



Palm shell activated carbon impregnated with task-specific ionic-liquids as a novel adsorbent for the removal of mercury from contaminated water

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HIGHLIGHTS

- PSAC modified with task-specific ionic liquid was used as a novel adsorbent.
- PSAC grafted with TOMATS enhances the adsorption of Hg(II) ions.
- The maximum adsorption of mercury at room temperature was 83.33 mg/g at pH 8.
- The higher removal of Hg indicates the chemisorptions nature of adsorption process.

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ABSTRACT

In this study, palm shell activated carbon modified with task-specific ionic liquids was used as a novel composition for the removal of mercury from aqueous solutions in water samples through a batch-adsorption technique. The effect of pH, adsorbent dosage, equilibrium time, temperature and initial concentration of mercury ions on the removal efficiency was examined. The maximum adsorption of mercury at room temperature was 83.33 mg/g at pH 8 with a contact time of 3 h and an initial concentration of 10–200 mg/L. The experimental data were fitted well with the linear form of Langmuir model. Adsorption kinetics followed a pseudo-second-order model. The calculated activation energy for adsorption was 68.21 kJ/mol, indicates that the adsorption process involves some chemical forces and occurs by a chemisorption process. Thermodynamic parameters, including ΔH° , ΔS° and ΔG° for the adsorption process of mercury from aqueous solutions onto palm-shell activated carbon impregnated with trioctylmethylammonium thiosalicylate (TOMATS) indicated that adsorption was endothermic and spontaneous process.

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1. Introduction

Heavy metals, primarily cadmium, nickel, copper, lead, zinc, mercury, arsenic and chromium, reach the environment through several anthropogenic sources. Low concentrations of heavy metals are toxic and harmful to humans, plants and animals due to their toxicity and non-biodegradability [1]. The contamination of water resources by heavy metals is a serious environmental problem worldwide. Numerous metals, including mercury, cadmium, chromium, and lead, are known to be significantly toxic [2]. Mercury is one of the most toxic heavy metals in aqueous solution and is usually found in the dissolved phase at low concentrations. According to the European Water Framework Directive, mercury and its compounds are classified as one of 20 priority hazardous substances. Mercury accumulates within the human brain and causes central nervous system (CNS) effects. Consequently, the development of

reliable methods for the removal of mercury in environmental samples is particularly significant. Several physical and chemical methods have been reported for the removal of mercury from aqueous solutions. The available methods include membrane separation [3–5], precipitation [6,7], solvent extraction [8,9], ion exchange [10,11], reverse osmosis [12], coagulation [13] and photo reduction [14]. However, these methods are associated with high operational costs and incomplete removal and consume either large amounts of energy or large quantities of chemicals. The adsorption technique is generally considered to be a promising method and has been studied for the removal of metals from solutions [15,16]. The adsorption method typically involves natural materials that are abundant and inexpensive. Among these materials, various adsorbents, such as modified biomaterials [17,18], modified cellulosic materials [19,20], extracellular biopolymers [21], quartz and gibbsite [22,23], zeolites [24,25], nanomaterials [26–28], carbon nanotubes [29] and activated carbon [30,31] have been investigated with respect to their adsorption of heavy metal ions from solutions.

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Recent research has focused on the removal of heavy metal ions by palm shell activated carbon [32]. Palm shell activated carbon is a waste produced during palm kernel oil production, which is an important sector of the Malaysian economy. This material is readily available at low cost, has a high surface area and is of high quality. These properties make palm-shell activated a good raw material for the adsorption of heavy metals from solutions.

Recent studies have focused on the modification of the surfaces of activated carbon with chelating groups to enhance the adsorption efficiency of activated carbon toward a particular metal species from aqueous solutions [33].

Similar to activated carbon, room temperature ionic liquids (RTILs) have also recently received a great deal of attention. Ionic liquids are salts that have very low melting temperatures (close to room temperature). Ionic liquids have become an extremely popular item in recent separation and purification research due to their unique properties [34]. The properties of ionic liquids (ILs) (e.g., high polarity, conductivity, viscosity, acidity and hydrophobicity) depend on the nature and the size of their cationic and anionic species. Task specific ionic liquids are a unique subclass of ionic liquids that possess a potential spectrum of utility extending far beyond that likely for more conventional IL [35]. A widespread field of research is the use of ionic liquids (ILs) as well as task-specific ionic liquids (TSILs) for the removal of heavy metals [36,37].

In this work, palm shell activated carbon impregnated with the task-specific ionic liquid (TOMATS) was used as a novel composition for the removal of mercury from aqueous solutions.

According to the theory of hard and soft acids and bases (HSAB theory), Hg(II) is classified as a soft Lewis acid and therefore forms strong bonds with soft base ligands, such as thiol groups (SH) [38]. Therefore, palm-shell activated carbon impregnated with a task specific-ionic liquid that contains SH groups should exhibit a high adsorption capacity for mercury ions. The unique adsorption of Hg(II) ions on the impregnated palm shell activated carbon with TOMATS depends on the coordinate interaction between the TOMATS and the Hg(II) ions, which may be explained by the chelating effect of the *ortho*-positioned carboxylate group on TOMATS molecules grafted onto palm shell activated carbon, in addition to the known formation of metal-thiolates.

The effect of several parameters, including the pH of the solution, the adsorption dosage, the equilibrium time, the initial concentration of the solution, the weight of impregnated TOMATS and the activated carbon particle size were studied. Isotherm results and kinetic and thermodynamic parameters for the adsorption process of mercury from aqueous solutions onto impregnated palm shell activated carbon were obtained.

2. Materials and methods

2.1. Chemicals and reagents

All chemicals used were of analytical-reagent grade and de-ionised water was used for the preparation of all aqueous solutions. Commercial granular palm shell activated carbon (PSAC) produced by a physical activation process was provided by Pacific Activated Carbon, a local manufacturer in Johor Bahru, Malaysia. Activated carbon was sieved into three series, with particle sizes that ranged from 0.50 to 0.60, 0.60 to 0.85 and 1.4 to 1.7 mm. The activated carbon was subsequently washed with distilled water to remove impurities and then dried in an oven at 110 °C for 24 h. Metal salts were purchased from Merck, and aqueous metal solutions were prepared by the dissolving of appropriate quantities of metal salts in de-ionised water. Trioctylmethylammonium thiosalicylate (TOMATS) was purchased from Sigma–Aldrich. The pH levels of the solutions were adjusted by the addition of the

appropriate amount of hydrochloric acid (1 M HCl) or sodium hydroxide (2 M NaOH).

2.2. Analytical methods

Mercury was analysed using inductively coupled plasma atomic emission spectrometer ICP-AES (PerkinElmer, model ICP Optima 7000DV). All pH measurements were made performed with a pH/ion meter (Metrohm-781, Germany) and pH module (Metrohm-867).

The surface morphology and elemental analysis of the palm-shell activated carbon were obtained using an AURIGA field-emission scanning electron microscope cross-beam workstation (Carl Zeiss) equipped with an EDAX unit (AMETEK, Advanced Microanalysis Solutions) operated at an accelerating voltage of 25 kV.

2.3. Preparation of impregnated palm shell activated carbon

To prepare impregnated palm shell activated carbon with task-specific ionic liquids, 1 g of each activated carbon series and 0.00–0.3 g of TOMATS were placed into 250 mL Erlenmeyer flasks with 50 mL of methanol. The solutions were subsequently agitated on an orbital shaker at 180 rpm at a constant temperature of 25 °C for 2 days. The solutions were then evaporated, and the remaining impregnated palm-shell activated carbon was weighed. The overall loading of TOMATS onto the palm shell activated carbon was calculated from weight-loss data of virgin and impregnated palm shell activated carbon.

2.4. Adsorption experiments

To optimise the adsorption efficiency, adsorption experiments were performed at various pH levels, contact times, adsorbent doses, adsorbate concentrations and temperatures.

Adsorption experiments were performed in 250 mL Erlenmeyer flasks at different temperatures between 10 and 35 °C. In each experiment, 50 mL of Hg(II) solution with a concentration between 10 and 200 mg/L was treated with a specified amount of 0.1 g of modified activated carbon (prepared from 1 g palm-shell activated carbon with average particle sizes of 0.50–0.60 mm loaded with 0.2 g of TOMATS), and the pH was adjusted to the desired value by the addition of either 1 M HCl or 2 M NaOH, as required. The solutions were then shaken on an orbital shaker at 180 rpm for a specified period of time. The adsorbent was subsequently separated from the solution using Whatmann No. 42 filter paper. The equilibrium concentrations of Hg(II) that remained in solution were measured using ICP. The mercury uptake by the modified activated carbon was calculated as follows:

$$q_e = \frac{V(C_o - C_e)}{m} \quad (1)$$

where q_e is the amount of mercury ion adsorbed per gram of modified activated carbon (mg/g), V is the sample volume (L), C_o is the initial Hg(II) concentration (mg/L), C_e is the equilibrium Hg(II) concentration (mg/L), and m is the weight of adsorbent.

The removal percentage of mercury ions (%) from aqueous solutions was calculated as follows:

$$\text{Removal}(\%) = 100 \frac{(C_o - C_e)}{C_o} \quad (2)$$

3. Results and discussion

3.1. Effect of pH

The influence of the pH of the solutions on the removal efficiency was examined at a Hg^{2+} concentration of 80 mg/L by following the

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