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Pb(II) Removal from Synthetic Wastewater Using *Kombucha* Scoby and Graphene Oxide/Fe_3O_4

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Kombucha Scoby is a colony consisting of bacteria, yeast and cellulosic pellicle that present fantastic performances in various fields. In addition to the anti-toxicity and antimicrobial specifications of Kombucha scoby, this unique colony can be used for waste water treatment and removal of heavy metals. Herein, efficiency of graphene oxide/Fe_3O_4 nanoparticles (GO/Fe_3O_4) and *Kombucha* Scoby in the removal of Pb(II) from synthetic wastewater were examined and compared. The characteristics of GO/Fe_3O_4 nanoparticles were analyzed using FTIR and SEM. Moreover, the effect of significant parameters such as pH (1-7), temperature (10-60 °C) and amount of adsorbent (1-200 g l^{-1}) on the removal of Pb(II) ion from aqueous solution was investigated. The maximum adsorption efficiency was obtained at a temperature of 50 °C and adsorbent amount of 0.3 and 15 g using GO/Fe_3O_4 and Kombucha Scoby, respectively. Additionally, pH_{PZC} values of 6.1 and 6.2 were obtained for GO/Fe_3O_4 and Kombucha Scoby, respectively. The maximum adsorption efficiencies for GO/Fe_3O_4 and Kombucha Scoby were found to be 98.08 and 99.73%, respectively. Likewise, the adsorption isotherm behavior of Pb(II) on adsorbents were investigated using Langmuir and Freundlich models. Achieved results showed that the Langmuir isotherm model is better fitted with experimental data. Furthermore, the maximum adsorption capacities by Langmuir model for GO/Fe_3O_4 nanoparticles and Kombucha Scoby were found to be about 114.9 and 126.6 mg/g, respectively. Generally, achieved results revealed that Kombucha Scoby, which is a cost affordable colony, can remove Pb(II) ions from water better than GO/Fe_3O_4.

Keywords: Graphene oxide, Magnetic nanoparticles, Pb(II), Kombucha Scoby, Adsorption

INTRODUCTION

Heavy metal ions in groundwater can be considered as a serious threat to public health and environmental systems [1]. These ions are not degradable and a vast majority of them have toxic and carcinogenic properties [2,3]. They can be stored within living tissues, and enter into food chains. Based on the United States Environmental Protection Agency (US EPA) and International Agency for Research on Cancer (IARC), metal ions such as lead (Pb (II)) are considered as human carcinogenic agents. It should also be noted that Pb(II) is a highly stable compound that cannot be easily degraded. In most cases, the toxico-kinetic behavior of Pb(II) ion in humans depends on its binding to the albumin protein [4]. To reduce or remove heavy metals from wastewater, various physical, chemical and biological procedures, such as coagulation, chemical oxidation,

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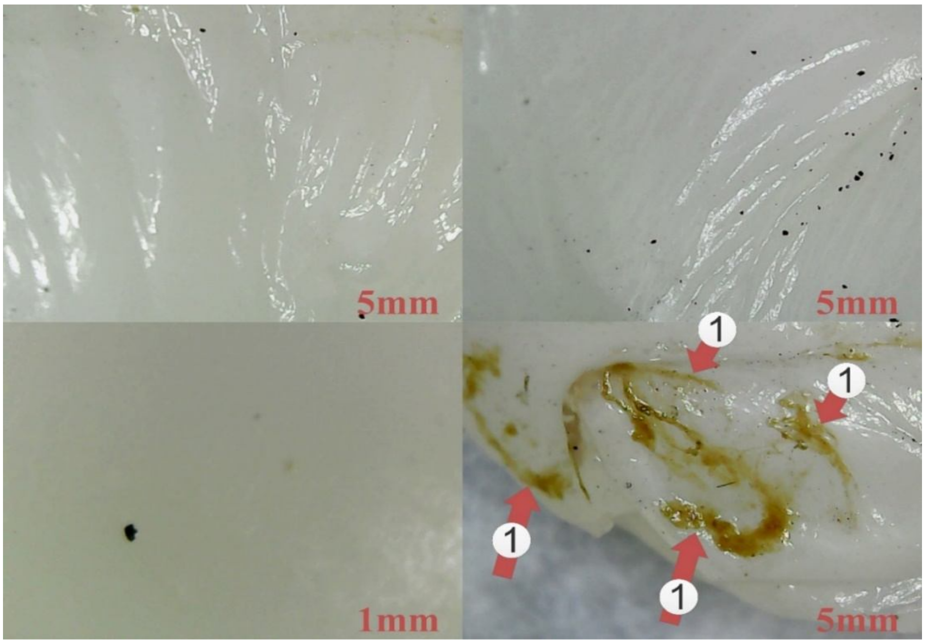

Fig. 1. Optical microscopic images of Kombucha Scoby (Number 1 shows the dead kombucha bacteria).

membrane filtration, electro dialysis, reverse osmosis, chemical precipitation and adsorption have been employed, however, often these methods are not very successful due to their high operating costs and inefficiencies. Among the above-mentioned methods, adsorption process has recently attracted lots of attention. This is a simple and efficient method for removal of heavy metals from water and wastewater with positive features such as high purification effect and no harmful by-products as well as significant properties such as availability and ease of use [5-7]. Therefore, the development of adsorbents with high adsorption capacity, low toxicity and efficient separation has attracted great attention [7,8]. Additionally, in order to remove heavy metals and organic pollutants from water, many attempts have been made to produce useful adsorbents with various chemical compounds and superficial properties [8]. The use of magnetic adsorbents (such as Fe_3O_4 nanoparticles) can be considered as a newly efficient technology in which magnetic nanoparticles are easily separated from sewage using an external magnetic field after adsorption [9]. To improve the magnetic properties of iron oxide, it can be combined with different materials such as clay, CaO, graphene oxide (GO), *etc.* [9,10]. In addition, some microorganisms such as algae, fungi and bacteria can be considered as an effective adsorbents to attenuate heavy metals [5,6].

The main aim of the present study is to deeply understand the removal percentage of Pb(II) ions from aqueous solution by means of two effective adsorbents such as modified graphene oxide with magnetite nanoparticles (GO/Fe_3O_4) and *Kombucha* Scoby (KS), and also comparing their performances with each other. To this end, the effect of different parameters such as pH, temperature and adsorbent concentration on the removal of Pb(II) ions from synthetic wastewater is investigated. In fact, Kombucha is a fermentation of sweetened tea that provides symbiosis of acetic acid bacteria and yeast species. Kombucha Scoby (floating solid part in the liquid media), so called "tea fungus" (*i.e.* symbiotic colony of bacteria and yeast [11]), consists of symbiosis of acetic acid bacteria, various kinds of yeasts, and cellulosic pellicle [12,13]. This unique colony is cost affordable, and can be developed through least funds to develop a highly effective waste water purification system. Besides, the adsorption isotherm models such as Langmuir and Freundlich were investigated to describe the equilibrium behavior of adsorption.

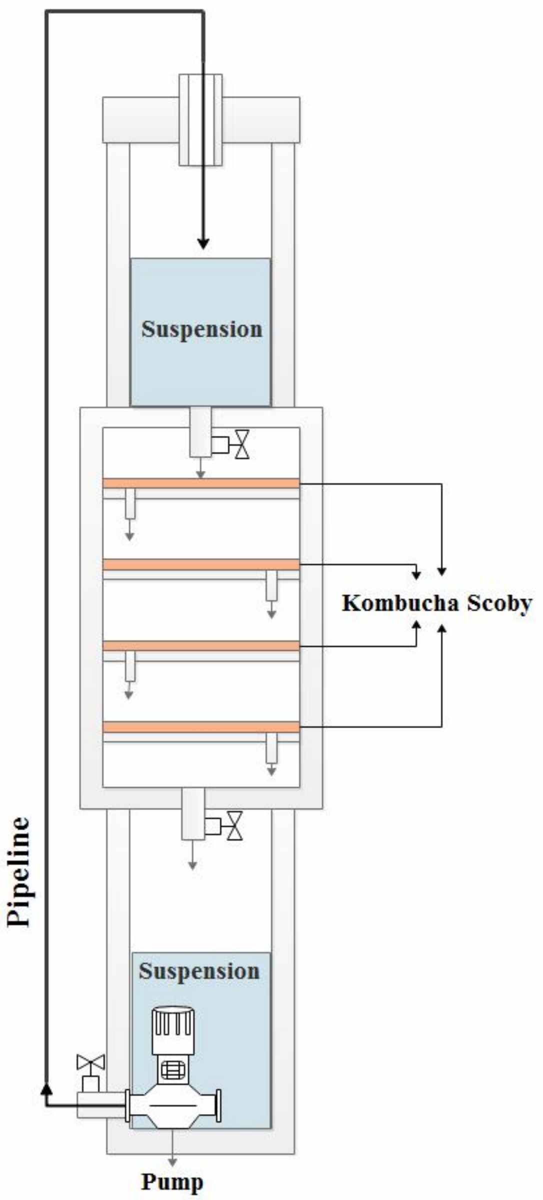

Fig. 2. Schematic of experimental setup for reactor operation.

EXPERIMENTAL

Materials and Methods

Kombucha Scoby was procurement from the Caucasus Mountains. The spectrophotometric measurements were performed with a Cintra 101 spectrophotometer (GBC, Australia). Scanning electron microscope (SEM, TESCAN) and digital pH meter (Metrohm 744) were used in these experiments. SEM was used to determine the morphology and structure of the adsorbent, and the pH meter was used to measure the pH value of solutions. Materials such as Ferric chloride ($FeCl_3.6H_2O$), $Pb(NO_3)_2$, $FeSO_4.7H_2O$ and graphene were purchased from Merck Co. (Germany). Furthermore, an optical microscope was used to observe Kombucha Scoby. Optical microscopic images of Kombucha Scoby is shown in Fig. 1.

Reactor setup and Operation

To perform experiments, anaerobic reactor made of polypropylene (PP) (4 cm in diameter, 63 cm in height and with an operating level of 58 cm) was used. The separation of Pb(II) from synthetic wastewater in the presence of microorganisms was done by a membrane module. A scheme of this bioreactor is shown in Fig. 2.

Preparation of Pb(II) Solution

A preliminary stock solution of lead was prepared at a concentration of 100 mg l^{-1} by dissolving 0.16 g of $Pb(NO_3)_2$ in 1000 ml deionized distilled water. Other solutions were prepared by diluting the base solution with double-distilled water. The pH of the working solution was adjusted by 0.1 M NaOH and 0.1 M HCl.

Preparation of Graphene Oxide (GO)

In this study, graphene oxide (GO) was prepared using a multi-step method. In this case, 10 g graphite was initially poured into a flat-bottom flask, and then, 2 l H_2SO_4 was added to the flask and stirred at 500 rpm for 30 min. Then, 60 g $KMnO_4$ was added to the suspension and stirred at 500 rpm for 1 h while the temperature was maintained at ~5 °C. Thereafter, 110 ml H_3PO_4 was added to the suspension, and then the temperature was smoothly increased to about 50 °C and stirred for 72 h (500 rpm). Then, the solution was poured into a vacuum Erlenmeyer flask and some ice (made by deionized water) was poured into the flask. Afterwards, 10 ml H_2O_2 was poured in the solution very slowly and the vacuum Erlenmeyer flask was filled with deionized water. After that, the solution was kept without stirring for 48 h for further filler sediment. The solution was then filtered, and remaining fillers on the filter were washed with deionized water, and the pH of the solution was then adjusted to 7. Eventually, materials were dried for 1 h at 80 °C in an oven and then placed in humidity reduction chamber for 48 h. A view of GO preparation steps is shown in Fig. 3. More details about the production procedure of GO have been documented in our previous works [14,15].

Preparation of the Modified Graphene Oxide with Fe_3O_4 (GO/Fe_3O_4)

GO nanoparticles were decorated with Fe_3O_4 nanoparticles through a multi-step method. In this case, 320 ml deionized water was poured into a round-bottom flask and its temperature was set to 80 °C. Then, 3.89 g $FeSO_4.7H_2O$ and 4.55 g $FeCl_3.6H_2O$ were poured into the flask and stirred for 1 h. In next step, 0.0844 g GO was ultrasonically mixed in 100 ml of deionized water for 20 min and then poured into the previous suspension. The resulting suspension was stirred for 2 h and then 40 ml NH_3 was slowly added to the suspension. The suspension was then stirred for further 2 h with simultaneous heat (80 °C). In addition, the suspension was filtered and simultaneously washed with deionized water in order to set the pH on 7 and afterward dried in an oven for 1 h at 100 °C. The resulting material was placed in a moisture absorbing chamber for 48 h. A schematic of this multi-step method is shown in Fig. 4.

Batch Adsorption Experiment

Pb(II) adsorption using prepared adsorbents was done within a 250 ml Erlenmeyer flask containing 100 ml lead solution with a certain initial concentration. In this case, the effect of significant parameters like pH, adsorbent dose and temperature on the adsorption efficiency was investigated. In order to investigate the effect of pH, the adsorption process was done at pH ranging between 1-7. After determining the optimal pH, the optimum amount of other parameters was determined. To investigate the effect of temperature on the adsorption efficiency, temperature evaluation in the range of 10-60 °C was performed. The effect of adsorbent dose on the adsorption efficiency of lead was studied over the range of 1-200 g l^{-1} of adsorbents in 100 ml solution. After performing the related assays, to separate the solid phase (sorbent) from the solution, about 10 ml of solution was centrifuged. The concentration of heavy metal after adsorption was determined by spectrophotometer. Adsorption efficiency (%) is determined by the following equation [16-18]:

$$\mathrm{Re}moval\,(\%) = \frac{(C_o - C_e)}{C_o} \times 100 \tag{1}$$

where, C_o and C_e are the initial concentration (mg l^{-1}) and the equilibrium concentration of Pb(II) ions in solution (mg l^{-1}), respectively.

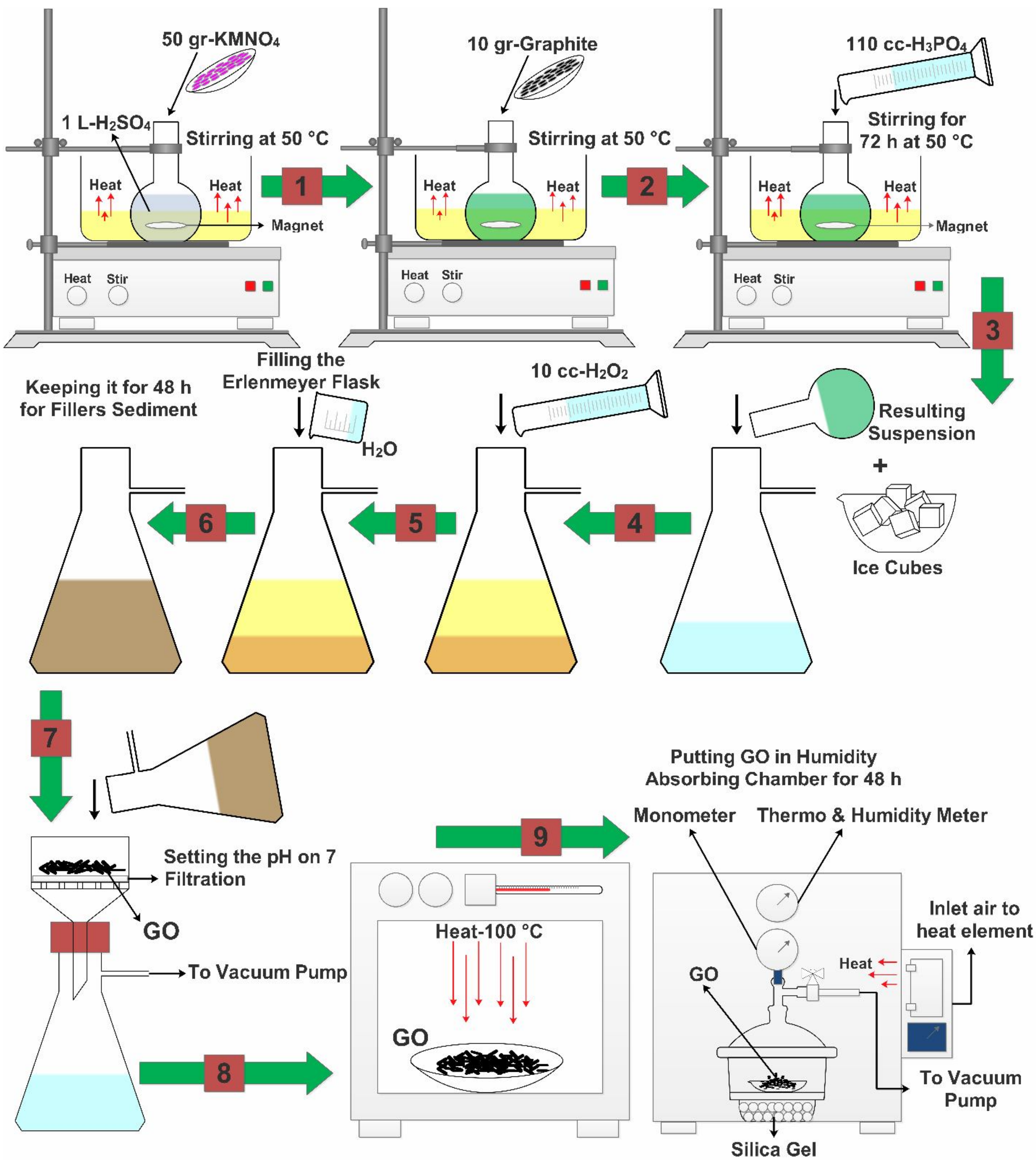

Fig. 3. Schematic of GO preparation steps.

RESULTS AND DISCUSSIONS

Characterization of Adsorbent

The FTIR spectra of Fe_3O_4 and GO-Fe_3O_4 nanoparticles are presented in Fig. 5. As can be seen in Fig. 5, peaks in the region between 530-630 cm^{-1}, related to the Fe_3O_4 spectrum, correspond to the stretching vibration mode of Fe-O. In this regard, a broad and strong peak at 575 cm^{-1} corresponds to Fe_3O_4 indicating the successful synthesis of Fe_3O_4 nanoparticles [19-21]. Furthermore, peaks at 923 and 1113 cm^{-1} are related to the sp^2 alkene C-H band (disubstituted-E) and stretching vibration of in-plane C-H,

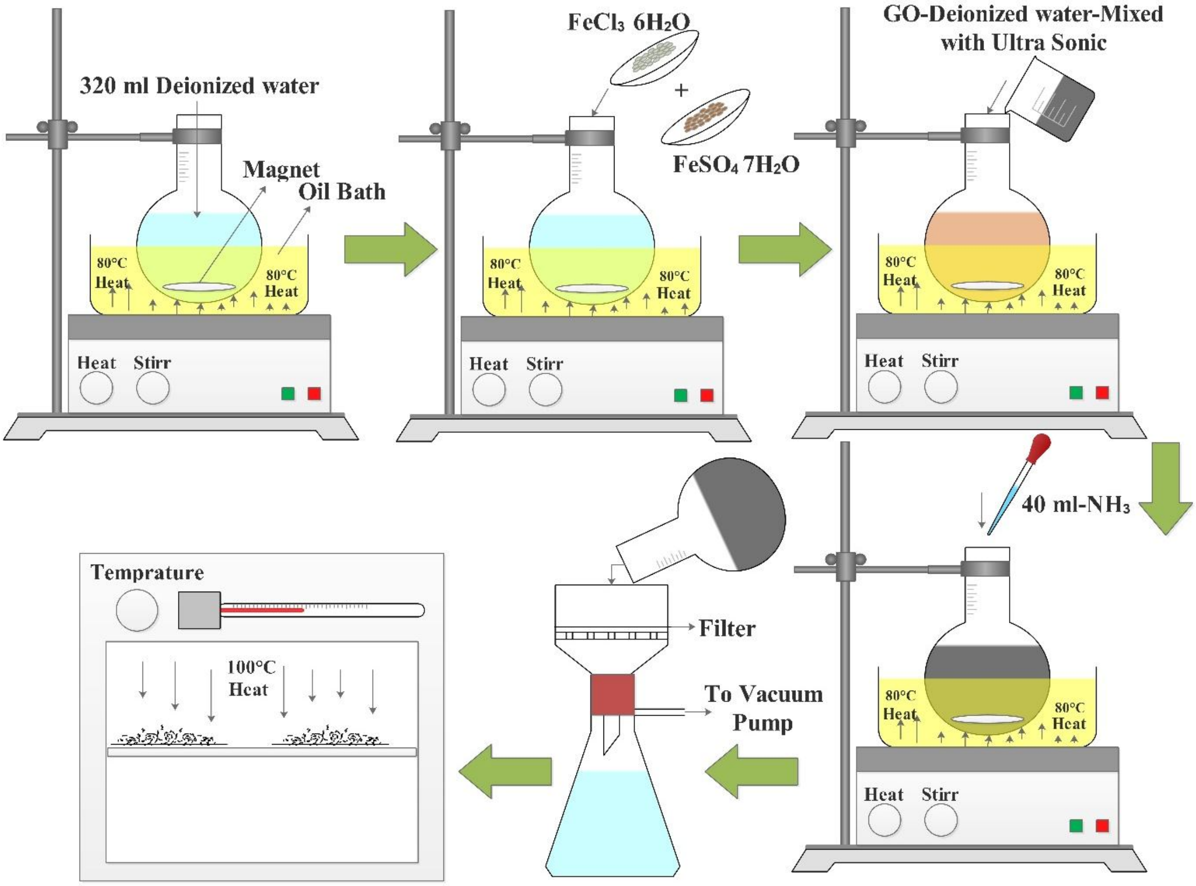

Fig. 4. Schematic of GO-Fe_3O_4 preparation.

respectively. Besides, peak with wavenumber 1627 cm^{-1} belongs to the FeOO- [20], while peaks in region between ~2838-2921 cm^{-1} are assigned to the C-H stretching vibration (sp^3 stretching of hexyl aliphatic side). Weak peaks in region between 3500-3700 cm^{-1} are attributed to the amine N-H functional groups. Furthermore, a broad and strong peak at 3424 cm^{-1} corresponds to the hydroxyl functional group (-OH) [20]. Evaluation of GO-Fe_3O_4 spectrum confirms the successful decoration of GO with Fe_3O_4 nanoparticles. In this matter, appearance of a strong peak at 584 cm^{-1}, known as the finger print of Fe_3O_4 nanoparticles [19-21], along with the peak at 1628 cm^{-1}, corresponding to the FeOO- [20], justifies the successful decoration of GO with Fe_3O_4 nanoparticles. Moreover, peaks at 945 and 3416 cm^{-1} correspond to the sp^2 alkene C-H bond and hydroxyl functional group, respectively. Appearance of Fe_3O_4 finger prints (577 and 1627 cm^{-1}) along with disappearance of the peaks corresponding to the carboxyl and epoxide functional groups, and decrease in the intensity of hydroxyl functional group in the GO-Fe_3O_4 spectrum compared to that in Fe_3O_4 spectrum, justify the successful decoration of GO with Fe_3O_4 nanoparticles. In fact, decrease in the intensity of hydroxyl functional group and disappearance of carboxyl and epoxide functional groups clarify the interaction between functional groups of GO and Fe_3O_4.

The SEM image of GO-Fe_3O_4 nanoparticles is also shown in Fig. 6, providing information about size and morphology of the developed nanoparticles. It can be observed that the adsorbent has heterogeneous surfaces with abundant pores and active sites for adsorption of Pb(II) ions.

Point of Zero Charge (PZC)

PZC could directly lead to a qualitative assessment of

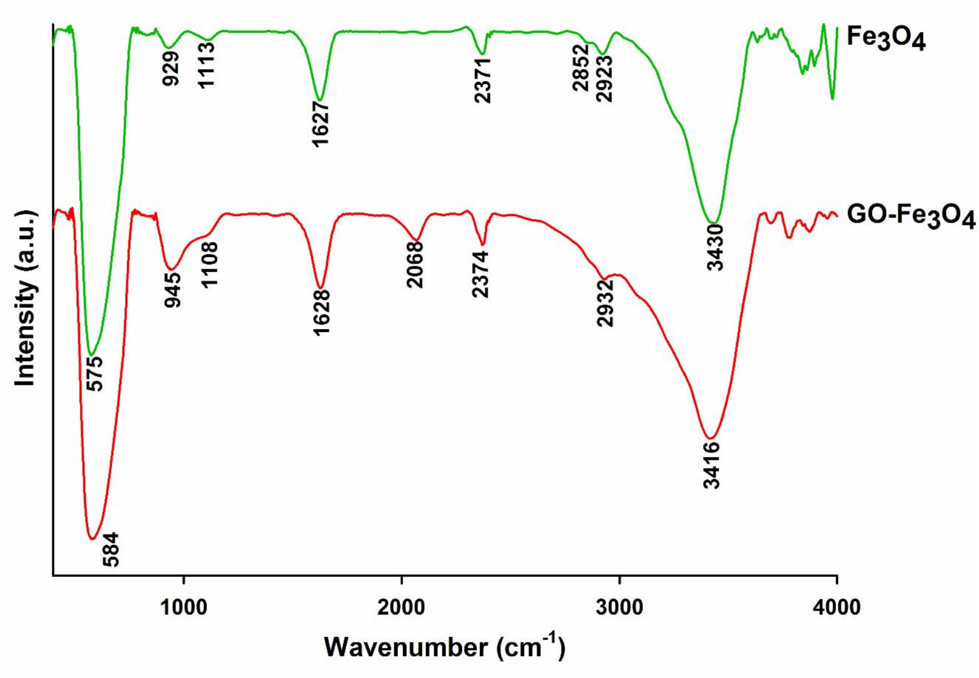

Fig. 5. FTIR spectra of Fe_3O_4 and GO-Fe_3O_4 nanoparticles.

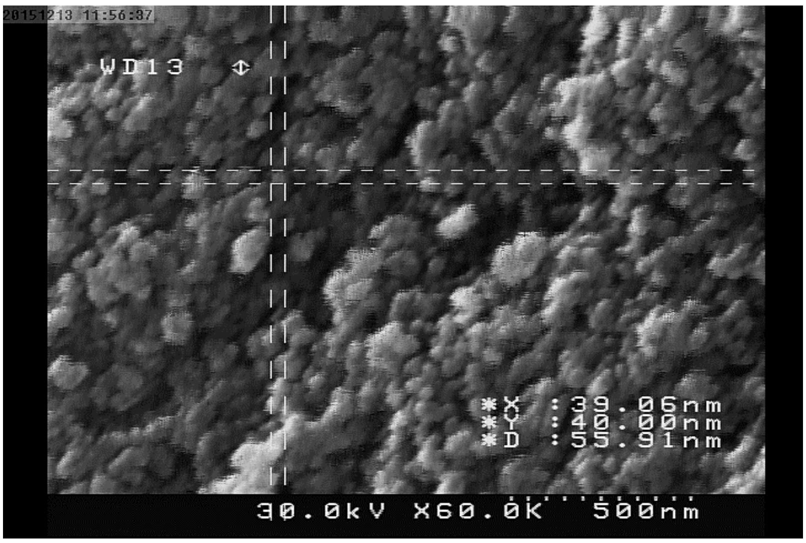

Fig. 6. SEM image of GO-Fe_3O_4.

the type of interactions (attraction or interaction) between *Kombucha* Scoby as a colony and GO-Fe_3O_4 if we know the type of charges on their surfaces. Unfortunately, it is experimentally difficult to directly measure the charges on the surface. Instead, indirect methods are available such as estimation of PZC from electrochemical impedance spectroscopy measurement. PZC is an expedient marker of surface charges, that is critically relevant to adsorption of an electrolyte. It denotes the potential when the surface carries zero charges. Normally, when electrode potential is larger

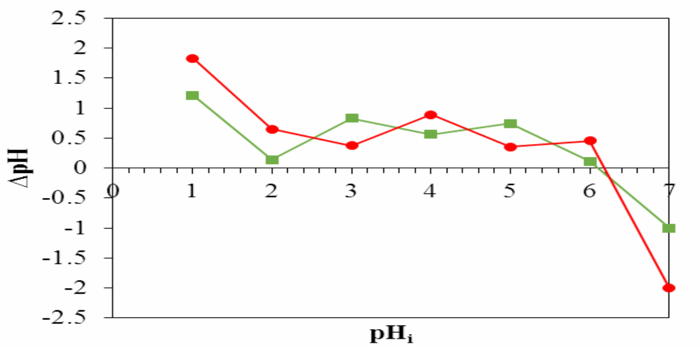

Fig. 7. pH_{ZPC} of GO-Fe_3O_4 (■) and *Kombucha* Scoby (●), and effect of initial pH on the sorption of Pb(II).

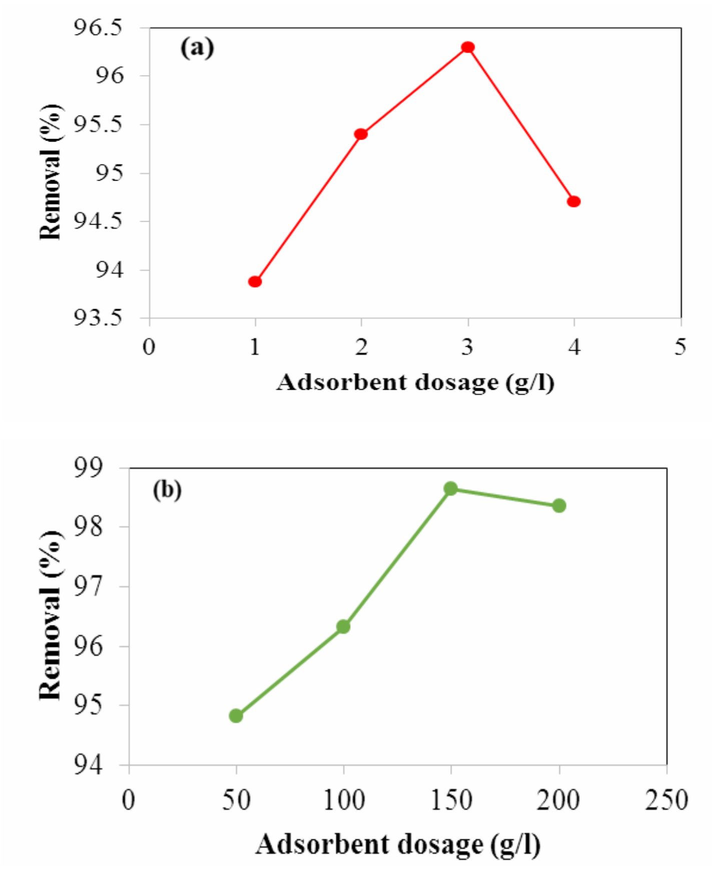

Fig. 8. The effect of the adsorbent dose on the Pb(II) sorption onto (a) GO-Fe_3O_4 and (b) *Kombucha* Scoby.

than PZC, the electrode carries positive charges and when it is smaller than PZC, the electrode carries negative charges. The pH value of the solution, affects the surface charge of the adsorbent and the uptake behavior and efficiency of adsorbent [22,23]. The difference between the initial and final pH values (pH = pH_f - pH_i) was plotted against the pH_i in Fig. 7. The pH_{PZC} was noted as the pH in which the initial pH and the final pH are equal [24]. The degree of Pb(II)

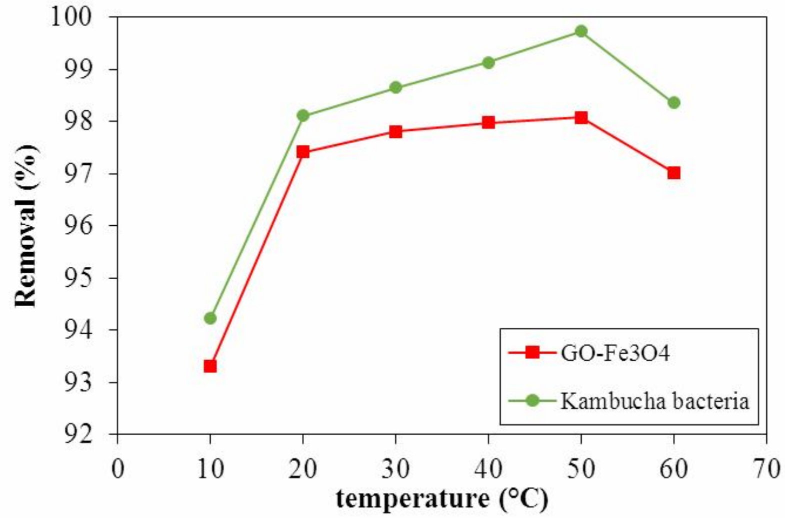

Fig. 9. The effect of the temperature on the sorption of Pb(II) onto GO-Fe_3O_4 and *Kombucha* Scoby.

adsorption onto the adsorbent surface is influenced by the pH zero-point charge (pHpzc) of the adsorbents. As can be seen from the results, the pH_{PZC} of GO-Fe_3O_4 and *Kombucha* Scoby were 6.1 and 6.2, respectively. When the pH value was less than 6.1, the surface of the GO-Fe_3O_4 was positively charged and repelled the cationic Pb(II) molecules at the surface of the adsorbent. This caused competition between protons and cations that resulted in the low adsorption. When the pH was greater than 6.1, negative charges were dominant on the surface of the GO-Fe_3O_4 that facilitated the adsorption of the Pb(II) cations through electrostatic attraction. Based on the results, decrease in the pH led to the increase of removal efficiency ; might be due to the increase in the H^+ ions of the solution ($pH < pH_{PZC}$) and in positive ions on the adsorbent surface [25].

Effect of Adsorbent Dosage on the Removal Rate of Pb(II) Ions

The effect of adsorbent dosage on the removal of Pb(II) ions is studied by changing the adsorbent dosage without any changes in other parameters. Figure 8 shows that removal efficiency was increased by increase in the adsorbent dose. This phenomenon is due to an increase in the available binding sites on the surface of the adsorbent for adsorption of Pb(II) ions [26]. The removal efficiency of Pb(II) ions increased dramatically from 93.88% to 96.3% and from 94.82% to 98.65% when the GO-Fe_3O_4 dosage increased from 1 to 3 g l^{-1} and Kombucha Scoby changed from 50 to 150 g l^{-1}, respectively. On the other hand, the efficiency decreased with further increase in the adsorbents dosage. According to the results, the optimum adsorbent dose for GO-Fe_3O_4 and Kombucha are 3 and 150 g l^{-1}, respectively. Based on the results, the percentages of metal ion removed by the Kombucha were higher than those removed by GO-Fe_3O_4, indicating that Kombucha is a better adsorbent because it is cheaper than GO-Fe_3O_4 and has higher efficiency.

Effect of Temperature

The effect of temperature on the adsorption capacity of GO-Fe_3O_4 and *Kombucha* Scoby for the removal of Pb(II) ions from an aqueous solution was studied at pH 6.0 and the results are shown in Fig. 9. As can be seen in this figure, increase in the temperature from 10 to 50 °C leads to increase in the removal efficiency of GO-Fe_3O_4 and *Kombucha* about 98.08% and 99.73%, respectively. This is due to the fact that higher temperature increases the interaction between Pb(II) and adsorbent. Likewise, it indicates the endothermic nature of the adsorption process [27]. In general, the adsorption process involves two successive processes, namely fast diffusion and slow complexation [28]. When the temperature is more than 50 °C, the adsorption capacity decreases, due to the instability of Pb(II) adsorption complex in high temperature.

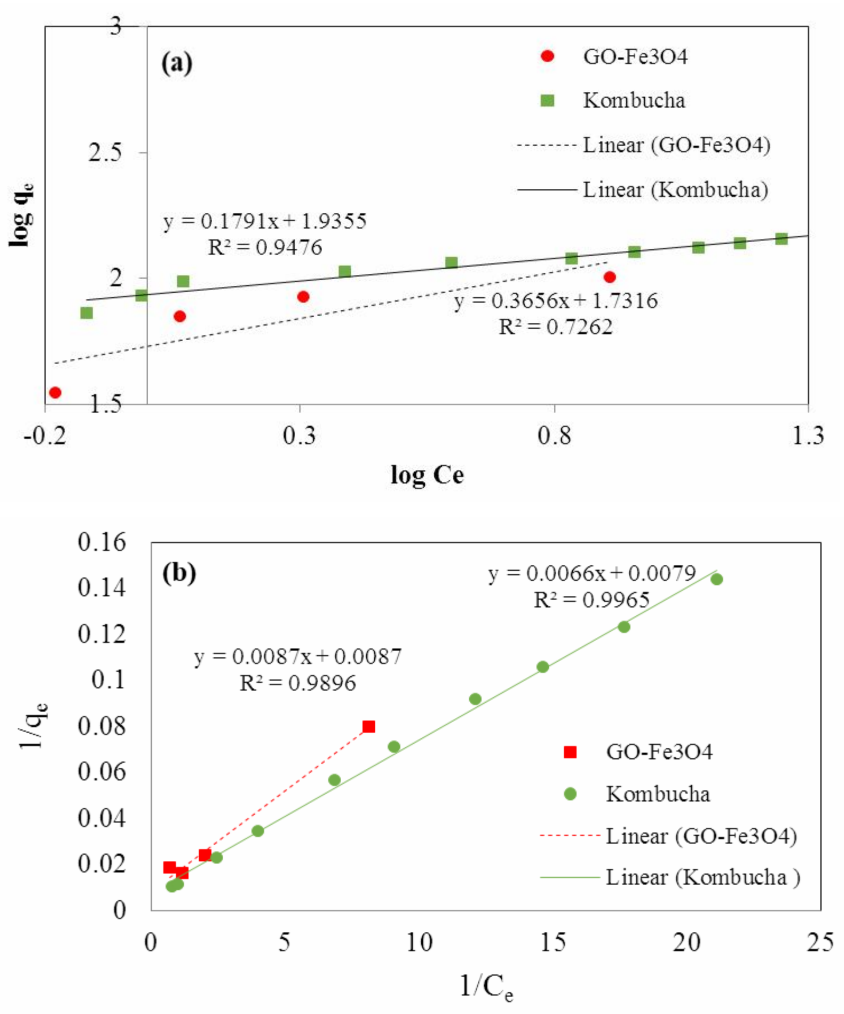

Fig. 10. Pb(II) adsorption using GO-Fe_3O_4 and *Kombucha* Scoby by Freundlich (a) and Langmuir (b) models.

Therefore, 50 °C was chosen as the optimum temperature for adsorption, and *Kombucha* Scoby proved to be a better adsorbent than GO-Fe_3O_4 due to cheaper and better performance.

Adsorption Isotherms

Adsorption isotherms display the interaction pathway of adsorbate with adsorbents [29]. Adsorption isotherms data were modeled using Langmuir and Freundlich models. Freundlich model assumes that the adsorbate uptake occurs on a heterogeneous surface of the adsorbent. Besides, the Langmuir model describes the monolayer sorption process onto the adsorbent surface [30]. The linearized form of the Freundlich model is given as follows [31]:

$$Lnq_e = LnK_F + \frac{1}{n}LnC_e \tag{2}$$

where, K_F and n are the model constants showing the relationship between adsorption capacity and adsorption intensity, respectively [24]. The plot of $\ln q_e$ *vs*. $\ln C_e$ was employed to determine K_F and n from the intercept and the slope, respectively, according to Fig. 10. The value of 1/n represents the isotherm type, that is irreversible (1/n = 0), favorable (0 < 1/n < 1) and unfavorable (1/n > 1) [23]. The linear form of the Langmuir model [32] can be seen in equation 3 and the results are plotted in Fig. 10,

$$\frac{1}{q_e} = \frac{1}{K_L q_m C_e} + \frac{1}{q_m} \tag{3}$$

Table 1. Constants and Parameters of Different Isotherm Models Using Kombucha Bacteria and GO-Fe_3O_4

Model	Parameter	Kombucha	GO-Fe_3O_4
	K_F	86.2	53.9
Freundlich	n	5.58	2.74
	R^2	0.9476	0.7262
	q_m	126.6	114.9
Longmuir	k_L	1.197	1
	R^2	0.9965	0.9896

where, q_e, C_e, K_L and q_m are the amount of adsorbed ion in equilibrium state per gram of adsorbent (mg g^{-1}), the equilibrium concentration of metal ions in solution (mg l^{-1}), the equilibrium constant related to the affinity of binding sites (l mg^{-1}) and the maximum amount of the ions per mass of adsorbent (mg g^{-1}), respectively [30]. The plot of $1/q_e$ against $1/C_e$ was applied to determine q_m and K_L from the intercept and the slope, respectively. Constants and parameters of Longmuir and Freundlich isotherm models using Kombucha Scoby and GO-Fe_3O_4 are tabulated in Table 1.

According to Fig. 10 and Table 1, the R^2 values for Kombucha Scoby were 0.996 and 0.947 and for GO-Fe_3O_4 adsorbent were 0.989 and 0.726 using the Langmuir and Freundlich isotherm models, respectively. The Langmuir isotherm model shows a higher correlation coefficient compared to the Freundlich isotherm model, suggesting that the Langmuir model has a close fitting to the experimental data. The maximum adsorption capacity (q_m) were found to be 126.6 and 114.9 mg g^{-1} for *Kombucha* and GO-Fe_3O_4, respectively, indicating more sorption of Pb(II) ions onto *Kombucha* Scoby.

CONCLUSIONS

In this study, *Kombucha* Scoby and GO/Fe_3O_4 were applied for Pb(II) ion removal from aqueous solution. In this case, the effect of pH, dosage of adsorbents and temperature were investigated. The sorption of Pb(II) ions and the capacity of adsorption raised by increase in the pH, amount of adsorbents, and temperature. The sorption rate of Pb(II) onto the adsorbents was rapid and Pb(II) was eliminated by 50% within the first 15 min. The maximum removal efficiencies were obtained to be 98.08% and 99.73% for GO-Fe_3O_4 and *Kombucha* Scoby, respectively. The equilibrium investigation of adsorption process was done and the results showed the experimental data were well fitted with the Langmuir isotherm model due to the higher correlation coefficient. The maximum adsorption capacity was higher for *Kombucha* Scoby (126.6 mg g^{-1}) than that for GO-Fe_3O_4 (114.9 mg g^{-1}), therefore, the *Kombucha* bacteria has an excellent potential in the removal of Pb(II) from wastewater, even better than magnetic GO.

Conflict of Interests Statement: The authors declare that there is no conflict of interests.

REFERENCES

[1] Abdelhafez, A. A.; Li, J., Geochemical and statistical evaluation of heavy metal status in the region around Jinxi River. *China. Soil Sediment Contam.* **2014**, *23*, 850-868. DOI: 10.1080/15320383.2014.887651.

[2] Nagajyoti, P. C.; Lee, K. D.; Sreekanth, T. V. M., Heavy metals, occurrence and toxicity for plants: a review. *Environ. Chem. Lett.* **2010**, *8*, 199-216. https://doi.org/10.1007/s10311-010-0297-8.

[3] Naseem, R.; Tahir, S. S., Removal of Pb(II) from aqueous/acidic solutions by using bentonite as an adsorbent. *Water Res.* **2001**, *35*, 3982-3986. https://doi.org/10.1016/S0043-1354(01)00130-0.

[4] Ayranci, E.; Duman, O., Binding of lead ion to bovine serum albumin studied by ion selective electrode. *Protein Pept. Lett.* **2004**, *11*, 331-337. DOI: 10.2174/0929866043406814.

[5] Mullen, M. D.; Wolf, D.; Beveridge, T. J.; Bailey, G. W., Sorption of heavy metals by soil fungi Aspergillus niger and Mucor rouxii. *Soil. Biol. Biochem.* **1992**, *24*, 129-135. https://doi.org/10.1016/0038-0717(92)90268-3.

[6] Foroutan, R.; Esmaeili, H.; Abbasi, M.; Rezakazemi, M.; Mesbah, M., Adsorption behavior of Cu(II) and Co(II) using chemically modified marine algae. *Environ. Technol.* **2017**. DOI: 10.1080/09593330.2017.1365946.

[7] Sarvestani, F. S.; Esmaeili, H.; Ramavandi, B., Modification of Sargassum angustifolium by molybdate during a facile cultivation for high-rate phosphate removal from wastewater: structural characterization and adsorptive behavior. *3 Biotech.* **2016**, *6*, 251. https://doi.org/10.1007/s13205-016-0570-z.

[8] Wang, Y.; Liang, S.; Chen, B.; Guo, F.; Yu, S.; Tang, Y., Synergistic removal of Pb(II), Cd(II) and humic acid by Fe_3O_4@mesoporous silica-graphene oxide composites. *PLoS One.* **2013**, *8*, e65634. https://doi.org/10.1371/journal.pone.0065634.

[9] Khoo, F.S.; Esmaeili, H., Synthesis of CaO/Fe_3O_4 magnetic composite for the removal of Pb(II) and Co(II) from synthetic wastewater. *J. Serb. Chem. Soc.* **2018**, *83*, 237-249. doi.org/10.2298/JSC170704098S.

[10] Peng, W.; Li, H.; Liu, Y.; Song, S., A review on heavy metal ions adsorption from water by graphene oxide and its composites. *J. Mol. Liq.* **2017**, *230*, 496-504. https://doi.org/10.1016/j.molliq.2017.01.064.

[11] Sreeramulu, G.; Zhu, Y.; Knol, W., Kombucha fermentation and its antimicrobial activity. *J. Agric. Food Chem.* **2000**, *48*, 2589-2594. DOI: 10.1021/jf991333m.

[12] Kappel, T.; Anken, R. H., The tea-mushroom. *Mycologist.* **1993**, *7*, 12-13. https://doi.org/10.1016/S0269-915X(09)80616-2.

[13] Steinkraus, K. H., Classification of fermented foods: worldwide review of household fermentation techniques. *Food Control.* **1997**, *8*, 311-317. https://doi.org/10.1016/S0956-7135(97)00050-9.

[14] Hashemi, S. A.; Mousavi, S. M.; Faghihi, R.; Arjmand, M.; Sina, S.; Amani, A. M., Lead oxide-decorated graphene oxide/epoxy composite towards X-Ray radiation shielding. *Radiat. Phys. Chem.* **2018**, *146*, 77-85. https://doi.org/10.1016/j.radphyschem.2018.01.008

[15] Mousavi, S. M.; Hashemi, S. A.; Arjmand, M.; Amani, A. M.; Sharif, F.; Jahandideh, S., Octadecyl amine functionalized graphene oxide towards hydrophobic chemical resistant epoxy nanocomposites. *ChemistrySelect.* **2018**, *3*, 7200-7207. https://doi.org/10.1002/slct.201800996

[16] Foroutan, R.; Esmaeili, H.; Derakhshandeh Rishehri, S. M.; Sadeghzadeh, F.; Mirahmadi, S. R.; Kosarifard, M.; Ramavandi, B., Zinc, nickel, and cobalt ions removal from aqueous solution and plating plant wastewater by modified Aspergillus flavus biomass: *A data set. Data Brief.* **2017**, *12*, 485-492. DOI: 10.1016/j.dib.2017.04.031.

[17] Teimouri, A.; Esmaeili, H.; Foroutan, R.; Ramavandi, B., Adsorptive performance of calcined Cardita bicolor for attenuating Hg(II) and As(III) from synthetic and real wastewaters. *Korean J. Chem. Eng.* **2018**, *35*, 479-88. https://doi.org/10.1007/s11814-017-0311-y.

[18] Mahini, R.; Esmaeili, H.; Foroutan, R., Adsorption of methyl violet from aqueous solution using brown algae Padina sanctae-crucis. *Turk. J. Biochem.* **2018**. DOI: https://doi.org/10.1515/tjb-2017-0333.

[19] Entezari Juybari, F.; Kamran-Pirzaman, A.; Ghorbani, M., Chemical modification of magnetite with polythiophene and characterization of formed core-shell nanocomposite. *Inorg. Nano-Met. Chem.* **2017**, *47*, 121-126. https://doi.org/10.1080/15533174.2016.1149733

[20] Wei, Y.; Han, B.; Hu, X.; Lin, Y.; Wang, X.; Deng, X., Synthesis of Fe_3O_4 nanoparticles and their

magnetic properties. *Procedia Eng.* **2012**, *27*, 632-637. https://doi.org/10.1016/j.proeng.2011.12.498.

[21] Baharin, S. N. A.; Muhamad Sarih, N.; Mohamad, S., Novel functionalized polythiophene-coated Fe_3O_4 nanoparticles for magnetic solid-phase extraction of phthalates. *Polymers.* **2016**, *8*, 117. https://doi.org/10.3390/polym8050117.

[22] Ghaneian, M. T.; Momtaz, M.; Dehvari, M., An investigation of the efficacy of Cuttlefish bone powder in the removal of Reactive Blue 19 dye from aqueous solutions: equilibrium and Isotherm studies. *J. Community Health Res.* **2012**, *1*, 68-78. http://jhr.ssu.ac.ir/article-1-46-en.html.

[23] Lv, X.; Xue, X.; Jiang, G.; Wu, D.; Sheng, T.; Zhou, H.; Xu, X., Nanoscale zero-valent iron (nZVI) assembled on magnetic Fe_3O_4/graphene for chromium(VI) removal from aqueous solution. *J. Colloid Interface Sci.* **2014**, *417*, 51-59. https://doi.org/10.1016/j.jcis.2013.11.044.

[24] Moawed, E.; Abulkibash, A.; El-Shahat, M., Synthesis and characterization of iodo polyurethane foam and its application in removing of aniline blue and crystal violet from laundry wastewater. *J. Taibah Univ. Sci.* **2015**, *9*, 80-88. https://doi.org/10.1016/j.jtusci.2014.07.003.

[25] Çiçek, F.; Özer, D.; Özer, A.; Özer, A., Low cost removal of reactive dyes using wheat bran. *J. Hazard. Mater.* **2007**, *146*, 408-416. https://doi.org/10.1016/j.jhazmat.2006.12.037.

[26] Elkady, M.; Ibrahim, A. M.; El-Latif, M. A., Assessment of the adsorption kinetics, equilibrium and thermodynamic for the potential removal of reactive red dye using eggshell biocomposite beads. *Desalination.* **2011**, *278*, 412-423. https://doi.org/10.1016/j.desal.2011.05.063.

[27] Kousalya, G.; Gandhi, M. R.; Meenakshi, S., Sorption of chromium(VI) using modified forms of chitosan beads. *Int. J. Biol. Macromol.* **2010**, *47*, 308-315. https://doi.org/10.1016/j.ijbiomac.2010.03.010.

[28] Liu, B.; Lv, X.; Meng, X.; Yu, G.; Wang, D., Removal of Pb(II) from aqueous solution using dithiocarbamate modified chitosan beads with Pb(II) as imprinted ions. *Chem. Eng. J.* **2013**, *220*, 412-419. https://doi.org/10.1016/j.cej.2013.01.071.

[29] Gurgel, L. V. A.; Gil, L. F., Adsorption of Cu(II), Cd(II) and Pb(II) from aqueous single metal solutions by succinylated twice-mercerized sugarcane bagasse functionalized with triethylenetetramine. *Water Res.* **2009**, *43*, 4479-4488. https://doi.org/10.1016/j.watres.2009.07.017.

[30] Sun, X.; Yang, L.; Li, Q.; Zhao, J.; Li, X.; Wang, X.; Liu, H., Amino-functionalized magnetic cellulose nanocomposite as adsorbent for removal of Cr(VI): synthesis and adsorption studies. *Chem. Eng. J.* **2014**, *241*, 175-183. https://doi.org/10.1016/j.cej.2013.12.051.

[31] Ahmadi, F.: Esmaeili, H., Chemically modified bentonite/Fe_3O_4 nanocomposite for Pb(II), Cd(II), and Ni(II) removal from synthetic wastewater. *Desalin. Water Treat.* **2018**, *110*, 154-167. DOI: 10.5004/dwt.2018.22228.

[32] Zadeh, B. S.; Esmaeili, H.; Foroutan, R., Cadmium(II) Removal from Aqueous Solution Using Microporous Eggshell: Kinetic and Equilibrium Studies. Indones. *J. Chem.* **2018**. https://doi.org/10.22146/ijc.28789.

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