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Species removal from aqueous radioactive waste by deep-bed filtration☆

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ABSTRACT

Performances of aqueous suspension treatment by deep-bed sand filtration were experimentally studied and simulated. A semiempirical deterministic model and a stochastic model were used to predict the removal of clay particles (20 μm) from diluted suspensions. Model parameters, which were fitted based on experimental data, were linked by multiple linear correlations to the process factors, *i.e.*, sand grain size (0.5 and 0.8 mm), bed depth (0.2 and 0.4 m), clay concentration in the feed suspension (1 and 2 kg_p/m^3), suspension superficial velocity (0.015 and 0.020 m/s), and operating temperature (25 and 45 °C). These relationships were used to predict the bed radioactivity determined by the deposition of radioactive suspended particles (>50 nm) from low and medium level aqueous radioactive waste. A deterministic model based on mass balance, kinetic, and interface equilibrium equations was developed to predict the multicomponent sorption of ^{60}Co, ^{137}Cs, ^{241}Am, and ^{3}H radionuclides (0.1–0.3 nm). A removal of 98.7% of radioactive particles was attained by filtering a radioactive wastewater volume of 10 m^3 (0.5 mm sand grain size, 0.3 m bed depth, 0.223 kg_p/m^3 suspended solid concentration in the feed suspension, 0.003 m/s suspension superficial velocity, and 25 °C operating temperature). Predicted results revealed that the bed radioactivity determined by the sorption of radionuclides (0.01 kBq/kg_b) was significantly lower than the bed radioactivities caused by the deposition of radioactive particles (0.5–1.8 kBq/kg_b).

1. Introduction

Deep-bed filtration (DBF) is a method widely used for separation of inorganic and organic particles from suspensions with low levels of suspended solids. When a diluted suspension flows through a filtering granular bed, the particles to be removed approach the filter grains (collectors) by various transport mechanisms (direct interception, inertia, sedimentation, molecular diffusion, electrostatic forces, hydrodynamic action) and they can further adhere to the collector surface (by van der Waals forces, electric double layer interactions, hydrogen bonding, mutual adsorption of polymeric species) (Cushing and Lawler, 1988; Ding et al., 2018; Dobre and Sanchez Marcano, 2007; Gitis et al., 2010; Ives, 1987; Jegatheesan and Vigneswaran, 2005; Rege and Fogler, 1988; Rushton et al., 2000; Šećerov Sokolović et al., 2009; Vaz et al., 2017; Zamani and Maini, 2009). DBF involves two stages, *i.e.*, an initial (clean bed) one, wherein the deposition of particles occurs on the clean filter grains, followed by a transient stage, wherein the particles are deposited on the filter grains which are partially covered by other particles (Jegatheesan and Vigneswaran, 2005; Zamani and Maini, 2009). In the transient stage both attachment and detachment forces should be considered.

DBF performances, usually evaluated in terms of suspended solid concentration in the effluent (filtrate quality) and head loss across the bed, are affected by various process factors. Accordingly, they depend on physical and chemical characteristics of both filtration medium (material, shape, size distribution, density, and surface charge of filter grains, depth and void fraction of fixed bed) and suspension (type, shape, size distribution, surface charge, density and concentration of particles, age and viscosity of suspension) as well as on the operating conditions (suspension flow rate, operating temperature and pressure).

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Sand, anthracite coal, granular activated carbon, garnet, natural zeolites, synthetic inorganic sorbents, ion exchange resins, glass and expanded polystyrene beads, are common granular materials used to remove TSS from a diluted suspension by DBF (Altmann et al., 2016; Cushing and Lawler, 1988; Gitis et al., 2010; Huang et al., 2012; Ives, 1987; Löwenberg et al., 2016; Rushton et al., 2000; Šećerov Sokolović et al., 2009). Rapid sand filtration is widely applied to water treatment (EPA, 1995; Ives, 1987).

In aqueous radioactive waste (ARW) processing, the knowledge that exists on water purification can be applied. However, there are some additional problems referring to the specialized requirements of radioactive waste management. ARW treatment by deep-bed sand filtration (DBSF) could lead to an effluent with an acceptable level of residual radioactivity for discharge or further processing by other separation methods (*e.g.*, ion exchange, fixed-bed adsorption, ultrafiltration, reverse osmosis) as well as a low amount of secondary radioactive waste (Abdel Rahman et al., 2011; Huang et al., 2012). Moreover, the rapid sand filters involve simple and relatively inexpensive operation and maintenance. The first application of DBSF in nuclear engineering dates from 1948, when this process was used to air purification (DOE HDBK, 2003).

The process control based on modeling could offer valuable information that may be applied to reduce the treatment costs and to protect the health of employees. A DBSF model for the removal of radioactive pollutants from medium and low level ARW could be very useful to design and optimize the process, because it determines a decrease in the number of experiments, which can be expensive, may require considerable time and, most important, increase the radiological dose of workers.

The quality of ARW treated by rapid sand filtration was measured and simulated in this paper. Mathematical models were adopted to predict the process performance and their adjustable parameters were fitted based on experimental data. The models developed to describe DBF process could be coupled with characteristic models of further ARW processing for predicting the performances of integrated separations/purifications.

2. Experimental

Two experimental studies of DBSF were conducted under various operating conditions using silica sand as a filtering medium. Diluted clay suspensions were processed in the first study, whereas ARW was treated in the second one.

2.1. DBSF of clay suspensions

DBSF experiments were performed using a pilot scale setup (Fig. 1) consisting of: (i) a metallic column (90 cm height and 5 cm inner diameter) equipped with manometers at its top and bottom, (ii) a pressurized vessel (400 L volume) for suspension storage, (iii) a compressed air supply for maintaining a constant pressure (1.5–2 bar) in the storage vessel, and (iv) flow rate control valves.

Gravel was placed on a sieve at the column bottom to support the granular bed and to distribute uniformly the suspension. Uniform sand grains, either 0.5 or 0.8 mm diameter (D), were packed in the column resulting in bed void fractions (ε_b) of 0.324 and 0.377 $\mathrm{m^3/m_b^3}$, respectively. Clay suspensions were obtained by adding 200 and 400 g of clay particles (20 μm mean diameter (d_p) and 1400 $\mathrm{kg_p/m_p^3}$ density (ρ_p)), respectively, in 200 L tap water. Pressure difference between the bed inlet and outlet ($\Delta p = p_{in}\text{-}p_{out}$) was continuously measured.

Effluent samples were collected at regular intervals and mass concentration of particles (c_p) was determined for each sample. A volume of sample was filtered on a Whatman® quantitative filter paper, ashless, Grade 40 (8 μm pore size). The filter paper with solid residue retained was dried into an oven at 105 °C for 2 h, placed into a desiccator to cool to room temperature, and then weighed. The effects of sand grain size (D=0.5, 0.8 mm), bed depth ($H = 0.2$, 0.4 m), mass concentration of particles in the influent ($c_{p0} = 1$, 2 $\mathrm{kg_p/m^3}$), suspension superficial velocity ($w = 0.015$, 0.020 m/s), and operating temperature ($t = 25$, 45 °C) on c_p/c_{p0} were evaluated.

2.2. DBSF of ARW

Experiments of DBSF were conducted in a column (Fig. 2) of a filtration module for ARW treatment. It consisted of three metallic columns (114 cm height and 35 cm inner diameter), tanks for ARW and effluent storage, a submersible pump for feed, piping, measuring and control equipment.

Samples from collecting tank were taken at regular intervals and mass concentration of particles (c_p^c) as well as activity concentration of radionuclides (^{60}Co, ^{137}Cs, ^{241}Am, and ^{3}H) in suspended particles and liquid phase were measured for each sample. TSS (c_p^c) was determined by filtering on a 50 nm Whatman membrane using a Varian DS 102A vacuum pump (Agilent). The membrane was further dried into an oven at 105 °C for 2 h, cooled to room temperature in a desiccator, and then weighed. Activity concentrations of γ-emitting radionuclides (^{60}Co, ^{137}Cs, and ^{241}Am) in the particles

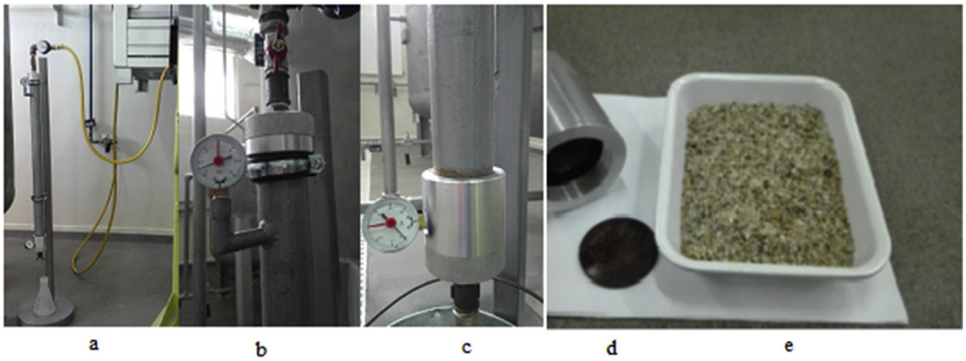

Fig. 1. Pilot scale setup for DBSF of clay suspensions:
(a) fixed bed column and filtrate collector vessel, (b) valve and manometer at the column top, (c) manometer at the column bottom, (d) sieve, and (e) gravel for supporting the fixed-bed sand.