



Mechanistic insights into the remediation of bromide ions from desalinated water using roasted date pits



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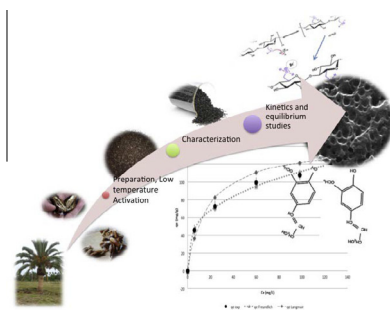
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HIGHLIGHTS

- Low temperature roasted date pits provides effective adsorbent for bromide ions.
- RODPs removal efficiency was higher than AC at weak acidic conditions.
- Adsorption processes were controlled by the surface and intraparticle diffusion.
- Potentially cost effective process with 87% cost reduction.

GRAPHICAL ABSTRACT



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ABSTRACT

Bromide ions (Br^-) are associated with toxic by-products that might be produced during the disinfection of drinking water. The removal of Br^- from water using roasted date pits (RODPs) and activated carbon was investigated. The percentage removal capacity of Br^- was studied under various experimental conditions including solution pH, adsorbent mass and particle size, and initial Br^- concentration. Surface characterisation was investigated using scanning electron microscopy (SEM) and Fourier transform infrared (FTIR). SEMs images showed the presence of fine particles (debris) on the surface of the adsorbent solids. The raw date pits (RDPs) surface images showed a marked difference in pore sizes upon roasting, whilst FTIR spectra showed a slight shifting in $-\text{O}-\text{H}$, $\text{C}=\text{O}$, and $-\text{C}-\text{O}$ stretching in addition to $-\text{O}-\text{H}$ bending for the RODPs. In addition to the Freundlich and Langmuir isotherm models, the experimental data were fitted to different adsorption kinetic models. It was found that the adsorption of Br^- onto RODPs followed pseudo-second order mechanism. Removal rates of around 54% were obtained at lower pH.

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1. Introduction

The availability of clean drinking water is a major concern in many countries around the globe. The problem is particularly acute in Middle Eastern countries including Qatar. Qatar has virtually no surface or ground fresh water resources, and accordingly is

classified as one of the water poorest regions in the world with less than 1% of freshwater availability. As a result, Qatar depends heavily on desalination for its fresh water needs [1]. Around 11 million cubic meters of drinking water were produced in 2012 forming slightly less than half the world's desalination capacity [1]. As a result of Qatar's booming economy, urbanisation and the steep increase in population, more stress has been exerted on the water resources, which resulted in building and commissioning of more desalination plants in Qatar to satisfy the growing demand. Before

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desalinated water reaches the customers, it is subjected to a number of quality control processes to ensure the highest drinking water quality. Disinfection is used to control the formation of bacteria and other pathogens in the water. Chlorination, ozonation, ultraviolet irradiation, chloramination are some of the widely used disinfection processes [2]. It has been reported that desalinated seawater contains around $650 \mu\text{g L}^{-1}$ and $<4 \mu\text{g L}^{-1}$ of Br^- and I^- , respectively [4,5]. Bromide ions concentration levels in surface water in the United States, for example, were found to be between 2 and $200 \mu\text{g L}^{-1}$ [5]. Concerns about the formation of brominated and iodinated toxic by-products, as a result of disinfection, have been highlighted and recently reported [3,4].

It is acknowledged that Br^- ions present in water can react differently with different disinfectants. The Br^- reaction with free Cl_2 is much faster than with chloramine to form HOBr. During the process of chlorination, Br^- can be oxidised to HOBr, which can react with organic matter in water to form brominated toxic by-products. Brominated by-products may have far more impact on human health than conventional chlorinated toxic by-products [6]. Bromate has been classified by the United States Environmental Protection Agency (US EPA [7]) as a potential carcinogenic substance to humans if orally ingested. Accordingly, the US has set the maximum contamination limit for bromate in drinking water at $10 \mu\text{g L}^{-1}$ [8]. As a result, an effective technique to eliminate bromate species, or limit its presence in drinking water, is pressing. Bromate is highly soluble and known to be very stable in aqueous solution upon formation; rendering its treatment difficult using traditional technologies [5,8].

In general, Br^- removal methods can be categorised into: membrane, electrochemical and adsorption remediation methods. Whilst membrane processes can positively remove Br^- from water, adsorption remediation methods form a key element in halide elimination. Membranes offer a barrier to most chemical molecules, though not permanently a comprehensive barrier. For example, the tendency of boron (as borate or boric acid), to pass through membranes raises the question as to what other anions and small organic molecules will pass through the membranes as well [6]. The capacity of metal oxides, activated carbons and other materials to adsorb inorganic ions, such as Br^- , are reported in literature but without offering insights into the adsorption mechanisms and surface interactions [9–11]. Activated carbon is widely used in water treatment for the removal of large variety of pollutants, however limited studies considered the removal of Br^- from water [13–15].

Date palm is a food-producing plant that has a long association with the culture, religion and history of the Arabian Peninsula. The date fruits are a major subsistence produced in most arid areas including Qatar. Date pits form nearly 15% (by weight) of the date fruits and are considered as a waste product with no economic value. In fact, date pits present a potential disposal problem [12]. This inexpensive, plentiful, and renewable waste material has been considered as a potential adsorbent for the treatment of a number of pollutants such as synthetic dyes, heavy metals and phenols [13]. In a recent article, Ahmed [14] reviewed the use of date stones, and the various methods of its preparation and activation, for the removal of pollutants from water. In his extended study, no reference was made to the removal of bromate/bromide. This supports our claim that, to our knowledge, no previous studies were undertaken for the application of date pits in the treatment of desalinated seawater and in particular, for the remediation of Br^- in aqueous solutions. Accordingly, the overarching aim of this work is to evaluate the results of adsorption performances of locally abundant date pits as an efficient adsorbent for the remediation of Br^- from desalinated seawater. The paper will report on the most prominent available functional groups on the surface of this low-cost, low-temperature activated adsorbent in addition to offering fundamental understanding of the equilibrium relation-

ships between the Br^- and the solid surface. Process kinetics will be considered by applying various models to offer an insight into possible adsorption mechanisms that governs the interaction between Br^- and the date pits adsorbent. The effects of experimental controlling conditions such as pH, initial Br^- concentration and solid particle size and mass are considered in order to examine and illustrate their impact on the adsorption isotherms and its related kinetics.

2. Materials and methods

2.1. Adsorbate solution

Standard procedures were used to prepare Br^- stock solution using KBr. HCl and NaOH were used to adjust the pH of the solutions as required. Dilutions were prepared to obtain KBr solutions of 10, 20, 30, 50, 70, 100, and 150 mg L^{-1} in order to elucidate the effect exerted by initial Br^- concentrations. Ion chromatography (850 Professional IC, Metrohm) was used to determine Br^- concentration in the aqueous solutions by following the ASTM standard testing procedures D6581-12 [15].

2.2. Preparation and characterisation of the adsorbents

The Qatari date fruits (*Phoenix dactylifera* L.) were purchased from a local market. The hard seeds of the date fruit (raw date pits = RDPs) were the only part used in this study. Two adsorbents were prepared from the RDPs namely: raw and roasted (RODPs) date pits. Both starting raw materials were hand washed with abundant distilled water to remove dirt and any other inherent pulp from the date pits. The RDPs were dried in an oven for 2 h at 65°C to remove excess water. RDPs were roasted in an electric oven for 3 h at 130°C , producing a brown coloured adsorbent, which will be referred to as roasted date pits (RODPs). The adsorbents were crushed mechanically and sieved separately using sieve shaker (Retsch) with three different particles size ranges namely; 0.125–0.25, 0.25–0.50, and 0.50–1.00 mm. No other chemical or physical treatments were carried out. Samples were then stored in dry sealed glass jars in the refrigerator for further use. Locally available commercial activated carbon (AC), with a specific surface area and cumulative pore volume of $890 \text{ m}^2 \text{ g}^{-1}$ and $0.51 \text{ cm}^3 \text{ g}^{-1}$, respectively, was used as a reference material given its wide spread application in the removal and remediation of numerous pollutants.

The characterisation of the adsorbents in any adsorption system offers vital insights into the processes involved and its governing mechanisms [9,11,13,16]. Accordingly, surface characterisation of the RDPs, RODPs and AC were carried out before and after adsorption. Fourier transform infrared (FTIR – Spectrum 400, PerkinElmer) was also used for the RDPs, RODPs and AC before and after adsorption. The elemental analyses of the RODPs were carried out using X-ray fluorescence (XRF) (JSX 3201M, JEOL). In addition, the specific surface area, pore diameter and cumulative pore volume of the RODPs were analysed using surface area analyser (Micromeritics ASAP 2420) (Barrett-Joyner-Halenda (BJH) Adsorption cumulative).

2.3. Equilibrium adsorption isotherm studies

In the equilibrium isotherm studies of Br^- , RODPs and AC were the only adsorbents used. To conduct this study, Here, different concentrations (10, 20, 30, 50, 70, 100, 150, 200 mg L^{-1}) of KBr solution were prepared and pH the solutions were adjusted as appropriate to 2, 4, 8, and 11. A 50 mL of each concentration was mixed with a constant mass (0.25 g) of adsorbent in a 100 mL glass

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