



Fast and efficient cesium removal from simulated radioactive liquid waste by an isotope dilution–precipitate flotation process



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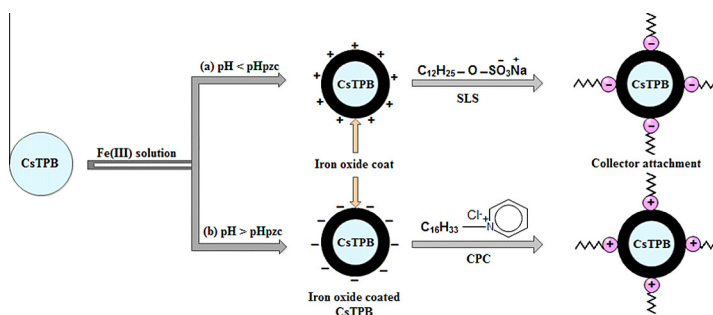
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HIGHLIGHTS

- Removal of Cs-137 by an isotope dilution–precipitate flotation was studied.
- Sodium tetraphenylborate precipitating agent was used with SLS and CPC surfactants.
- Lower remaining Cs-137 activity values than the permissible value are obtained.
- CsTPB precipitate was efficiently separated within half a minute.
- The removal mechanism of cesium by SLS and CPC was proposed at different pH values.

GRAPHICAL ABSTRACT



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ABSTRACT

The performance of an isotope dilution–precipitate flotation process for Cs-137 removal from low-level liquid radioactive waste is evaluated in this study using sodium tetraphenylborate (NaTPB) as the precipitating agent. In absence of Fe(III), neither the anionic sodium lauryl sulfate (SLS) nor the cationic cetylpyridinium chloride (CPC) could float the CsTPB precipitate over the studied pH range of 2.5–11.9. It is proposed that iron oxide coated CsTPB particles are formed upon addition of Fe(III) solution and their floatability is greatly dependent on the solution pH and the Fe(III) content. At the optimum conditions, remaining Cs-137 activity values <1.3 nCi/L (SLS, pH ~6.8) and <2.3 nCi/L (CPC, pH ~10.2) are achieved from radioactive wastewater within half a minute which are below its discharge limits. The mechanism of flotation is proposed and cesium removal is suggested to proceed via flotation of iron oxide coated CsTPB. Flotation technique is compared to other conventional solid–liquid separation processes (decantation, centrifugation and filtration) for separation of the iron oxide coated CsTPB particles. The results showed that the best separation was attained by flotation process. The data obtained by the present combined process, precipitation followed by flotation, for Cs-137 from radioactive wastewater are also compared to those reported by other treatment methods. Based on the data obtained, the combined process has great potential as a radioactive wastewater treatment technology.

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1. Introduction

Due to its long half-life ($T_{1/2} = 30.17$ years), high fission yield (6.5%), high solubility/mobility and high radiotoxicity, the Cs-137

radionuclides-contaminated radioactive liquid wastes must be effectively treated prior its release into the environment. Chemical precipitation, ion exchange, membrane separation solvent extraction and adsorption are the usual methods used for the treatment of radioactive liquid wastes [1–4]. Among them, chemical precipitation method is a well-established method for the removal of radionuclides from radioactive wastewaters and is

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in regular use [5]. Unfortunately, precipitation of alkali metals is mostly difficult since their salts usually are very water soluble [6]. Moreover, this method is inappropriate for treatment of wastewaters having very low concentrations of the radionuclide. Herein, an isotope dilution-precipitation process has been used in this work. This process is based on the utilization of stable cesium (Cs-133) as the carrier and sodium tetraphenylborate (NaTPB) as the precipitating agent. When coexisted with Cs-137 in the aqueous solution, the stable cesium will increase the total cesium concentration and thus the solubility product for cesium tetraphenylborate (CsTPB), $K_{sp} = 1.76 \times 10^{-9} \text{ mol}^2/\text{L}^2$, can be exceeded [7].

Numerous previous studies reported that NaTPB forms poorly soluble CsTPB with cesium in aqueous solutions [1,6–9] and thus providing a promising method for removal of radiocesium from radioactive liquid wastes. Other precipitating agents are also reported in literature for cesium [6]. Nevertheless, CsTPB had the lowest K_{sp} of all those cited. Based on this finding, NaTPB was used in this study as a precipitating agent for cesium.

For precipitation of Cs-137 from nuclear wastes, much more precipitating agent than necessary is required. This is because not only CsTPB is formed, but also other alkali metal salts (e.g. KTPB). The radioactivity of the nuclear waste, especially the high level radioactive one, causes radiolytic decomposition of TPB and primarily environmentally harmful benzene is formed [10]. Therefore, the solid phase (CsTPB) should be separated as soon as formed, instead of storage, to avoid such decomposition. To achieve this goal, a fast and efficient solid-liquid separation process should be used. Generally, separation of the precipitate during chemical precipitation treatment is often problematic. This is because the traditional solid-liquid separation processes (decantation, centrifugation, evaporation and membrane filtration) facing many challenges during their application particularly when dealing with large volumes of wastewaters. Consequently, foam separation (or simply flotation) has been evaluated in this study as an alternative technique.

Precipitate flotation is a foam separation process that involves the precipitation of the ionic species to be removed, using a suitable precipitating agent, prior to its flotation with an adequate surfactant (collector). The interaction between the collector and the precipitate produces hydrophobic particles which then become able to attach to the air bubbles, introduced into the suspension, and is then removed in the foam phase [11]. In this work, NaTPB is used as a precipitating agent for cesium and two types of surfactants, namely, anionic sodium lauryl sulphate (SLS) and cationic cetylpyridinium chloride (CPC), are used as collectors. Besides its prosperous use for the treatment of water and wastewater [12,13], flotation has been also broadly applied as a solid-liquid separation technique [11,14–16]. Though, no publications are reported on the removal of cesium radionuclides from aqueous solutions by precipitate flotation process, suggested in the present work, using the NaTPB precipitating agent.

The aim of the present work was to determine the adequate conditions for the removal of Cs-137 from low-level liquid radioactive waste (LLRW) by precipitate flotation. To achieve this purpose, the effects of the solution pH, collector type and concentration and stable cesium concentration were studied. The employed process, flotation, was also compared to the conventional solid-liquid separation processes for Cs-137 removal from LLRW.

2. Experimental

2.1. Flotation apparatus and reagents

The flotation system used in this work was previously described in detail [14]. In essence, it is consisted of a pure nitrogen cylinder

connected to a flotation column through a fine pressure reduction unit, manometer and filtered flasks containing baryta solution and water. The flotation column, in which the flotation experiments were carried out, is supported over a filter flask and is made of a G4 sintered glass disc 4 cm in diameter fused to a Pyrex glass column about 40 cm in height, drawn at the bottom in the form of a funnel. A tap is sealed to the flotation column at a distance of 0.8 cm above the fritted disk to enable sample withdrawing from the bulk solution for radioactivity and pH measurement. Nitrogen gas was introduced through the fritted disk at a rate of $25 \text{ cm}^3/\text{min}$.

Sodium tetraphenylborate, NaTPB, was obtained from Merck. Fresh solution of the desired concentration prepared with NaTPB was kept in a dark bottle to avoid photochemical decomposition. Stable cesium chloride having a purity of 99.99% and ferric nitrate nonahydrate were obtained from Sigma-Aldrich. The radioactive cesium, Cs-137, was purchased from the Amersham Radiochemical Center and was used for solution spiking. The collectors, sodium lauryl sulfate (SLS) and cetylpyridinium chloride (CPC), were obtained from Merck. The collector solutions were freshly prepared daily. Absolute ethanol, provided by Merck, was used as a solvent for CPC as well as a frother in order to decrease the bubble size and increase the foam stabilization. Because SLS is not soluble in absolute ethanol, it was dissolved in 50% (v/v) solution of ethanol and water to allow the simultaneous addition of the collector and the frother. During all the experimental work, ethanol was added at a dose rate of 1 mL per 250 mL aqueous suspension. Adjusting the solution pH was done with reagent grade sodium hydroxide and hydrochloric acid. The chemical composition of the synthetic low-level liquid radioactive waste (LLRW) tested in this work is represented in Table 1.

2.2. Operational procedure, analysis and data presentation

In the present work, the experimental procedure for cesium removal involves two steps: (i) precipitation and (ii) flotation. For the first step, precipitation, NaTPB was added to a solution of stable cesium spiked with Cs-137 and CsTPB precipitate was immediately formed according to the following reaction:



For all experiments, the initial Cs-137 activity was 30 nCi/L and NaTPB was added to produce a 1:1 M ratio of cesium to tetraphenylborate ion. The initial stable cesium concentrations were varying from 5×10^{-5} to $7.5 \times 10^{-4} \text{ mol/L}$. After NaTPB addition, the suspensions were stirred at 200 rpm for 5 min using a magnetic stirrer. Ferric nitrate solution, of the desired concentration, was then added and the pH of the mixture (Cs-TPB/Fe(III)) was adjusted to the desired value. Except for studying the effect

Table 1
Characteristics of LLRW.

Characteristics	Value
Total dissolved solids	9.35
pH	6.85
Chloride	4
Sulfate	58
Phosphate	0.3
Carbonate	158.5
Oxalate	0.028
Potassium	16.5
Sodium	34
Magnesium	18
Calcium	32
Iron	0.1

All units are in mg/L except pH.

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