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Jonas, Bayuo

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# Adsorption and desorption ability of divalent mercury from an interactive bicomponent sorption system using hybrid granular activated carbon

Jonas Bayuo · Mwemezi J. Rwiza ·  
Kelvin Mark Mtei

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**Abstract** The sequestration of heavy metals from multicomponent sorption media has become critical due to the noxious effects of heavy metals on the natural environment and subsequently on human health as well as all life forms. The abatement of heavy metals using bio-adsorbents is one of the efficient and affordable approaches for treating water and wastewater. Therefore, the interactive effect of arsenic [As(III)] ions on the sorption and desorption ability of mercury [Hg(II)] from a binary sorption system was conducted. More so, the impact of reaction time, solution pH, bio-adsorbent particle size, bio-adsorbent dose, initial mono-metal, and binary-metal concentration as well as reaction temperature on the individual and competitive sorption of Hg(II) was explored. The study showed that Hg(II) could be removed effectively from the single-component system and competitively from the aqueous phases by the bio-adsorbent in the coexistence of As(III) species in

the bicomponent medium. The adsorptive detoxification of Hg(II) from the monocomponent and bicomponent sorption media showed dependence on all the studied adsorption parameters. The occurrence of As(III) species in the bicomponent sorption medium affected the decontamination of Hg(II) by the bio-adsorbent and the major interactive mechanism was found to be antagonism. The spent bio-adsorbent was effectively recycled using 0.10 M nitric (HNO<sub>3</sub>) and hydrochloric (HCl) acids solutions and the multi-regeneration cycles showed a high removal efficiency in each cycle. The first regeneration cycle was found to have the highest Hg(II) ions removal efficiencies of 92.31 and 86.88% for the monocomponent and bicomponent systems, respectively. Thus, the bio-adsorbent was found to be mechanically stable and reusable up to the 6.00 regeneration cycle. Therefore, this study concludes that the bio-adsorbent not only has a higher adsorption capacity but also a good recycling performance pointing to good industrial applications and economic prospects.

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J. Bayuo (✉) · M. J. Rwiza · K. M. Mtei  
School of Materials, Energy, Water, and Environmental  
Sciences (MEWES), The Nelson Mandela African  
Institution of Science and Technology (NM-AIST), P.O.  
Box 447, Arusha, Tanzania  
e-mail: bayuoj@nm-aist.ac.tz; jbayuo@cktutas.edu.gh

J. Bayuo  
School of Science, Mathematics and Technology  
Education (SoSMTE), Department of Science Education,  
C. K. Tedam University of Technology and Applied  
Sciences (CKT-UTAS), Postal Box 24, Navrongo, Upper  
East Region, Ghana

**Keywords** Adsorption · Desorption · Heavy metal · Sorption system · Wastewater

## Introduction

Heavy metals are harmful contaminants that exist in aquatic environments due to human activities such as industrialization, urbanization, technological

advancement, and agriculture. Metal plating, mining, metal smelters, alloying, storage batteries, plastics, wood preservatives, and textiles manufacturing as well as agricultural sources where fertilizers, pesticides, and fungicidal spray are heavily utilized contribute to heavy metals pollution in the aqueous systems (El-Bouhy et al., 2021). Due to their inability to degrade or be destroyed, these hazardous heavy metals persist in the ecosystem and cause serious health problems to all living things. These toxic metal ions constantly bioaccumulate in the bodies of all lifeforms through the food web (Dawodu et al., 2020). Different from organic contaminants, heavy metals are unable to break down chemically or biologically into less harmful forms. However, they could only undergo transformation to become less harmful species. For instance, highly toxic Hg(II) is usually converted to a less toxic Hg(0) species (Priyadarshane & Das, 2021), As(III) to As(V) (Biswas & Sarkar, 2019), and Cr(VI) to Cr(III) (Mohamed et al., 2020). When excessive heavy metal ions accumulate in biological cells, they can result in cancer and cause harm to the brain, kidney, liver, and reproductive and neurological systems, which can lead to death (Ali et al., 2019).

As emphasized by the Agency for Toxic Substances and Disease Registry, divalent mercury [Hg(II)] and trivalent arsenic [As(III)] are among the “First Top Hazardous Substances” and are required to be reduced to an acceptable level when present in aqueous solutions due to their detrimental consequences on the ecosystem, which comprises humans, animals, and all other living species (Wang et al., 2021). The sequestration of heavy metals from aquatic environments has been explored using a variety of approaches including adsorption, ion exchange electrocoagulation, membrane filtration, and oxidation (Naga et al., 2021). Due to its high removal performance and related flexible operating characteristics, the adsorption technique has become the most attractive remediation approach for the depollution of toxic heavy metals from aquatic systems (Tekin & Unsal, 2022).

Mercury [Hg(II)] is a lethal heavy metal, and when ingested into the body through the food web, it bioaccumulates in the tissues of living organisms and causes various organ malfunctions (Abou Taleb et al., 2021). Therefore, the amount of Hg(II) that is permitted in industrial effluent is strictly controlled, while the ability of bio-adsorbents derived from agro-based

waste to adsorb Hg(II) ions is still limited. Several possible sources of inexpensive agro-based bio-adsorbents are presently being explored to see how well they perform in removing Hg(II) ions from single-component adsorption systems (Abou Taleb et al., 2021; Egirani et al., 2021; El-Bouhy et al., 2021). However, very few studies are dedicated to binary and multi-metal adsorption, which is holistic and practical to study since the existence of one metal species in wastewater is unfeasible.

To the best of our knowledge, the competitive adsorptive removal of Hg(II) ions in the coexistence of As(III) species using a bio-adsorbent produced from different parts of a single agricultural product has not been reported yet. Therefore, the present study provides quantitative data relating to the sorption and desorption of Hg(II) ions from binary sorption systems in the company of competitive As(III) ions using hybrid granular activated carbon produced from maize plant biomass. More so, the impact of reaction time, bio-adsorbent dose, solution pH, bio-adsorbent particle size, initial mono- and binary-metal concentration, and reaction temperature on the individual and competitive sorption of Hg(II) was studied. In Tanzania, huge numbers of maize cobs, stalks, and tassels are produced throughout the season posing environmental nuisance and disposal issues. Hence, the application of these maize plant biomass as bio-adsorbents provides a cost-effective technological solution to heavy metal contamination and disposal challenges.

## Materials and methods

### Bio-adsorbent preparation

Maize plant wastes including cobs, stalks, and tassels were sampled from farmers in Nambala in the Arusha Region, Tanzania. To get rid of the dust and other soluble impurities, these agricultural wastes were thoroughly treated with distilled water. The washed carbon precursors were then dried at room temperature (23 °C) for 1 week and later dried again in an oven at 105 °C for 6 h to attain constant weight. The dried cobs, stalks, and tassels were crushed and ground into powder separately. The powdered residues were then sieved, measured equally, and mixed homogeneously. The homogeneous powdered mixed maize

wastes were chemically treated with H<sub>3</sub>PO<sub>4</sub> acid into hybrid granular activated carbon (HGAC) following the steps documented by Bayuo et al. (2023). The produced HGAC was then sieved with standard steel sieves in the size range of 90–450 μm and applied as a bio-adsorbent in the batch mode sorption studies.

#### Preparation of single-metal and binary-metal stock solutions

All of the reagents and chemicals meant for the laboratory experimentations were of analytical grade and were acquired from Alpha Chemika in Mumbai, India, and utilized exactly as they were supplied. These reagents and chemicals include the following: phosphoric acid (H<sub>3</sub>PO<sub>4</sub>, 85%), mercuric chloride (HgCl<sub>2</sub>, 99%), sodium arsenite (NaAsO<sub>2</sub>, 98%), starch, hydrochloric acid (HCl, 37%), sodium borohydride (NaBH<sub>4</sub>), nitric acid (HNO<sub>3</sub>, 68%), sodium hydroxide (NaOH, 97%), sulphuric acid (H<sub>2</sub>SO<sub>4</sub>, 98%), ethylenediaminetetraacetic acid (EDTA, 99%), deionized and distilled water.

The single and binary sorption experiments were performed using analytical grade solid mercuric chloride (HgCl<sub>2</sub>) and sodium arsenite (NaAsO<sub>2</sub>) in preparing the synthetic wastewater of mercury and arsenic. In the preparation of the mono- and binary-metal synthetic wastewater of mercury and arsenic, respectively, procedures specified by Bayuo et al. (2023) were duly followed.

#### Batch adsorption experiments

The mono-metal [Hg(II)] and binary-metal [Hg(II)+As(III)] sorption experiments were conducted in batch systems. For both the sorption systems including the mono- and bicomponent sorption media, the removal rate and uptake rate were examined as a function of varying reaction time, solution pH, bio-adsorbent particle size, bio-adsorbent dose, initial mono- and binary-solutes concentration, and reaction temperature on the individual and competitive sorption of Hg(II). The required pH of non-competitive and competitive solutions was maintained by applying 0.1 M NaOH and HCl solutions, respectively. Working solutions of 100 mL in 250-mL flasks served as the single and binary sorption media for both the single and interactive depollution

of Hg(II) ions from the liquid phase using the HGAC. The Erlenmeyer flasks were positioned in a rotatory flask shaker that was set to agitate at a consistent rate of 120 rpm with the tops covered to prevent the solution from evaporation. After reaching equilibrium conditions, the residuals were then collected from the flasks and analyzed by a cold vapour atomic absorption spectrophotometer (CVAAS) to determine the quantity of Hg(II) ions eliminated from the mono-solute and bi-solute sorption media as well as its uptake capacity by the bio-adsorbent using Eqs. (1–3), respectively (Pasgar et al., 2022; Tho et al., 2021). To ensure the authenticity and dependability of the experimental measurements, each analysis was performed thrice and the average values were recorded.

$$\text{Removal efficiency (\%)} = \left( \frac{C_0 - C_e}{C_0} \right) \times 100 \quad (1)$$

The sorption rate (q<sub>t</sub>) of Hg(II) ions by HGAC at a certain time is expressed by Eq. (2):

$$q_t = \left( \frac{C_0 - C_t}{W} \right) \times V \quad (2)$$

Besides, the sorption capacity (q<sub>e</sub>) at the point when equilibrium is established is given in Eq. (3):

$$q_e = \left( \frac{C_0 - C_e}{W} \right) \times V \quad (3)$$

where C<sub>0</sub> and C<sub>e</sub> are initial Hg(II) and equilibrium concentration, and q<sub>t</sub> and q<sub>e</sub> denote Hg(II) ions adsorbed per bio-adsorbent load at specific and equilibrium times, correspondingly. W and V are the bio-adsorbent weight (g) and solution volume (L), correspondingly.

#### Competitive adsorption of heavy metals

In multi-solute systems, competitive adsorptive removal of heavy metals typically takes place, and three primary types of interactions are feasible including synergism, antagonism, and no interaction (Anna et al., 2015; de Morais França et al., 2021). The interaction among diverse metal species in the bicomponent sorption medium was determined by considering the ratio of the sorption rate in the bicomponent sorption medium to the sorption rate in the monocomponent medium as expressed in Eq. (4) (Shivangi et al., 2022).

$$R_{Q_e} = \frac{Q_{e, \text{Binary system}}}{Q_{e, \text{Single system}}} \quad (4)$$

where  $R_{Q_e}$  is the ratio of the sorption rate,  $Q_{e, \text{Single}}$  is the maximum sorption capacity of the monocomponent medium, and  $Q_{e, \text{Binary}}$  is the maximum sorption rate of the bicomponent medium.

When  $R_{Q_e} = \frac{Q_{e, \text{Binary}}}{Q_{e, \text{Single}}} > 1$ , then the adsorptive reduction of the specific water contaminant is enhanced by the occurrence of other competing cations or anions in the bicomponent sorption medium, indicating synergism (Touihri et al., 2021).

Also, when  $R_{Q_e} = \frac{Q_{e, \text{Binary}}}{Q_{e, \text{Single}}} = 1$ , then the occurrence of other ionic species does not alter the decontamination of the targeted heavy metal from the binary system, indicating non-interaction (Du et al., 2021).

However, when  $R_{Q_e} = \frac{Q_{e, \text{Binary}}}{Q_{e, \text{Single}}} < 1$ , then the presence of other competing ionic species inhibits the decontamination of a specific heavy metal from the binary system, implying antagonism (Sellaoui et al., 2018).

Subsequently, Eq. (5) was used to determine the rate of adsorption reduction ( $\Delta\gamma$ ) after the identification of the category of interactions among the ionic species and bio-adsorbent present in the bicomponent sorption medium (Liu & Lian, 2019).

$$\Delta\gamma = \frac{Q_{e, \text{Single}} - Q_{e, \text{Binary}}}{Q_{e, \text{Binary}}} \times 100 \quad (5)$$

### Selectivity

The selectivity of the HGAC for Hg(II) ions over As(III) ions was determined by agitating 0.5 g of the bio-adsorbent with 100 mL solution containing binary-metal [Hg(II)-As(III)] ions with varied initial concentrations of 5–100 mg/L at pH of 4.0 and temperature of 298 K. After agitating at 120 rpm for a predetermined contact time of 120 min, the concentration of metal ions in the supernatant was determined by the CVAAS. Then, the ratio of the quantity of metal ions adsorbed on the HGAC to the equilibrium concentrations of metal ions in the liquid phase for both the mono- and bicomponent systems was calculated. Subsequently, the selectivity factor (SF) was calculated in the binary sorption systems at different initial concentrations using Eq. (6) (An et al., 2019).

$$SF = \frac{\left[\frac{Q}{C_e}\right]_a}{\left[\frac{Q}{C_e}\right]_b} \quad (6)$$

where  $Q$  is the concentration of adsorbate in bio-adsorbent (mg/g),  $C_e$  is the adsorbate concentration at equilibrium (mg/L), and subscripts  $a$  and  $b$  represent monocomponent and binary component systems, respectively.

## Results and discussion

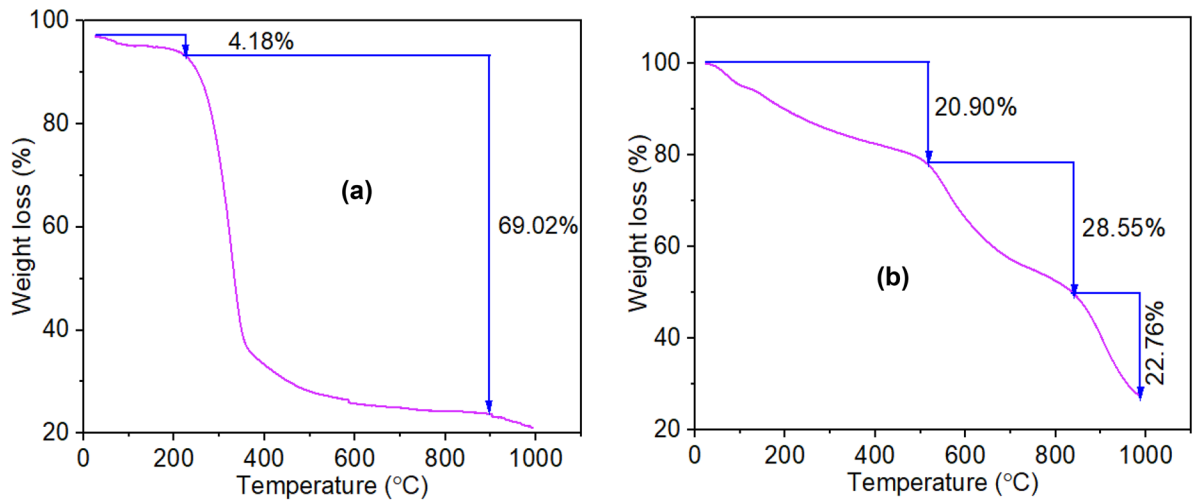
### Bio-adsorbent physicochemical properties and characterization

The physicochemical properties of the bio-adsorbent including moisture content, ash content, bio-adsorbent pH, fixed carbon content, point of zero charge pH, volatile matter, yield of carbon, iodine number, and bulk density as well as the BET specific surface area, pore volume, and the surface functional groups were determined in our previous study (Bayuo et al., 2023). In the current study, the characterization of the bio-adsorbent using thermogravimetric analysis (TGA), x-ray fluorescence spectrometry (XRF), and scanning electron microscopy (SEM) was investigated.

### Thermogravimetric analysis (TGA)

The thermogravimetric analysis (TGA) makes it feasible to discover potential weight or constituent losses due to temperature action while also determining the sorbent material stability throughout a range of temperatures. Figure 1(a) and (b) represent the TGA curves of the thermal degradation process of the carbon precursors attained at 10.00 °C min<sup>-1</sup> heating rate from 25 to 1000 °C in an inert (nitrogen) environment to prevent combustion.

Figure 1(a), which depicts the unmodified carbon precursor, demonstrates that the TGA took place in two steps. The first stage, which occurred between 25 and 230 °C, had a weight loss of 4.18% that was attributable to water loss in the carbon precursor. The second stage, which took place between 230 and 900 °C and resulted in 69.02% weight loss, was attributable to lignocellulose degradation. However, the TGA curve of the activated carbon presented in Fig. 1 (b)



**Fig. 1** TGA of the raw carbon precursor (a) and the hybrid granular activated carbon (b)

can be divided into three stages of weight losses in the temperature ranges of 50–500 °C, 500–830 °C, and 830–975 °C, respectively. The first weight loss of 20.90% at the temperature range of 50–500 °C is due to the desorption of physically adsorbed and interlamellar water molecules as well as the devolatilization of organic matter from the activated carbon (Zhao et al., 2019). In phase 2, the weight loss of 28.55% from 500 to 830 °C is a result of the degradation of hemicellulose, cellulose, lignin, and surface functional groups. The third weight loss of 22.76% which occurred at 830–975 °C is attributed to the disappearance of the carbon skeleton. More importantly, it was found that there is an increase in weight loss starting at 500–975 °C suggesting the existence of carbonyls, basic, and phenols functional groups in the hybrid activated carbon (Labied et al., 2018). The TGA results showed that the hybrid granular activated carbon is thermally stable and can be applied as a possible bio-adsorbent for the decontamination of Hg(II) ions from aquatic environments.

*Scanning electron microscopy (SEM)*

The SEM was utilized to study the morphological features and surface properties of the HGAC employed in the elimination of Hg(II) from the individual and bicomponent sorption systems. The SEM micrographs of the HGAC were determined at a scale of 50 μm before and after the abatement of the Hg(II) ions

from both sorption systems as presented in Fig. 2(a, b, and c), respectively.

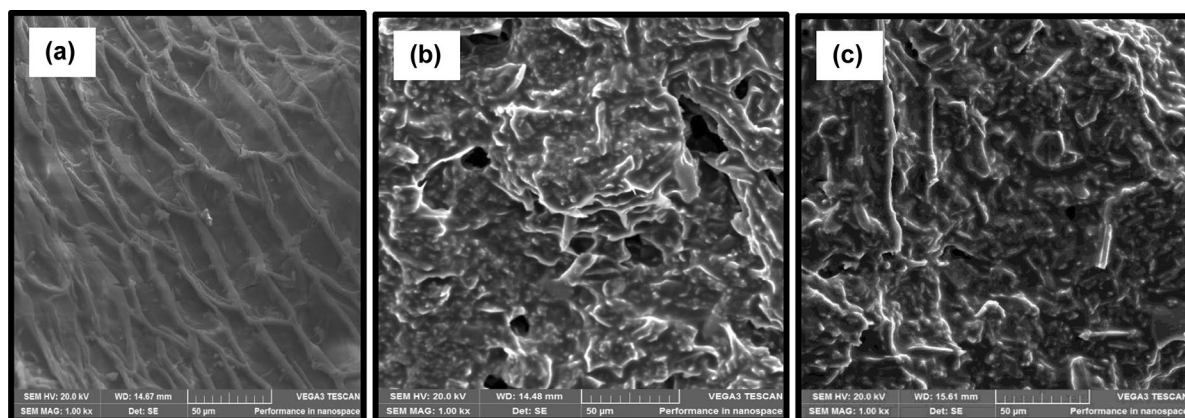
The image of the HGAC before the sorption of Hg(II) is presented in Fig. 2(a). In Fig. 2(a), the rough microstructure, intergranular pores, and presence of numerous voids, and fissures in the SEM image may make it easier for Hg(II) ions to interact with the bio-adsorbent surface, resulting in the efficient elimination of Hg(II) ions from both sorption systems. The micro- and mesopores development in the HGAC was triggered by the volatilization of various constituents including hemicellulose, cellulose, lignin, and other organic components found in the maize residues during the impregnation and activation process of the carbon precursor using H<sub>3</sub>PO<sub>4</sub>.

The disappearance of the cavities or the pores from the HGAC surface is an indication that the depollution of Hg(II) ions from the mono- and bicomponent sorption media have taken place effectively as displayed in Fig. 2(b) and (c), correspondingly.

*X-ray fluorescence spectrometry (XRF)*

The XRF results of the HGAC before and after the elimination of the Hg(II) ions from the mono- and bicomponent media are presented in Table 1. In Table 1, the elemental compositions that were found in the pristine bio-adsorbent are silicon (Si), calcium (Ca), potassium (K), sulphur (S), and magnesium (Mg). However, upon the activation process, the





**Fig. 2** SEM images of the HGAC before (a) and after the elimination of Hg(II) ions from monocomponent (b) and bicomponent (c) systems

**Table 1** Elemental compositions of the pristine bio-adsorbent, activated carbon before and after monocomponent [Hg(II)] and bicomponent [Hg(II)+As(III)] adsorption

Element	Percentage composition (wt. %)			
	Pristine bio-adsorbent	Activated carbon	Activated carbon loaded with Hg(II)	Activated carbon loaded with Hg(II) + As(III)
Calcium (Ca)	0.30	0.01	0.00	0.00
Potassium (K)	0.06	0.00	0.00	0.00
Magnesium (Mg)	0.01	0.00	0.00	0.00
Silicon (Si)	26.29	32.70	19.25	15.72
Sulphur (S)	0.15	0.01	0.00	0.00
Phosphorous (P)	0.00	24.86	24.81	22.37
Mercury (Hg)	0.00	0.00	15.11	13.44
Arsenic (As)	0.00	0.00	0.00	10.98

element phosphorus (P) was found to be present in the HGAC including the other elements with 24.86 wt% compositions. The percentage compositions of the silicon and calcium were discovered to be higher than those of the other elements, with respective values of 26.29 wt% and 0.30 wt%. Due to absorption from the soil, the carbon precursor has a high silicon and calcium concentration (Labied et al., 2018). After the sorption processes, the percentage compositions of the elements in the HGAC were found to reduce while new elements appeared including mercury (Hg) and arsenic (As), which is indicative of the decontamination of Hg(II) ions from the mono- and bicomponent sorption systems in the presence of the competing As(III) ions.

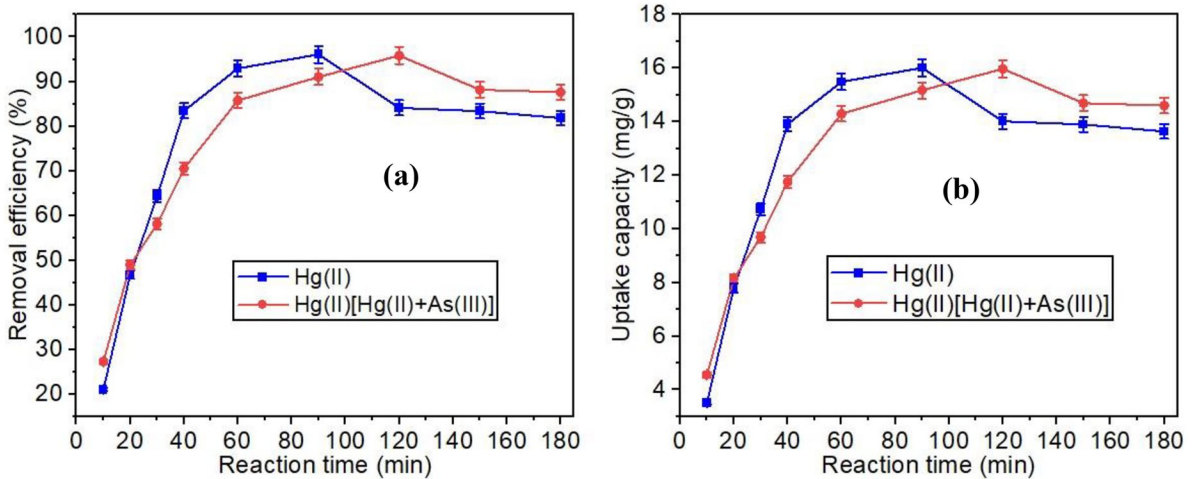
## Influencing adsorption factors

### *Effect of reaction time*

The abatement of Hg(II) ions from the mono- and bicomponent sorption systems was carried out at varied contact times ranging from 10 to 180 min and at initial mono-metal [Hg(II)] and binary-metal [Hg(II)+As(III)] concentration of 25 mg/L in flasks containing 100 mL solutions, while other operating parameters including agitation speed (120 rpm), pH (6), bio-adsorbent particle size (90 μm), bio-adsorbent dose (1.5 g/L), and reaction temperature (21±0.5 °C) remained constant.

The percentage removal and uptake capacity of Hg(II) ions by the HGAC from the monocomponent





**Fig. 3** Effect of reaction time on Hg(II) percentage removal (a) and uptake rate (b) by the HGAC from mono-metal [Hg(II)] and binary-metal [Hg(II)+As(III)] systems

and bicomponent media was observed to increase with increasing reaction time during the initial stage and later plateaued as presented in Fig. 3. The fast uptake of Hg(II) ions by the HGAC in both the single and bicomponent systems during the initial stages of the sorption process was attributable to the existence and accessibility of active binding sites on the bio-adsorbent surface and the initial high amount of Hg(II) ions present in the sorption media (Adio et al., 2019). However, as contact time increased, the percentage removal and uptake capacity decreased because the metal ions covered the active binding sites of the bio-adsorbent, causing repulsion as agitation time increased (Iggerase et al., 2017).

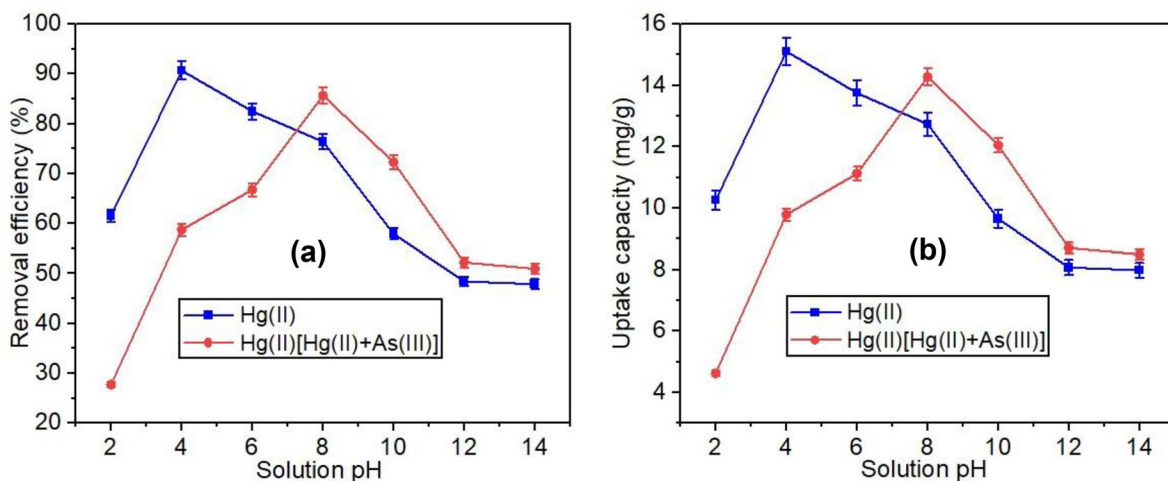
As can be seen from Fig. 3 (a) and (b), respectively, the competitive elimination of Hg(II) ions in the coexistence of As(III) ions in the bicomponent sorption system [Hg(II)+As(III)] exhibited a similar trend compared with the single-component system [Hg(II)]. Nonetheless, in comparison to the mono-component sorption system, the maximal removal rate and Hg(II) uptake rate by the HGAC in the bicomponent sorption medium were found to be lower. At the agitation time of 90 and 120 min, optimum percentage elimination and uptake capacity of 96.09 and 95.84% and 16.02 and 15.97 mg/g were attained by the HGAC for Hg(II) removal from the monocomponent and bicomponent sorption media, correspondingly. The coexistence of As(III) species

in the bicomponent sorption medium inhibited the detoxification of Hg(II) ions from the aqueous solution. The ratio ( $R_{Q_e}$ ) of both the percentage removal and Hg(II) uptake rate by the HGAC in the bicomponent sorption medium to the single sorption medium was found to be approximately equal to 1. Thus,  $R_{Q_e} = \frac{Q_{e, Binary}}{Q_{e, Single}} \approx 1$ , which indicated no interactive effect with only a 0.31% rate of adsorption reduction ( $\Delta\gamma$ ).

*Effect of pH of the solution*

The sequestration of Hg(II) ions in the mono- and bicomponent sorption media at different solution pH values (2–14) is shown in Fig. 4(a) and (b), respectively. The sorption experiments were agitated at an equilibrium agitation time of 90 and 120 min for Hg(II) ions detoxification by the HGAC from the mono-metal and binary-metal sorption media, respectively. Other parameters such as the agitation speed (120 rpm), bio-adsorbent particle size (90 μm), bio-adsorbent dose (1.5 g/L), and reaction temperature (21±0.5 °C) remained constant.

In Fig. 4, it was found that the removal rate and sorption capacity of Hg(II) by the HGAC in the monocomponent sorption medium increased with decreasing pH of the aqueous solution. The percentage removal and uptake capacity of the Hg(II) ions by the bio-adsorbent increased during the first stage



**Fig. 4** Effect of solution pH on Hg(II) percentage removal (a) and uptake capacity (b) by the HGAC from mono-metal [Hg(II)] and binary-metal [Hg(II)+As(III)] systems

because there is no precipitation of the metal ions but at higher pH values, the metal ions will precipitate in the form of hydroxyl ions, which will compete for the binding sites on the bio-adsorbent surface. Therefore, due to the high competition between the metal and hydroxyl ions in the mono-solute system, the removal rate and sorption capacity of Hg(II) by the HGAC declines drastically as displayed in Fig. 4 (a) and (b), respectively. Meanwhile, the decontamination of Hg(II) from the bicomponent sorption system augmented at higher pH values.

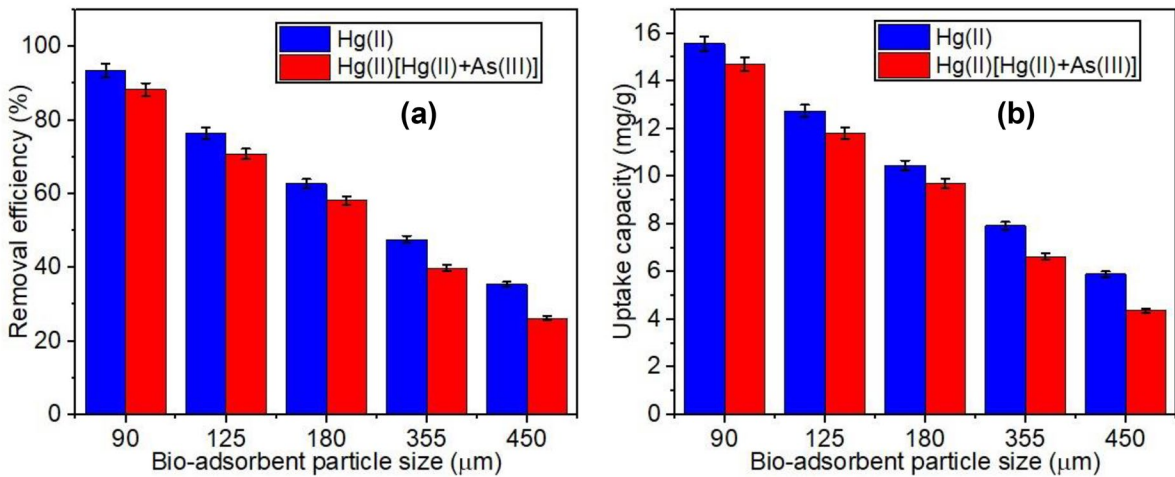
This is because the solution pH facilitated the Hg(II) ions to bind to the available functional groups present on the HGAC surface. The active HGAC sorption sites were better able to deprotonate when the solution pH increased and created a more negative charge on the HGAC surface, which has a strong affinity for the Hg(II) ions (Egirani et al., 2021). The surface charge of every bio-adsorbents plays a vital role in heavy metal ions adsorption and this is determined by the pH point of zero charge (pHpzc). The pHpzc is the pH at which the bio-adsorbent surface is universally neutral with the same number of negatively and positively charged surface functions. At solution pH values below the pHpzc, the bio-adsorbent surface is positively charged, and at solution pH values beyond the pHpzc, the bio-adsorbent is negatively charged (Sajjadi et al., 2018). The pHpzc of the HGAC was found to be 3.20, which is below

the optimum solution pH values of 4 and 8 obtained for the mono- and bicomponent sorption systems, thereby creating a negatively charged surface. Therefore, it was much easier to adsorb metal cations such as the Hg(II) ions on the negatively charged bio-adsorbent surface. A similar trend was observed by Bashir et al. (2019).

In comparison, Hg(II) removal from the mono-component medium was greater than that from the bicomponent sorption medium. While Hg(II) maximum removal efficiency of 90.67% and uptake rate of 15.11 mg/g were accomplished at the pH of 4 from the single sorption system, optimal elimination of Hg(II) from the bicomponent system was attained using a pH of 8 with removal rate and uptake capacity of 85.68% and 14.28 mg/g, correspondingly. The interactive effect of the As(III) species on the adsorptive sequestration of Hg(II) ions from the binary system could be described as an antagonistic effect since the ratio of Hg(II) percentage removal from the bicomponent system to the single-component system was lower than one ( $R_{Q_c} < 0.95$ ) with an uptake rate reduction ( $\Delta\gamma$ ) of 5.81%.

#### *Effect of bio-adsorbent particle size*

The impact of bio-adsorbent particle size on the non- and competitive depollution of Hg(II) species from monocomponent and bicomponent sorption media



**Fig. 5** Effect of bio-adsorbent particle size on Hg(II) percentage removal (a) and uptake capacity (b) by the HGAC from mono-metal [Hg(II)] and binary-metal [Hg(II)+As(III)] systems

was investigated at different particle sizes ranging from 90 to 450 μm and the experimental results are displayed in Fig. 5 (a) and (b), respectively. The experiment was operated at the following constant conditions: 120 rpm speed of agitation, 25 mg/L mono- and binary-solute initial concentration, solution temperature of 21 ± 0.5 °C, pH values of 4 and 8, and 90 and 120 min agitation time for the depollution of Hg(II) ions from the monocomponent and bicomponent sorption media, respectively.

The results demonstrated that the percentage removal and Hg(II) ions uptake rate by the HGAC from both sorption media were similar and decreased with increasing particle size of the HGAC as presented in Fig. 5.

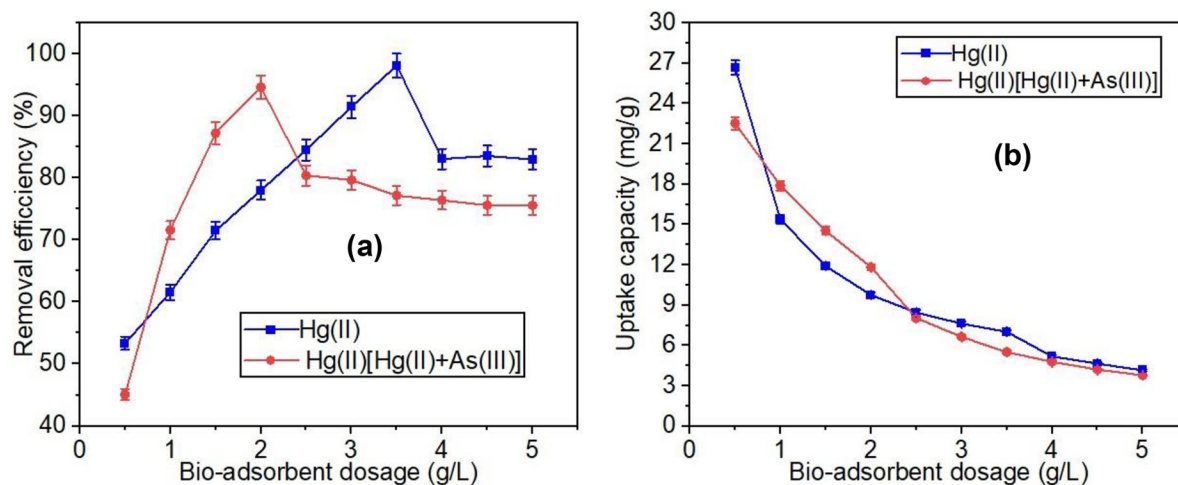
Due to higher access to pores and larger surface area for bulk sorption per unit weight of the bio-adsorbent, decreasing particle size enhances Hg(II) ions uptake at a constant bio-adsorbent dosage (Krishna & Swamy, 2012; Memon et al., 2021). The optimum removal percentage and uptake rate of Hg(II) ions obtained from the bicomponent medium were lower than those attained from the mono-metal sorption medium. This might be because the occurrence of the As(III) species in the bicomponent system lowered the uptake ability of the Hg(II) ions. It was found that the ratio of the Hg(II) removal efficiency in the bicomponent sorption medium to the monocomponent sorption medium is lower than

1 ( $R_{Q_c} < 0.94$ ) suggesting an antagonistic effect. Respectively, the Hg(II) maximum removal of 93.45 and 88.25% and uptake capacity of 15.58 and 14.71 mg/g were accomplished for the non-competitive and competitive systems at a bio-adsorbent particle size of 90 μm. The Hg(II) ions uptake rate ( $\Delta\gamma$ ) by the bio-adsorbent was observed to be reduced by 5.91% in the binary solute system due to the competitive As(III) ions coexisting in the solutions.

#### Effect of bio-adsorbent dose

The impact of the bio-adsorbent dose on the detoxification of Hg(II) ions individually and competitively in the mono- and binary-metal sorption media was conducted at different loads of the HGAC in the range of 0.5–5.0 g/L and the results are presented in Fig. 6(a) and (b), correspondingly. Similarly, the operating conditions that remained constant include agitation speed (120 rpm), bio-adsorbent particle size (90 μm), initial concentration of mono- and binary-metal (25 mg/L), temperature (21 ± 0.5 °C), solution pH (4 and 8), and agitation time (90 and 120 min) for the sequestration of Hg(II) ions from the monocomponent and bicomponent sorption media, respectively.

As observed in both monocomponent and bicomponent sorption media, upon increasing the bio-adsorbent dose, Hg(II) ions removal rate increased while the uptake capacity decreased as presented



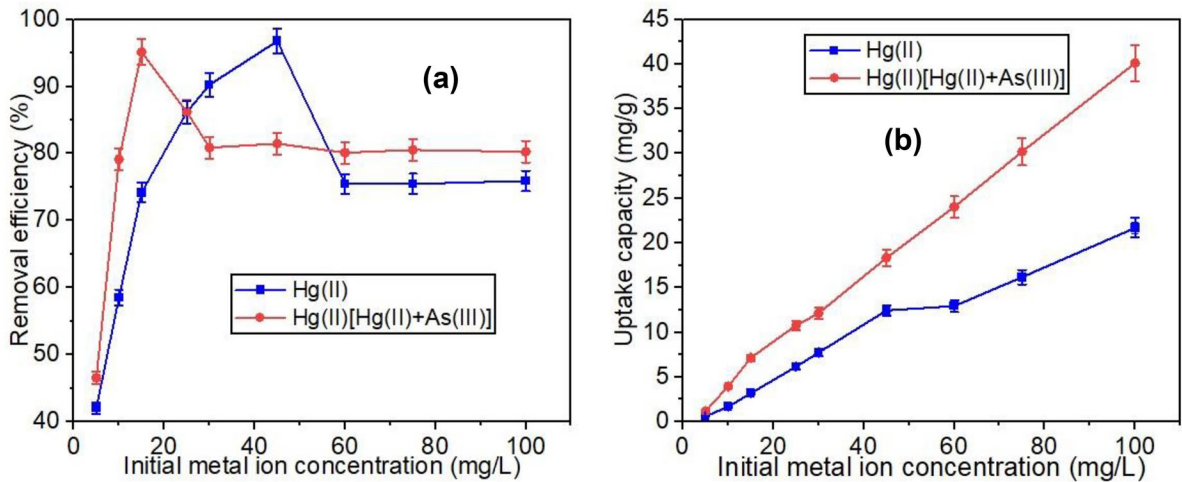
**Fig. 6** Effect of bio-adsorbent dose on Hg(II) percentage removal (a) and uptake capacity (b) by the HGAC from mono-metal [Hg(II)] and binary-metal [Hg(II)+As(III)] systems

in Fig. 6(a) and (b), respectively. For both sorption systems, when the bio-adsorbent load is augmented, there is a corresponding upsurge in the available active sites and the bio-adsorbent surface area leading to an increase of Hg(II) ions removal (Hiew et al., 2021). The optimal Hg(II) decontamination efficiencies from the monocomponent and bicomponent sorption media were 98.13 and 94.63% attained at 3.5 and 2.0 g/L, respectively, as best bio-adsorbent loads. It was found that the removal efficiencies of Hg(II) attained from both sorption media decreased at higher bio-adsorbent doses. This is because, at excessive doses, particle agglomeration is likely to occur leading to the availability of fewer Hg(II) ions to be adsorbed on the active sites of the HGAC. As a result, the uptake rate is reduced by the non-occupancy of the HGAC active binding sites by the Hg(II) ions (Peng et al., 2018). The maximal Hg(II) uptake rates of 26.67 and 22.54 mg/g were realized from the monocomponent and bicomponent sorption media, correspondingly, at the lowest HGAC dose of 0.50 g/L. Comparatively, the Hg(II) ions uptake rate by the HGAC became much lower in the competitive mode than that of the mono-metal sorption medium because of the antagonistic effect among Hg(II) and As(III) species in the solution. The ratio ( $R_{Q_e}$ ) of the uptake capacity of Hg(II) ions by the HGAC in the binary system to the single system was less than 1 ( $R_{Q_e} < 0.85$ ) with adsorption rate reduction ( $\Delta\gamma$ ) of 18.32%.

#### *Effect of mono- and binary-solute initial concentration*

The influence of mono- and binary-metal initial concentration on the removal of Hg(II) ions from the monocomponent and bicomponent sorption media are shown in Fig. 7(a) and (b), correspondingly. The mono- and binary-metal initial concentrations were varied from 5 to 100 mg/L alongside other constant conditions such as speed of agitation (120 rpm), bio-adsorbent particle size (90  $\mu\text{m}$ ), solution temperature ( $21 \pm 0.5$   $^{\circ}\text{C}$ ), pH of 4 and 8, a bio-adsorbent dose of 3.5 and 2.0 g/L, and agitation time of 90 and 120 min for the decontamination of Hg(II) ions from mono-component and bicomponent sorption media.

The batch experiments of the binary systems were carried out using pairs of the metal ions [Hg(II)+As(III)] with the same initial concentration in the ratio of 1:1 [Hg(II):As(III)]. The percentage removal and Hg(II) ions uptake rate by the HGAC followed an identical trend in both monocomponent and bicomponent sorption media as displaced in Fig. 7(a) and (b), respectively. As the mono- and bi-solute initial concentration in the sorption media augmented, Hg(II) ions uptake rate by the HGAC also increased while the removal efficiency decreases in the reverse order. The percentage removal of Hg(II) ions from the mono-metal [Hg(II)] and binary-metal [Hg(II)+As(III)] systems decreased at elevated initial concentrations due to surface-reactive sites



**Fig. 7** Effect of initial concentration on Hg(II) percentage removal (a) and uptake capacity (b) by the HGAC from mono-metal [Hg(II)] and binary-metal [Hg(II)+As(III)] systems

getting occupied steadily and reaching saturation point (Pasgar et al., 2022). Likewise, Igberase et al. (2017) investigated the removal of Pb(II), Zn(II), Cu(II), Ni(II), and Cd(II) by modified ligand as an adsorbent and it was observed that at lower initial concentrations, Pb(II), Zn(II), Cu(II), Ni(II), and Cd(II) removal efficiencies were very high. However, at higher initial concentrations, lower removal efficiencies were recorded for all heavy metals. This was because as the initial concentration upsurges, more metal ions are present in the aqueous solution, which leads to more ions being adsorbed to the same amount of adsorbent. This causes the adsorbent to become saturated, which lowers the removal efficiencies of the heavy metals. Similar trends were observed by Hosseini-Bandegharai et al. (2011) and Chen et al. (2017), in which a decrease in concentration increased the removal efficiency.

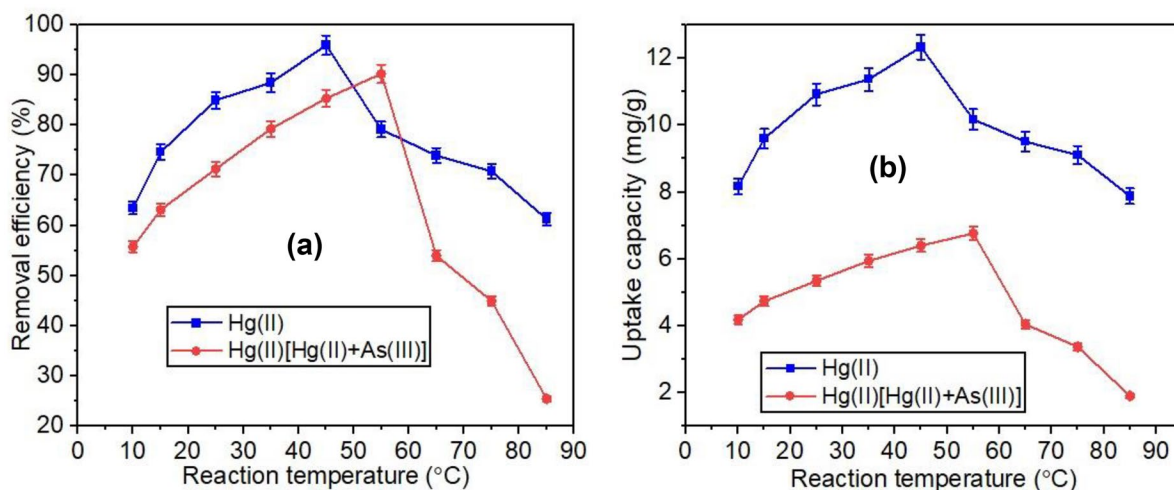
At initial metal ions concentrations of 45 and 15 mg/L, correspondingly, maximum uptake of Hg(II) ions from the monocomponent and bicomponent sorption media was achieved by the bio-adsorbent. The maximum removal percentage of Hg(II) ions from the monocomponent and bicomponent sorption media attained at 45 and 15 mg/L was 96.78 and 95.13%, respectively. However, by increasing initial concentration, the sorption capacities of Hg(II) in both mono- and bi-solute sorption systems also upsurge. This is because the interactive behavior between the ionic species and active bio-adsorbent

sites became easier with the diffusion of the ionic species to the sorption sites occurring more quickly (Abou Taleb et al., 2021). The maximum sorption capacities of 21.69 and 40.13 mg/g for Hg(II) were attained at 100 mg/L from the single-metal and binary-metal sorption systems, respectively. The ratio of the uptake capacity attained from the binary sorption system to the single solute system was much greater than 1 ( $R_{Q_e} > 1.85$ ) signifying synergistic effect of As(III) species on Hg(II) removal by the bio-adsorbent. The Hg(II) ions reduction uptake rate ( $\Delta\gamma$ ) by the bio-adsorbent was found to be increased to 45.95% in the bi-solute sorption system in the coexistence of As(III) ions in the solution.

*Effect of reaction temperature*

The influence of solution temperature on the confiscation of Hg(II) ions from the monocomponent and bicomponent sorption media by the HGAC was studied within the temperature range of 10–85 °C as shown in Fig. 8, respectively. The experimental conditions that remained constant include agitation speed (120 rpm), bio-adsorbent particle size (90 μm), initial concentration of mono- and binary-metals (25 mg/L), a bio-adsorbent dosage of 3.5 and 2.0 g/L, solution pH of 4 and 8, and agitation time of 90 and 120 min for the elimination of divalent mercury from monocomponent and bicomponent sorption media, respectively.





**Fig. 8** Effect of solution temperature on Hg(II) percentage removal (a) and uptake rate (b) by the HGAC from mono-metal [Hg(II)] and binary-metal [Hg(II)+As(III)] systems

In comparison, the elimination of Hg(II) from the mono- and bicomponent sorption media was found to be analogous as presented in Fig. 8(a) and (b), respectively. The decontamination of Hg(II) species in both sorption media augmented with rising temperature. Therefore, the removal of Hg(II) by the HGAC is an endothermic process, which also indicates the mechanism of Hg(II) adsorption from the mono- and bicomponent sorption media was mainly chemisorption. The upsurge of temperature in the sorption system increases Hg(II) uptake rate due to the enhanced kinetic energy of the ionic species through the bio-adsorbent and the subsequent adsorption on the active surface (Lenka et al., 2021). However, it was observed that at higher levels of reaction temperature, both the removal and uptake rates decreased drastically and this might be because of the deterioration of the surface chemistry of the adsorbent through excessive heating.

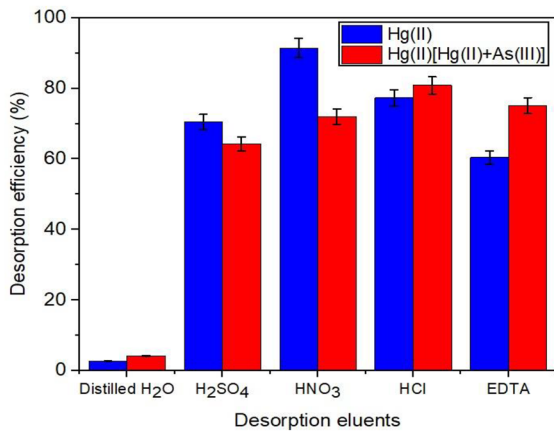
At a temperature of 45 and 55 °C, Hg(II) maximum removal of 95.91 and 90.20% and uptake capacity of 12.33 and 6.77 mg/g were attained, respectively, from the mono- and binary-solute sorption media. The optimum uptake of the Hg(II) ions from the bicomponent medium by the bio-adsorbent was observed to be smaller than that obtained from the monocomponent medium. Since the ratio of Hg(II) ions uptake ability by the HGAC from the binary system to the single

system was lower than  $1(R_{Q_c} < 0.55)$ , the As(III) ions exhibited antagonistic effect on Hg(II) removal in the bicomponent sorption system. Hence, the Hg(II) ions uptake rate ( $\Delta\gamma$ ) by the bio-adsorbent was found to be reduced by 82.13% in the binary-solute system.

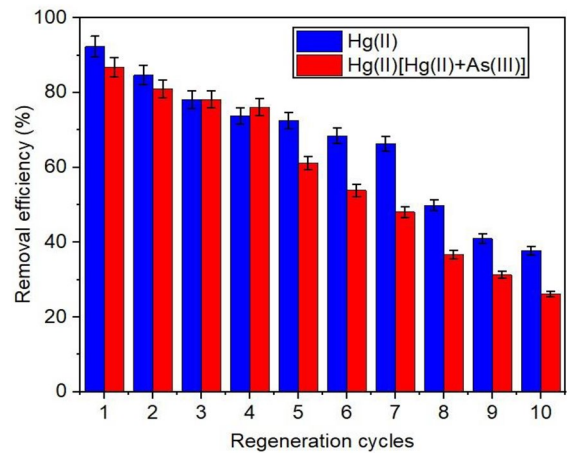
#### Desorption, regeneration, and reusability studies

The efficiency of any sorbent material is largely assessed based on its recyclability and reusability. Hence, it is indispensable that the adsorbent maintains its original sorption capacity even after multiple cycles of adsorption-desorption studies (Dahake et al., 2021). Furthermore, renewability and reusability are imperative to safeguard the economic viability of using the bio-adsorbent for sorption process scale-up and industrial applications.

Before the desorption studies, the bio-adsorbent was initially placed in 100 mL aqueous solutions containing optimum 45 and 15 mg/L initial mono- and bi-solutes concentration, solution pH of 4 and 8, an agitation time of 90 and 120 min, bio-adsorbent doses of 3.5 and 2.0 g/L with 90  $\mu\text{m}$  particle size, and a temperature of 45 and 55 °C, respectively, which was operated at 120 rpm agitation speed for the elimination of Hg(II) species from monocomponent and bicomponent sorption media, respectively.



**Fig. 9** Desorption eluents for Hg(II) ions desorption from the HGAC surface



**Fig. 10** Renewability of the spent HGAC and its reusability

Thereafter, the mono- and binary-metal ions-loaded bio-adsorbents were separated from the liquid phase through filtration and thoroughly treated using deionized water until neutral pH was achieved. The spent bio-adsorbent was dried at 110 °C to a fixed weight using an oven operated for 24 h. Afterward, the dried spent bio-adsorbent was treated separately with different desorption solutions such as distilled water, 0.1 M of H<sub>2</sub>SO<sub>4</sub>, HNO<sub>3</sub>, HCl, and EDTA to test the retrievability of the Hg(II) ions adsorbed from the mono- and binary components sorption media. The desorption tests were also performed in a batch process by employing the same experimental conditions as that of the adsorption studies wherein 50 mL of each desorption eluent was used to desorb Hg(II) from the spent HGAC and solutions containing the desorbed Hg(II) ions were analyzed using the CVAAS.

The desorption percentage removal was computed by Eq. (7) given below (Kavand et al., 2020).

$$\text{Desorption efficiency} = \frac{C_{de}}{C_{ad}} \times 100 \tag{7}$$

where C<sub>ad</sub> and C<sub>de</sub> are the amount of Hg(II) ions adsorbed on the HGAC and desorbed from the HGAC active binding sites, respectively.

The results of the reclamation of Hg(II) ions accumulated on the bio-adsorbent surface after the mono-component and bicomponent sorption processes are displaced in Fig. 9.

Among all the desorption eluents tested, the study found that 0.1 M HNO<sub>3</sub> and HCl acids were the best desorption eluents and could retrieve about 91.47 and 80.85% of Hg(II) ions adsorbed onto the bio-adsorbent surface in the monocomponent and bicomponent sorption media, respectively. The HNO<sub>3</sub> and HCl acids are the best desorption eluents because, in an HNO<sub>3</sub> solution, H<sup>+</sup> can replace adsorbed ions on the surface of bio-adsorbent while Cl<sup>-</sup> from HCl can easily form a complex with Hg(II) ions and then release to the solution (Kim et al., 2011; Vakili et al., 2019). However, distilled water was observed not to be a suitable desorption eluent in desorbing Hg(II) ions adsorbed onto the surface of the bio-adsorbent in both mono- and bi-solutes sorption systems and have the lowest desorption efficiencies of 2.65 and 4.13%, respectively.

Therefore, 0.1 M HNO<sub>3</sub> and HCl solutions were chosen for the regeneration of the spent bio-adsorbent for its reusability in successive multicycles of adsorption-desorption as shown in Fig. 10.

Figure 10 demonstrates that the decontamination efficiencies of divalent mercury from the mono- and binary-metal ions-loaded bio-adsorbent decreased slowly from 92.31 to 68.48% and 86.88 to 53.85%, respectively, as the regeneration cycles were elevated from one to six. This could be due to the decrease in the availability of binding sites for Hg(II) ions uptake and partial desorption of Hg(II) ions chemically adsorbed on the HGAC surface (Hiew et al., 2021).



**Table 2** Adsorption selectivity as a function of initial concentrations of Hg(II) in the binary sorption system

Concentration (mg/L)	Single system $[q/c_e]_a$	Binary system $[q/c_e]_b$	Selectivity factor (SF) $SF = \frac{[q/c_e]_a}{[q/c_e]_b}$
5	0.296	0.435	0.681
10	0.469	0.593	0.790
15	0.912	1.025	0.890
25	1.674	1.653	1.013
30	2.598	1.896	1.370
45	2.938	1.911	1.538
60	4.010	2.115	1.896
75	4.600	1.964	2.343
100	5.267	1.388	3.795

Since there was no noticeable weight loss in the bio-adsorbent load even after the fifth cycle, this suggests good mechanical stability of the bio-adsorbent as supported by the TGA results presented in Fig. 1 (Gupta et al., 2021). However, the percentage removal of Hg(II) ions reduced drastically after the sixth to tenth regeneration cycles. This was probably due to the collapse of the HGAC microstructure and continuous pore microstructure distortion owing to repeated adsorption-desorption cycles (Gupta et al., 2021; Liu et al., 2020).

### Selectivity

The adsorption selectivity as a function of the initial concentrations of Hg(II) ions in the binary system, using a 0.5 g/L bio-adsorbent dose at a pH of 4.0, was examined. The selectivity study was performed to confirm the preference for the metal ions removal by the HGAC from the binary sorption media as summarized in Table 2.

Table 2 shows that the selectivity of Hg(II) increases with the increase in initial concentration. Therefore, the HGAC was confirmed to be an effective adsorbent in the removal of Hg(II) ions from competitive aqueous systems. Similarly, the competitive adsorption of Cu(II), Ni(II), and Zn(II) ions onto synthetic zeolite was conducted by de Morais França et al. (2021). While it was found that the selectivity of Cu(II) ions in the binary and ternary sorption systems upsurged with the rise of initial concentration, the adsorbent has low selectivity for Ni(II) and Zn(II)

ions in both sorption systems. This could be due to the competition between the heavy metal ions for specific adsorption sites causing a decrease in affinity for the Ni(II) and Zn(II) ions (Anna et al., 2015). Also, the same trend was observed by Hosseini-Bandegharai et al. (2014), in which the selectivity factor values for Pb(II) in the competitive systems were very high which indicated that the adsorbent had a high affinity for Pb(II) over other metal ions.

### Conclusion

The interactive effect of As(III) species on the sorption and desorption potential of divalent mercury from the binary sorption medium was investigated. The study discovered that the HGAC has excellent regeneration efficiency in addition to maximum adsorption capacity, demonstrating the possibilities of industrial applications. The adsorption studies showed that the Hg(II) ions can be removed effectively from the single system and competitively from the aqueous phases by the HGAC in the presence of As(III) species in the bicomponent sorption medium. The best operating experimental conditions obtained for the single decontamination of Hg(II) ions from non-competitive solutions were found to be 90 min reaction time with maximum removal and uptake rates of 96.09% and 16.02 mg/g, solution pH of 4 with maximum removal and uptake rates of 90.67% and 15.11 mg/g, 90  $\mu\text{m}$  bio-adsorbent particle size with maximum removal and uptake rates of 93.45% and 15.58 mg/g, 3.5 g/L bio-adsorbent dosage with a maximum removal rate of 98.13%, 45 mg/L initial mono-metal concentration with a maximum removal rate of 96.78%, and 45 °C solution temperature with maximum removal and uptake rates of 95.91% and 12.33 mg/g, respectively.

Besides, the presence of As(III) species in the bicomponent sorption system affected the decontamination of Hg(II) ions by the bio-adsorbent, and the major interactive mechanism was found to be antagonism. The suitable experimental conditions for efficient Hg(II) ions detoxification from the bicomponent sorption medium were observed to be 120 min reaction time with maximum removal and uptake rates of 95.84% and 15.97 mg/g, pH of 8 with maximum removal and uptake rates of 85.68% and 14.28 mg/g, 90  $\mu\text{m}$  bio-adsorbent particle size

with maximum removal and uptake rates of 88.25% and 14.71 mg/g, 2.0 g/L bio-adsorbent dosage with a maximum removal rate of 94.63%, 15 mg/L initial mono-metal concentration with a maximum removal rate of 95.13%, and 55 °C reaction temperature with maximum removal and uptake rates of 90.20% and 6.77 mg/g, respectively.

Among all the desorption eluents tested, the study found that 0.1 M HNO<sub>3</sub> and HCl acids were the best desorption eluents and could retrieve about 91.47 and 80.85% of Hg(II) ions adsorbed onto the bio-adsorbent surface in the monocomponent and bicomponent sorption media, respectively. The used bio-adsorbent was effectively regenerated and reused in ten adsorption-desorption cycles, in which the removal efficiencies declined from 92.31 to 68.48% and 86.88 to 53.85%, respectively, as the regeneration cycles were elevated from one to six. The first regeneration cycle was found to have the highest Hg(II) ions removal efficiencies of 92.31 and 86.88% for the monocomponent and bicomponent sorption media. Therefore, the HGAC was found to be mechanically stable and reusable up to the sixth regeneration cycle suggesting its economic prospects and application at a larger scale for heavy metals removal from monocomponent and multi-component aqueous systems.

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**Author contribution** Jonas Bayuo was in charge of conceptualization, funding acquisition; methodology; investigation; data curation; formal analysis; and writing — original draft. Mwemezi J. Rwiza and Kelvin Mark Mtei were also in charge of supervision; validation; writing — review and editing.

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**Data availability** Data will be made available upon reasonable request.

#### Declarations

All authors have read, understood, and have complied as applicable with the statement on “Ethical responsibilities of Authors” as found in the Instructions for Authors.

**Conflict of interest** The authors declare no competing interests.

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