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ORIGINAL PAPER

Removal of zinc oxide nanoparticles in aqueous environment using functionalized sorbents derived from sago waste

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Abstract

The increase in sago processing industries especially in Malaysia has inadvertently created its waste effect on the environment. The study therefore modified the raw sago waste residue (*hampas*) generated by three (3) chemical processes; esterification, acetylation and phosphorylation. The as-prepared sorbents were characterized by surface area analyser (BET), Fourier transform infrared (FTIR) spectroscopy, field emission-scanning electron microscopy (FE-SEM) and energy-dispersive X-ray spectroscopy (EDX). Thereafter, their sorption efficiencies were evaluated on zinc oxide nanoparticles in lake water in a laboratory-scaled column-bed sorption study. Spectra analysis confirmed the presence of ester and ether functionality in the esterified and acetylated samples, whereas phosphoric ester and amine functionality were present in the phosphorylated sample. The acetylated sample recorded the largest surface area (29.02 m^2/g) and degree of substitution (1.99). The experimental data established that acetylated sago residue recorded the maximum column capacity of 4.50 mg/g in all the parameters examined. Meanwhile, increase in influent concentration (1.51–5.04 mg/L) and flow rate (4.20–6.60 ml/min) decreased the breakthrough time while increase in column-bed height (3–7 cm) increased the breakthrough time for the three sorbents. However, Adams–Bohart model adequately described the behaviour of the sorption process better than Thomas and Yoon–Nelson models. The results suggested that acetylated sago residue is potentially scalable for removing zinc oxide nanoparticles from water.

Keywords Sago 'hampas' · Acetylation · Phosphorylation · Esterification · Column sorption

Introduction

Waste generation and its management has been a focus of attention to most countries globally in the last 2 decades. The Global Waste Management Outlook (2015), estimated that, 7–10 billion tonnes of waste are generated from households, commerce, industry and construction activities (Wilson and Velis 2015), out of which 998 million tonnes are generated from agricultural waste (Neh 2020). Nevertheless, the nanotechnology industry has also been projected to increase in growth rate (CAGR) of 10.5% from 2019 to 2025 (Broomfield et al. 2016; Allied Market Research 2018–2019). The industry encompasses the production and application of nanoscale devices which are beneficial to various industrial domains including communication, medicine, transportation, agriculture, energy, materials and manufacturing, consumer products and households (Husham et al. 2017; Jeem et al. 2017; Sarmah and Pratihar 2017). This implies that, as market share and production volume increases, their prevalence and potential threat to public health is a concern to stakeholders when they exceed their threshold concentrations (Rosenfeldt et al. 2014; Aljeboree et al. 2017; Sahu and Hayes 2017). It was estimated that 9–37% of engineered nanomaterials (ENMs) are released into the environment with the remaining 63–91% ending up in the landfills (Nowack et al. 2015).

The morphology and other unique properties (electronic, optical, mechanical, magnetic) of nanocrystalline ZnO make

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them suitable for many applications (Uikey and Vishwakarma, 2016; Anuraj and Gopalakrishnan 2017; Singh et al. 2017). After their utilization, they eventually end up in the various environmental media/sinks through various physical, chemical and biological means (Bansal et al. 2006; Handy et al. 2008; Auffan et al. 2010; Kaegi et al. 2010; Liu and Hurt 2010; Fabrega et al. 2011; Rabe et al. 2011). When these particles get into aqueous media, relevant environmental conditions such as aqueous chemistry (pH, temperature), system complexity (competing ions and biological components) as well as ion concentration affect their transformation which eventually affect their removal.

The use of agricultural by-products in adsorption studies has been proven to be an efficient, cost-effective and environmentally friendly technique for various pollutant removal from aqueous media (Bhatnagar et al. 2015; Gisi et al. 2016). Owing to the high amount of cellulose in these materials, they can be modified by different techniques to enhance their efficiency in pollutants removal (Kumar et al. 2017; Nag et al. 2017; Sulyman et al. 2017; Belalia et al. 2018). According to literature, cellulose materials can undergo different modification processes such as halogenation, esterification, amination, etherification and oxidation to increase their sorption sites (Melo et al. 2015). Prior to chemical modification of the biosorbent, pre-treatment process exposes the quality and quantity of cellulose as well as increases the porosity and surface area of the material (Thakur et al. 2020). For instance, when neem bark was pre-treated with concentrated H_2SO_4 (98%), the surface area was found to be 36.64 m^2/g which was higher as compared to other raw biosorbents such as tamarind seeds and tea factory waste (Maheshwari and Gupta, 2015). In another report, pre-treatment of pine with phosphoric acid (95%) recorded a specific surface area and iodine number of 734 m^2/g and 483.5 mg/g, respectively (Samarghandi et al. 2014).

After the pre-treatment process, the cellulosic biomass can now be chemically modified or functionalized by grafting technique (Lizundia et al. 2016). This mechanism introduces new functional groups (carboxylic, acetyl, amine, phosphoryl, ether) onto the sorbent surface as a result of chemical reaction between the hydroxyl (O–H) group of the cellulose material and the chemical species which further enhances their sorption efficiencies (Reddy et al. 2012; Qin et al. 2016; Luo et al. 2017).

The high volumes of sago waste residue (*hampas*) generated from sago starch extraction in Malaysia have raised much environmental pollution concerns which contravene the Environmental Policy Act, 1974. The literature documented the utilization of these waste materials in the area of water remediation studies (Ngaini et al. 2014, 2021; Amode et al. 2016; Wahi et al. 2017; Fauzia et al. 2018; Dahlan et al. 2019; Karthika and Vasuki 2019; Rajan et al. 2019; Beh et al. 2020; Makshut et al. 2020; Lim et al. 2021). However, there is no available literature on the utilization of sago waste residue (*hampas*) in the sorption of nanocrystalline ZnO species in natural waters like lake water.

Based on the previous studies, the current study modified raw sago waste residue (*hampas*) through three (3) chemical processes and their sorption efficiencies were evaluated on zinc oxide nanoparticles (ZnO NPs) in lake water sample in a laboratory-scale column-bed sorption study. Lake water was used for this study, because natural environmental conditions affect nanoparticle transformation and their stability (Peng et al. 2017; Parsai and Kumar, 2019). Also, lakes serve as drinking water sources but are usually used as places for waste discharge (Gasim et al. 2006). Some physico-chemical parameters of the natural water were determined and the chemically modified sorbents were also characterized using different techniques (FE-SEM, EDX, FTIR, BET) based on the available resources at the time of study. The sorption behaviour was described in terms of breakthrough curve and the effects of column-bed height, flow rate and influent sorbate concentration were investigated. Three (3) model types, Thomas, Yoon–Nelson and Adams–Bohart, were applied to the experimental data to stimulate the breakthrough curves and to find the column capacity in order to predict the scale-up for industrial purposes.

Materials and methods

Chemical preparation

Analytical reagent grade chemicals were used in the study. All solutions used were prepared using deionized water (DW) prepared in the Geochemistry laboratory of the Faculty of Resource Science and Technology, Universiti Malaysia Sarawak, Kota Samarahan.

Collection and preparation of sorbent

Raw wet sago residue (*hampas*) was supplied by Herdsen Sago Industries Sdn Bhd, a local sago industry in Sarawak, Malaysia. The sample was air-dried for 2 weeks at room temperature to minimize agglomeration of the cellulosic material (hornification) which could result in the reduction of water retention (Beaumont et al. 2017), and also to prevent the deformation of the samples structural and mechanical properties such as crystallinity and biodegradability (Vasconcellos & Farinas, 2018). The air-dried sample was oven-dried at 70 °C for 24 h. It was then blended into fine powder and sieved with Retsch Analysensieb mesh of size 150 µm (DIN-ISO 3310/1) mounted on a mechanical shaker (Endecotts EFL 2 mk3 Test Sieve Shaker). The prepared sample (RSH) was then stored for chemical modification.