Introduction

Environmental pollution is the natural consequence of humankind’s activities; therefore, it is impossible to inhibit the pollution of the Earth as long as a single person lives on this planet (Alattabi et al., 2017; Hashim, Shaw & Al-Shamma’a, 2021). The by-products that resulted from the activities of a single person apparently harmless for both environment and public health, but the collective effects of large populations, such as crowded cities, cause extensive pollution phenomena, such as the discharge of liquid and solid wastes to the surrounding environment (Goel, 2006; Abdulla et al., 2020; Khalid et al., 2020a). Unfortunately, there is an irretrievable relationship between the environmental pollution and the existence of humankind because the human needs to use the natural resources to survive and also to develop the infrastructures, which in turn result in the production of different forms of pollutants, including gaseous, liquid and solid pollutants (Salah, Ortega-Martorell, Abdellatif, Gharghan & Ahmed, 2020; Abdulhadi et al., 2021). For instance, the literature shows that the increasing global demand for food has resulted in a dramatic increase in the consumption of fertilizers and pesticides. Zhang et al. (2014) reported that 300 million kg of pesticides are annually used in China to increase crop production, which is 2.5–5 times the average global usage of pesticides, while the usage of fertilizers in China has risen to 2.8–3 times the global average (Han, Currell & Cao, 2016; Alenezi, Hasan, Amoako-Attah, 2020).
Gkantou & Abdulhadi, 2020; Al-Marri, AlQuzweeni, AlKizwini, Zubaidi & Al-Khafaji, 2020). The rapid increase in the worldwide population has also resulted in a substantial expansion of industrial activities, such as the textile and pharmaceutical industries, which substantially contributes to the rise of water pollution (Aqeel et al., 2020; Emamjomeh et al., 2020a). In Japan, the literature confirms that concentrations of some heavy metals in the groundwater and soil in Kunugi-yama area also reached alarming levels; zinc concentration in groundwater was about 90 times higher than its reference level, while the concentrations of copper, zinc, and lead in soils were about 2.3–3.4 times higher than their reference levels (Ozaki et al., 2019). Besides the significant water pollution, the planet of Earth faces the dilemma of the increasing generation of municipal solid wastes (MSW) that is seriously polluting both soils and sources of groundwater (Alenazi et al., 2020; Alhendal et al., 2020), and it also generates a substantial amount of greenhouse gases that results in climate changes (Alenazi et al., 2020; Alhendal et al., 2020). This short literature survey indicates that water pollution and the generation of MSW represent direct threats to both eco-system and public health. At the same time, the literature shows many proposed solutions to each one of these dilemmas. For example, a broad spectrum of treatment methods was used to minimize or eliminate water/wastewater pollution, such as electrocoagulation (Hashim et al., 2020b), adsorption (Alyafei et al., 2020), bio-reactors (Khalid et al., 2020a), membranes and chemical precipitations (Al-Saati et al., 2019). In terms of MSW, many solutions were suggested to minimize its impacts on the environment, these solutions mainly focused on the recycle, reuse, sort, incineration, and disposal into landfills (Idowu, Atherton, Alo & Shaw, 2019). It can be seen that the majority of the proposed solutions focused on each dilemma separately. In addition, many of the proposed solutions, especially for water pollution, are expensive and complex, which limits their wide range of applications. Hence, the current study aims at providing a partial solution to these dilemmas by utilizing the wasted cow bones (a kind of MSW) that generated from houses and hospitality facilities to produce a green sorbent that was applied to remediate water from heavy metal pollution (lead as a case study). Heavy metal pollution was considered in the present study as the concentrations of some heavy metals in freshwater bodies have increased by 1,000 folds of the allowable limits.

Methodology

Preparation of the green sorbent

Samples of cow bones were collected from local butcher shops and restaurants, in the city of Babylon, Iraq. Initially, the collected sample was manually cleaned to remove the residuals of meat and fat, then bones were chopped into small pieces (2.0–5.0 cm) (Alquzweeni & AlKizwini, 2020). The cleaned pieces were used to prepare the biochar according to the recommended method by Iriarte-Velasco, Sierra, Zudaire and Ayastuy (2016) as follows: the cleaned
pieces were subjected to a pre-carbonization process at a temperature of 450°C in a nitrogen atmosphere for 1 h to remove any residual of meat and fat. In this step, the heating rate and nitrogen flow were kept constant at 10°C·min⁻¹ and 120 cm³·min⁻¹, respectively, until the required level was reached. The pre-carbonized sample was cooled down, and then it was sieved to separate particles with a size range of 0.25–0.35 mm. The collected particles were divided into two parts. The first part was subjected to the coating process, while the second part was not further treated to be used later as a reference (for comparison purposes). Analytical grade MgCl₂·6H₂O, AlCl₃·6H₂O and NaOH, provided by Sigma-Aldrich, were used in the preparation of the MgAl-LDH-biochar. Synthesis of the MgAl-LDH-biochar has been carried out according to the stated procedures by Meili et al. (2019) as follows: 3.62 g of AlCl₃·6H₂O and 6.09 g of MgCl₂·6H₂O were added into a flask containing 20 mL of deionized water, and stirred for 30 min and speed of 150 rpm to ensure that the added chemicals were completely dissolved. Then, 1 g of the biochar was added to this solution; the pH of the solution was increased to 10 by adding 3M of NaOH. The mixture was stirred again for 240 min before centrifuge it using a Sigma machine (model: 3-16 PK) at speed of 3,000 rpm for 5 min. The separated solids were washed with deionized water and dried using an electric oven (model: 101-0BS) at a temperature of 60°C for 16 h. According to the literature, a molar ratio of Mg : Al was kept at 4 : 1 showed a good performance (Meili et al., 2019).

Characterization of the MgAl-LDH-biochar

The physicochemical properties of the MgAl-LDH-biochar were characterized using the X-ray diffractometer (XRD) (Aeris: PANalytical). The characterization process was carried out at λ of 0.1542 nm and a scanning rate of 2°·min⁻¹ in 2θ range of 5–65°. Additionally, metals concentration was measured using an Inductively Coupled Plasma-Mass Spectroscopy (ICP-MS) on a Varian Vista Pro.

Batch experiments

A stock lead solution (1,000 mg·L⁻¹) was prepared by dissolving 1.598 g of Pb(NO₃)₂ (provided by Sigma-Aldrich) in 1.0 L of deionized water. Fewer concentrations were diluted from this concentrated solution. Batch experiments were carried out by mixing different doses of the MgAl-LDH-biochar (0.1–0.3 g·L⁻¹) with 100 mL of lead solution in a 200 mL flask. The latter was shaken using a mechanical shaker for 3 h at a speed of 150 rpm (Alquzweeni & Alkizwini, 2020). To attain the best removal of lead; the effects of lead concentration (Lc) (10–50 mg·L⁻¹), solution initial pH (2.0–7.0), contact time (Ct) (5–180 min), and solution temperature (St) (15–45°C) on the adsorption efficiency were measured. To measure the residual lead concentration, a 5 mL sample was taken from the mixture at the required time and instantaneously filtered on a 0.22 nylon membrane filter (provided by Hach-Lange Ltd); the filtrate was examined using a Thermo atomic absorption spectrophotometer (model: ICE 3300). All
experiments were repeated three times. Adsorption isotherm and kinetics of lead on the MgAl-LDH-biochar has been measured by contacting different doses of MgAl-LDH-biochar (0.1–0.3 g·L⁻¹) with 100 mL of lead solution (30 mg·L⁻¹). The mixture was continuously shaking for 24 h at a speed of 150 rpm to attain the equilibrium condition. The adsorption isotherm experiments were carried out at room temperature. Langmuir isotherm model (Eq. 1) was used to assess the adsorption of lead on the MgAl-LDH-biochar not only because this model is commonly used in such studies, but also because it provides the necessary information to calculate the separation factor (\(R_L\)) (Eq. 2), which is a reliable tool to assess the affinity between the adsorbrates and adsorbents. The adsorption process is unfavorable when \(R_L\) is more than 1, linear when \(R_L = 1\), favorable when the \(R_L\) is within the range 1–0 or irreversible when \(R_L\) equals 0 (Dada, Olalekan, Olatunya & Dada, 2012).

\[
\frac{C_e}{q_e} = \frac{1}{Q_o \cdot b} + \frac{C_e}{Q_o} \quad (1)
\]

\[
R_L = \frac{1}{1 + b \cdot C_i} \quad (2)
\]

where:

- \(q_e\) – adsorbed amount of lead at equilibrium [mg·g⁻¹],
- \(Q_o\) – hypothetical monolayer adsorption [mg·g⁻¹],
- \(b\) – energy of adsorption [L·mg⁻¹].

Additionally, pseudo-second-order model (P-S-O) was also used to examine the adsorption kinetics. Removal efficiency was measured basing on the initial and final concentrations of lead as follows (Emamjomeh et al., 2020b; Hashim, Andrew, Al-Jumeily, Alwash & Aljefery, 2020a):

\[
\text{removal [%]} = \frac{\text{initial concentration} - \text{final concentration}}{\text{initial concentration}} \cdot 100\%
\]

**Results and discussion**

Figure 1 shows the XRD patterns of the developed MgAl-LDH-biochar adsorbent and the reference (pure biochar). The XRD pattern of the MgAl-LDH-biochar shows a number of characteristic peaks, in 2θ range of 100–250, which confirm the successful deposition of the MgAl-LDH on the surfaces of the biochar particles as these peaks were absent in the reference. A number of unidenti-
fied peaks were noticed in the XRD patterns that indicate the presence of minor amounts of other minerals, which are usually detected in biochars that derived from organic origins. Additionally, minor crystallinity was observed in the biochar that could be resulted from the degradation of carbonaceous materials. In summary, the obtained results from the XRD test confirmed the presence of the MgAl-LDH on the surfaces of the biochar particles.

**Lead adsorption experiments**

**Kinetics of lead adsorption on the MgAl-LDH-biochar**

To assess the kinetics of lead removal on both MgAl-LDH-biochar and the reference. Figure 2 shows the results of fitting experimental results to the P-S-O, which reveals a set of key facts about the adsorption of lead ions on both MgAl-LDH-biochar and the reference. Firstly, a glance on Figure 2 reveals that the P-O-S is suitable for describing the adsorption of lead on MgAl-LDH-biochar and the reference; $R^2$ values for the relationships of reference – P-S-O and MgAl-LDH-biochar-P-S-O were 0.947 and 0.932, respectively, which indicate the suitability of P-S-O for describing the adsorption of lead on coated and uncoated adsorbents. Secondly, it can be seen from Figure 2 that the equilibrium condition was attained after 80 min of contact. Finally, the obtained results show that the MgAl-LDH-biochar is superior to the reference (untreated biochar) in terms of lead removal from water. Figure 3 shows the outcomes of Langmuir isotherm model application to the experimental data obtained from mixing different amounts of MgAl-LDH-biochar with the lead solution. It can be seen from this figure that the constant of Langmuir isotherm model

![FIGURE 2. Experimental data of the reference and MgAl-LDH-biochar adjusted to the kinetic model of P-S-O](image2)

![FIGURE 3. Langmuir isotherm for lead adsorption on MgAl-LDH-biochar](image3)
was 0.1332, which means that the $R_L$ is 0.07. The latter indicates high affinity between leads ions and MgAl-LDH-biochar (the $R_L$ is within the range 1–0). Basing on the outcomes of this part of the study, the $C_t$ will be kept constant at 80 min.

**Effects of solution pH on lead removal**

Lead adsorption on the MgAl-LDH-biochar has been examined at a pH range of 2.0–7.0 at MgAl-LDH-biochar dose of 0.2 g·L$^{-1}$, $C_t$: 80 min, $L_c$: 30 mg·L$^{-1}$, $S_t$: 25°C, and stirring speed of 150 rpm. Figure 4 indicates that lead adsorption was enhanced with the increase of the initial pH, but a negligible enhancement in the removal of lead when the pH increased from 6.0 to 7.0. At low pH values, the surfaces of the MgAl-LDH-biochar adsorbent develop positive charges that result in a repulsion of the positively charged lead ions, which minimizes the removal efficiency. Additionally, the competition between the accumulated hydrogen/hydronium ions and lead ions for the available active sites also minimizes the removal efficiency. Increasing the pH of the solution to the vicinity of 5 results in the development of positive charges on the surfaces of the MgAl-LDH-biochar adsorbent that helps to develop an electrostatic attraction between the adsorbent and lead ions. It is also believed that the inner surfaces of MgAl-LDH-biochar develop coordinate covalent bonds between the surfaces oxide’s oxygen and the metal forming monodentate and bidentate complexes. It noteworthy to highlight that the predominant path of lead removal at neutral or slightly acidic pH range is the precipitation of lead in the form of lead(II) hydroxides. The rest of the batch experiments, therefore; were run at a pH of 5.0.

**Effects of $C_t$ on lead removal**

Effects of $C_t$ on the adsorption of lead on the MgAl-LDH-biochar was examined at 10, 20, 30, 40, 50, 60, 70, and 80 min at $S_t$: 25°C, pH: 5.0; MgAl-LDH-biochar dose: 0.2 g·L$^{-1}$, $L_c$: 30 mg·L$^{-1}$ and stirring speed: 150 rpm. Lead removal increased from about 59.8 to 92.3% as the $C_t$ increased from 20 to 120 min, respectively (Fig. 5). The majority of the lead uptake happened during the first 100 min. Additionally, it was
noticed that the uptake rate of lead was rapid during the first 80 min, but it was slowed down during the last 40 min. The rapid uptake rate of lead, during the first 80 min, may be attributed to the fact that the initial uptake of lead mostly happens on the available external sites on the surfaces of MgAl-LDH-biochar. With the progress of the $C_t$ the available binding sites became limited, which results in slowing down the uptake rate of lead.

**Effects of $St$**

Solution temperature ($St$) plays a significant role in the adsorption process not because it governs the driving force of the pollutant ions onto the surface of the MgAl-LDH-biochar, but it also governs other key parameters, such as the strength of the energy barrier of reaction between the adsorbent and the adsorbate and it governs the size of the binding sites. Hence, the effect of the $St$ on the uptake of lead by the MgAl-LDH-biochar was investigated by commencing the adsorption experiments at different temperatures. Lead adsorption was examined at $St$ of 15, 25, 35, and 45°C keeping the initial pH, MgAl-LDH-biochar dose, $Lc$, $C_t$, and stirring speed constant at 5, 0.2 g·L$^{-1}$, 30 mg·L$^{-1}$, 80 min, and 150 rpm, respectively. The results of these experiments showed that the lead removal was enhanced at high temperatures, thereby, confirming that the nature of the lead adsorption process is endothermic (Fig. 6). The increase in the removal of lead at high temperatures is attributed to the reasons mentioned above; the increase in both sizes of the active sites and the diffusion of lead ions, which enhances the removal of lead. Additionally, some studies indicated that increasing the $St$ expands the size of the adsorbent particles that result in the breaking of some internal bonds near the surfaces of the particles, which produces more binding sites (Alquzweeni & Alkizwini, 2020).

**Effects of MgAl-LDH-biochar dose**

Adsorption of lead has been carried out at three different doses of MgAl-LDH-biochar (0.1, 0.2, and 0.3 g·L$^{-1}$) at initial pH: 5.0, $C_t$: 80 min, $St$: 45°C, $Lc$: 30 mg·L$^{-1}$, and stirring speed of 150 rpm. The effects of the MgAl-LDH-biochar
dose on the removal of lead are shown in Figure 7, which indicates that the lead removal is positively influenced by the increase of the MgAl-LDH-biochar dose. It was noticed that the removal of lead increased vicinity from 72 to 100% when the MgAl-LDH-biochar dose increased from 0.1 to 0.3 g·L⁻¹, respectively. The enhancement of lead removal with the increase of the MgAl-LDH-biochar dose is related to the increase in the number of the adsorption sites; the more the dose of the MgAl-LDH-biochar, the higher the number of active sites, which results in better removal of lead. However, increasing the dose of MgAl-LDH-biochar from 0.2 to 0.3 g·L⁻¹ increased the removal of lead by only 1%. Thus, to maintain the cost-effectiveness of the treatment method, MgAl-LDH-biochar dose of 0.2 g·L⁻¹ was used to run the rest of the experiments.

**Effects of \( L_c \)**

Basing on the fact that the discharged concentration of any pollutant is varying according to the season and/or the industrial activity. Thus, a set of experiments were commenced to measure the effects of
different \( Lc \) (10, 20, 30, 40 and 50 mg·L\(^{-1}\)) using the optimum conditions obtained in the previous sets of experiments (pH: 5.0, \( St \): 45°C, Ct: 80 min, MgAl-LDH-biochar dose: 2 g·L\(^{-1}\) and stirring speed: 150 rpm). Figure 8 depicts the effect of the \( Lc \) on the removal of lead using the MgAl-LDH-biochar; it can be seen that the higher the \( Lc \), the lower the removal of lead, where, it was noticed that increasing the \( Lc \) from 10 to 50 mg·L\(^{-1}\) has decreased the removal of lead by about 30%. This decrease in the removal of lead is related to the availability of the active sites on the surfaces of the MgAl-LDH-biochar. Simply, a constant amount of MgAl-LDH-biochar (0.2 g·L\(^{-1}\)) is available for the adsorption process, therefore, at high lead concentrations, it is not sufficient to absorb all lead ions, i.e. the available active sites will not be enough to occupy all lead ions. Generally, these obtained results in this study follow the same trends in the published literature (Gong et al., 2012).

**Conclusions**

In conclusion, the present study explored the adsorption of lead ions on green absorbent that was made by coating biochar particles with MgAl-LDH. The results of this study revealed that the MgAl-LDH-biochar is suitable for adsorbing of lead ions. Generally, it was observed that the performance of the MgAl-LDH-biochar was enhanced when the solution pH became slightly acidic, and also when the solution temperature increased. Additionally, it was noticed that increasing the dose of the MgAl-LDH-biochar more than 2 g·L\(^{-1}\) was not beneficial for the studied concentrations of lead. In contrast, increasing lead concentration has negatively influenced the performance of the MgAl-LDH-biochar. Thus, based on the obtained results from the commenced experiments, the efficiency of the MgAl-LDH-biochar could be enhanced by keeping the pH of the solution at the vicinity of the neutral range and by avoiding low water temperatures. The kinetic study confirmed a good affinity between lead ions and the MgAl-LDH-biochar. In summary, it could be concluded that the MgAl-LDH-biochar is a potential eco-friendly partial solution for two manger environmental concerns, which are the municipal solid wastes and water/wastewater pollution.

![FIGURE 8. Effects of \( Lc \) on lead removal by MgAl-LDH-biochar](image-url)
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**Summary**

**MgAl-LDH-biochar composites as a green sorbent for lead removal from water.**

The present study aims at the development of a green sorbent (MgAl-LDH-biochar) from the municipal solid wastes (cow bones) to remediate water from heavy metals. MgAl-LDH-biochar was characterized using X-ray diffraction (XRD), and it was used to remove lead at different pH, doses, contact time (*Ct*), and solution temperatures (*St*). The obtained results proved MgAl-LDH-biochar removes 99% of lead when the initial pH, *Ct*, *St*, and dose were 5.0, 80 min, 45°C, and 0.2 g·L⁻¹, respectively.

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