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An environmentally friendly process; Adsorption of radionuclide Tl-201 on fibrous waste tea

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ABSTRACT

This work presents an investigation of the adsorption of the radionuclide of Tl-201 from waste water on the fibrous tea factory waste. The experimental parameters were chosen as temperature, pH, stirring speed, adsorbent dose and nominal particle size in the ranges of 10.0–40.0 °C, 2.0–10.0, 300–720 rpm, 1.0–15.0 g/L and 0.15–0.71 mm, respectively. The most effective parameter on the adsorption yield was found to be pH of the solution. Fourier transforms infrared and electron paramagnetic resonance spectroscopy studies were performed for the characterisation of the adsorption on tea waste. The experimental data were found to be in good agreement with the isotherm models of Freundlich, Halsey, Handerson and Dubinin–Radushkevich. Thermodynamic analysis showed that the values of ΔG and ΔH are negative. It was obtained that the adsorption rate can be represented very well by second-order pseudo homogeneous kinetic model. All the results proved that fibrous tea plant waste makes an excellent adsorbent for Tl-201 radionuclide.

1. Introduction

There are three fundamental ways for the protection from radiation; the application of the rules of time, distance and shielding. Radiation causes very serious biological effects. The radiation from ionised radioactive sources can cause some changes in molecular level in the cell through which they pass, in similar ways to those caused by X, gamma, alpha and beta rays. These changes can cause temporary (curable) or permanent (non-curable) damages depending upon the type, amount and period of suffered radiation. There is no cell which has absolute resistance to radiation [1].

In nuclear medicine, the production of some solid or liquid radioactive wastes is unavoidable; these wastes can have different chemical structures, activities and concentrations. Liquid radioactive wastes can be classified according to their sources, chemical compositions or salt contents [2].

For the intravenous application in nuclear medicine, the sterilised apyrogenic thallium chloride is prepared from thallium-201 formed by degeneration of Pb-201 radionuclide produced by the bombardment of Tl-203 in cyclotron. Tl-201 radionuclide converts to Hg-201 as follow:

$$Tl^{201} \rightarrow Hg^{201} + \text{gamma ray}[135 - 167\text{Kev}(12\%)] + \text{X-ray}[69 - 83\text{Kev}(88\%)] \quad t_{1/2} = 73.1\,\text{h} \tag{1}$$

The discharge of the radionuclides by patient or of the unused radioactive substances as a waste to the environment can cause serious radioactive problems. The precautions to reduce this problem are to keep the liquid wastes in lead tanks and the solid wastes in lead chambers until their radioactivity level reduces to an allowable value to discharge into the environment. This method is expensive, cumbersome and far away being practical and avoiding the harmful effect of radioactivity [3]. Moreover, this method cannot provide a complete shielding for radioactivity besides the tanks or chambers occupy large areas. Finally, the diluted radionuclides are left to the environment before their activity is reduced to a harmless level. In addition to the radioactive contamination, it causes heavy metal contamination and poisoning because radionuclides convert to stable metal ions in their steady states. When they mix with underground water, their harmful effects become unavoidable [1]. Therefore, the removal of these ionised radioactive substances from liquid wastes is of vital importance. There are various ways of removing these ionised species from waste water such as reverse osmosis, ion exchange, precipitation and coagulation. However, these methods are quite expensive and are not so

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Nomenclature

B_r	resonance field
C	concentration of adsorbate in the solution at equilibrium ($mg\,L^{-1}$)
E	adsorption free energy ($J\,mol^{-1}$)
E_A	activation energy ($J\,mol^{-1}$)
G	Gibbs free energy ($J\,mol^{-1}$)
H	enthalpy ($J\,mol^{-1}$)
h	Planck constant ($6.626068 \times 10^{-34}\,J\,s$)
k	kinetic rate constant
K	constant related to adsorption capacity ($L\,g^{-1}$)
K_1	constant related to adsorption energy ($mol^2\,kJ^{-2}$)
n	constant related to adsorption intensity
q	adsorbed amount per amount adsorbent at equilibrium ($mg\,g^{-1}$)
R	regression coefficient, ideal gas constant ($8314\,J\,K^{-1}\,mol^{-1}$)
S	entropy ($J\,mol^{-1}\,K^{-1}$)
T	absolute temperature (K)
t	time (min)

Subscript

t	any time
t_0	initial period
e	equilibrium

Greek letters

Δ	change
ε	Polany potential
ν	microwave frequency

effective if the waste contains radionuclides in low concentrations [4]. Therefore, after treatment of the liquid waste with one of the above methods, the waste still contains poisonous materials. The use of sorbents for removing radionuclides has some advantages over the other methods [5]. Many microorganisms such as spores, yeasts, bacteria and sea organisms can adsorb anionic and cationic species in liquid medium [6,7].

To remove radioactive species in liquid medium, an ion exchange process was applied, and Iodine-131 was removed by the use of amberlite as ion exchanger [8]. Clinoptilolite, which is a zeolite, was used to adsorb and successful results were obtained [9]. Cr-51 radionuclide used in nuclear medicine was removed by the use of water plants such as *Eichhornia crassipes*, *Pista* sp., *Nymphea alba*, *Menhta aquatica*, *Ephorbia* sp. and *Lemna minor* as adsorbents [10]. Activated carbon was also used for the adsorption of radionuclides in aqueous medium such as uranium [11] and iodine [12].

The aim of this work is to investigate the adsorption of the radioactive ions of Thallium-201 from waste water by using the fibrous tea factory waste as adsorbent and to study the effect of some experimental parameters on the adsorption yield.

2. Materials and method

2.1. Adsorbent

Solid wastes in large amounts are produced in the factories which process agricultural products. These wastes, sometimes, occupy large areas in the operation site of factories, creating some difficulties in the regular operation of the factories. Approximately, 20,000 tonnes tea waste is produced in the official tea factories in Turkey. When the private sector is considered, this amount rises to about 30,000 tonnes [13]. The tea factory waste used as adsorbent was provided from Cumhuriyet tea plant located in Rize in Black Sea Region of Turkey. There are two types of the tea factory waste in Cumhuriyet tea plant such as subjacent of sieve and chimney. The fibrous tea factory waste (FTFW) of the chimney subjacent was used in this research. The solid waste first was ground and then sieved to obtain desired particle size fractions.

Table 1
Some properties of waste tea used in experiments

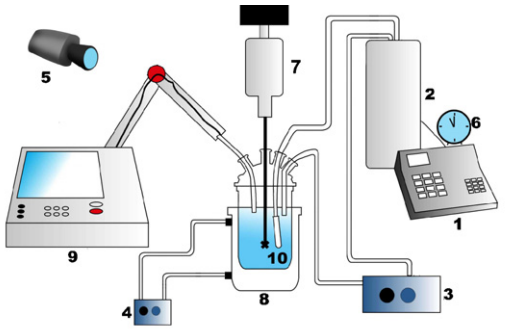
Bulk density (g/cm^3)	0.119
Particle size (mm)	0.212–0.150
BET surface area (m^2/g)	0.702

Some properties of the tea factory waste are presented in Table 1. Prior to the experiments, other soluble contaminants and coloured components were removed from the waste tea by washing it with distilled water for a few times until a colourless filtrate of the washing water was spectrometrically observed at room temperature. The decolourized and cleaned tea waste was dried at room temperature for a few days, and then used in the adsorption experiments.

2.2. Experimental system

The system shown in Fig. 1 was employed in the adsorption experiments. In the design of this experimental system, all the necessary safety considerations were taken into account so that the person who carries out the experiments will expose to radioactivity at possible minimum level. A 1000-mL jacketed vessel was used as the adsorption medium. A mechanical mixer was attached to the system to stir the content of the vessel during the adsorption process. A circulating bath was employed to keep the adsorption medium at constant temperature by circulating the cooling/heating fluid through the jacket. A master flex pump was used to circulate the aqueous solution through the radiation dosimeter to measure automatically the radioactivity of the solution. The circulation of the adsorption solid together with the solution was avoided by using a suitable filter, which does not adsorb the radioactive substance, just at the outlet of the solution from the vessel. All the necessary precautions were taken for an accurate measurement with the dosimeter. The experimental readings of digital screens were recorded by a video camera to minimize the involvement of the researcher with the radioactive medium.

The experimental parameters were chosen as temperature, pH, stirring speed, adsorbent dose and nominal particle size. The ranges of these parameters are given in Table 2. When the effect of one parameter on the adsorption was investigated, the values of the



1. Radioactivity dosimeter
2. Radioactivity reading chamber
3. Pump
4. Circulating bath
5. Video camera
6. Clock
7. Mechanical stirrer
8. Jacketed adsorption vessel
9. pH meter
10. Filter

Fig. 1. Experimental system.