ADSORPTION STUDIES OF FULLERS EARTH NANOCOMPOSITES FOR THE REMOVAL OF COPPER AND REACTIVE YELLOW 18

M. JAMEEL[a], S. A. KHAN[b,*], A. AFZAL[c]
aDepartment of Chemistry, School of Science, University of Management and Technology, Lahore 54770, Pakistan
bCenter of Super-Diamond and Advanced Films (COSDAF) and Department of Chemistry, City University of Hong Kong, 83 Tat Chee Avenue, Kowloon 999077, Hong Kong
cDepartment of Chemistry, University of Agriculture Faisalabad-38000, Pakistan

Fuller's earth of D. G. Khan was used as a low-cost adsorbent to remove copper and reactive yellow 18 from aqueous solutions as it has the capacity of adsorption of toxic particles in its structure. Because of this capacity to take up the ionic component, utilization of fuller's earth has experimented for the purification of wastewater in the laboratory. Sampling and physical processing by grinding and sieving/classification were conducted. Characterization of fuller's earth (adsorbent) was carried out by X-Ray Fluorescence (XRF), Scanning Electron Microscope (SEM), and Fourier Transform Infrared (FTIR). The result of XRF revealed the presence of large proportion of metal oxides like TiO_2 (0.78%), Fe_2O_3 (3.13%), Al_2O_3 (12.38%), MgO (2.16%), CaO (10.73%), Na_2O (0.22%), P_2O_5 (0.11%), Cl (0.03%), K_2O (2.63%), MnO (0.03%), C (1.30%) and SiO_2 (66.31%) in the fullers earth. SEM images show the morphology, porous nature, and different micro size particles of the adsorbent. FTIR results show the presence of different functional groups. The batch adsorption process was performed, and different operating parameters such as contact time, the concentration of fuller's earth, adsorbate concentration, pH values, and temperature were evaluated to find the maximum level of adsorption. Contact time of 100 minutes, 100 mg/L initial adsorbate concentration, 0.5 g adsorbent dosage at 65 ^{o}C temperature are the optimum values at which percentage removal is maximum, i.e., 96% for copper at pH 6 and 68% for reactive yellow 18 at pH two by Fullers earth was achieved. The solid addition method describes the pH point of zero charges, which is 4 for fuller's earth. Maximum adsorption at high temperature indicates that this adsorption process is endothermic.

1. Introduction

Pollution of heavy metals and dyes has turned out to be among today's most serious ecological problems. Because of their stubbornness and laboriousness in design, the therapy of substantial metals and dyes are of great concern [1]. Due to several benefits like working under gentle circumstances, giving vivid colors, and sustainable systems, reactive dyes are by far the most prevalent dyes. Reactive colorants are nitrogen, comprising heterocyclic bodies comprising halogen substitutes consequently receive a nucleophilic replacement interaction with wool fibers [2]. Waste effluent from passive dyeing procedures has developed a specific issue since the dyes can display small fiber fixation concentrations. Highly water-soluble, the strongly tinted unpatched colors, will not be withdrawn by standard therapy technologies. The wastewaters of textiles are incredibly poisonous and cancerous. Coloration is one of these sewage flows' features and decreases photosynthetic activity by hindering the permeability of sunlight. From this current perspective, the colors and dyes trigger severe ecological issues as they absorb the dissolved oxygen and dismantle flora and fauna. Neurologic and respiratory diseases, nasal sinusitis and

---

* Corresponding author: shakilahmad56@gmail.com

psoriasis, infectious melanoma of interaction, adult neurogenesis, teratogenicity, cancer, and genotoxicity are caused by reactive dyes [2].

From the perspective of water contamination, the most significant heavy metals are Zn, Cu, Pb, Cd, Hg, Ni, and Cr. Several of these metals (Zn, Cr, Ni, and Cu) are vital decipher metals for living systems, but at high levels, they become poisonous. As a consequence of leaching from landfill sheds, polluted water, and land, metals are accelerated and transported to the energy chain [3]. Once they access the biosphere, these poisonous chemicals acquire massive quantities of this component in individuals and livestock, triggering countless negative environmental impacts [4]. Their withdrawal from sewage is, therefore, essential until dissipating into the aquatic setting.

Numerous techniques were utilized to remove dyes from wastewater, such as physiological, biological, and chemical procedures. Biological and chemical methods are efficient for discoloration, but they involve power and specific machinery; they sometimes produce large side products. Physical techniques like adsorption, interchange of ions, and reverse osmosis are usually efficient for discoloration without generating illegitimate by-products [5].

Adsorption is an effective method of environmental segregation for effluent medium purification. One of its primary and significant advantages of such a demarcation method is that it can be easy to manage or regulate and efficient in eliminating harmful chemicals even in very trace amounts [6]. Based on the type of factors concerned, adsorption procedures can be categorized as chemical and physical. The adsorption method is influenced by several chemical and physical variables, including solute-solvent association, the curvature of adsorbent and membrane composition, external layer morphology, adsorbent properties, specific particle impact, column magnitude, heat, pH, interaction period, and so forth. Adsorption procedures provide such a viable therapy, mainly when the adsorbent is cheap and easily accessible [7].

Though the adsorbent selection is focused on economic and convenient factors, and clay mineral resources are limited-cost organic products Latest study has indicated that clays can remove around seventy percent of water pollution in a sewage extraction scheme. In comparison, the remaining can remove more than thirty percent utilizing other adsorbent products [8]. Because of its accessibility and simple implementation, clay has gained specific consideration as an appropriate and budget-effective adsorbent for sewage removal of toxic chemicals, even though it is hard to disentangle from drainage [9]. In Fullers Earth, there are three or four significant clay groups mineral components. Low concentrations of several other clay minerals like illite, chlorite, and smectite deposits are also present. Many raw clays, together with other sedimentary ores, are combinations of such distinct kinds. Clay relates to a spontaneously generated product consisting mainly of coarse-grained raw materials that might have been fragile in texture and firm if washed or fired at suitable salinity.

My research aim is to determine the adsorption capability of fuller's earth against contaminants like copper and reactive yellow 18 for water purification and to discover a proper solution to reduce the toxin levels in our surroundings. By using the batch adsorption technique, fuller's earth will be utilized to withdraw copper and reactive yellow 18 dye from wastewater.

2. Materials and methods

2.1. Sample collection
Samples from the Dalana region, 15 Kilometers from D.G. Khan, were gathered in this research. Analyses have been conducted on the regionally available samples to check fuller's earth's adsorption characteristics.

2.2. Preparation of adsorbent
Initial specimens were crushed with a jaw crusher to decrease the particle size. By utilizing a roller grinder, particles of 1-inch size transform to particles of 0.5 to 0.2-inch size. Particles of very standardized size were taken for the experimental job by the sieving technique. To remove moisture, the fuller's earth was dried in the oven at 65 ^{o}C.

2.3. Batch studies

A stock solution of 1000 mg/L of copper and reactive yellow 18 was formulated, and more dilutions of various levels were designed. By adding 0.1 M HCl or 0.1 M NaOH, the pH was set from 2 − 10 using a pH meter. Batch adsorption was performed by adding 0.1 g of the adsorbent fuller's earth to 10ml of a present RY-18 dye and Cu metal ion solution at 300 K room temperature. The influence of different working requirements like interaction period (40-120 mins), pH (2-10), the quantity of adsorbent (0.1-2g), initial Cu ions, and RY-18 dye concentration (100ppm-25ppm) and temperature (25-65 ^{o}C) was observed. One parameter has been modified in each experiment, while the others have been held steady. When a specified interaction period was completed, the contact sample was separated, and the UV visible spectrophotometer (UV-1800 Series) was used to evaluate the quantity of Cu ions and RY-18 dye left in the filtrate. Each test was conducted repeatedly, and the average sum was calculated, and the amount of metal ions and dye adsorbed was evaluated using the following equations:

$$qe = V(Co - Ce)/m \qquad \qquad (1)$$

$$\% \; Removal = 100[(Co - Ce)/Co] \qquad \qquad (2)$$

where qe (mg/g) is the adsorption capacity of the adsorbent for the adsorbate at equilibrium, Co (mg/L) and Ce (mg/L) are the initial and equilibrium concentrations of metal ions in solution respectively, m (g) is the mass of adsorbent, and V (L) is the volume of solution used.

2.4. Characterization of adsorbent

Adsorbent (Fullers Earth) were characterized by X-Ray Fluorescence (XRF), Scanning Electron Microscope (SEM), and Fourier Transform Infrared Spectroscopy (FTIR).

2.5. Determination of element percentage

The elemental analysis was carried out by using X-Ray Fluorescence Spectrometer (WD-XRF- Philips Axios Max).

2.6. Determination of surface morphology

Scanning Electron Microscope (JSM-6380A JEOL Japan) was helpful in determining the adsorbents surface essential physical characteristics and its adequate distribution of size.

2.7. Determination of functional groups

Fourier Transform Infrared spectroscopy (FTIR) of fuller's earth and sample loaded Reactive Yellow 18 dye, and Copper Sulphate was performed to determine the functional groups responsible for adsorption.

2.8. Evaluation of optimum contact time

To evaluate the impact of contact time (0.1 g) adsorbent was applied in the corresponding flasks with 100 mg/L of RY-18 dye and Cu (II) metal solutions for different time intervals (40-120 minutes). After that interaction period, the blend was screened, and by using a UV visible spectrophotometer (UV-1800 Series), absorbance was measured.

2.9. Evaluation of optimum adsorbate concentration

Solutions of various levels of RY-18 dye and Cu (II) metal (100, 75, 50, and 25 mg/L) have been designed. 10mL of these solutions were taken separately by adding a fixed amount of adsorbent (0.1g). By using a UV visible spectrophotometer (UV-1800 Series), absorbance was observed after the blend was stirred and screened.

2.10. Evaluation of optimum adsorbent concentration

To determine the optimal level of adsorbent, 100 mg/L of RY- 18 dye and Cu (II) metal solutions (adsorbate) was passed in a series of flasks with varying proportions of adsorbent 0.1,

0.5, 01, 1.5, and 2 g. By utilizing a UV visible spectrophotometer (UV-1800 Series), absorbance was observed after the blend was carefully screened and stirred.

2.11. Determination of pH point of zero charges (pHpzc)

To evaluate the point of zero charge (pHpzc) for fuller's earth, the Solid Addition method was used. A set of 0.1M KNO_3 solutions were designed (50 mL each), and their pH was customized from 01 – 12 by adding 0.1N HCl or NaOH. 0.1g of adsorbent (Fullers earth) was appended to each sample unit with continuous shaking of suspensions and was held for the duration of 48 hours with constant stirring. The final pH of the solution was estimated. A graph was drawn between the solution's initial pH on X-axis and the value obtained by the difference between initial and final pH (Δ pH) on Y-axis. The point of zero charges (pHpzc) of fuller's earth was yielded by the point of intersection of the resulting curve.

2.12. Evaluation of optimum pH

The pH of the mixtures would be customized to (2-10) by adjusting either 0.1 M HCl or 0.1M NaOH with adsorbent dose of 0.1g and 100mg/L RY-18 dye and Cu(II) metal solution concentration to determine the impact of ideal pH. By using a UV visible spectrophotometer (UV-1800 Series), absorbance was observed after the blend was screened.

2.13. Evaluation of optimum temperature

0.5 g of fuller's earth was applied in the corresponding flasks with 100mg/L solution of Cu (II) and RY 18 with constant stirring. The sample mixture after 100 min of interaction period was heated at temperature (25, 35, 45, 55, and 65 ^{o}C). UV Visible spectrophotometer (UV-1800 Series) was used to measure the absorbance of the sample solution.

4. Results and discussions

4.1. Characterization of adsorbent
4.1.1. Element analysis

The element analysis and estimation of significant and minor mineralogy of fuller's earth were performed utilizing an X-Ray Fluorescence spectrometer (WD-XRF- Philips Axios Max), and the outcomes provided are given in Table 1. X-Ray Fluorescence findings indicate the presence of more significant metal oxide proportions in the fuller's earth.

4.1.2. Surface morphology

Using a scanning electron microscope (JSM-6380A JEOL Japan), the surface morphology and particle size of fuller's earth at different magnifications were explored. The findings in Fig. 1 indicate that the adsorbent show irregular structure. The micrograph confirms that the material is forming micron-size agglomerates which conglomerate into larger size particles. The surface of fuller's earth is rough and contains a considerable number of pores of different shapes and sizes, which trapped and adsorbed the RY-18 dye and copper metal ions.

Table 1. Fullers earth chemical composition (Wt. %).

Element	Wt. %
TiO_2	0.78
Fe_2O_3	3.13
Al_2O_3	12.38
MgO	2.16
CaO	10.73
Na_2O	0.22
P_2O_5	0.11
Cl	0.03
K_2O	2.63
MnO	0.03
C	1.30
SiO_2	66.31

(a)   (b)
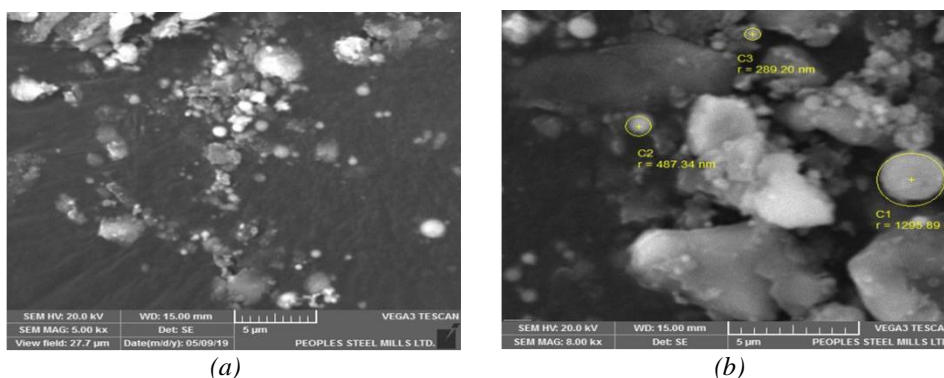
*Fig. 1. SEM images of Fullers earth nanocomposites (a and b) showing
the porous nature and micron-sized agglomerates.*

4.2. Characterization through FTIR studies

The FTIR profile of reactive yellow 18 dye before adsorption has shown different vibration peaks at 3070 cm$^{-1,}$ which was due to C-H stretching, 1594 cm^{-1} for N-H bending, 1574 cm^{-1} for C=C stretching, 1369 cm^{-1} for S=O stretching, 1275 cm^{-1} for CO stretching, 1071 cm^{-1} for C-N stretching, 1023 cm^{-1} for S=O stretching, 870 cm^{-1} for C=C bending and 786 cm^{-1} for C-Cl stretching. The FTIR spectrum of reactive yellow 18 is shown in Fig. 2 (a). The FTIR profile of copper sulfate pentahydrate before adsorption has shown different vibration peaks at 1418 cm^{-1} for O-H bending and 1369 cm^{-1} for S=O stretching. The FTIR spectrum of copper sulfate pentahydrate is shown in Fig. 2 (b). The spectra after adsorption showed excellent variety by breaking distinct functional groups, aromatic ring destruction, and dye molecule. After adsorption of the dye solution, the spectrum showed some minor peaks at 2101 cm^{-1} due to N=C=S stretching and 1635 cm^{-1} for C=C stretching. Similarly, after adsorption of copper sulfate, the spectrum shows some peaks at 2110 cm^{-1} due to N=C=S stretching and 1635 cm^{-1} for C=C stretching. The FTIR spectra for RY 18 and $CuSO_4.5H_2O$ are shown in Figs. 2 (c) and (d) respectively.
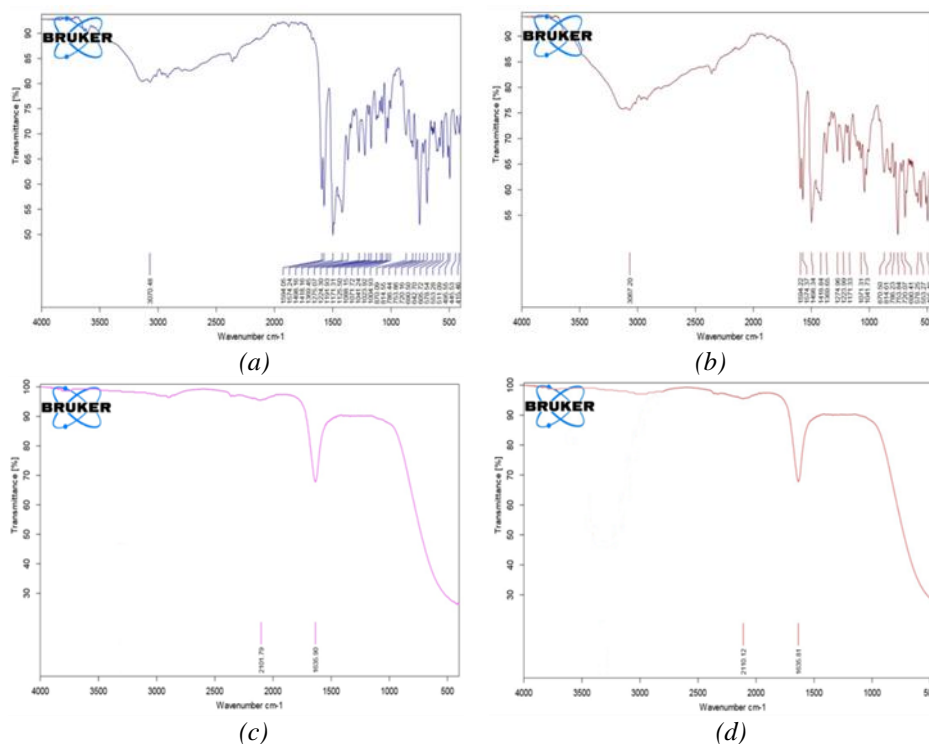
Fig. 2. FTIR spectra of (a) reactive yellow 18 dye, (b) copper sulfate pentahydrate, (c) reactive yellow 18 after adsorption, and (d) copper sulfate pentahydrate after adsorption.

4.3. Optimizing of fundamental batch process variables
4.3.1. Maximum wavelength (λ max)

The maximum wavelength was determined by UV Visible Spectrophotometer (UV-1800 Series). The λ max for the RY 18 dye was found to be 411 nm, and copper sulfate is 227 nm.

4.4. Impact of contact time

Contact time experiments were conducted to comprehend the pattern of copper and reactive yellow 18 dye adsorption by fuller's earth through a period of (40-120 min) by employing a fixed number of adsorbent (0.1 g) with 100 mg/L adsorbate quantity.

Fig. 3 (a) demonstrates the concentration of adsorbed copper and reactive yellow 18 dye per unit adsorbent mass enhanced with increasing time before a plateau was reached. The adsorption was improved by 40-120 minutes of contact time from 61-82% for copper and 43-62% for reactive yellow 18 dye. The equilibrium is accomplished after 100 minutes of contact time. When the contact time was extended, there was no noticeable improvement. During the earlier phases, fast adsorption may be owing to the existence of numerous clay surface active sites, which, over time, became saturated [10].
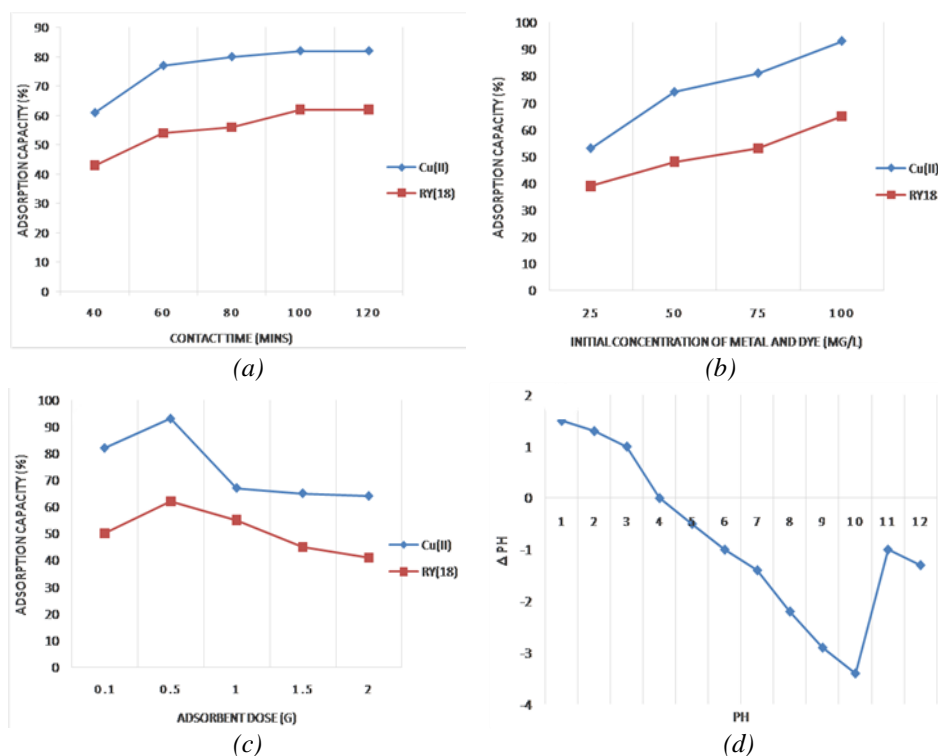
Fig. 3. Influence of contact time for the removal of copper and reactive yellow 18 by fuller's earth (a), Influence of initial concentration for the removal of copper and reactive yellow 18 by fuller's earth (b), Influence of adsorbent dose for the removal of copper and reactive yellow 18 by fuller's earth (c), and pH point of zero charges of fuller's earth (d).

4.5. Impact of initial copper and reactive yellow 18 dye concentration

The observations were conducted at an adjusted adsorbent dose (0.1 g) and varying initial copper and reactive yellow 18 dye concentrations (25, 50, 75, and 100 mg/L) after a contact period of 100 minutes.

Fig. 3 (b) shows that there was an increase in adsorption potential for Cu (II) and RY 18 by fuller's earth by increasing the concentration of metal and dye. By increasing the concentration of Cu (II) and RY -18 from 25 mg/L to 100 mg/L, the percentage removal for copper ions risen from 53% to 93% and for reactive yellow 18 dye from 39% to 65%. This is due to the growing concentration gradient, which functions as an improved accelerating power to fix the obstacles to the transfer of mass of metal ions between the aqueous stage and the adsorbent. A high solution concentration means more significant metal ion to be deposited on the surface of the adsorbent [11]. This rise in adsorption capability might be due to the greater rate of adsorption and usage of all accessible active spaces for adsorption at a high concentration of dye. In fact, by increasing the initial dye quantity, removal was also improved owing to the adsorbent constant quantity and availability of absorbed regions, and increased mass transfer results in the rising of adsorption capability [12].

4.6. Impact of adsorbent dose

Copper and Reactive Yellow 18 dye adsorption on fullers earth was investigated by varying the amount of fullers earth (0.1, 0.5, 1.5, and 2 g) in the sample mixture with a constant proportion of Cu(II) and RY-18 (100 mg/L).

Fig. 3 (c) indicates that by increasing the adsorbent dose from (0.1-2g), a decline in the capability of adsorption for copper and reactive yellow 18 dye after a particular amount of fuller's earth was achieved. The maximum adsorption of 93% for copper and 62% for RY18 was observed at 0.5 g of adsorbent, but after that, it decreases to 64% and 41% for Cu and RY 18 at 2g adsorbent dose. This enhancement is primarily associated with an increase in the amount of accessible

active nodes with an improvement in adsorbent dosage. Still, then it reduces, and this could be attributed to a decrease in the entire adsorption region available for metal ions triggered by merging or aggregating adsorption sites. Another consequence could be greater adsorbent dosage offering more significant active sites for adsorption resulting in unsaturated active sites left during adsorption [11,13].

4.6. Determination of pH Point of Zero Charge (pHpzc)

To comprehend the method of adsorption, the point of zero charges is determined. At pH greater than pHpzc, cations adsorption ability is supported while the ability to adsorb anions is supported at lesser pH than pHpzc. The influence of pH on the surface of adsorbent by protonation or deprotonation processes was measured by pHpzc [14]. Thus, based on the system's individual features (adsorbate and adsorbent), acid and basic pH interferes in the adsorption process.

Fig. 3 (d) indicates that the pH point of zero charges for fuller's earth was about 4. In other words, the fuller's earth surface has zero charges at this pH. The fuller's earth surface would be charged negatively owing to the presence of OH^- ions at above four pH and show an affinity for cationic species. But at pH less than four, the surface will be positively charged due to the presence of H^+ ions and show an affinity for anionic species.

4.7. Impact of pH

The impact of the initial pH on the fuller's earth adsorption capacity for copper and reactive yellow 18 was investigated by varying solution pH from 2-10 with 0.1g dose of adsorbent using 100 mg/L initial concentration of Cu (II) and RY 18.

Fig. 4 shows that the maximum percent removal of 85% was achieved at pH 6 for copper and 66% at pH 2 for reactive yellow 18 dye and then decreased.

Fig. 4 indicates that in the case of copper ions, at low pH, a high quantity of hydrogen ions (H^+) are formed in solution, creating a struggle between the metal ions and protons for the adsorbents active sites and show reduced adsorption due to the saturation of active sites of fuller's earth with H^+ ions. The enhanced absorption of metals at high pH could be owing to low struggle among metal particles and protons for the active sites and a decline in the positive surface charge resulting in low electrostatic repulsion among the metal ions and surface before the exchange of ion [10]. Hence copper shows high adsorption at pH 6 (85%). At a high pH of more than 6, owing to the presence of negative charges (OH^-) in the solution, Cu (II) ion gets precipitated as their hydroxide, which reduces the adsorption rate [15]. In the case of RY 18 dye, at less pH (acidic media), the active sites of adsorbents are charged positively due to the existence of (H^+) ions and due to the electrostatic attraction among negatively charged dye anions and positively charged adsorption sites, dye adsorption increased (Fig. 5). At acidic pH of 2, maximum removal of RY-18 with fuller's earth was achieved. While in basic media, the surfaces are likely negatively charged (Fig. 5). Due to the increased Hydroxyl ions (OH^-) on adsorbent causing repulsion among the anionic dye molecules and negatively charged surface. In addition, the exterior surface of the adsorbent does not contain exchangeable anions at greater pH values, and eventually, the adsorption of RY 18 reduces [16].
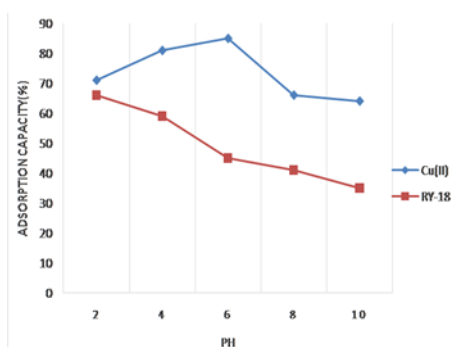

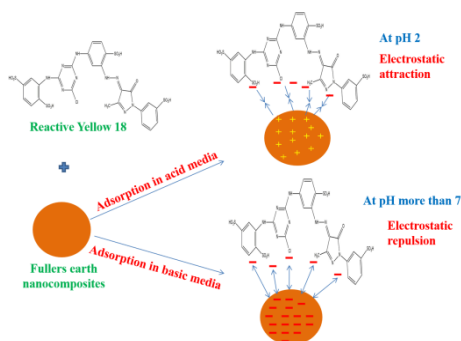*Fig. 4. Influence of pH for the removal of copper and reactive yellow 18 by fuller's earth*

Fig. 5. Proposed mechanism for the adsorption of reactive yellow 18 by the Fullers earth nanocomposites at acid and alkaline pH.

4.8. Impact of temperature

The influence of temperature on the adsorption potential of fullers earth for the removal of copper and reactive yellow 18 was investigated by varying the temperature from (25, 35, 45, 55, 65 °C) of the mixture at an adsorbent dosage of 0.5g with 100 mg/L initial concentration of copper and reactive yellow 18 with a contact time of 100 min.

Fig. 6 shows that the adsorption potential of fuller's earth for the removal of copper and reactive yellow 18 increased by increasing temperature. When the temperature increased from 25 – 65 °C, adsorption capacity increased from 63 to 96% for copper and from 34 to 68% for RY-18. This increase in adsorption capacity indicates the endothermic nature of this process. This rise in adsorption is primarily due to an increased proportion of adsorption channels generated by ripping a few of the inner bonds close to the edge of the adsorbent surface-active sites. This condition also enhanced the accessibility of active surface sites, enhanced porosity, and overall adsorbent membrane density [17]. Subbareddy *et al*., [18] analyzed that when temperature increased, the inner framework of fuller's earth produces a swelling impact that allows the molecules of dyes to further migrate into the pores.
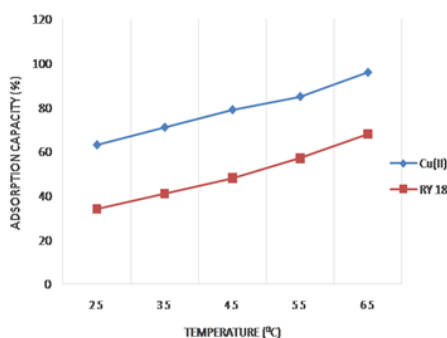

Fig. 6. Influence of temperature for the removal of copper and reactive yellow 18 by fuller's earth

5. Conclusion

The present research work was designed to investigate the adsorption capability of fuller's earth against contaminants like copper and reactive yellow 18 for water purification and to discover a proper solution to reduce the toxin levels in our surroundings. The results of the present research revealed that low cost and natural adsorbent (fuller's earth) could proficiently exclude fundamental colorants from the aqueous medium, and it can be used as an alternative to more costly materials.

Characterization of fuller's earth (adsorbent) was carried out by X-Ray Fluorescence (XRF), Scanning Electron Microscope (SEM), and Fourier Transform Infrared (FTIR). The result of XRF revealed the presence of large proportion of alkali oxides like TiO_2 (0.78%), Fe_2O_3

(3.13%), Al$_2$O$_3$ (12.38%), MgO (2.16%), CaO (10.73%), Na$_2$O (0.22%), P$_2$O$_5$ (0.11%), Cl (0.03%), K$_2$O (2.63%), MnO (0.03%), C (1.30%) and SiO$_2$ (66.31%) in the fullers earth. SEM images show the morphology, porous nature, and different micro size particles of the adsorbent. FTIR results show the presence of different functional groups.

The experimental results of the batch adsorption technique revealed that different parameters such as contact time, initial concentration of adsorbate, adsorbent dosage, pH, and temperature shows the optimum values and conditions for maximum adsorption phenomenon. Contact time of 100 minutes, 100 mg/L initial adsorbate concentration, 0.5 g adsorbent dosage at 65 ^{o}C. Temperature is the optimum values at which percentage removal is maximum, i.e., 96% for copper at pH 6 and 68% for reactive yellow 18 at pH two by fuller's earth was achieved. The solid addition method describes the pH point of zero charges at 4 for fuller's earth. Maximum adsorption at high temperature indicates that this adsorption process is endothermic.

Acknowledgements

References

[1] S. A. Kazi, H. H. Iqbal, N. Shahid, G. M. Shah, N. Jameel, Bulletin of Environmental Studies **1**(2), 43 (2016).

[2] R. S. Gowri, R. Vijayaraghavan, P. Meenambigai, International Journal Current Microbiology and Applied Sciences **3**, 421 (2014).

[3] C. R. Wang, X. Ren, W. X. Li, Z. F. Hou, C. Ke, Q. Geng, Polish Journal of Environmental Studies **22**(6), (2013).

[4] H. S. Silva, D. L. Granados, S. V. Ruiz, Latin American applied research **42**(3), 229 (2012).

[5] E. Sayan, O. N. Ata, M. E. Edecan, Journal of the Chemical Society of Pakistan **36**(2), 232(2014).

[6] H. Tahir, U. Hammed, M. Sultan, Q. Jahanzeb, African Journal of Biotechnology **9**(48), 8206 (2010).

[7] U. A. Isah, A. I. Gatawa, Clay Science **12**(2), 106 (2006).

[9] M. A. El-Latif, M. El-Kady, M. Ibrahim, M. Ossman, Journal of American Science **6**(5), 280 (2010).

[10] A. Emam, L. F. M. Ismalia, M. A. Abdel khalek, A. Rehana, International Journal of Advancement in Engineering Technology, Management and Applied Science **3**(7), 152 (2016).

[11] K. G. Akpomie, F. A. Dawodu, Journal of Taibah University for Science **8**(4), 343 (2014).

[12] H. Nadi, M. Alizadeh, M. Ahmadabadi, A. R. Yari, S. Hashemi, Archives of Hygiene Sciences **1**(2), 41 (2012).

[13] M. Kamranifar, A. Naghizadeh, Iranian Journal of Chemistry and Chemical Engineering **36**(6), 127 (2017).

[14] M. P. D. Silva, M. D. S. F. Santos, M. R. M. C. Santos, S. Júnior, L. de Sousa, M. G. D. Fonseca, E. C. D. Silva Filho, Materials Research **19**(6), 1232 (2016).

[15] L. M. Ahmed, F. T. Tawfeeq, M. H. A. Al-Ameer, K. A. Al-Hussein, A. R. Athaab, Journal of Geoscience and Environment Protection **4**(11), 34 (2016).

[16] M. Mirjalili, M. B. Tabatabai, L. Karimi, African Journal of Biotechnology **10**(65), 14478 (2011).

[17] E. Kalkan, H. Nadaroğlu, N. Celebi, G. Tozsin, Desalination and Water Treatment **52**(31-33), 6122 (2014).

[18] Y. Subbareddy, C. Jayakumar, S. Valliammai, K. S. Nagaraja, B. Jeyaraj, International Journal of Research in Chemistry and Environment **4**(3), 16(2014).