

**EREM 82/3**

Journal of Environmental Research,  
Engineering and Management  
Vol. 82 / No. 3 / 2026  
pp. 114–131  
10.5755/j01.erem.82.3.44857

**Removal of Copper Ions from an Aqueous Solution Using *Salvia officinalis* and *Alchemilla vulgaris* Leaves as Natural Materials**

Received 2026/04

Accepted after revisions 2026/08

<https://doi.org/10.5755/j01.erem.82.3.44857>

# Removal of Copper Ions from an Aqueous Solution Using *Salvia officinalis* and *Alchemilla vulgaris* Leaves as Natural Materials

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This study demonstrated the use of two plant leaves, *Salvia officinalis* (SO) and *Alchemilla vulgaris* (AV), as natural absorbents to remove the copper ions ( $\text{Cu}^{2+}$ ) from an aqueous solution. The adsorption process was investigated under different operating conditions, including the initial concentration of the  $\text{Cu}^{2+}$  ion, contact time, pH, particle size, and adsorbent dose, at room temperature. The adsorbent materials were characterised using Fourier transform infrared spectroscopy (FTIR), Brunauer-Emmett-Teller (BET) and scanning electron microscope (SEM) tests. The best  $\text{Cu}^{2+}$  ion removal efficiency was 95.864% on SO leaves under the following optimal conditions: pH 6, a contact time of 75 min, an adsorbent material dose of 10 g/L, an initial  $\text{Cu}^{2+}$  ion concentration of 10 mg/L and a particle size of 75  $\mu\text{m}$ . The maximum removal efficiency for  $\text{Cu}^{2+}$  ions was 91.566% on AV leaves with optimum parameters of pH 6, a contact time of 90 min, an adsorbent material dose of 15 g/L, an initial  $\text{Cu}^{2+}$  ion concentration of 10 mg/L and a particle size of 75  $\mu\text{m}$  at  $25 \pm 5^\circ\text{C}$  temperature. It was also found that the equilibrium results from the adsorption procedure were well fitted by the Freundlich isotherm model. The maximum adsorption capacity for  $\text{Cu}^{2+}$  ions was 16.949 mg/g on SO leaves and 10.101 mg/g on AV leaves. It was observed that the pseudo-second-order model provided a good fit to the  $\text{Cu}^{2+}$  ion removal rate per dry mass of plant leaves. The results conclude that both SO and AV leaves can be effectively applied as adsorbents for the removal of  $\text{Cu}^{2+}$  ions from an aqueous solution.

**Keywords:** adsorption process, biosorbents, copper ions removal, adsorption isotherms and kinetics.

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## Introduction

The contamination of natural water supplies is one of the most important problems facing humanity today. It has

grown exponentially over the past few years, and its impacts on living things are now alarmingly high (Hussein et al., 2023; Kayani and Mohammed, 2025). Heavy metals that are hazardous to humans and animals are

considered to have a direct impact on both species and can slowly accumulate in the body (Hussein, 2019b). The heavy metals are defined as cations with a density above  $5 \text{ g/cm}^3$  and an atomic number greater than 20, and at low doses, they are hazardous (Carolin et al., 2017). There are many examples of heavy metals like copper (Cu), nickel (Ni), cadmium (Cd), mercury (Hg), chromium (Cr), antimony (Sb), arsenic (As), and others (Tekin and Acikel, 2023; Bahouch et al., 2026). For instance, the discharge of industrial effluents containing these heavy metals and chemicals can contribute significantly to water pollution. The increased use of heavy metals over the last few decades has inevitably resulted in a greater flux of metallic pollutants in groundwater, surface water and drinking water, which has posed serious problems for the environment and human health (Carolin et al., 2017). In particular, excessive amounts of  $\text{Cu}^{2+}$  ions can be harmful to humans, animals and plants. For humans, it can produce severe irritation of the mucous membranes and the digestive system, liver and kidney damage, and widespread problems in the capillaries and the central nervous system (Ojedokun and Bello, 2016).

It is important to control the excess amount of copper in the soil, as it has a toxic effect on microorganisms, disrupting the nutrient cycle and inhibiting the mineralisation of essential nutrients such as nitrogen and phosphorus (Yirga et al., 2022). It is of the utmost importance to develop effective processes to remove copper from aquatic environments. Among the various techniques that can be used for the treatment of polluted water are lime coagulation, precipitation, exchange of ions, adsorption and solvent extraction (Yirga et al., 2022; Hussein, 2023). Adsorption is the most common method used because it is efficient in removing a wide range of contaminants. It is widely recognised as an effective technique for the removal of contaminants, including heavy metals, from wastewater. The adsorption method, when compared with other technologies, is highly effective because it produces purified water of a high standard (Tran, 2023). Adsorption is a reliable alternative to more traditional approaches because it has several benefits: it is a cost-effective process, has low sediment production, and efficiently treats wastewater containing heavy metals at a relatively cost. Due to the process reversibility, the adsorbent used in the adsorption process can be retrieved throughout the disposal process; moreover, it is utilised for removal of organic

and inorganic pollutants (Ince and Ince, 2017). The metal adsorption efficiency depends on many factors like pH, adsorbent particle size, dosage, surface area, adsorbate concentration and solution temperature. This means that it is possible to control the removal efficiency by any change in the previous factors (Tekin and Acikel, 2023). As a direct result of this, considerable efforts have been made to detect adequate materials that are both cost-effective and efficient (Czelej et al., 2016).

Adsorption, as a sustainable treatment method, is a cost-effective technique to remove heavy metals. The reason for that is the use of many natural materials, including microorganisms such as bacteria and fungi, simple organisms such as algae and yeasts, and agricultural wastes such as peat, wood, pine bark, fruit peel, seeds, plant leaves, and nut shells (Sirijaree and Praipipat, 2023). For instance, the majority of agricultural waste or by-products are not seen as high-value products in the marketplace (Mohsen et al., 2023). Plant leaves have been widely investigated as natural, affordable, acceptable and effective adsorbent materials (Rahimpour et al., 2017). Bani-Atta et al. (2021) investigated the removal of methylene blue dye from an aqueous solution using chemically modified *Ocimum basilicum* leaf powder (CMOBLP) under different experimental conditions, including initial methylene blue dye concentration, pH value, contact time and temperature. The results demonstrated that the adsorption capacities of 714.290 mg/g, 666.670 mg/g, 625.000 mg/g, and 555.560 mg/g were gained at 20, 30, 40, and 50°C temperature steps, respectively. The methylene blue dye adsorption onto the CMOBLP was best suited by the second-order kinetic model and the Langmuir isotherm model. The porosity and surface area were determined by a Brunauer-Emmett-Teller (BET) surface analyser.

Darweesh et al. (2022) studied the possibility of using adsorbents, such as banana leaves (BL) and banana-leaf-derived activated carbon (BLAC), to remove copper(II) from aqueous solutions in order to investigate the impact of many testing parameters, such as temperature, initial copper(II) concentration, pH, adsorbent dosage, and agitation speed. The maximum removal efficiency was 52% for BL and 83% for BLAC at a pH value of 5, with a contact time of 40 min and 60 min for BL and BLAC, respectively. For both adsorbents (the initial copper(II) concentration was 250 mg/L, the adsorbent dose was 0.250 g, the temperature was 40°C, and

the agitation speed was 400 rpm). The Langmuir isotherm model fitted well, and the maximum capacity of copper(II) was 66.200 mg/g and 48.700 mg/g for BLAC and BL, respectively. Kazemi et al. (2021) assessed the use of synthesised *Origanum majorana*-capped silver nanoparticles for the removal of Ni(II) and Co(II) ions from wastewater. The maximum removal of Ni(II) and Co(II) of 88.600% was achieved within the optimum time of 60 min. The pH was 9 and 7 and the dose of biosorbent was 40 and 50 mg for Ni(II) and Co(II), respectively. The initial concentration of both metal ions was 30 mg/L. To evaluate the surface characteristics of the Ni(II) and Co(II) ion-loaded biomass, scanning electron microscopy (SEM), X-ray diffraction, transmission electron microscopy and Brunauer-Emmett-Teller (BET) were used. The results showed that the adsorption of Ni(II) and Co(II) ions fit the pseudo-second-order model. The Langmuir model fitted the experimental results better than the Freundlich, Dubinin-Radushkevich, and Temkin isotherms. It was concluded that the use of such natural plant leaves as adsorbents might be promising because of their non-toxic nature, low cost, and lower operating costs when compared with other adsorbents (Liakos et al., 2021).

In addition, other plant leaves have the quality to provide a rational means for the treatment of polluted water by adsorption. *Salvia officinalis* plant is known by various common names, and the most well-known are Sage and Meramia (in Arabic). It belongs to the genus *Salvia*, which comprises about 900 species. It is a member of the family Labiatae/Lamiaceae (Khiya et al., 2019; Jakovljević Kovač et al., 2021). It is cultivated in the eastern Mediterranean region and southern Europe but has been spread in many places throughout the world. The best time to plant the *Salvia officinalis* plant is in the spring and summer months (Bendifallah et al., 2018). Recently, most cosmetic and food factories continue to widely use the plant; moreover, this herb has a clear place in medicinal uses to treat some pathological conditions with antimicrobial activity, like Salmonella (Hayouni et al., 2008). It has positive health effects, and because of that, it is used in popular medications for the treatment of numerous health problems, like respiratory problems, inflammatory conditions, and nervous and mental illnesses (Sobhanardakani and Zandipak, 2017). In addition to the medicinal uses of sage, it is also favourable in the cosmetics industry (Calimli et al., 2020). Bingul and

Adar (2021) investigated the use of spent *Salvia officinalis* as an adsorbent for the removal of maxilon yellow and methylene blue dyes from aqueous solutions. The maximum adsorption capacities for the dyes (maxilon yellow and methylene blue) were found to be 11.790 mg/g and 16.530 mg/g, respectively. The Freundlich isotherm model was in good fit with the experimental data. The adsorption process suited the pseudo-second-order kinetic model, according to the kinetic model analysis. A pH of 5.700 for methylene blue and 6.500 for Maxilon yellow, a dye concentration of 100 mg/L, a revolution speed of 200 rpm, an adsorbent dose of 0.050 g/100mL and a particle size of 180 µm at 25°C were the optimum experimental parameters. The highest removal efficiencies of methylene blue and maxilon yellow were 78% and 56%, respectively.

In Arabic, *Alchemilla vulgaris* L known as lion's foot and lady's mantle is a member of the family Rosaceae (Vlaisavljevic et al., 2019). It includes nearly 250 species growing in Europe, North America, and western Asia (Kanak et al., 2022). Some researchers confirmed that it has medicinal significance because of its biological properties (Tadiæ et al., 2020). Recently, research has demonstrated that some of *Alchemilla vulgaris* L.'s biological properties can be used for wound healing, dysmenorrhea, mild diarrhoea, muscle spasms, and high blood pressure (Takır et al., 2015). *Alchemilla vulgaris* L. has many different components, like phenolic acids and tannins (Kovač et al., 2022). Methylene blue dye removal by adsorption on *Alchemilla vulgaris* leaves was investigated by Reçber (2022). The highest removal efficiency was found to be 87.850% in the experiments at optimum parameters of pH (5), adsorbent dose (0.500 g/L), initial methylene blue dye concentration (10 mg/L), particle size (75 µm) and contact time (60 min).

Few studies have dealt with the use of *Salvia officinalis* (SO) and *Alchemilla vulgaris* (AV) leaves as adsorbents for heavy metal treatment in polluted waters. SO and AV leaves are attractive adsorbents because of their abundance, low cost, environmental friendliness, ease of storage, high contents of particulate carbon, cellulose, hemicellulose, lignin, and surface functional groups (e.g., hydroxyl and carboxyl groups). This study aims to achieve the copper ion removal with SO and AV leaves, which were optimised by the use of various parameters (pH, adsorbent dosage, contact time, particle size, and initial copper ion concentration) at room temperature.

Scanning electron microscopy (SEM), Fourier transform infrared spectrophotometer (FTIR), and Brunauer-Emmett-Teller (BET) were used to characterise adsorbent materials. Furthermore, isotherm (Freundlich and Langmuir) and kinetics (pseudo-first order and pseudo-second order) models were also investigated.

## Methodology

Synthetic wastewater containing copper(II) chloride dihydrate ( $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ ) with an atomic weight of 63.550 g/mol and a molecular weight of 170.480 g/mol was used. To prepare a  $\text{Cu}^{2+}$  ions stock solution of 1000 mg/L, 2.68 g of  $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$  was dissolved in one litre of distilled water. The mass of metal salt, under the presumption that the metal salt was completely dissolved in the water, was determined by following Eq. (1) (Hussein, 2019a):

$$W = V \times C_o \times \frac{M.wt}{At.wt} \quad (1)$$

where  $W$  is the  $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$  mass (g),  $V$  is the volume of solution (L),  $C_o$  is the initial concentration of  $\text{Cu}^{2+}$  ions (mg/L),  $M.wt$  is the metal salt  $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$  molecular weight (g/mol), and  $At.wt$  is the atomic weight of  $\text{Cu}^{2+}$  ions (g/mol).

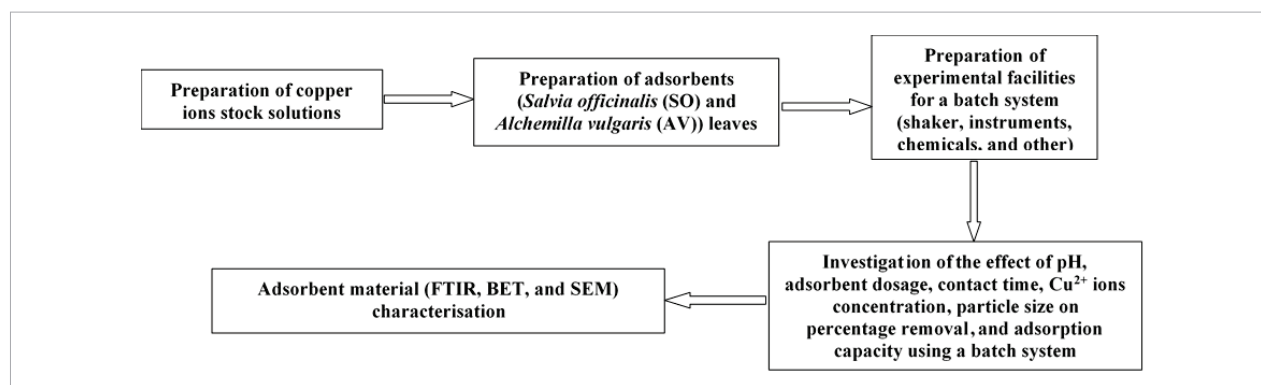
*Salvia officinalis* (SO) and *Alchemilla vulgaris* (AV) leaves as natural adsorbents were supplied from a local market in Baghdad, Iraq. They were washed several times with tap water and then rinsed with distilled water to get rid of any impurities, such as sand. The solution was then dried using an electric oven for one hour at 80°C. The materials were ground with an electrical

blender to obtain powder and then sieved with different diameters of 75, 100, 300, 600, 700, and 800  $\mu\text{m}$  to be utilised in the experiments.

## Experimental work

A batch of experiments was conducted to investigate the removal of  $\text{Cu}^{2+}$  ions from an aqueous solution using SO and AV leaves to find the best adsorbent material dose, pH, contact time, initial concentration of  $\text{Cu}^{2+}$  ions and particle size. The experiments were carried out in a conical flask of 250 mL volume, and then the flask was filled with 100 mL of a copper solution. The pH of the copper solution in each flask was adjusted by adding either 0.100 M NaOH or 0.100 M HCl using a pH meter (pH tester, China) to reach the desired pH value and measured by pH meter. A known weight of adsorbent material (SO and AV) leaves was added into each flask, the flask was placed in a shaker (Heidolphunimax 2010, Germany), and the mixture was agitated continuously at a speed of 200 rpm for the desired time at a temperature of  $25 \pm 5^\circ\text{C}$ . The withdrawn copper samples were filtered using filter paper (qualitative filter paper, 15 cm) and then an atomic absorption spectrophotometer (AAS) (A-shimadzu aa-7000f, Japan) was employed to determine the equilibrium concentration of  $\text{Cu}^{2+}$  ions in the tested samples. The effects of pH (2 to 12), initial  $\text{Cu}^{2+}$  concentration (10 to 200 mg/L), contact time (0 to 180 min), dose of adsorbents (1 to 30 g/L), and particle size (75 to 800  $\mu\text{m}$ ) variables were calculated to determine the  $\text{Cu}^{2+}$  ion removal using SO and AV leaves. The flowchart for methodology work is shown in Fig. 1. Eq. (2) was used to calculate the removal percentage ( $R\%$ ) of  $\text{Cu}^{2+}$  ions from an aqueous solution and Eq. (3) was used to calculate the adsorption equilibrium

Fig. 1 Flow chart of the methodology work



capacity ( $q_e$ ) of the adsorbents (Darweesh et al., 2022). These equations were expressed as follows:

$$R(\%) = \frac{(C_o - C_e)}{C_e} \times 100 \quad (2)$$

$$q_e = \frac{V(C_o - C_e)}{m} \quad (3)$$

where  $C_o$  is the initial  $\text{Cu}^{2+}$  ion concentration (mg/L),  $C_e$  is the equilibrium  $\text{Cu}^{2+}$  ions concentration after adsorption (mg/L),  $V$  is the volume of the solution sample (L), and  $m$  is the adsorbent material amount (g).

### Characterisation of adsorbent materials

To identify the main functional groups of the adsorbent (SO and AV) leaves, Fourier transform infrared spectrophotometer (FTIR) was used before and after the adsorption process. The FTIR (Shimadzu, Japan) with a range of 400–4000  $\text{cm}^{-1}$  was used in the present study. The images were obtained using scanning electron microscopy (SEM) (VEGA II SBH, Czech) to provide details on the surface morphology of the adsorbent materials (SO and AV) leaves. Average pore diameter (nm), pore volume ( $\text{cm}^3/\text{g}$ ), and surface area ( $\text{m}^2/\text{g}$ ) of adsorbent leaves (SO and AV) were identified from Brunauer-Emmett-Teller (BET) (BEL, BELSORP MINI II, Japan).

### Isotherm Models

Langmuir and Freundlich isotherm models were used in this study. According to the Langmuir isotherm model, it is assumed that there is monolayer adsorption of adsorbate on a homogeneous adsorbent surface, all adsorption sites are identical and limited in number, with no interaction between adsorbed molecules and a uniform energy distribution on the adsorption surface (Yousef et al., 2020). The equation of the Langmuir isotherm is expressed as in Eq. (4) (Shamkhi and Hussein, 2022):

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e} \quad (4)$$

The above equation can be written as in Eq. (5):

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

where  $q_e$  is the adsorption capacity of  $\text{Cu}^{2+}$  ions at equilibrium (mg/g),  $q_m$  is the maximum adsorption capacity on the surface of the adsorbent (mg/g),  $C_e$  is the equilibrium  $\text{Cu}^{2+}$  ions concentration (mg/L), and  $K_L$  is

the Langmuir isotherm constant, which expresses the affinity of the binding site (L/mg). The values of the  $q_m$  and  $K_L$  parameters were determined from the line slope, and an intercept was drawn between  $C_e/q_e$  and  $C_e$ . The Langmuir isotherm equation may be represented by the dimensionless constant separation factor ( $R_L$ ) (Meshkat et al., 2022) as demonstrated in Eq. (6):

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

$R_L$  value refers to the isotherm nature, which is either unfavourable ( $R_L > 1$ ), linear ( $R_L = 1$ ), favourable ( $0 < R_L < 1$ ), or irretrievable ( $R_L = 0$ ). The Freundlich isotherm model is the oldest known model that calculates the adsorption isotherm. It assumes the adsorption of multilayers of adsorbate on a heterogeneous adsorbent surface where adsorption sites are not identical, with a non-uniform energy distribution on the adsorption surface (Kumar et al., 2019). The Freundlich isotherm model is illustrated by Eq. (7) (Mohammed et al., 2016):

$$q_e = K_f C_e^{1/n} \quad (7)$$

To simplify Eq. (7), it can be written as in Eq. (8):

$$\ln q_e = \ln K_f + \frac{1}{n} \ln C_e \quad (8)$$

where  $q_e$  and  $C_e$  are as referred to before,  $K_f$  is the constant of Freundlich adsorption, which represents adsorption capacity (mg/g), and  $n$  is the intensity factor of adsorption. The values of the  $1/n$  and  $K_f$  parameters were estimated from the line slope and the intercept that was drawn between  $\ln q_e$  and  $\ln C_e$ . The value of  $1/n$ , which ranges between 0 and 1, represents the favourable adsorption process of metal ions. If the value of  $1/n$  is less than 1, it refers to normal adsorption, whereas a value of  $1/n$  greater than 1 refers to the cooperative adsorption process (Dada et al., 2012).

### Kinetic models

To compute kinetic adsorption data, pseudo-first-order and pseudo-second-order models were used (Hidayata et al., 2024). The pseudo-first-order kinetic model is predicated on the idea that the alteration rate of solute uptake with time is directly proportional to the amount of solid uptake and the difference in saturation concentration with time (Lopez-Luna et al., 2019). Eq. (9)

expresses the pseudo-first-order kinetic model (Tan and Hameed, 2017):

$$\frac{dq_t}{dt} = K_1(q_e - q_t) \quad (9)$$

where  $q_e$  is the pollutant quantity at equilibrium (mg/m), and  $q_t$  is the pollutant quantity at time  $t$  (mg/m).  $K_1$  is the pseudo-first-order equilibrium rate constant (1/min). By integrating Eq. (9) with the boundary conditions of  $q_t=0$  at  $t=0$  and  $q_t = q_t$  at  $t = t$ , Eq. (10) is given as follows:

$$\ln(q_e - q_t) = \ln q_e - K_1 t \quad (10)$$

$K_1$  and  $q_e$  values were obtained from the slope of the line and the intercept by plotting  $\ln(q_e - q_t)$  against  $t$ .

A pseudo-second-order kinetic model is predicated on the idea of the adsorbate chemisorption on the adsorbent material. In addition, this model predicated the hypothesis that chemical sorption involving valence force through sheering or electron exchange between the adsorbent and the adsorbate could be a limiting step (Ebelegi et al., 2020). This model can be shown in Eq. (11):

$$\frac{dq_t}{dt} = K_2(q_e - q_t)^2 \quad (11)$$

After integrating Eq. (11) at boundary conditions,  $q_t = 0$  at  $t = 0$  and  $q_t = q_t$  at  $t = t$ , Eq. (12), which represents the pseudo-second-order kinetic model, can be obtained (Kazemi et al., 2021):

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e} \quad (12)$$

where  $K_2$  is the rate constant of adsorption at the equilibrium for pseudo-second-order (g/mg.min). The  $K_2$  and  $q_e$  parameters are determined from the line slope and the intercept by drawing  $t/q_t$  versus  $t$ .

## Results and Discussion

### FTIR results

The  $\text{Cu}^{2+}$  ion adsorption presence on the surface of SO and AV leaves was demonstrated by the FTIR spectra of the leaves before and after sorption of  $\text{Cu}^{2+}$  ions, as shown in Figs. 2 and 3 as well as Tables 1 and 2. The FTIR of SO and AV leaves was gained within the 400

to 4000  $\text{cm}^{-1}$  range of wave numbers. There are a few different adsorption peaks that are associated with the complexity of the surface. The heavy metal ions tend to get stuck with various functional groups that are found in the organic compounds that cover the sorbents surfaces (Hussein and Jasim, 2019).

**Table 1.** Functional groups for the SO leaves before and after adsorption of  $\text{Cu}^{2+}$  ions

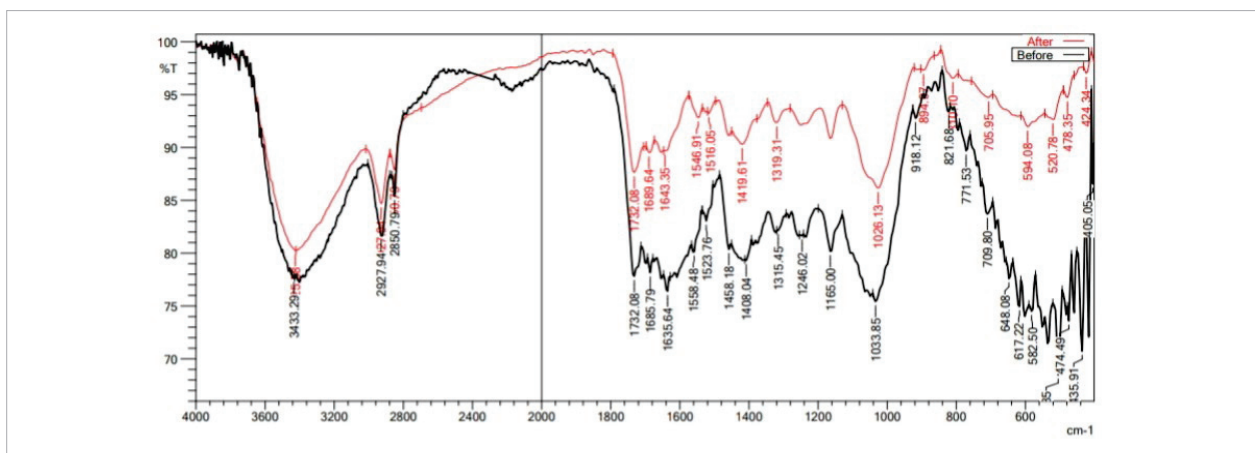
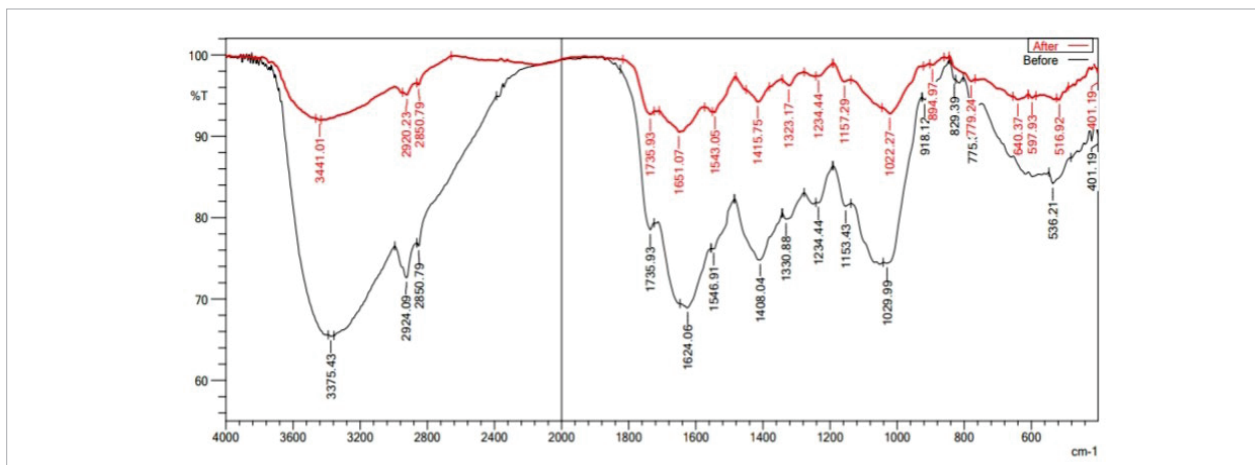
| Functional group                        | Before adsorption | After adsorption |
|-----------------------------------------|-------------------|------------------|
| Hydroxyl group (Alcohols) (O-H) stretch | 3433              | 3425             |
| Alkane (C-H) stretch                    | 2927              | 2927             |
| Carbonyl (C=O) stretch                  | 1732              | 1732             |
| Alkane (-C-H) bend                      | 1315              | 1319             |
| Alcohol (C-O) stretch                   | 1033              | 1026             |
| Alkene (C-H) bend                       | 918               | 894              |
| Amine (N-H) bend                        | 709               | 705              |

The spectra before adsorption of SO leaves show peaks at 3433  $\text{cm}^{-1}$ , 2927  $\text{cm}^{-1}$ , 1732  $\text{cm}^{-1}$ , 1315  $\text{cm}^{-1}$ , 1033  $\text{cm}^{-1}$ , 918  $\text{cm}^{-1}$ , and 709  $\text{cm}^{-1}$ , and those for AV leaves show peaks at 3375  $\text{cm}^{-1}$ , 2924  $\text{cm}^{-1}$ , 1735  $\text{cm}^{-1}$ , 1330  $\text{cm}^{-1}$ , 1029  $\text{cm}^{-1}$ , 918  $\text{cm}^{-1}$ , and 775  $\text{cm}^{-1}$ . The existence of a broad peak at 3433  $\text{cm}^{-1}$  and 3375  $\text{cm}^{-1}$  shows the existence of an intermolecularly bonded hydroxyl group (alcohols) (O-H) stretch (Mosoarca et al., 2022). The existence of peak at 2927  $\text{cm}^{-1}$  and 2924  $\text{cm}^{-1}$  indicates a saturated C-H chemical bond, which can be likened to alkane (C-H) that are found in the pulp (Bello et al., 2017; Mosoarca et al., 2022). The peaks at 1732  $\text{cm}^{-1}$  and 1735  $\text{cm}^{-1}$  show the existence of the carbonyls (C=O) stretch (double bonds) (Rahimpour et al., 2017). The peaks absorbed at 1315  $\text{cm}^{-1}$  and 1330  $\text{cm}^{-1}$  indicate the alkane group (-C-H) bend (Çelebi et al., 2020). The peaks detected at 1033  $\text{cm}^{-1}$  and 1029  $\text{cm}^{-1}$  are related to the alcohol group (C-O) stretch (Shamkhi and Husssein, 2022). The peaks that appear at 918  $\text{cm}^{-1}$  indicate alkene group (C-H) bend (strong), the peaks at 709  $\text{cm}^{-1}$  are related to amine (N-H) bend (wag), 775  $\text{cm}^{-1}$  is alkene group (C-H) bend (strong) and amine (N-H) bend (wag) (Liu et al., 2017). The high quantity existence of hydroxyl (3000–4000  $\text{cm}^{-1}$ ) and carboxyl groups (1000–2000  $\text{cm}^{-1}$ ) on the biosorbent surface indicates the tendency of SO and AV leaves to adsorb metal ions with positive charge.

**Table 2.** Functional groups for AV leaves before and after adsorption of  $\text{Cu}^{2+}$  ions

| Functional group                             | Before adsorption | After adsorption |
|----------------------------------------------|-------------------|------------------|
| Hydroxyl group (Alcohols) (O-H) stretch      | 3375              | 3441             |
| Alkane (C-H) stretch                         | 2924              | 2920             |
| Carbonyl (C=O) stretch                       | 1735              | 1735             |
| Alkane (-C-H) bend                           | 1330              | 1323             |
| Alcohol (C-O) stretch                        | 1029              | 1022             |
| Alkene (C-H) bend                            | 918               | 894              |
| Alkene (C-H) bend and Amine (N-H) bend broad | 775               | 779              |

The existence of such functional groups plays an important role for the adsorption of heavy metals, which occurs as a result of the affinity of metal ions to the functional groups. Figs. 2 and 3 compare the FTIR spectra of SO and AV leaves before and after adsorption. The two curves appear similar because the same functional groups exist on the surface before and after adsorption. The difference is the important shifts and stretches in the peaks and intensities of the spectrum, which contribute to the interaction between the functional groups on the adsorbent surface and  $\text{Cu}^{2+}$  ions in the solution. The important shift happens after absorption in the O-H, C-O, and C-H peaks, which are peculiar to the  $\text{Cu}^{2+}$  ion adsorption. The outcomes show that carboxylic and hydroxylic groups perform a major role in the adsorption of  $\text{Cu}^{2+}$  ions (Santos et al., 2022).

**Fig. 2.** Results of FTIR analyses of SO leaves before (black line) and after (red line) adsorption of  $\text{Cu}^{2+}$  ions**Fig. 3.** Results of FTIR analyses of AV leaves before (black line) and after (red line) adsorption of  $\text{Cu}^{2+}$  ions

### Scanning electron microscopy (SEM) analysis

Scanning electron microscopy (SEM) for studying the surface of SO and AV leaves, respectively shown in Figs. 4 and 5, could provide important information on the interaction degree between the  $\text{Cu}^{2+}$  ions and the adsorbent particles. SEM showed the morphology and surface texture of SO and AV leaves before and after adsorption. The morphology of the adsorbent surface revealed porous and convoluted areas before adsorption, indicating the presence of active sites. The porous and irregular adsorbent surface structure may be easily detected in the SEM image of SO leaves (Fig. 4(a)). Moreover, the morphology of the surface of AV leaves (Fig. 5(a)) is rough and homogeneous; the adsorbent

scanning electron micrograph demonstrates and seems to verify the homogeneous structure and the presence of different grooves that aid in its effective application in the various compound adsorptions. SEM images of SO and AV leaves after  $\text{Cu}^{2+}$  ion adsorption are presented in Figs. 4(b) and 5(b). The adsorbent surface morphology shows that the active sites were occupied by  $\text{Cu}^{2+}$  ions after adsorption. The homogenous surface manifests that all cavities on the surface were occupied by  $\text{Cu}^{2+}$  ions (Padma and Sastry, 2022). In addition, on the adsorbent surface after the adsorption process of  $\text{Cu}^{2+}$  ions, the grooves were completely covered with  $\text{Cu}^{2+}$  ions, and the adsorbent structure was changed (Padma and Sastry, 2021).

Fig. 4. SEM images of SO leaves (a) before and (b) after adsorption of  $\text{Cu}^{2+}$  ions

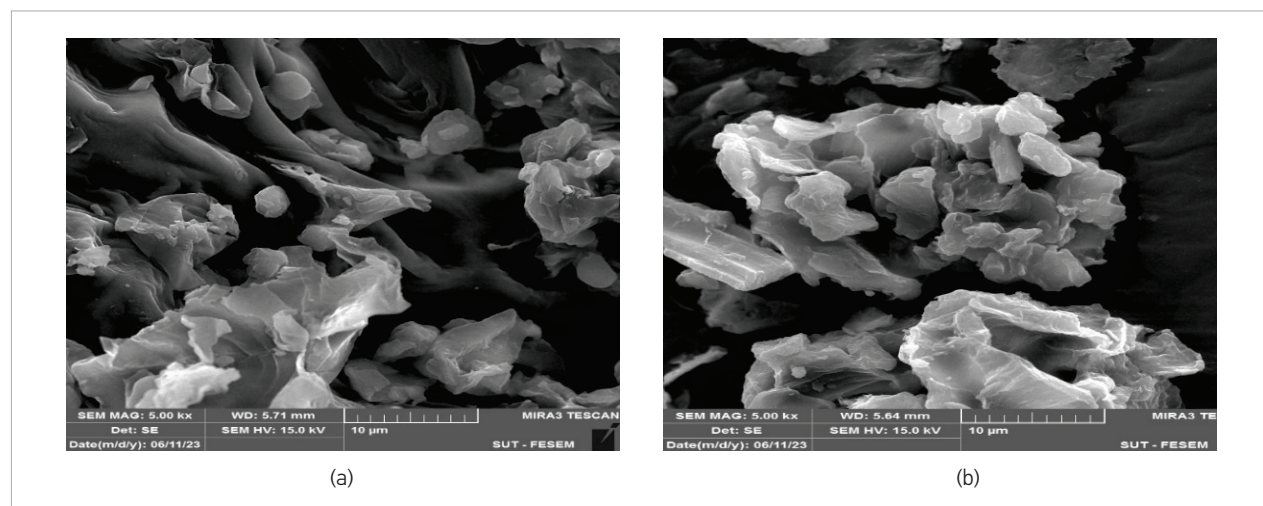
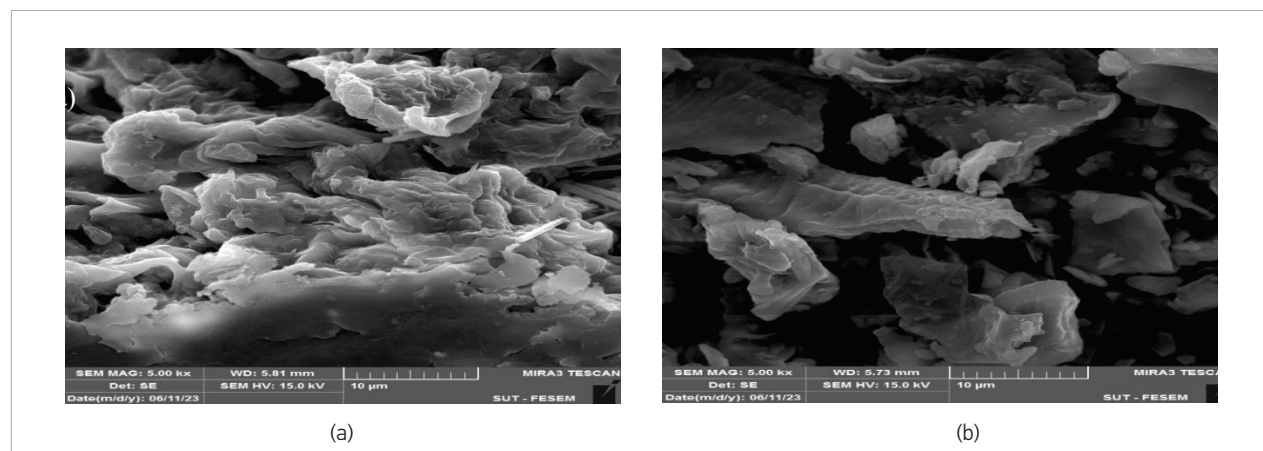


Fig. 5 SEM images of AV leaves (a) before and (b) after adsorption of  $\text{Cu}^{2+}$  ions



### Brunauer–Emmett–Teller (BET) analysis

The specific surface areas were determined using the Brunauer–Emmett–Teller (BET) equation at a relative vapor pressure of 88 KPa and 77 K using nitrogen adsorption/desorption tests. The relative pressure ( $p/p_0$ ) was adjusted from 0.001 to 0.984 before the measurements of gas adsorption (Liakos et al., 2021). The surface area was calculated by the standard BET equation at a relative pressure ( $p/p_0$ ) of 0.010 to 0.459 for SO leaves and 0.021 to 0.458 for AV leaves. The results of surface area by the BET method were 6.004 m<sup>2</sup>/g and 5.395 m<sup>2</sup>/g for SO and AV leaves, respectively. The total pore volumes for SO and AV leaves were 0.067 cm<sup>3</sup>/g and 0.042 cm<sup>3</sup>/g, respectively.

The average pore diameter of SO and AV leaves was within the range of 2 < d < 50 nm, and the adsorbents were categorised as mesoporous materials (Ain et al.,

2020). The N<sub>2</sub> adsorption/desorption isotherm of both SO and AV leaves is demonstrated in Fig. 6. The adsorption of BET isotherms fitting line in the relative pressure ( $p/p_0$ ) was drawn to calculate the SO and AV leaves of a specific surface area. The curves reveal that the isotherms of the SO and AV leaves follow a hysteresis loop, which extends to 0.296  $p/p_0$  for SO and AV leaves according to the categorisation of the International Union of Pure and Applied Chemistry (IUPAC). The extensive hysteresis loop means that there are thriving and active sites for adsorption because there are sufficient pores as suitable recipients for the pollutant molecules (Sirijaree and Praipipat, 2023). Moreover, the Barrett-Joyner-Halenda (BJH) technique was used to calculate the pore size distribution and is revealed in Figs. 7(a) and 7(b) for SO and AV leaves, respectively. Therefore, the nitrogen adsorption/desorption isotherms and pore volume

Fig. 6. N<sub>2</sub> isotherms of adsorption/desorption: (a) SO and (b) AV leaves

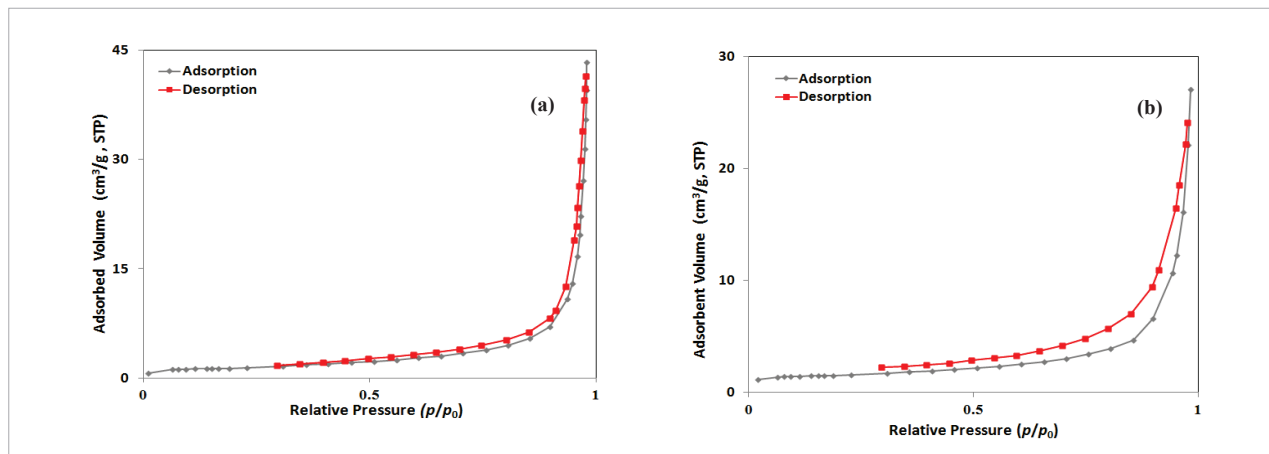
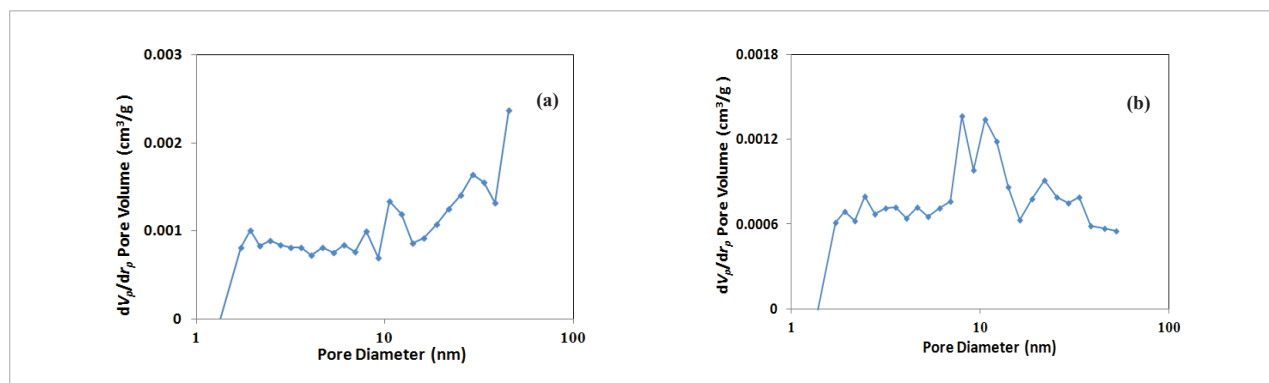


Fig. 7. BJH pore size distribution of (a) SO and (b) AV leaves



measurements would indicate that SO and AV leaves have porous structures. Furthermore, these results proved that the surface area enhanced the adsorption/desorption capacity and was compatible with the literature study (Alnasrawi and Mohammed, 2023). The BET analysis results (pore size, surface area, and pore volume) for SO and AV leaves are shown in *Table 3*.

**Table 3.** Details of textural parameters (BET) for SO and AV leaves

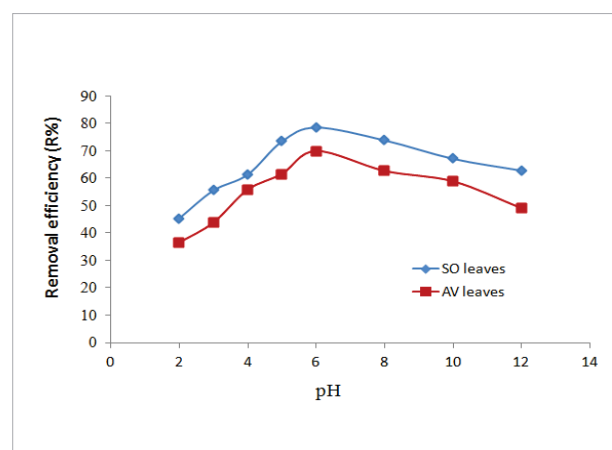
| Samples   | BET surface area (m <sup>2</sup> /g) | Total pore volume (cm <sup>3</sup> /g) | Average pore diameter (nm) |
|-----------|--------------------------------------|----------------------------------------|----------------------------|
| SO leaves | 6.004                                | 0.067                                  | 48.625                     |
| AV leaves | 5.395                                | 0.042                                  | 31.019                     |

### Impact of pH

The pH values of the Cu<sup>2+</sup> ion solution have an impact on the adsorption results due to it being the main controller of the charges that are found on the sorbent particles (Shamkhi and Husssein, 2022). In this study, pH ranged from 2 to 12, an adsorbent dosage of 5 g/L of SO and AV leaves with 50 mg/L of an initial Cu<sup>2+</sup> ion concentration for 60 min, a particle size of 300 µm while the shaker was set to 150 rpm and a temperature of 25 ± 5°C were investigated. According to *Fig. 8*, the adsorbed Cu<sup>2+</sup> ion percentage increases when the pH increases. The pH meter showed the maximum removal efficiency at pH 6. At optimum pH (6), the maximum removal efficiency is due to chemisorption, or electrostatic attraction, which will occur more readily as pH increases because the active group in SO and AV leaves, such as –OH groups, are gradually deprotonated, accompanied by the negatively charged (Alnasrawi and Mohammed, 2023). After reaching this maximum, the percentage of Cu<sup>2+</sup> ions that is adsorbed begins to fall as the pH keeps rising. The lowest pH values show the lowest amount of adsorption, which may be because the hydrogen ions (H<sup>+</sup>) start competing with the active surface sites of SO and AV leaves by competition with metal ions (Padma and Sastry, 2022). Furthermore, below pH 6, the results recorded low adsorption due to the repulsion forces between the positive particles of the adsorbent material and the metal ions (Meseldzijaa et al., 2019). A pH greater than 6 leads to metal ion precipitation on the adsorbent surface because of the insolubility of Cu(OH)<sub>2</sub> (Didaran et al., 2025). In order

to conduct accurate sorption studies, it is essential to maintain a pH of 6 to prevent metal precipitation during the removal process. This pH level allows for a maximum removal reach of 78.561% and 69.777% using SO and AV leaves, respectively, for Cu<sup>2+</sup> ion removal. Cu<sup>2+</sup> ions have been removed by adsorption using potato peels; the maximum removal was at pH 6 and reached 73%; at values higher than pH 6, the maximum removal value decreased (El-Maghraby, 2019).

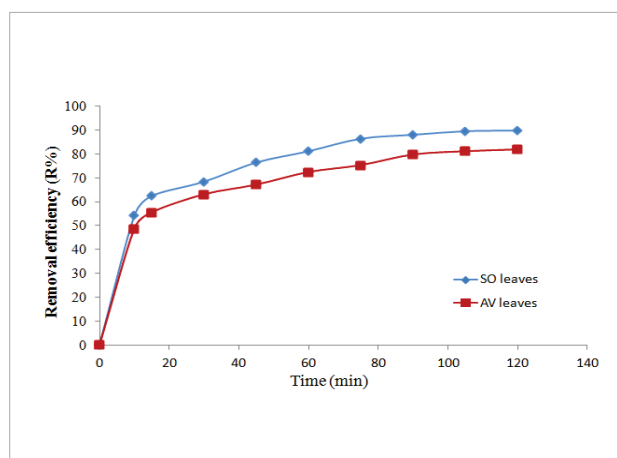
**Fig. 8.** Impact of pH values on the Cu<sup>2+</sup> ions removal using SO and AV leaves



### Impact of contact time

The influence of contact time on the Cu<sup>2+</sup> ion adsorption by SO and AV leaves was assessed using a variety of time intervals that ranged from 0 to 120 minutes with 50 mg/L of Cu<sup>2+</sup> ions in a solution of pH 6, a concentration of 5 g/L for SO and AV leaves, a particle size of 300 µm, a stirring speed of 150 rpm at 25 ± 5°C temperature to obtain the best results for the adsorption time. *Fig. 9* depicts the relation between the quantity of metal ions removed and contact time. As shown, the removal rate of Cu<sup>2+</sup> ions increased with prolonged contact time, reaching equilibrium at 75 min for SO leaves and 90 min for AV leaves. This is due to the presence of abundant active sites on the sorbent surfaces, facilitating metal ion sorption. Subsequently, the sorption rate slowed down due to the depletion of vacant surface sites (Akpomie et al., 2015). Based on these findings, the best contact time for the experiments was calculated to be 75 min with a maximum removal efficacy of 86.346% for SO leaves and 90 min with a maximum removal efficiency of 79.694% for AV leaves.

**Fig. 9.** Impact of contact time on the  $\text{Cu}^{2+}$  ions removal using SO and AV leaves



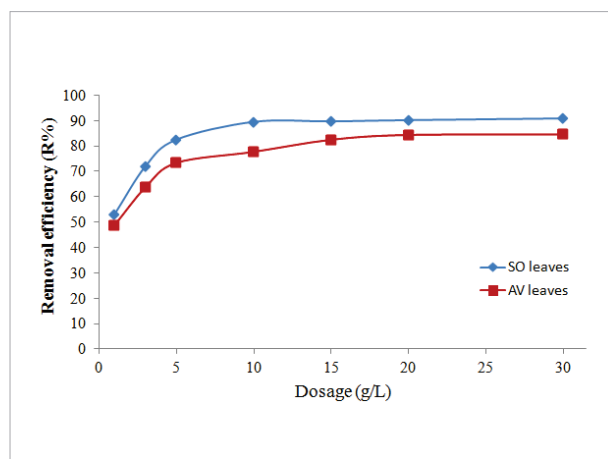
The contact time reached 60 min when industrial wastewater polluted with lead, copper, and cobalt ions was treated by *Solamen Vaillantii* snail shell as a biosorbent; the maximum removals for lead, copper, and cobalt ions were 86%, 83%, and 79%, respectively (Esmailia et al., 2020). The higher removal rate of copper ions by adsorption onto cassava leaves (*Manihotesculenta* Crantz), mangosteen peels (*Garciniamangostana* L.), and Thai copper pod leaves (*Sennasiamea*), respectively, was 49%, 35%, and 25% at a contact time of 120 minutes (Paksamut and Boonsong, 2018).

### Impact of Adsorbent Dosage

To assess the relation between the  $\text{Cu}^{2+}$  ion adsorption and the quantity of adsorbent, the amounts of SO and AV leaves were varied from 1 to 30 g/L with a 50 mg/L  $\text{Cu}^{2+}$  ion solution at a pH of 6, a contact time of 75 min for SO leaves and 90 min for AV leaves, an agitation rate of 150 rpm, a 300  $\mu\text{m}$  particle size and a temperature of  $25 \pm 5^\circ\text{C}$ . As per the observations, the efficiency of removing the adsorbate generally rose with an increase in the dosage of adsorbent materials. According to Fig. 10, the highest attainable removal efficiency of  $\text{Cu}^{2+}$  ions was recorded at 89.341%, achieved with 10 g/L of SO leaves, whereas with 15 g/L of AV leaves, it was 82.454%. This behaviour can be explained due to the reality that the adsorbent surface areas increase as the adsorbent mass increases, in addition to the more effective site availability for adsorption. However, beyond that, the adsorption capacity plateaued due to a blockage effect occurring between the adsorbent

particles (Mohsen et al., 2023). Yirga et al. (2022) studied the  $\text{Cu}(\text{II})$  adsorption onto dried orange peels; the maximum removal recorded was 85% at 25 g/L adsorbent dosage. Moreover, the same results were observed when cotton stalk was used to remove  $\text{Cu}^{2+}$  ions from an aqueous solution; the rate of removal increased from 32.580% to 54.950% when the cotton stalk dose increased from 0.150 to 0.600 g/100mL (Cabarcas et al., 2024).

**Fig. 10.** Impact of biosorbent dosage on the  $\text{Cu}^{2+}$  ions removal using SO and AV leaves

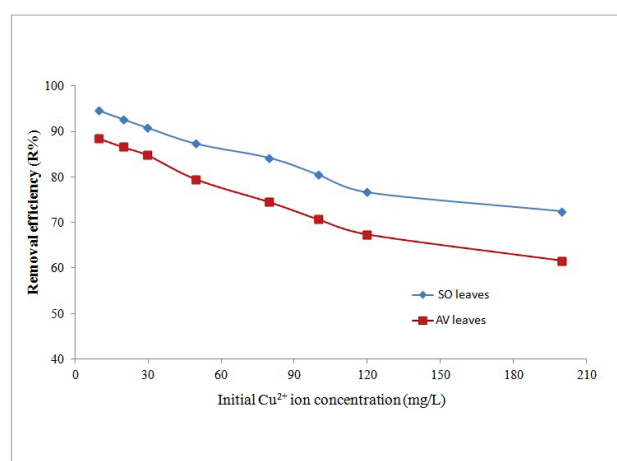


### Impact of the initial $\text{Cu}^{2+}$ ion concentration

To investigate the impact of initial  $\text{Cu}^{2+}$  ion concentration on removal efficiency, experiments were conducted with varying initial  $\text{Cu}^{2+}$  ion concentrations, ranging from 10 to 200 mg/L, maintaining a pH of 6, with SO and AV leaves used at dosages of 10 g/L and 15 g/L, respectively, and contact times of 75 min for SO leaves and 90 min for AV leaves. The mixture was stirred at a rate of 150 rpm, a particle size 300  $\mu\text{m}$  and a temperature  $25 \pm 5^\circ\text{C}$ . According to the findings illustrated in Fig. 11, the  $\text{Cu}^{2+}$  ions removal percentage decreased as the initial concentration of  $\text{Cu}^{2+}$  ions increased for both SO and AV leaves. This may be because of the limited availability of sorption sites on the surface area of the adsorbent materials; only a limited amount of  $\text{Cu}^{2+}$  ions can be absorbed (Kumar et al., 2019). The ratio between the initial number of metal ions and the available adsorption sites was observed to increase with higher metal ion concentrations, leading to a lower sorption percentage (Carreira et al., 2023). Conversely,

at low initial concentrations, the removal efficiency of  $\text{Cu}^{2+}$  ions increased, reaching its peak because of the abundance of sorption sites available on the adsorbent's surface area (Mohammed et al., 2016) with the maximum removal reaching 94.545% and 88.466% for SO and AV leaves, respectively, at the initial  $\text{Cu}^{2+}$  ion concentration of 10 mg/L. The maximum  $\text{Cu}^{2+}$  ion removal was 98.780% at an initial  $\text{Cu}^{2+}$  ion concentration of 20 mg/L using mallet flower leaf powder (MFLP) as a biosorbent (Padma and Sastry, 2021).

**Fig. 11.** Impact of initial  $\text{Cu}^{2+}$  ion concentration on the  $\text{Cu}^{2+}$  ion removal using SO and AV leaves

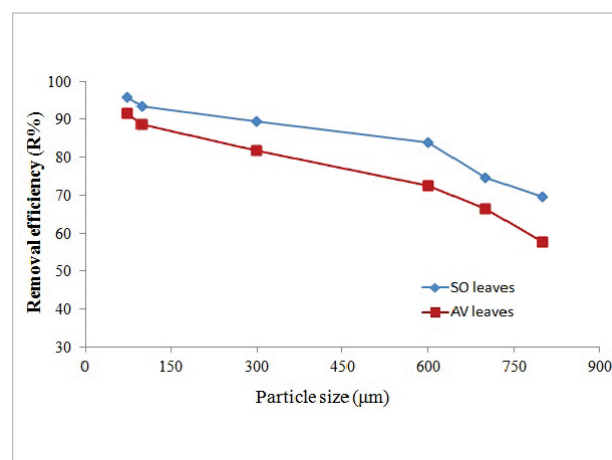


### Impact of particle size

The particle size plays a worthy role in the adsorption study. If the particle size is smaller, the surface area is larger, and therefore, the removal efficiency is high. Sieve analysis was carried out, and various particle sizes of SO and AV leaves ranging from 75 to 800  $\mu\text{m}$  were investigated using a 10 g/L dosage for SO leaves with a contact time of 75 min and for AV leaves using a 15 g/L dosage with a 90 min contact time. The experiments were done at a pH of 6, with an initial metal ion concentration of 10 mg/L, a shaking speed of 150 rpm, and a  $25 \pm 5^\circ\text{C}$  temperature. Fig. 12 demonstrates that the percentage of adsorption increased with the reduction in the biosorbent particle size. This resulted from a larger surface area available for bulk adsorption/unit mass of the biosorbent and the greater accessibility to the pores (Rukayat et al., 2021). The higher removal percentages at a particle size of 75  $\mu\text{m}$  were 95.864% and 91.566% for SO and AV leaves, respectively, as can

be seen in Fig. 12. The same trend was also seen on the  $\text{Cu}^{2+}$  ion adsorption using powdered orange peel (POP) and powdered activated carbon (PAC); the maximum percentage removal was 78% using POP with a particle size less than 0.075 mm, and it was 91.300% using PAC with a particle size less than 75  $\mu\text{m}$  (Malkhede and Rao, 2021).

**Fig. 12.** Impact of particle size on the  $\text{Cu}^{2+}$  ions removal utilizing SO and AV leaves



### Adsorption Isotherms Study

Figs. 13 (a and b) present the  $\text{Cu}^{2+}$  ion adsorption according to Langmuir and Freundlich isotherms on adsorbent materials (SO and AV leaves) with optimal parameters. Table 4 provides all parameters and the correlation coefficient ( $R^2$ ) values. The  $R^2$  levels gained from the Langmuir model were 0.919 and 0.934, while the  $R^2$  levels gained from the Freundlich model were 0.997 and 0.993 for  $\text{Cu}^{2+}$  ion removal using SO and AV leaves, respectively. Consequently, the data did not fit well with the Langmuir isotherm, as evidenced by relatively lower  $R^2$  compared with the Freundlich isotherm model, showing that the Freundlich isotherm model was a better agreement for  $\text{Cu}^{2+}$  ion adsorption. This shows that the adsorption is heterogeneous and occurs on multilayer adsorption surfaces (Kamaraj et al., 2017). According to the  $R_L$  values obtained, 0.242 and 0.342 for SO and AV leaves, respectively, in the range ( $0 < R_L < 1$ ), the adsorption procedure of  $\text{Cu}^{2+}$  ion is favourable. For the Freundlich isotherm model, adsorption intensities ( $1/n$ ) were 0.578 and 0.608 for SO and AV leaves, respectively, indicating favourable  $\text{Cu}^{2+}$  ion adsorption.

**Table 4.** The Langmuir and the Freundlich models' parameters of  $\text{Cu}^{2+}$  ion adsorption using SO and AV leaves

| Adsorbents | Langmuir     |              |       |       | Freundlich |       |       |
|------------|--------------|--------------|-------|-------|------------|-------|-------|
|            | $q_m$ (mg/g) | $K_L$ (L/mg) | $R_L$ | $R^2$ | $K_f$      | $1/n$ | $R^2$ |
| SO leaves  | 16.949       | 0.063        | 0.242 | 0.919 | 1.443      | 0.578 | 0.997 |
| AV leaves  | 10.101       | 0.039        | 0.342 | 0.934 | 1.639      | 0.608 | 0.993 |

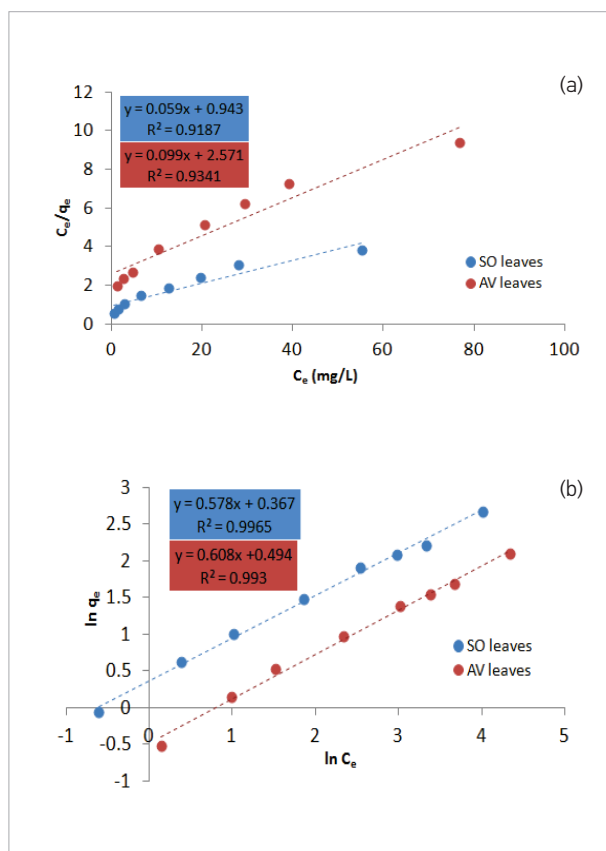
Some research has been conducted to identify the  $\text{Cu}^{2+}$  ion binding properties of various adsorbent materials. As illustrated in Table 5, several adsorbents exhibit effective adsorption of  $\text{Cu}^{2+}$  ions. SO and AV leaves possess good to moderate adsorption capacities when compared with other materials. This could be due to the abundance of active groups in SO and AV leaves, which enhance the adsorption process.

**Table 5.** Comparison of the adsorption capacities of several adsorbents for  $\text{Cu}^{2+}$  ions

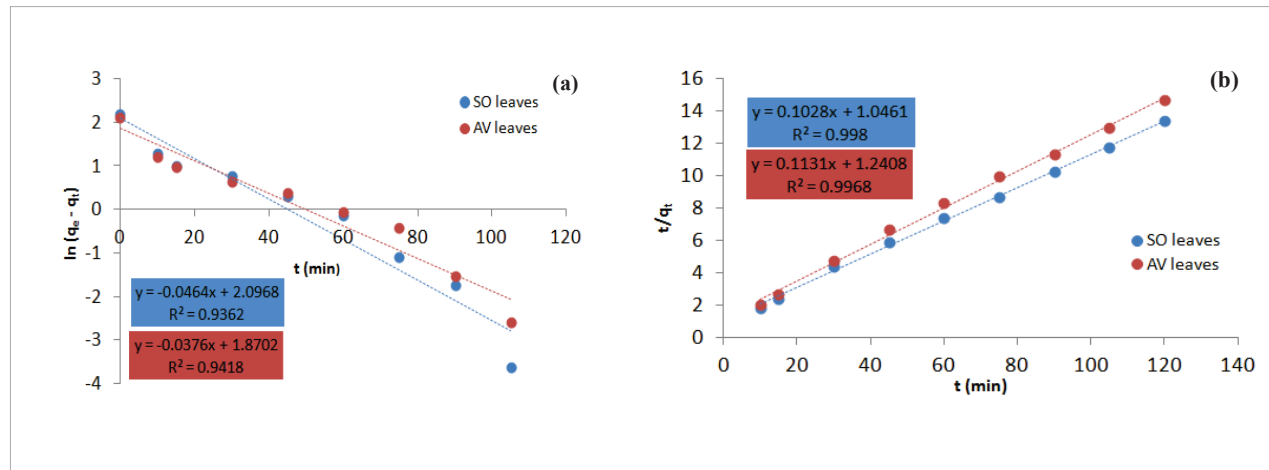
| Adsorbents                 | $q_m$ (mg/g) | Reference               |
|----------------------------|--------------|-------------------------|
| Orange peels               | 38.180       | Afolabi et al., 2022    |
| Tea leaves                 | 71.200       | Shrestha et al., 2016   |
| Banana leaves              | 48.700       | Darweesh et al., 2022   |
| <i>Lemna minor</i>         | 2.860        | Al-Baldawi et al., 2022 |
| <i>Azolla filiculoides</i> | 1.490        | Al-Baldawi et al., 2022 |
| Arborvitae leaves          | 7.940        | Shi et al., 2016        |
| Cabbage leaves             | 5.490        | Kamar et al., 2016      |
| Banana peel                | 1.940        | Ince et al., 2017       |
| Chestnut shell             | 2.250        | Ince et al., 2017       |
| Tea leaf waste             | 3.360        | Ince et al., 2017       |
| SO leaves                  | 16.949       | Present study           |
| AV leaves                  | 10.101       | Present study           |

### Adsorption kinetics study

The kinetic models of  $\text{Cu}^{2+}$  ion sorption onto SO and AV leaves using pseudo-second-order and pseudo-first-order models were examined, as illustrated in Figs. 14 (a and b) and Table 6. Across all adsorbents, the pseudo-second-order kinetic model demonstrated the best agreement with experimental data compared with the pseudo-first-order kinetic models. The data illustrated that  $R^2$  for the pseudo-second-order kinetic model for  $\text{Cu}^{2+}$  ion adsorption was 0.998 and 0.996 onto SO and AV leaves, respectively, while  $R^2$

**Fig. 13.** Adsorption isotherm models for the  $\text{Cu}^{2+}$  ion removal onto SO and AV leaves: (a) Langmuir adsorption isotherm and (b) Freundlich adsorption isotherm

for pseudo-first-order for the  $\text{Cu}^{2+}$  ion adsorption was 0.936 and 0.941 onto SO and AV leaves, respectively, and the  $q_e$  experimental value closely matched the  $q_e$  calculated value from the pseudo-second-order model, indicating the chemisorption involvement in the sorption procedure. This suggests that chemical interactions likely play a significant role, leading to ions being bound to the adsorbent material surface through bonding as strong as covalent binding (Afolabi et al., 2022).

**Fig. 14** Adsorption kinetic models of  $\text{Cu}^{2+}$  ion sorption using SO and AV leaves: (a) pseudo-second-order and (b) pseudo-second-order**Table 6.** Parameters of kinetic models (pseudo-first order and pseudo-second order) of  $\text{Cu}^{2+}$  ions sorption using SO and AV leaves

| Adsorbents | $q_e$ (exp.) (mg/g) | Pseudo-first-order |                     |       | Pseudo-second-order |                     |       |
|------------|---------------------|--------------------|---------------------|-------|---------------------|---------------------|-------|
|            |                     | $K_1$ (1/min)      | $q_e$ (cal.) (mg/g) | $R^2$ | $K_2$ (1/min)       | $q_e$ (cal.) (mg/g) | $R^2$ |
| SO leaves  | 8.974               | 0.046              | 8.134               | 0.936 | 0.009               | 9.804               | 0.998 |
| AV leaves  | 8.183               | 0.037              | 6.488               | 0.941 | 0.010               | 8.849               | 0.996 |

## Conclusions and Recommendations

The  $\text{Cu}^{2+}$  ion adsorption from an aqueous solution using SO and AV leaves as natural adsorbents was experimentally studied in batch mode. Different operating parameters, such as pH, duration time, ground leaves dosage, initial metal ion concentration, and particle size, were assessed. Throughout the experimental runs, SO leaves consistently had an adsorption capacity higher than AV leaves. The highest  $\text{Cu}^{2+}$  ion adsorption occurs at a pH of 6. The removal percentage of  $\text{Cu}^{2+}$  ions increased with the increasing adsorbent dosage and also increased as the contact time increased, reaching equilibrium for SO and AV leaves at 75 min and 90 min, respectively. The  $\text{Cu}^{2+}$  ion removal rate decreased with the increasing initial  $\text{Cu}^{2+}$  ion concentration and particle size. The  $\text{Cu}^{2+}$  ion removal rate reached 95.864% using SO leaves and 91.566% using AV leaves at optimum conditions. The FTIR analysis showed multiple functional groups, including C-H, O-H, and C-O. BET analyses revealed that the measured surface area of SO and AV leaves was  $6.004 \text{ m}^2/\text{g}$  and  $5.395 \text{ m}^2/\text{g}$ , respectively. The SEM images depicted many porous

and convoluted areas on the adsorbent surface. The Freundlich and pseudo-second-order models fit well the equilibrium and kinetic experimental results. This means that adsorption occurred in multilayers and that chemisorption was high during the adsorption process. This study suggests that SO and AV leaves are efficient adsorbents suitable for the  $\text{Cu}^{2+}$  ion removal from an aqueous solution. Finally, the following are recommendations for future research: (i) studying the capability of using SO and AV leaves biomass for removing organic pollutants from wastewater, (ii) conducting future research to evaluate the influence of ionic strength on the behaviour of the solution under different experimental conditions, (iii) studying different configurations for column studies, and (iv) investigating the desorption behaviour of  $\text{Cu}^{2+}$  ions and the reusability or regeneration capabilities of SO and AV leaves over multiple adsorption-desorption cycles.

## Acknowledgements

The author acknowledges Mustansiriyah University ([www.uomustansiriyah.edu.iq](http://www.uomustansiriyah.edu.iq)) Baghdad, Iraq, for its support in the present work.

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