection of flowers, segregation of flowers, sun-drying, and conversion into powder form. The developed biosorbents (marigold and rose flowers) were characterized by analytical instruments for detailed features. Finally, the potential of both types of floral-based biosorbents was assessed for the remediation of MR dye from the aqueous solutions. Such approaches for wastewater treatment are quite economical due for several reasons, firstly, less energy is required for the manufacturing of biosorbents, and secondly, biosorbents originated from the waste material. The current novel approach utilizes biological waste as an economical biosorbent without affecting the environment due to its biodegradable nature.

2. Materials and Methods

2.1. Materials. Flowers were collected from temples (Uttam Nagar, New Delhi, India) and mosques (Hazrat Nizamuddin, New Delhi, India), double distilled water (ddw), ceramic mortar pestle (Laboratory Scientific, Haryana, India), methyl red dye (99% purity, AR grade) (SRL, New Delhi, India), methanol (98% purity, AR grade) (SRL, New Delhi, India), potassium bromide, 99.98% IR grade (Sigma Aldrich, Darmstadt, Germany), pulverizer (Kudarat Enterprises, Rajkot, Gujarat, India), sieve sets (Laboratory Scientifics, Haryana, India), and 100 ml round bottom flask (Borosil, Gujarat, India).

2.2. Methods

2.2.1. Processing of Sacred Recycled Flowers. Sacred flowers were collected from the temples, mosques, and other events in plastic bags. The flowers were manually segregated to remove wrappers of incense sticks, incense cones, candles, paper pieces, and other items. Then, the rose flowers and petals were separated from the marigold flowers. The flower petals for both flowers were collected by detaching them with other flower parts like sepals, carpels, and sometimes leaves. The detached floral petals were then spread on a plastic sheet and kept under sunlight for several days. This ensures the complete drying and removal of moisture content from the floral petals. Once the sun-drying was over, the floral petals were collected in a plastic bag, which in turn was kept in an airtight container. Both marigold and rose petals were passed separately through a pulverizer machine for grinding. Both types of floral petal powders were collected separately. The floral petal powder was then sieved by using sieve sets to remove larger particles. The floral powder was then analyzed by using sophisticated instruments for detailed morphological and elemental properties. The floral petal powders were then stored for future use.

Figure 1 is showing the sequential steps involved in the development of biosorbents from floral waste. Figure 1(a) is the collected flowers from religious places, whereas Figure 1(b) is the segregated marigold flowers, which are free from all other parts of a flower. While Figure 1(c) is the powdered form of the sun-dried marigold flower petals. Figure 1(d) is showing dried rose petals while Figure 1(e) is showing the powdered form of rose petals obtained after passing through pulverizer. While Figure 2 is showing a mini

pulveriser machine used in this experiment for grinding floral petals, here the powdered flowers are collected in the stainless-steel collecting drum placed at the bottom. Figure 3 is showing a schematic flow chart for the development of biosorbents from floral waste.

2.2.2. Preparation of Aqueous Solution of Dyes. About 3 mg of dye was dissolved in a 200 ml beaker and was continuously stirred on a magnetic stirrer at 500 rpm for the complete dissolution of the dye particles. Further, the aqueous solution of dye was filtered through a Whatman filter paper no 42 to remove the impurities. Finally, the methyl red (MR) dye stock solution was stored in a reagent bottle for future use.

2.2.3. Remediation of Dyes by Using Bioadsorbents. The flower powder obtained previously was used as an adsorbent for the remediation of dyes (methyl red) from the aqueous solution prepared in the above step. The stocks of an aqueous solution of MR dye were prepared by weighing about 0.2 mg of MR dye. The weighed quantity of the MR dye solute was both the dyes were mixed with the 100 mL distilled water. About 50 mL of MR dye solution was transferred to a 100 mL plastic beaker. About 1 mg of each bioadsorbent/flower powder was added to the aqueous solution of MR dye in two separate beakers. Further, an initial sample of MR dye aqueous solution was kept for analysis. Further, both the beakers containing an aqueous solution of dyes and rose petal powders and marigold petal powders were kept for stirring at 350 rpm at room temperature (RT). Further, an aliquot of about 5 mL was collected from both beakers after a regular interval of 10 minutes and kept in a centrifuge tube.

The dye adsorption study was carried out in the batch shake flask method in an incubator shaker under controlled conditions. The effect of time on the remediation of MR dye from the aqueous solution was analyzed. To settle down the bioadsorbents from the mixture, centrifugation of the sample was done at 5000 rpm for 10 minutes. Finally, the centrifuged samples were analyzed by using a Carry 60, UV-Vis spectrophotometer (Agilent Technologies, Santa Clara, USA). The spectral analysis was done to reveal the concentration of the MR dye in the various samples. The removal percentage of MR dye and adsorption capacity of MGP and RPP at any time (q_t) were calculated using the following equation.

$$\% \text{Decolorization} = \frac{A_0 - A_t}{A_0} \times 100, \quad (1)$$

$$q_t (\text{mg/g}) = \frac{(A_0 - A_t)V}{M},$$

where A_0 is the initial concentration of MR (mg L^{-1}), A_t is the concentration of MR at a given time (mg L^{-1}), V is the volume of solution (liter), and M is the mass of bioadsorbents.

Figure 4 is showing the mechanism involved in the removal of methyl red dye from the aqueous solutions by using floral biosorbents.

2.2.4. Characterization of the Bioadsorbents. Both the bioadsorbents were analyzed by various sophisticated instruments like Fourier-transform infrared spectroscopy (FT-IR), Field



FIGURE 1: Steps involved in the processing and transformation of floral waste into biosorbents.

emission scanning electron microscopy (FESEM), and X-ray diffraction spectroscopy (XRD). The FT-IR analysis of both bioadsorbent powders was done by using the solid KBr pellet method, where KBr and each bioadsorbents were separately mixed in a ratio of 98:2 and mixed thoroughly in a mortar and pestle. Preparation of the solid pellet of KBr and samples were done by using a hydraulic press for both samples separately. Finally, both pellets were analyzed by using a blank KBr pellet, and measurements were done by using Bruker Tensor 27 FT-IR Spectrometer (Billerica, Massachusetts, USA), instrument at a resolution of 2 cm^{-1} . To find the mineral phases, present in the biosorbent powder, both the powders were analyzed by the D-8 Advance Bruker (USA) made instrument. The scanning of the powder samples was done in the range of 10-80 degrees with a scanning rate of 5/sec. To find the morphological and elemental features, both powders were analyzed by using NOVA, NanoSEM, 450 FEI FESEM (USA). The powder samples were sprayed on the carbon tape which in turn adhered to the Al stub. The sample imaging was done in the variable range at 5 keV. The electron diffraction spectroscopy (EDS) was done by using the attached Oxford-made elemental analyzer in the FESEM. Finally, the different dye samples in aqueous solutions were analyzed by using a Carry 60, UV-Vis spectrophotometer (Agilent Technologies, Santa Clara, USA).

3. Results and Discussion

Figure 4 is showing the simplest possible mechanism of adsorption of MR dye molecules on the surface of the bioadsorbents. The dye molecules get adsorbed on the surface of the bioadsorbents due to the various functional groups like hydroxyl group and carboxyl group present in the bioadsorbents. The presence of these functional groups was confirmed by the FT-IR.

The FESEM analysis of both sorbents will in determining the size of the sorbents. From the literature, it is well established that the smaller the size of the particle, the more will be the surface area to volume ratio (SVR), so the more will be the adsorption sites for the pollutants. This feature will increase the efficiency of the sorbents for the remediation of pollutants from the aqueous solutions. Moreover, the elemental analysis by the EDS analyzer will of the biosorbents reveals the purity as an impurity in the sorbent will minimize the efficiency of adsorption and ultimately the remediation of pollutants. FT-IR measurement of the biosorbents will reveal the various functional groups present on the surface of the biosorbents like hydroxyl and carbonyl which make them specific for the adherence of the pollutants like MR dyes. Methyl red is an azo dye, so the FT-IR analysis will reveal the possibility of adherence of azo dye on the biosorbents based on the functional groups. The selection of XRD



FIGURE 2: Showing the mini pulveriser machine, where the powdered flowers are collected in the stainless-steel collecting drum placed at the bottom.

analysis of the biosorbents was mainly used to find the amorphous or crystalline phase. Since the original of both biosorbents was biological plant material, so there is the possibility of carbon in an amorphous form.

3.1. FESEM-EDS of Flower Powders

3.1.1. Marigold Flower Powder. FESEM was carried out to reveal the morphological details of the developed bioadsorbents. The imaging was done at various magnification scales. Figures 5(a)–5(c) show FESEM micrographs of marigold petal powder at different magnifications. The size of the MGP is in microns where the size is irregular in shape and highly aggregated. The size of lumps is 20–70 microns, which are visible in the FESEM image. There are a few rectangular-shaped particles also surrounded by smaller irregular spherical lumps. The irregular shape indicates the high carbon in the sample, which is quite obvious due to the organic nature of the powder. The particles are porous in nature, which is evident from the FESEM image. The EDS analysis of the MGP shows the EDS spectra in Figure 4(d) and the elemental composition. The spectra have various elements like C, O, P, Na, Mg, Al, and Si which are dominated by mainly C and O. The carbon is present in the highest amount, i.e., 63.5% and O is 34.46%, which alone constitutes more than 97%; while remaining 2% is contributed by Si, Mg, Al, and Na which are present in trace amount in the MGP. The high percentage of carbon and oxygen proves the organic nature of the bioadsorbent.

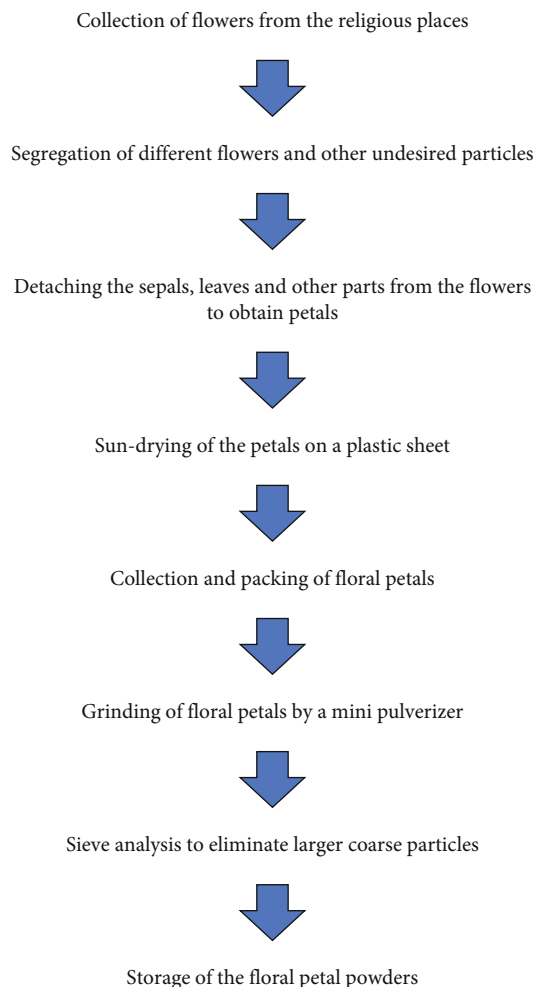


FIGURE 3: Schematic diagram for the development of floral petal powders/bioadsorbents.

Figures 6(a) and 6(b) show FESEM micrographs of rose petal powders that are obtained from pulverizing the dried rose petals. The particle's layered structure whose size is in the range of several microns, i.e., 8–17 microns. In Figure 6(a), there are several holes and spots in the structure. Such holes and spots were also reported by Saranya et al. in the floral powders of *Tegetis errata* and others [33]. The particles appear white in color, which indicates the carbon-rich region. While Figure 6(c) shows the EDS region of the RPP, analyzed for elemental detection. EDS spectra in Figure 4(d) and the elemental composition show that the powder is having mainly elements like C, O, Ca, Mg, P, Cl, and S. The major dominating element is C, i.e., 55.1% and O is 39.4%, while the remaining is in trace amounts. The percentage of C and O indicates the purity of the bioadsorbents. Several investigators have also reported similar properties of the RPP.

Among both MGP and RPP, the former is an irregular spherical shape while the latter is a layered sheet-like structure. MGPP has a slightly higher amount of carbon than the RPP, while O was marginally higher in MGPP. Besides this, both of them have traces of elements which constitutes 1–2% of the biosorbent. These elements may be present in the flowers from the soil via roots, etc.

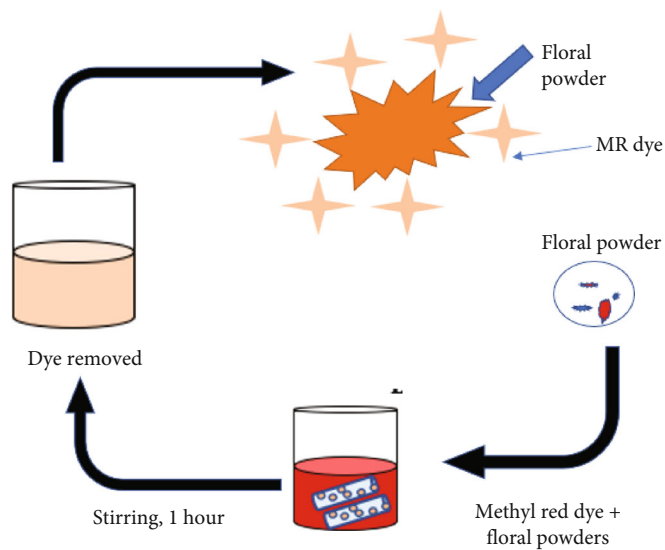
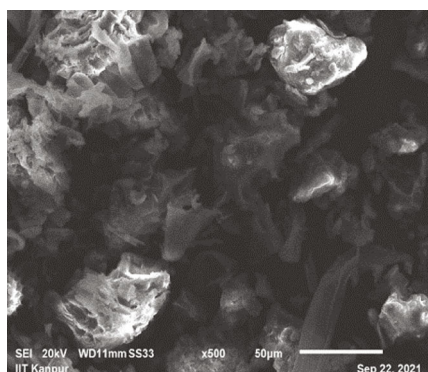
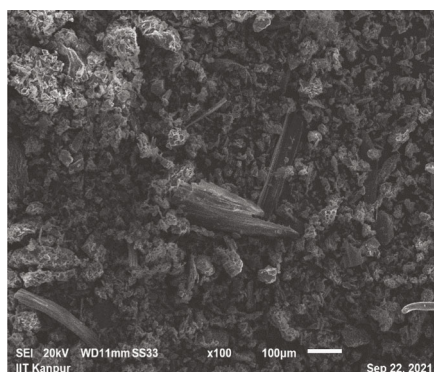


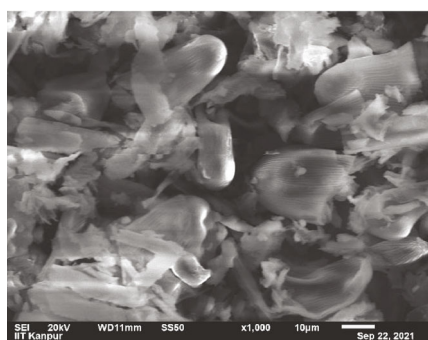
FIGURE 4: Steps involved in the floral waste based biosorbent development and dye removal from aqueous solutions.



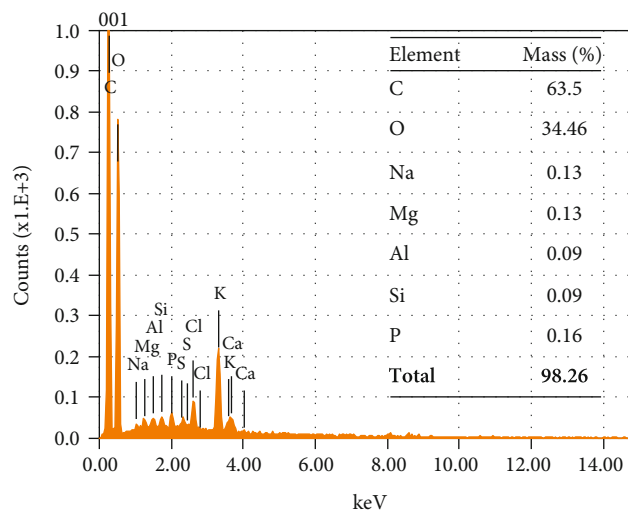
(a)



(b)



(c)



(d)

FIGURE 5: FESEM micrographs of marigold flowers (a, b), EDS spot (c), and EDS spectra (d).

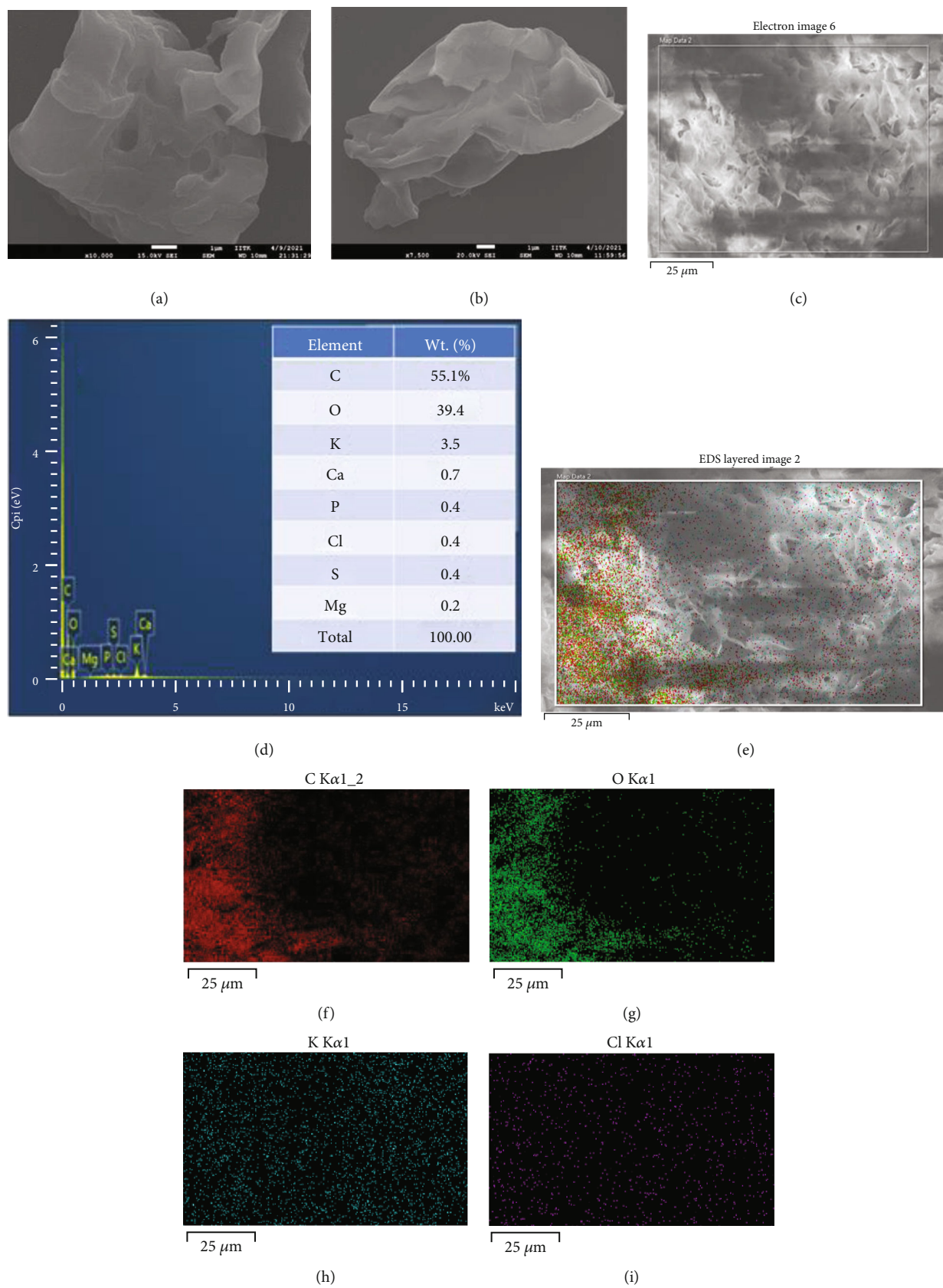


FIGURE 6: FESEM micrographs of rose petal powder (a, b), EDS spot (c), EDS spectra (d), and elemental mapping of rose powders (e-i).

TABLE 1: Comparative morphological and elemental studies of floral bioadsorbent by FESEM.

Biosorbent	Shape	Size	Application	References
Flower waste	Irregular	Micron sized	Pb ²⁺ removal	[62]
<i>Acroptilon</i> response flower powder	Rectangular, sheet-shaped	Micron sized	Cr VI removal	[60, 61]
Crushed marigold flowers	Powder has pansy's stripes, fritillary's checks, or tiger-lily's dots	Micron sized	Compost preparation	[3]
Mixed floral waste	Heterogeneous surface, having cellulose, hemicellulose, and lignin	Micron sized	For biofuel and compost manufacturing	[63]

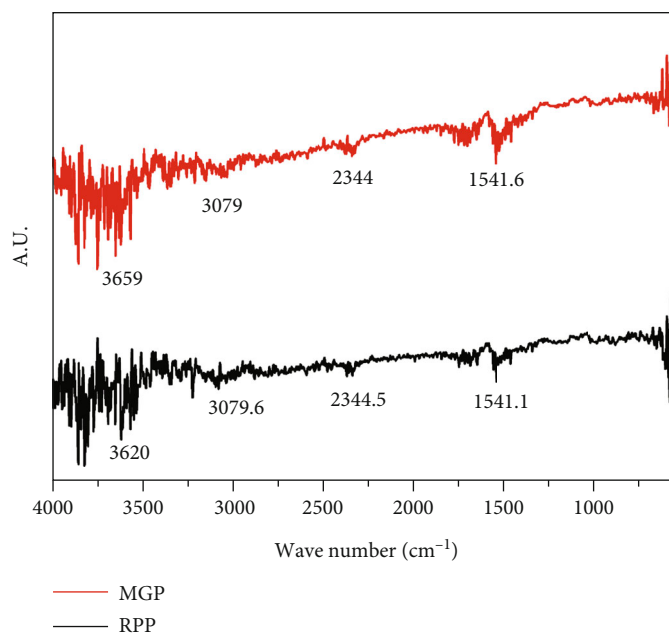


FIGURE 7: FT-IR spectrum of Rose Petal powder and Marigold flowers.

TABLE 2: FT-IR peak assignments of MGP and RPP along with previous work done.

Peaks	Functional group	Attributions	References
1735	C=O, (in all plants)	Stretching vibrations (SV)	[33, 62]
2851–2850 cm ⁻¹ and 2920–2919 cm ⁻¹	Alkanes	SV of C–H.	[33, 62]
3403.65–3421.19 cm ⁻¹	O–H, of the carboxylic group	SV	[33, 62]
3409.63–3416.57 cm ⁻¹	Alcohol and phenol compound of O–H vibration		[33, 62]
3404.97–3414.65 cm ⁻¹	N–H of amides	Symmetric and asymmetric vibrations	[33, 47, 61]
1297.74–1243.96 cm ⁻¹ and 1056–1054 cm ⁻¹	C–N of aliphatic amines	SV	[47, 61]
1413.29–1419.33 cm ⁻¹	C–C of the aromatic ring	SV	[33, 47, 66]
1337–1441 cm ⁻¹	C–H of alkanes group	Bending and rock vibration	[33, 47, 60]
1,526 and 1,528 cm ⁻¹	COO and C=O groups		[47, 61]
571–605 cm ⁻¹	Aromatic compounds	Bending modes	[47]

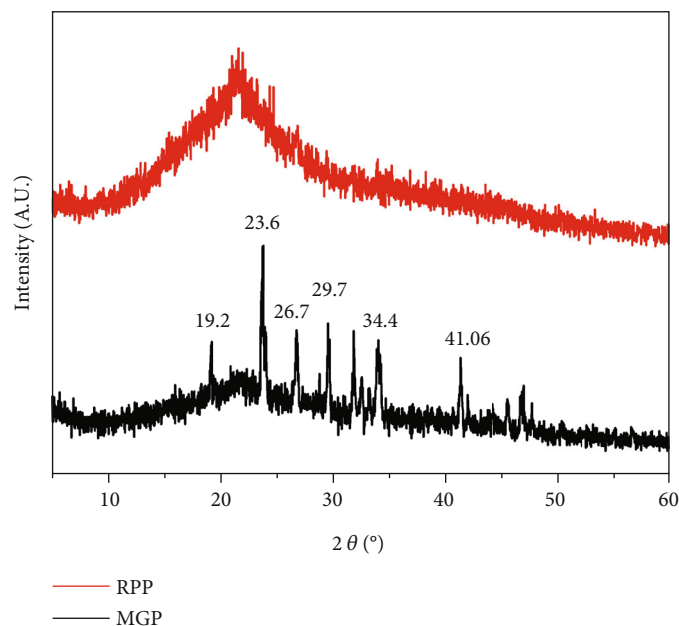


FIGURE 8: X-ray diffraction spectra of marigold and rose petal powders.

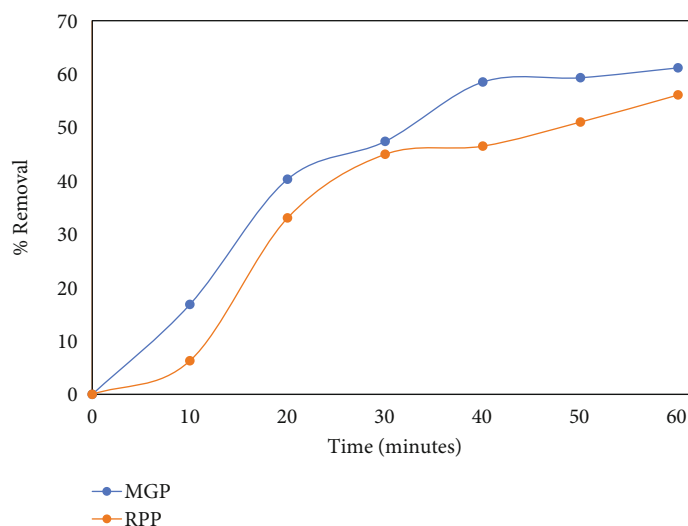


FIGURE 9: Removal percentage of methyl red dye by marigold and rose petal powder as biosorbents.

Figure 6(e) is showing the elemental mapping of RPP by EDS, and Figure 6(f) shows the mapping region of carbon in the EDS, while the green area is of oxygen (Figure 6(g)) while the blue zone is of potassium (Figure 6(h)). So, from the EDS elemental mapping, it is also confirmed that the bioadsorbent is highly pure in nature, i.e., made up of only carbon and oxygen along with traces of elements like K, etc.

Srinivasulu et al. also used flower-based bioadsorbent for the remediation of pollutants from the wastewater. The investigators have used the floral-based bioadsorbent for the removal of heavy metals. The size of the bioadsorbent was also in the micron range which was mainly rectangular and sheet-shaped [60]. Moreover, Ghaneian et al. also used floral-based bioadsorbents for the remediation of pollutants

from the wastewater and reported almost similar morphology of the bioad [61].

Similar elemental mapping of flowers was also reported by investigators whose details are provided in Table 1.

3.2. FT-IR Study of Flower Powders. Figure 7 shows typical FT-IR spectra of floral powders as bioadsorbents where (a) rose powder and (b) marigold powder revealed that the presence of some functional groups like $-OH$, $-NH$, and $C=O$ were identified by the peaks with specific band range. The functional groups in the floral powder samples help in the remediation of contaminants like heavy metals, dyes, and others. So, the presence of different functional groups in the floral powders makes them more suitable for the adsorption of different pollutants from wastewater. The functional

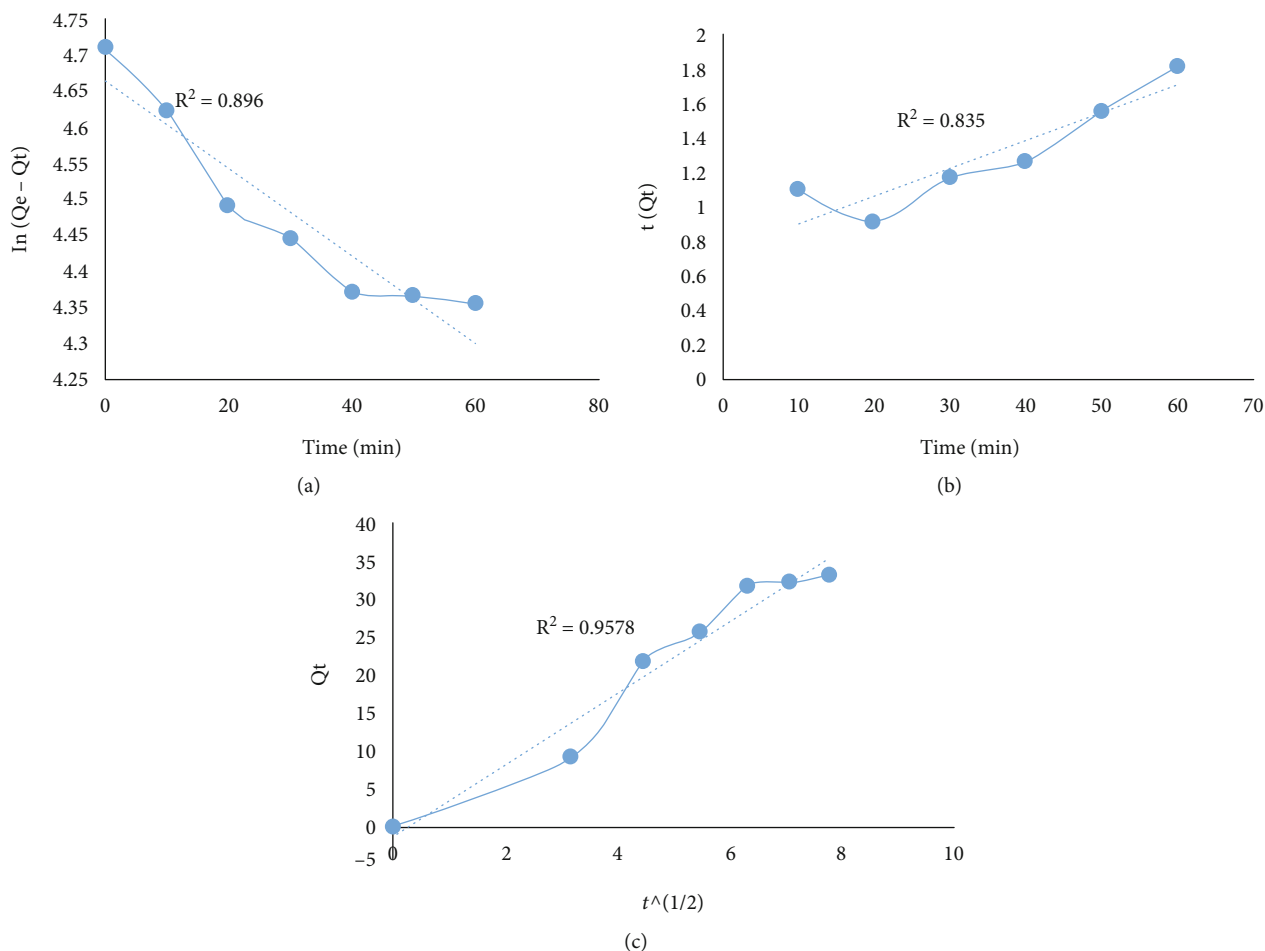


FIGURE 10: (a) Pseudo-first-order, (b) pseudo-second-order, and (c) intraparticle diffusion study of methyl red removal by MGP.

group-based removal of pollutants by floral wastes depends on the factors like the chemical state, accessibility, site quantity, and affinity among bioadsorbents surfaces and dyes or pollutants [64]. The variety of absorption bands was observed as overlapped and shifted so it was not feasible to confirm the specific stretching frequency for a functional group, due to their different environment and molecular structure. The factors which influence the shift of absorption bands are the electronic and mass effect of adjacent substitution, intra-molecular and intermolecular H-bonding, physical state, and conjugation and ring strain [65].

The weak and overlap bands do not provide much more data about the nature of the samples. The FT-IR spectrum of bioadsorbent revealed weak and overlap bands in the region of $550\text{--}4000\text{ cm}^{-1}$, which specified the presence of $\text{C}=\text{O}$ and --OH of carbonyl, --COOH , and phenols. The band near 1541 cm^{-1} is attributed to the $\text{C}=\text{O}$ stretching mode in $\text{--C}=\text{O}$ and --COOH , while very weak and overlap bands between $1500\text{--}1400\text{ cm}^{-1}$ were directed to the C--O stretching and O--H bending modes for instance phenolic and --COOH . The weak band around 3079 cm^{-1} was assigned to aromatic stretching. The band near $3620\text{--}3659\text{ cm}^{-1}$ is the N-H stretching of aromatic compounds. Besides this, the band near $3620\text{--}3659\text{ cm}^{-1}$ is also attributed to the --OH group present in the samples. Mondal et al. also obtained

TABLE 3: Kinetic parameters for methyl red removal by MGP.

Kinetic	Parameter	Values
Pseudo-first-order	$Q_e(\text{mg/g})$	105.86
	$k_1(\text{min}^{-1})$	0.0338×10^{-7}
	R^2	0.896
Pseudo-second-order	$Q_e(\text{mg/g})$	62.89
	$k_2(\text{g/mg} \cdot \text{Min})$	0.341×10^{-3}
	R^2	0.835
Intraparticle diffusion	$k_{id}(\text{mg/g} \cdot \text{min}^{3/2})$	4.7216
	$C(\text{mg/g})$	1.1332
	R^2	0.957

bands at the same position and attributed it to the N-H stretching of amides [47].

Mondal et al. reported this while working on marigold petal powder as a bioadsorbent for the remediation of heavy metals (Cd(II) and Cr(VI)) from wastewater [47]. The higher bands were assigned to O--H vibrations of free hydroxyl groups and overtones by 3600 cm^{-1} and above while some lower bands (under 800 cm^{-1}) could be assigned to nitrogen-containing sites in the bioadsorbent. So, it may

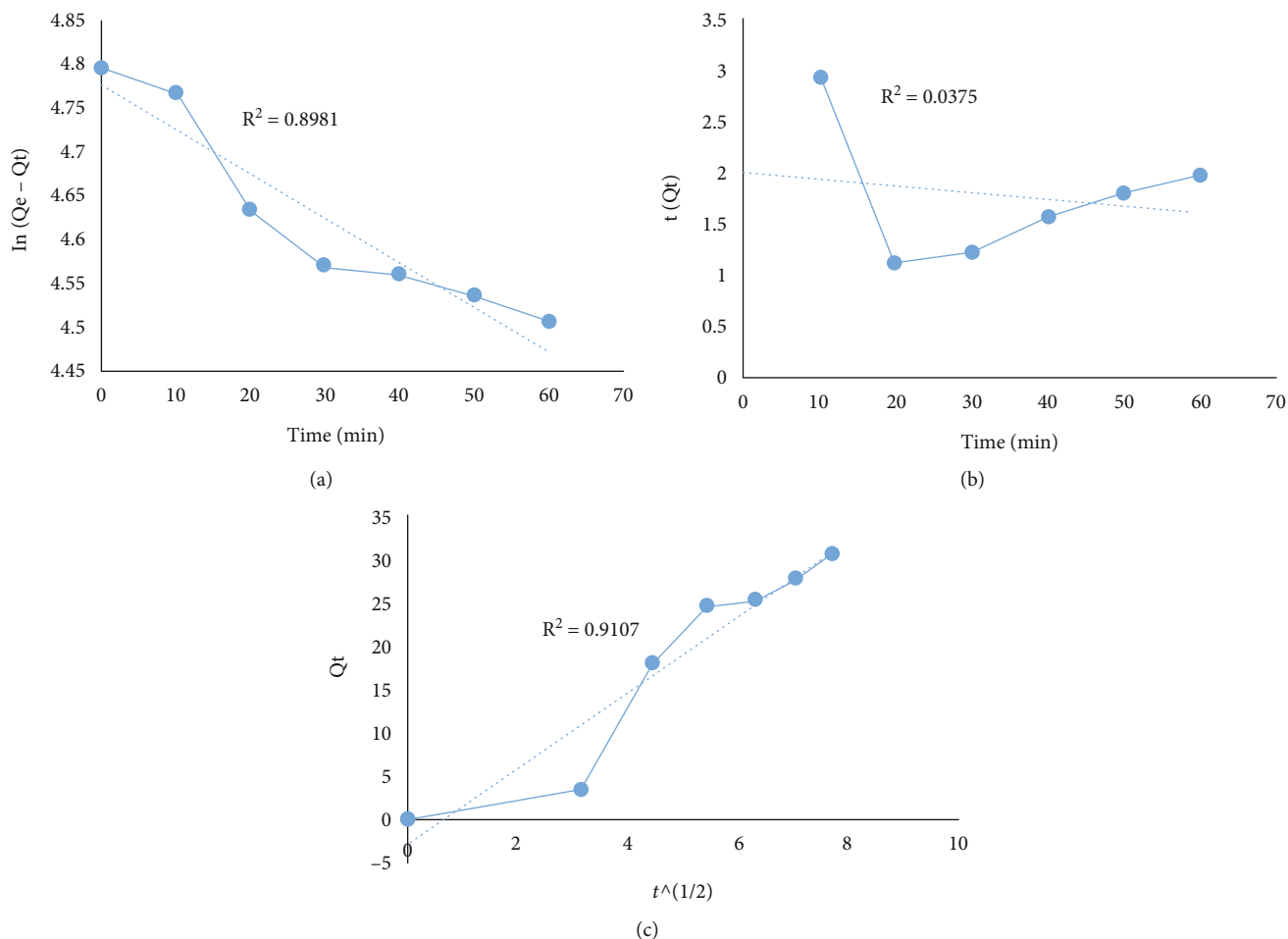


FIGURE 11: (a) Pseudo-first-order, (b) pseudo-second-order, and (c) intraparticle diffusion study of methyl red removal by RPP.

TABLE 4: Kinetic parameters for methyl red removal by rose petal powder.

Kinetic	Parameter	Values
Pseudo-first-order	Q_e (mg/g)	118.54
	k_1 (min^{-1})	2.78×10^{-5}
	R^2	0.898
Pseudo-second-order	Q_e (mg/g)	147.058
	k_2 (g/mg. min)	2.29×10^{-5}
	R^2	0.0375
Intraparticle diffusion	k_{id} (mg/g. $\text{min}^{3/2}$)	4.3399
	C (mg/g)	2.8001
	R^2	0.910

be concluded from spectrum results that functional groups like $-\text{CO}$, $-\text{COO}$, $-\text{NH}$, and $-\text{OH}$ are present in the bioadsorbent with a binding site. The dried floral powder from the marigold showed the presence of polymeric hydroxyl group, C-H, and carboxylic groups of polysaccharides [47]. The major FT-IR assignments of both the floral powders are given below in Table 2.

3.3. XRD Study of Flower Powders. The XRD patterns of the bioadsorbent are presented in Figure 8. The diffractogram of the bioadsorbent indicates the presence of a cellulose or carbon structure in the crystalline phase along with the presence of some amorphous material which is confirmed due to the peaks present at $2\theta \sim 19.2^\circ$, 23.6° , 26.7° , 29.7° , 34.4° , and 41.06° . This peak suggests the arrangement of cellulose molecules in ordered lattices where certain functional groups like $-\text{OH}$ were available. Moreover, these peaks are absent at higher 2θ values which confirmed a decrease in the degree of crystallinity, i.e., the presence of more amorphous than crystalline.

3.4. Bioremediation of Methyl Red Dye by Using Bioadsorbents. The removal of MR dye was observed against contact time for both types of biosorbents. The MR dye removal percentage by both types of biosorbents was calculated and is displayed in Figure 9. There was a continuous gradual increase in the percentage removal of MR dye by both types of biosorbents from initial to 60 minutes. After 60 minutes of contact time, the marigold powder (MGP) and rose petal powder (RPP) showed decolorization of 61.16% and 56.08% for 2 ppm of MR dye. There are several pieces of literature where MR dye removal was carried out

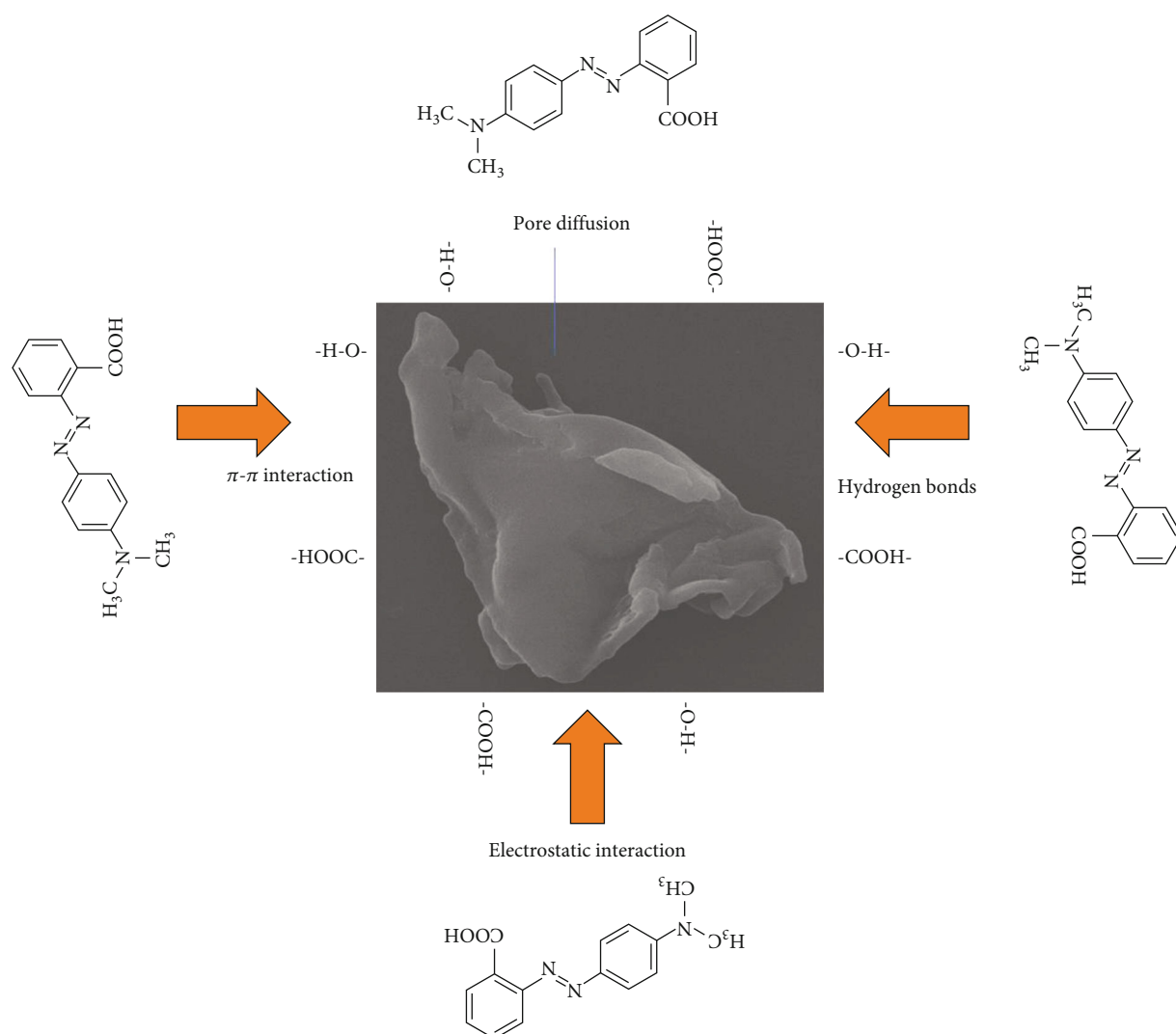


FIGURE 12: Summary of possible adsorption mechanism of methyl red dye on the floral waste powders.

by using different microorganisms like bacteria and mixed culture; for instance, Takkar et al. used actinobacterium *Zhihengliuella* sp. ISTPL4 for the remediation of MR dye [67]. Vijaya and Sandhya used mixed culture for the removal of MR dye from the wastewater [68].

Echavarria-Alvarez and Hormaza-Anaguano reported the utilization of floral waste as a low-cost adsorbent for the remediation of acid blue 19 dye from the aqueous solutions. The batch adsorption study was conducted at 2 pH, and removal efficiency was 90% when the concentration of acid blue 19 dye was 15.0 mgL^{-1} and the dosage of bioadsorbent of 4.0 gL^{-1} [69].

3.5. Kinetic Studies of MGP and RPP. To investigate the removal rate of the dye by MGP, different kinds of the kinetic model were used to perform the kinetic studies. Pseudo-first-order and pseudo-second-order kinetic model was examined on primary bases, which clearly showed that it does not follow pseudo-first-order as well as pseudo-second-order (Figures 10(a) and 10(b)). Intraparticle diffusion was applied for the same, it follows the intraparticle

diffusion reaction (Figure 9(c)) as it followed a linear fitted curve ($R^2 = 0.96$). Table 3 depicts the kinetic parameters for the calculated pseudo-first- and second-order kinetics along with the intraparticle diffusion.

In addition, kinetic models were used to study the removal rate of dye by rose petal powder and plots are shown in Figures 11(a)–11(c). Both the pseudo-first and second-order kinetic models were examined on primary bases. This case also does not follow pseudo-first-order as well as second-order (Figures 11(a) and 11(b)), while intraparticle diffusion was found to be significant for the removal rate of dye, Figure 11(c). Intraparticle diffusion has also been mentioned by Igwegbe et al. while remediation of various dyes by using biosorbents [70]. Table 4 indicates the kinetic parameters for all models used in kinetics studies, where the intraparticle diffusion showed $R^2 = 0.910$.

Bhattacharjee et al. also reported kinetics and isotherms for the removal of dyes and heavy metal from the wastewater using watermelon rind as a bioadsorbent. The investigator concluded that the dyes are being adsorbed on the surface

of bioadsorbent due to the presence of carboxyl and hydroxyl groups in the watermelon rind [71].

3.6. Adsorption Mechanism. Figure 12 is showing the possible mechanism of uptake of MR dye on the surface of floral powders based on the findings of modelling studies. There are several reports where it has been proved that the dye uptake is because of both the chemisorption and physisorption processes. Based on the parameters, like pH and temperature, the properties of various functional groups present on the surface of biosorbents also changes. The pKa value of MR is 5.1, it is red in color in acidic conditions (4.4), and yellow in color over 6.2 pH. The MR gets protonated below pKa value, i.e., 5.1, and there is generally electrostatic interaction. The biosorbent has mainly negative charge due to OH, and COOH, which interacts with dye due to electrostatic attraction. There are several forces like pi-pi interactions among the functional groups on the sorbents and the dye molecule. Besides this, there are electrostatic interactions like London dispersion interactions, van der Waal forces, and dipole-induced dipole bonds. The MR dye has two benzene rings, which are electron-rich areas that can induce could induce a donor-acceptor relationship. This will lead to a stacking effect of the dyes onto the surface of biosorbent. Sometimes, the dye molecules get inside the biosorbent via pore diffusion since the biosorbents are highly porous, which was evident from the FESEM. Similarly, a group of investigators led by Igwegbe et al. and Balarak predicated similar types of possible adsorption mechanism dyes like Congo red, malachite green, acid orange 7, and AB92. Igwegbe et al. used carbon as a biosorbents developed from rubber seed shells (*Hevea brasiliensis*) and used them for the removal of Congo red and malachite green dye from aqueous solutions. Balarak et al. also shown the summary of adsorption mechanism of AB92 dye onto single walled carbon nanotubes (SWCNTs). Balarak et al. also shown the acid orange 7 dye removal from the aqueous solutions by using synthesized mesoporous goethite [70, 72, 73].

4. Conclusion

The processing of sacred flowers offered at religious places for the development of value-added materials can prove to be an eco-friendly approach. The developed bioadsorbents are carbon-rich, micron-sized, and free from other elements. The sacred flowers from religion can be utilized as an efficient and economical adsorbent for the remediation of methyl red dye. Out of both, rose petal powder and marigold petal powder, rose petal powder was found slightly efficient in comparison to the marigold petal powder. The development of value-added materials from waste sacred flowers will reduce solid waste as well as water pollution leading due to the dumping of flowers in the water bodies.

Data Availability

All relevant data are included within the article.

Conflicts of Interest

There are no conflicts to declare.

Authors' Contributions

Abdelfattah Amari and Virendra Kumar Yadav contributed equally to this work.

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