

Surface Modification of Activated Carbon for the Removal of Congo Red Dye from Aqueous Media.

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Abstract: *The objective of the research was to examine the effect of increase in surface basicity of activated carbon on removal of Congo Red dye by activated carbon, the efficiency of carbon based adsorbents F400 and Lurgi were investigated for the removal of noxious Congo Red (CR) dye from aqueous environment. SEM shows presence of highly developed microporous nature of both adsorbents responsible for removal of Congo red from aqueous media. FTIR spectroscopic analysis found multiple functional groups on adsorbents and their interactive effects played important roles in dye removal. Batch experiments were carried out to determine adsorption isotherms. Experimental results showed that the rate of adsorption increased with increase in surface modification using SDS. Adsorption equilibrium was achieved within 3 h with efficiency of up to 91% at equilibrium. Batch experiments were carried out to determine adsorption isotherms. Langmuir adsorption isotherm model and Freundlich model gave a good fit to the experimental data. Results furnish evidences that both adsorbents are capable and efficacious, for dye removal from industrial effluents after surface modification with SDS as compared to bare carbon. In present work efficiency of different carbon-based materials for removal of Congo red dye from aqueous media have been studied, activated carbon's surface has been modified to improve the removal of Congo red dye from aqueous environments and characterized for proximate and ultimate analysis, iodine number, N₂-BET surface area SEM and FTIR*

Keywords: Activated carbon, adsorption, Langmuir Adsorption isotherm, Freundlich Adsorption isotherm, Congo red dye

I. INTRODUCTION

Water is the most important natural resource for sustaining life. Groundwater contributes only 0.6% of the total water resources on earth which is the major and preferred source of drinking water in rural as well as urban areas. It is getting polluted due to urbanization and industrialization in addition to geogenic contamination. Anions are commonly present in water; however, some of the anions including oxy-anions are undesired and often responsible for serious environmental and health problems. Effluents from textile industries are characterized by severe variations in many parameters such as BOD, turbidity, color, COD, pH, odor and salinity. The effects of water pollution on human health comprise hazardous heavy metals and dyes, besides other contaminants [1]. The textile, paper, plastic, leather, culinary, cosmetic, and industrial sectors of both natural and synthetic fibers, dyes and pigments are frequently used to color Products [2]. The Azo dyes are one of the most widely used classes of synthetic organic pigments used in colored manufactured goods worldwide, and as a result, they are commonly identified in industrial effluents [3]. The primary components of textile effluent are dyes. According to certain reports, some dyes might cause inflammatory dermatitis, skin rashes, cancer, and genetic mutations in people [4, 5]. These colors and their precursors are poisonous and carcinogenic, endangering the environment. Additionally, as their breakdown frequently produces extremely carcinogenic aromatic amines [6], adequate treatment for this contaminated water is required before reusing colored wastewaters. One significant dye found in wastewater, congo red, has a higher solubility in water, roughly 1 g/30 mL [7].



The removal of organic and inorganic pollutants from waste water by activated carbon depends mostly upon surface area, pore volume and pore size distribution. Various Inorganic contaminants such as Congo red dye which is most common effluent from industrial waste of various dyes industries like textiles, paper, plastics, leather, foodstuff, cosmetic. As reported the concentration of congo red dye in surface water is very low i.e., 0.15-0.5 mg/l, whereas higher concentration has been reported in ground waters up to 20 mg/L.

Worldwide, environmental scientists' focus has been developing efficient and sustainable technologies for water and wastewater management [8,9,10]. To overcome the shortcomings of more traditional approaches, cheaper and more efficient techniques to improve the quality of treated effluent have been proposed. Some of the widely used methods include adsorption, membrane filtration, coagulation and flocculation, chemical precipitation, ion exchange, electrochemical removal, biosorption, reverse osmosis, and oxidation processes [11,12,13–18]. However, most of those methods involve high operational and maintenance costs. Among the techniques mentioned, the adsorption process using local biowastes is a cost-effective and efficient technique for removing toxic dyes and metal ions from wastewater [12]. Dyes are widely used in textile, food, printing, leather, and pharmacology industries [19–21]. The presence of dyes in aqueous solutions and the environment can affect the photosynthetic functions of plants in water by blocking the sunlight with its aromatic compounds and reducing dissolved oxygen [19,21–24]. Plus, some dyes, especially cationic and anionic, are also carcinogenic and mutagenic that can affect digestive tract irritation, skin irritation, and cyanosis [18–22]. In addition, cationic dyes like MB and anionic dyes such as CR are the most widely used dyes in industries [21]. Thus, it is crucial to control the release of these compounds to the environment. The adsorption method typically uses activated carbon from natural resources that have high carbon content. Activated carbon is widely used to remove dyes from wastewater because of its convenience, ease of operation, simplicity of design, and reusability [25–27,28–30]. However, despite its extensive use in wastewater treatment, some commercial activated carbon remains an expensive material [28–30]. Therefore, interest in lower-cost alternatives to commercially available activated carbon that would still provide safe and economical methods of removing dyes from contaminated water has increased. For this reason, low-cost by-products of agriculture that able to act as an adsorbent have been recognized as a sustainable solution for wastewater treatment that minimizes waste, recovery, and reuse [31,32].

On the other hand, Fig. 16 shows the Langmuir and Freundlich linear plots for CR adsorption onto AC. As can be observed, both models match experimental data well, with R^2 values around 0.90. The application of both isotherms demonstrates that monolayer homogeneous adsorption and heterogeneous energetic distribution of active sites on the adsorbent's surface occur at the same time as adsorption onto the FA. One of the key reasons for heterogeneous adsorption and monolayer process is high-energy adsorption sites. Another factor is the surface condensation of liquid adsorbates. The first two levels interact with the surface, whereas molecules beyond the first two layers interact with one other, resulting in multilayer adsorption. The mechanism of dye adsorption is complicated, and both homogeneous and heterogeneous adsorption occur simultaneously in this adsorption process, according to the aforementioned arguments. The adsorption process can, however, be better described using the Langmuir model.

The solid surface can be modified by the surfactant to form micelle-like structures in different size and stabilities on its surface having the potential to solubilize organic molecules within the structures formed. These micelles are called hemimicelle or admicelle and the phenomenon is called adsolubilization [13,14]. Surfactants are used very often to modify the surface properties (e.g., hydrophobicity, hydrophilicity) of various materials. This paper aims to compare the efficiencies of two activated carbons for removing of methylene blue dye from aqueous solutions using treated and untreated activated carbons. This paper aims to compare the efficiencies of two activated carbons for removing of methylene blue dye from aqueous solutions using treated and untreated activated carbons. The activated carbon (AC) used in this study is a commercially available carbon F400 (filtrisorb-400) agent. The other carbon is surface modified F400 (filtrisorb-400) using sodium dodecyl sulphate. It has been shown that surface modified activated carbon has better adsorption capacity than unmodified activated carbon.

Adsorption technology has been identified as a potential broad spectrum treatment option. The adsorption of Congo Red dye on impregnated carbon was dependent upon both the pH of the impregnating solution and the temperature of



calcination. Impregnated carbon was shown to have a dye adsorption capacity of 3 to 4 times that of plain activated carbon.

Nomenclature

| | |
|----------------------|--|
| <i>b</i> | adsorption energy constant of Langmuir adsorption isotherm (mg ⁻¹) |
| <i>C₀</i> | initial liquid phase concentration (mg l ⁻¹) <i>C_e</i> equilibrium liquid phase concentration (mg l ⁻¹) |
| <i>k₁</i> | rate constant of first-order adsorption (min ⁻¹) |
| <i>k₂</i> | rate constant of second-order adsorption (g g ⁻¹ min ⁻¹) |
| <i>K_F</i> | Freundlich isotherm constant related to adsorption capacity [(mg ⁻¹)(mg ⁻¹) ^{1/n}] |
| <i>n</i> | Freundlich isotherm constant related to adsorption intensity |
| <i>q_e</i> | equilibrium solid phase adsorbate concentration (mg g ⁻¹) |
| <i>Q_t</i> | amount of adsorption at time <i>t</i> (mg g ⁻¹) |
| <i>Q</i> | the maximum surface coverage (formation of monolayer) of sorbent (mg g ⁻¹) |
| <i>R_L</i> | dimensionless separation factor |
| <i>R₂</i> | correlation coefficient |
| <i>V</i> | volume of solution (l) |
| <i>W</i> | mass of adsorbent (g) |

Subscripts

| | |
|-----|--------------|
| exp | experimental |
| cal | calculated |

II. MATERIALS AND METHODS

2.1 Materials

Filtrisorb-400(F400) activated carbon from Pittsburgh, USA and Hydrarffin UV 43 (Lurgi) , Aktivkolhe , Germany were used in this study. The carbons were crushed to desired mesh size 72BS mesh, washed with distilled water and dried in a moisture oven at 108±20C .Physico chemical properties of Activated carbon are shown in Table I. Sodium dodecyl sulphate (SDS) of GR grade, Merck, USA were procured from local market.

Modification of activated carbon: About 10g of washed F400 & Hydrarffin UV 43 activated carbon of size72 BS mesh (212mm) were impregnated with 100ml of 0.1% of Sodium dodecyl sulphate (SDS). The impregnated carbons were washed with distilled water till no leachate was observed and dried in a moisture oven at 108±20C.

Estimation of Congo Red Dye (CR): The concentration of CR in the supernatant solution after and before adsorption was determined using a double beam UV/VIS spectrophotometer (Lambda 35) at 620 nm by Spectroscopic method.

Method for adsorption: Batch equilibrium studies: Adsorption isotherms were performed in a set of 10 BOD bottles (250 mL) where 50 ml solution of CR in the concentration range 10-100 ppm, accurately weighed 0.100 ± 0.0001g carbon sample was then introduced in each bottle and kept in an isothermal shaker (30 ± 1 °C) for 24 hrs to reach equilibrium of the solid-solution mixture. After shaking for predetermined time intervals, samples were withdrawn from the bottles and dye were separated from the adsorbent by centrifugation. CR concentrations in the supernatant solutions were estimated by measuring absorbance at 620 nm with Perkin Elmer UV/VIS spectrophotometer.

Adsorption isotherms: The amount of adsorption at equilibrium, *Q_e* (mg g⁻¹), was calculated by:

$$Q_e = \frac{(C_0 - C_e)V}{W} \quad (1)$$

where *C₀* and *C_e* (mg l⁻¹) are the liquid-phase concentrations of Congo Red at initial and equilibrium, respectively. *V* is the volume of the solution, and *W* (g) is the mass of dry adsorbent used.

Langmuir isotherm can be used as a model to describe the adsorption isotherm. The Langmuir equation is given as;



Where C_e is the concentration of adsorbate solution at equilibrium (mg l⁻¹), q_e is the amounts of adsorbate adsorbed per mass of adsorbent (mg g⁻¹), b is the equilibrium constant related to the sorption energy between the adsorbate and adsorbent (dm³ mg⁻¹) and Q_0 is limiting amount of adsorbate that can be taken up per mass of adsorbent. The well-known logarithmic form of Freundlich model is given by the following equation:

$$\frac{C_e}{q_e} = \frac{1}{(Q_0 \times b)} + \frac{C_e}{Q_0} \quad (2)$$

where q_e is the amount adsorbed at equilibrium (mg g⁻¹), C_e the equilibrium concentration of the adsorbate and KF and n are Freundlich constants, n giving an indication of how favorable the adsorption process and KF is the adsorption capacity of the adsorbent.

$$\log q_e = \log KF + \left(\frac{1}{n}\right) \log C_e \quad (3)$$

Effect of Contact Time: The value of optimum contact time was varied in the range of 1-4 hour in a series of experiments in which the initial Congo red dye concentration 100 ppm, the temperature 20°C and adsorbent amount 1g per 100ml. The effect of contact time on the removal of CR by the adsorbent is illustrated in Table 2. It is observed that dye is rapid adsorption at the first time (first hour) and then the process gradually it becomes slowly and finally the process continuous to becomes almost constant, this effect result from coverage the surface of adsorbents by the molecules of Congo red dye [5,12]. Table.1 Effect of contact time Contact time (hour) 1 ,2, ,3, 4 Congo Red dye removal % 81.4 ,91.9 ,94.8 ,95.8 respectively.

Table.1 Effect of contact time Contact time on CR removal.

| Contact time (hour) | 1 | 2 | 3 | 4 |
|-------------------------|------|------|------|------|
| Congo Red dye removal % | 80.1 | 89.9 | 91.8 | 92.3 |

Effect of pH

The following procedure was adopted to verify the effect of pH on the removal of Congo Red dye. The maximum capacity of the sorbents for the sorption of dye was measured as a function of sample pH 3,4,5,6,7,8,9,10 &11. pH was adjusted accordingly using 0.1 N NaOH or 0.1N HCL. 100 ml. of each pH adjusted sample was transferred into 250 ml capacity glass bottle. An adsorbent dose of 0.5 gm was added to each glass bottle. Bottles were stoppered and fixed to shaking machine and agitated for a desired period. Samples were withdrawn from the machine, and arranged serially. The supernatant liquid was centrifuged and filtered through Whatman paper no.41. Filtrate was used for the analysis of CR Dye remaining. The initial pH of dye solution plays an important role for adsorption process because the initial pH has direct influence on the dye and adsorbent in aqueous solution [16]. the effect of pH is studied between 4 and 9 because reported that at strong acidic medium, the solution of Congo Red changes its colour from red to dark blue and the original red colour is different above pH 10. pH values the removal Congo red percentage the maximum removal efficiency is achieved in the acidic medium and gradually reduced to the basic medium.

III. RESULT AND DISCUSSION

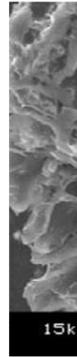
The calculated Langmuir constants are given in Table II. As shown in the table II, the resulting Q_0 value for modified F-400 is 7.58 mg g⁻¹ compared to 2.52 mg g⁻¹ for unmodified and modified Hydrffin UV 43 is 7.05 mg g⁻¹ compared to 3.86 mg g⁻¹ for unmodified. The results indicated that modified carbon shows slightly higher adsorption capacity of fluoride than unmodified AC. The plot of C_e/Q_e vs C_e in Fig: 3 a and b for F400 and fig. 3 c and d for Lurgi gave straight line implies that the adsorption for both adsorbents well fitted to Langmuir adsorption model.



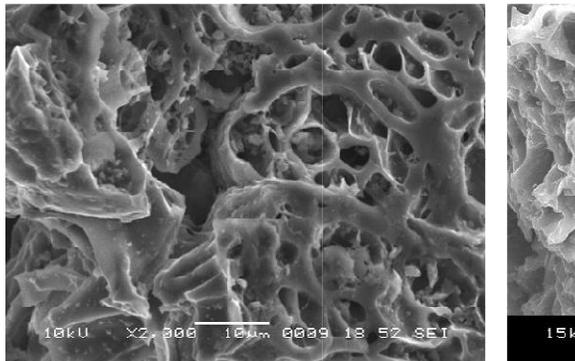
SEM Morphology:



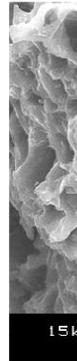
Electron Micrograph Lurgi



Electron Micrograph Lurgi



Electron Micrograph F400



Electron Micrograph F400

Figure 1: SEM morphology

Table I: Physico-chemical properties of Activated carbons

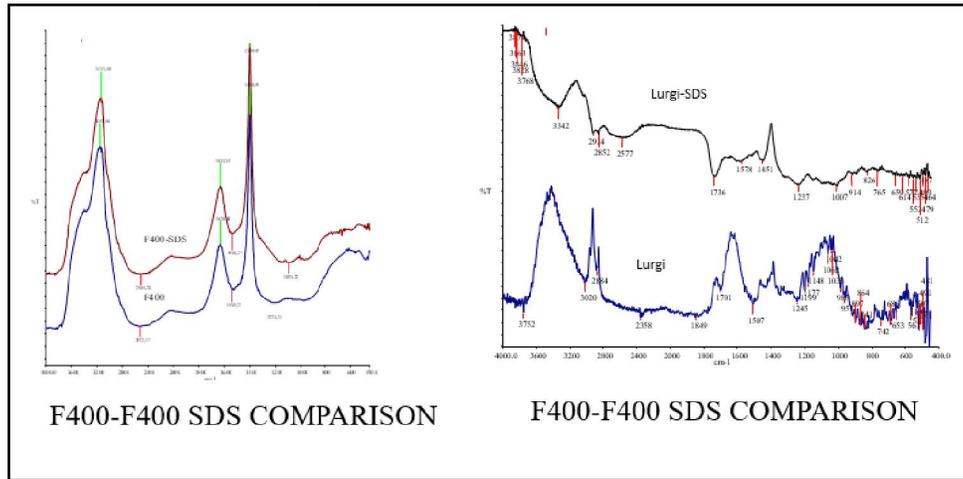
| carbon | M% | Ash% | V.M% | F.C% | C % | H % | Iodine Value | N2-BET surface area | Pore volume |
|-----------------|-----|------|------|------|-------|------|--------------|---------------------|-------------|
| F-400 | 1.9 | 6.1 | 2.7 | 89.3 | 96.04 | 0.29 | 1193 | 998 | 0.825 |
| Hydraffin UV 43 | 2.9 | 3.6 | 4.9 | 88.6 | 97.09 | 0.87 | 1024 | 1112 | 0.802 |



Table II: Adsorption isotherm data

| Adsorbent | Langmuir adsorption constants | | Freundlich adsorption constants | |
|-------------------|-------------------------------|---------|---------------------------------|-------|
| | Q ₀ | B | K _f | 1/n |
| Activated carbons | | | | |
| F400 | 37.03 | 925.92 | 19.37 | 0.167 |
| F400-SDS | 100 | 58823.5 | 43.35 | 0.467 |
| Lurgi | 76.92 | 1165.5 | 857.03 | 0.564 |
| Lurgi-SDS | 90.90 | 2392.34 | 1377.20 | 0.598 |

FTIR:



Figures:

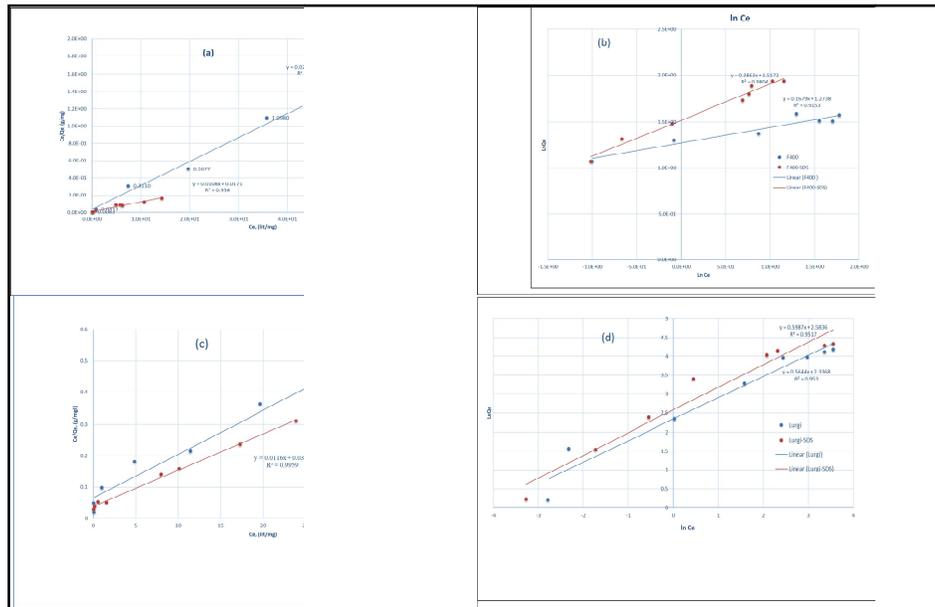


Figure 3: a) Langmuir Adsorption isotherm CR-F400 b) Freundlich adsorption isotherm CR-F400 c) Langmuir Adsorption isotherm CR-Lurgi d) Freundlich Adsorption isotherm CR-Lurgi



| Adsorbent | Langmuir adsorption constants | | Freundlich adsorption constants | |
|------------------|-------------------------------|----------|---------------------------------|-------|
| | Q_0 | B | K_f | 1/n |
| Activated carbon | | | | |
| F400 | 37.03 | 925.92 | 19.31 | 0.167 |
| F400-SDS | 100 | 58823.5 | 43.35 | 0.467 |
| Lurgi | 76.92 | 1165.5 | 857.03 | 0.564 |
| Lurgi-SDS | 90.90 | 2392.34 | 1377.20 | 0.598 |
| ABAC | 250 | 6410.25 | 216.77 | 0.692 |
| ABAC-SDS | 333.3 | 25641.02 | 382.82 | 0.650 |

IV. CONCLUSIONS

In the present study it has been conclude that;

1. Adsorption isotherms of Congo Red on modified and unmodified activated carbons are favorable.
2. Modified activated carbons with SDS have better adsorption capacity than unmodified activated carbons. SDS modification enhances the adsorption capacity 2 times in case of Lurgi, 3 times for F 400.
3. As shown in the table II, the resulting Q_0 value for modified F-400 is 100 mg g⁻¹ compared to 37.03 mg g⁻¹ for unmodified and modified Lurgi is 90.90 mg g⁻¹ compared to 76.92 mg g⁻¹ for unmodified... The results indicated that modified carbon shows slightly higher adsorption capacity of dye than unmodified AC.
4. The plot of C_e/q_e vs C_e in Fig: 1.1 for F400 & Fig.1.3 for Lurgi gave straight line implies that the adsorption for both adsorbents well fitted to Langmuir adsorption model.
5. SEM analysis shows the morphology of both the carbon samples before and after modification. It was observed that morphology of modified carbon changed slightly as compared to unmodified carbon.
6. Scanning Electron Microscopy (SEM), Iodine value, Proximate and Ultimate analysis show that the source material used for preparation and modification of activated carbon samples has significant effect on its pore structure, surface texture and characteristics.
7. Different characterization methods were applied such as SEM, FTIR, Iodine Value and N₂ BET surface area.
8. The influences of contact time and pH were studied. The results indicated that the CR dye adsorption capacity improved with increasing contact time and achieved maximum adsorption after 4 hours. pH is significant factor in adsorption process and gives maximum uptake at pH 10 thereafter it is not showing any significant change in adsorption capacity.
9. The Langmuir model fit better the adsorption results compared to the Freundlich model, in both conditions before and after surface modification.
10. The presence of various acidic groups on the adsorbent surface formed during the activation and surface modification process is corroborated by FTIR spectra.

REFERENCES

- [1] Adam, A. M. A. et al. Preparation and characterization of new CrFeO₃-carbon composite using environmentally friendly methods to remove organic dye pollutants from aqueous solutions. *Curr. Comput.-Aided Drug Des.* 11(8), 960. <https://doi.org/10.3390/cryst 11080960> (2021).
- [2] Alzaydien. A.S. Adsorption of methylene blue from aqueous solution onto a Low-cost Natural Jordanian Tripoli, *Am.J. of Environ. Sci.* vol5. pp197-208. 2009.
- [3] Rys .P. and Zollinger .H. *Fundamentals of Chemistry and Applications of Dyes*, John Wiley & Sons Inc. USA 1972.
- [4] Alau. K.K. *Archives of Applied Science Research*vo, Vol. 12, No. 5. pp. 456-461. 2010.
- [5] Dawood .S.;SenT.K. Removal of anionic dye Congo red from aqueous solution by raw pine and acid-treated pine cone powder as adsorbent: Equilibrium, thermodynamic, kinetics mechanism and process design, *Water Res.* Vol. 46.pp1933-1946. 2012.



- [6] Somasekhara .R.; Sivarama K. L.; and Varada.R. A, The use of an agricultural waste material, Jujuba seeds for the removal of anionic dye (Congo red) from aqueous medium J Haz. Mats. Vol. 203. pp118-127. 2012.
- [7] Gupta .V.K.; Mittal .A. R.; Jain .M.; MathurSikarwar.S, Photochemical degradation of hazardous dye- Safaranin-T using TiO₂ catalyst, J. Colloid. Interface Sci. Vol. 309, pp460-465. 2007.
- [8] Masoudian, N., Rajabi, M. & Ghaedi, M. Titanium oxide nanoparticles loaded onto activated carbon prepared from bio-waste watermelon rind for the efficient ultrasonic-assisted adsorption of congo red and phenol red dyes from wastewaters. Polyhedron 173, 114105 (2019).
- [9] Gupta, V. K. & Nayak, A. Cadmium removal and recovery from aqueous solutions by novel adsorbents prepared from orange peel and Fe₂O₃ nanoparticles. Chem. Eng. J. 180, 81–90 (2012).
- [10] Ranjan, B., Pillai, S., Permaul, K. & Singh, S. Simultaneous removal of heavy metals and cyanate in a wastewater sample using immobilized cyanate hydratase on magnetic-multiwall carbon nanotubes. J. Hazard. Mater. 363, 73–80 (2019).
- [11] Burakov, A. E. et al. Adsorption of heavy metals on conventional and nanostructured materials for wastewater treatment purposes: A review. Ecotox. Environ. Safe. 148, 702–712 (2018).
- [12] Hanafah, M. M., Hashim, N. A., Ahmed, S. T. & Ashraf, M. A. Removal of chromium from aqueous solutions using a palm kernel shell adsorbent. Desalin. Water Treat. 118, 172–180 (2018).
- [13] Al-Raad, A. et al. Treatment of saline water using electrocoagulation with combined electrical connection of electrodes. Processes. 7, 242 (2019)
- [14] Banch, T. J., Hanafah, M. M., Alkarkhi, A. F., Amr, A. & Salem, S. Factorial design and optimization of landfill leachate treatment using tannin-based natural coagulant. Polymers 11, 1349 (2019).
- [15] Fu, F. & Wang, Q. Removal of heavy metal ions from wastewaters: a review. J. Environ. Manage. 92, 407–418 (2011).
- [16] Im, D., Nakada, N., Fukuma, Y. & Tanaka, H. Effects of the inclusion of biological activated carbon on membrane fouling in combined process of ozonation, coagulation and ceramic membrane filtration for water reclamation. Chemosphere 220, 20–27 (2019).
- [17] Rybak, M., Drzewiecka, K., Woźniak, M., Ratajczak, I., Joniak, T. Iron-induced behavioural and biochemical responses of charophytes in consequence of phosphates coagulant addition: Treats to lake ecosystems restoration. Chemosphere. 126844 (2020).
- [18] Yao, F. et al. Electrochemical Cr (VI) removal from aqueous media using titanium as anode: Simultaneous indirect electrochemical reduction of Cr (VI) and in-situ precipitation of Cr (III). Chemosphere 260, 127537 (2020).
- [19] Foroutan, R., Peighambaroust, S. J., Aghdasinia, H., Mohammadi, R. & Ramavandi, B. Modification of bio-hydroxyapatite generated from waste poultry bone with MgO for purifying methyl violet-laden liquids. Environ. Sci. Pollut. Res. 27(35), 44218–44229 (2020).
- [20] Pashaei-Fakhri, S., Peighambaroust, S. J., Foroutan, R., Aرسالani, N. & Ramavandi, B. Crystal violet dye sorption over acrylamide/graphene oxide bonded sodium alginate nanocomposite hydrogel. Chemosphere 270, 129419 (2021).
- [21] Esvandi, Z., Foroutan, R., Peighambaroust, S. J., Akbari, A. & Ramavandi, B. Uptake of anionic and cationic dyes from water using natural clay and clay/starch/MnFe₂O₄ magnetic nanocomposite. Surf. Interfac. 21, 100754 (2020).
- [22] Foroutan, R. et al. Performance of montmorillonite/graphene oxide/CoFe₂O₄ as a magnetic and recyclable nanocomposite for cleaning methyl violet dye-laden wastewater. Adv. Powder Technol. 31(9), 3993–4004 (2020).
- [23] Foroutan, R., Peighambaroust, S.J., Esvandi, Z., Khatooni, H. and Ramavandi, B. Evaluation of two cationic dyes removal from aqueous environments using CNT/MgO/CuFe₂O₄ magnetic composite powder: A comparative study. J. Environ. Chem. Eng. 104752 (2020).
- [24] Foroutan, R. et al. Calcined alluvium of agricultural streams as a recyclable and cleaning tool for cationic dye removal from aqueous media. Environ. Technol. Innov. 17, 100530 (2020).



- [25] Chowdhury, S., Pan, S., Balasubramanian, R. & Das, P. Date palm based activated carbon for the efficient removal of organic dyes from aqueous environment. *Sustain. Agric. Rev.* 34, 247–263 (2019).
- [26] Masoudian, N., Rajabi, M. & Ghaedi, M. Titanium oxide nanoparticles loaded onto activated carbon prepared from bio-waste watermelon rind for the efficient ultrasonic-assisted adsorption of congo red and phenol red dyes from wastewaters. *Polyhedron* 173, 114105 (2019).
- [27] Wang, G., Li, G., Huan, Y., Hao, C. & Chen, W. Acrylic acid functionalized graphene oxide: High-efficient removal of cationic dyes from wastewater and exploration on adsorption mechanism. *Chemosphere* 261, 127736 (2020).
- [28] Foroutan, R., Mohammadi, R., Razeghi, J. & Ramavandi, B. Performance of algal activated carbon/Fe₃O₄ magnetic composite for cationic dyes removal from aqueous solutions. *Algal Res.* 40, 101509 (2019).
- [29] Foroutan, R., Mohammadi, R. & Ramavandi, B. Elimination performance of methylene blue, methyl violet, and Nile blue from aqueous media using AC/CoFe₂O₄ as a recyclable magnetic composite. *Environ. Sci. Pollut. Res.* 26(19), 19523–19539 (2019).
- [30] Bonyadi, Z. et al. Ultrasonic-assisted synthesis of *Populus alba* activated carbon for water defluorination: application for real wastewater. *Korean J. Chem. Eng.* 36(10), 1595–1603 (2019).
- [31] Charola, S., Patel, H., Chandna, S. & Maiti, S. Optimization to prepare porous carbon from mustard husk using response surface methodology adopted with central composite design. *J. Clean. Prod.* 223, 969–979 (2019).
- [32] Said, M., Mohammad, A. W., Nor, M. T. M. & Abdullah, S. Investigation of three pre-treatment methods prior to nano filtration membrane for palm oil mill effluent treatment. *Sains Malays.* 44, 421–427 (2015).

