Journal of Materials Science and Engineering A 5 (7-8) (2015) 313-317
doi: 10.17265/2161-6213/2015.7-8.006

Study of Coprecipitation of Strontium (Sr^{2+}) with Barium Sulphate ($BaSO_4$) in Aqueous Solution

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Abstract: The objective of this work is to highlight the phenomenon of coprecipitation of strontium at low concentrations and in varying proportions, in order to establish and optimize the parameters that affect this process of decontamination of radioactive liquid effluents containing strontium. The co-precipitation method is one of the most used in the nuclear industry for the treatment of radioactive liquid waste as it can be applied to all effluent whatever their composition. This process consists of forming in situ by precipitation solid particles in order to selectively capture one or more radioisotopes. This study allowed defining the best operating conditions for treatment of synthetic wastewater containing strontium. In fact, various experimental parameters such as stirring speed and stirring time, pH, conductivity of the solution and the initial effluent concentration of strontium, were selected for the experimental study of the phenomenon of coprecipitation in semi-batch reactor. The decontamination technology by coprecipitation is an interesting and alternative method for treating liquid radioactive effluents.

Key words: Liquid waste treatment, coprecipitation, strontium, barium sulphate.

1. Introduction

A variety of radioactive wastes is generated in a broad range of activities involving the use of radioactive materials in medicine, universities, industrial applications and research institutions. Among these, radioactive liquid effluents are the subject to special monitoring. Most of these radioactive wastes are referred to as LLWs (Low-level wastes) [1]. Like many LLWs can greatly harm humans, they must be treated to reduce their content of radioactive contaminants to such an extent that the decontaminated volume of aqueous waste can be either released to the environment or recycled [2]. Our study consists in using the coprecipitation as decontamination technique to reduce or eliminate radioactive chemical elements, including strontium, which contains a radioactive effluent.

Coprecipitation is a phenomenon where a solute that would normally remain dissolved in a solution precipitates out on a carrier that forces it to bind together, rather than remaining dispersed. It is typically defined as the simultaneous removal of both the solute and carrier constituents from an aqueous solution without regard to the specific mechanisms involved [3]. It also involves in situ formation of solid particles by precipitation in order to selectively capture one or more radionuclides. This coprecipitation phenomenon involved in the chemical treatment of radioactive liquid waste can be decomposed into two processes taking place simultaneously: the formation of the solid phase host by precipitation and incorporation therein of the compound present in trace amounts in the solution as strontium [4]. Indeed, 90Sr is one of the major heat producers and biohazards in nuclear wastes. The removal of radioactive strontium is essential for reducing the risk of human exposure to radiation and for realizing considerable cost savings through the minimization of the storage-space requirements for nuclear wastes [5].

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In this work, experiments were conducted in inactive with synthetic solutions containing a low concentration of strontium in variable proportions, to establish and optimize the parameters affecting this process of decontamination of radioactive liquid effluents. The operating parameters will be determined, taking into account, physicochemical conditions are similar to those of real effluents and particle characteristics. For this, we used a synthetic solution containing non-radioactive strontium, since, when the element has stable isotopes, the analogy of behavior between its stable and radioactive isotopes is implicitly accepted, it does not therefore seem exist significant difference between the behavior of radioactive strontium and that of stable isotope. Thus, mobility, geochemistry, speciation of radioactive strontium is the same as that of another non-radioactive strontium isotope.

2. Materials and Methods

In the industrial plants, significant quantities (2-20 g/L) of barium sulphate are used to decontaminate effluents containing the radioactive strontium [7]. This adsorbent is the most used in the effluents treatment plant, it is also a compound studied in depth, often used as a model substance for the study of precipitation. Liquid nuclear effluents often contain a high concentration of soluble salts (10-100 g/L), particularly sodium nitrate [6, 7]. To avoid dilution of the liquid waste, sodium sulphate is dissolved into the effluent and a high concentration solution of barium nitrate is used [7].

Taking these particularities into account, our decontamination experiments are conducted with a simulated effluent containing sodium nitrate, sodium sulphate and nitrate strontium as the source of strontium. This solution of strontium is treated with barium sulphate in a semi-batch reactor.

The experimental apparatus used to perform the coprécipitation experiences of strontium by barium sulphate in semi-batch reactor consists of a glass beaker with a useful volume of 1,000 ml, used as a reactor in which we introduced a given volume of effluent of determined concentration of strontium. We conduct a stirring to ensure the contact solution / precipitate. Stirring is provided by a glass propeller which rotates by a mechanical motor. A conductivity probe and a pH probe are immersed in the reaction medium. The conductivity probe allows to monitor the evolution conversion of the reaction. The pH probe is used to control the gap at the initial pH.

The operating protocol consists, in the first instance, to fill the reactor with 500 ml of simulated effluent of concentration determined out of strontium at which a given concentration of barium nitrate is added thereby established volume. This addition constitutes the initial moment of the co-precipitation experience. Once the injection is complete, the reactor is stirred. The separation of solid and liquid phases is achieved by centrifugation at 3,000 rpm for 15 min. [8]. All coprecipitation experiments of strontium with barium sulphate were performed at room temperature (23-25 °C). The pH of the simulated effluent is initially adjusted to 9 with sodium hydroxide solution at 0.1 mol/L. All chemicals used in this study were of analytical quality and the deionized water used to prepare aqueous solutions. Different concentrations of solute were considered during experiments.

3. Results and Discussion

The method used for the analysis of liquid samples is the flame Atomic Absorption Spectrometry which allows to be assayed essentially the metals in solution. The measurement of the concentration of strontium before and after coprecipitation reaction, permit to determine the DF (Decontamination factor). The latter allows the assessment of treatment efficacy.

3.1 Kinetic Study of Agitation

In order to study the effect of stirring time and stirring speed, kinetic studies of co-precipitation were performed to determine and optimize the stirring time and stirring speed. For this, a volume of 500 ml of simulated effluent of 8.638 mg/L was introduced into a beaker and a predetermined volume of the solution of barium sulphate is added after adjusting the pH to 9. The mixture was stirred at various times and agitation speeds. The range of agitation speeds considered varies from 100 to 400 rpm. The results on the influence of stirring time, stirring speed and the corresponding decontamination factor, are shown in Fig. 1.

3.1.1. Influence of Stirring Time

The results show that the residual concentration of strontium decreases with the stirring time; this implies an increase in the percentage of strontium incorporated into the precipitate of barium sulphate formed, until reach a plateau indicating time required to have the maximum of strontium coprecipitate with barium sulphate. A time of 60 minutes appeared optimum for the co-precipitation of strontium with barium sulphate. This result is in agreement with previous studies [1], for which a stirring time of 1 h was used. The decontamination factor achieved for this time of 60 min is 93, 80, 192 and 64 for stirring speeds 100, 200, 300 and 400 rpm, respectively.

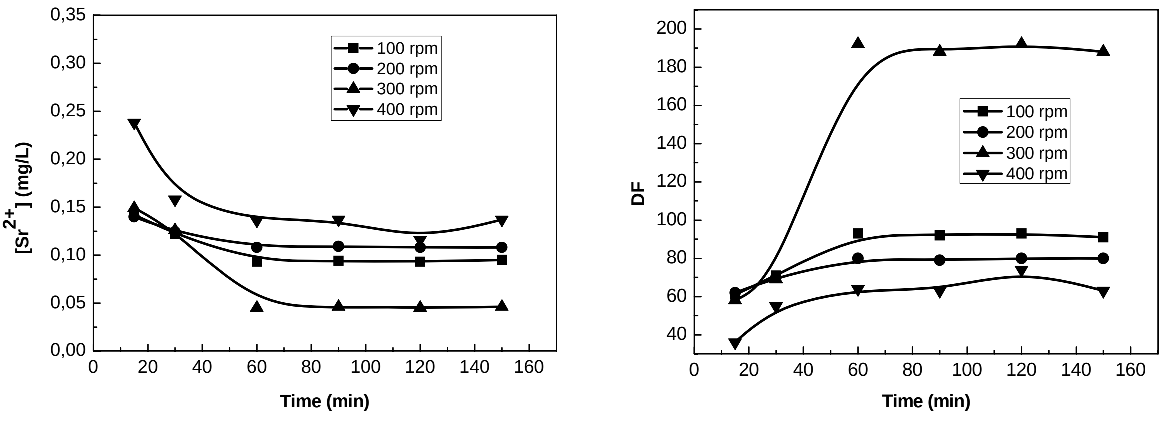
3.1.2. Influence of Stirring Speed

The stirring speed plays a crucial role in the effectiveness of the method of coprecipitation in discontinuous system, it must be sufficient to allow a good homogenization of the two phases (the solution and the precipitate) and also to maintain the solid particles in suspension.

According to the results obtained, we notice that the best decontamination factor is obtained at a stirring speed of 300 rpm. This result agrees with the study [7] for which a speed of 300 rpm was taken as the reference speed for all experiments of coprecipitation.

3.2 Evolution of PH and Conductivity as a Function of Time

The pH determines the acidity or alkalinity of the water that corresponds to the activity of H^+ ions in the water. Conductivity allows an overall assessment of the water content of dissolved salt and therefore provides a good assessment of the material in solution. Fig. 2 presents the evolutions of the pH and conductivity during the experiment of co-precipitation in semi-batch reactor. Before adjusting the pH of the solution, the conductivity is of 39.9 ms/cm and after the adjustment, it drops to 39.8 µS/cm (not shown on graph). After the injection of the solution of barium nitrate, it still falls to 38.5 µS/cm then no longer changes significantly thereafter. The fall of the conductivity is caused by the consumption of barium and sulphate ions by formation of barium sulphate. For the pH, after its adjustment and adding the solution



(a) (b)

Fig. 1 (a) Effect of stirring time and stirring speed on the co-precipitation of strontium; (b) Decontamination factors obtained at different stirring times and stirring speeds.

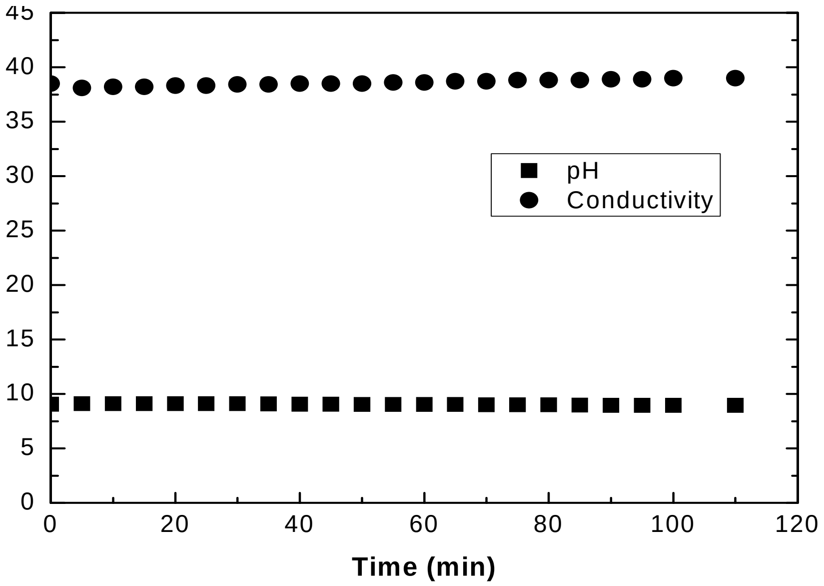
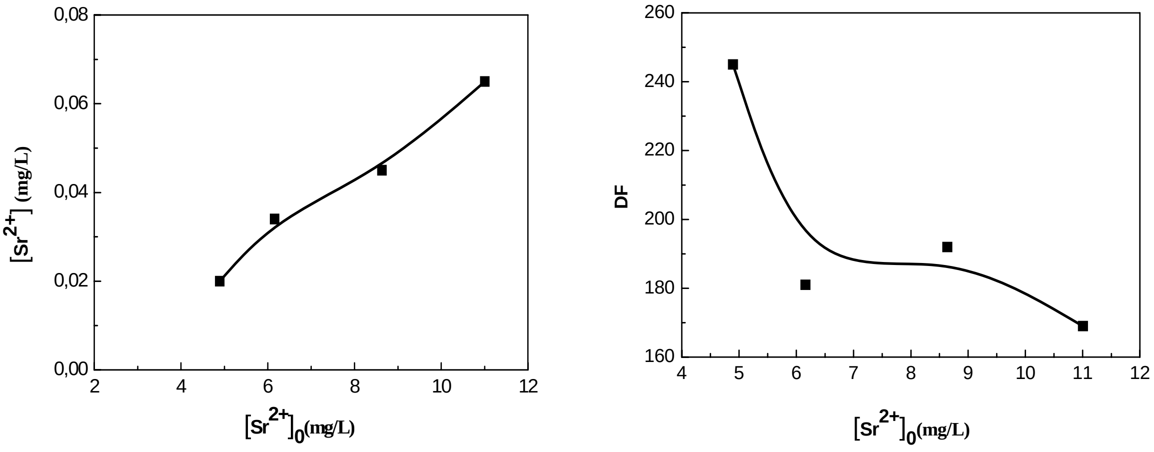


Fig. 2 Evolution of pH and conductivity as a function of time during the experiment of coprecipitation.



(a) (b)

Fig. 3 (a) Influence of the initial concentration of strontium; (b) Evolution of the decontamination factor depending on the initial concentration of strontium.

of barium nitrate, the pH of the solution does not vary appreciably. It oscillates between 9 to 8.9.

3.3 Influence of the Initial Concentration of Strontium

In order to study the effect of the initial concentration of strontium, the solutions of simulated effluents with various initial concentrations of strontium (4.895, 6.160, 8.638 and 11.007 mg/L) were prepared. For each solution, a given volume of barium sulphate of concentration thus established is added, after having adjusted the pH to 9. These solutions are mechanically agitated at 300 rpm for 1 h. Then, the residual concentration of strontium was analyzed by flame Atomic Absorption Spectrometry.

Figs. 3a and 3b, respectively, represent the influence of the initial concentration of strontium and the decontamination factor changing depending on the initial concentration of strontium. According to the results, it was found that the amount of residual strontium solution increases with the increase of the initial concentration of strontium in the effluent, which means that the percentage removal of strontium decreases with increasing of the initial concentration. Indeed, the decontamination factor decreases with the increase of the initial concentration of strontium in the effluent.

4. Conclusions

The co-precipitation method is universally used to remove the radionuclides contained in the radioactive liquid effluents, and it was still the subject of a great number of researches in the world. The coprecipitation of strontium with barium sulphate was studied in order to examine the effect of various operating parameters on the reduction of strontium. The tests were carried out in different operating conditions on synthetic water-containing strontium. The results obtained allow to determine a time of 60 min required to have the maximum of strontium coprecipitated with barium sulphate; the best decontamination factor is obtained at a stirring speed of 300 rpm; pH and conductivity of the solution do not vary significantly as a function of time during the experience of coprecipitation. In the end, the decontamination factor decreases with the increase of the initial concentration of strontium in the effluent. These results allow us to affirm that the decontamination process by coprecipitation is an interesting and alternative method for liquid radioactive wastes treatment.

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