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Sequential separation of Fe and Sr from liquid samples by using Sr resin and rapid determination of ^{55}Fe and $^{89,90}\text{Sr}$

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HIGHLIGHTS

- Fe (III) and Sr (II) ions can be separated by using Sr resin and mixture of HCl and HNO_3 .
- The optimal binding strength is achieved in 6 M HCl + 3 M HNO_3 .
- Varying of concentrations of acids in the mixture allows their selective separation.
- Separation of Sr and Fe on Sr resin enables simple determination of $^{89,90}\text{Sr}$ and ^{55}Fe .

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ABSTRACT

The obtained results showed that the Sr resin can be used for the chromatographic separation of Fe (III) and Sr (II) ions by using a mixture of HCl and HNO_3 . It was shown that the binding strength of Fe (III) decreases with increasing concentration of HNO_3 while the binding strength of Sr (II) increases. It was found that the optimal bonding strength is achieved in 6 mol/L HCl:3 mol/L HNO_3 and varying concentrations of acid in the mixture allow their selective separation. On basis of these results, a method for the separation of ^{55}Fe and $^{89,90}\text{Sr}$ from liquid samples which consists of binding of Fe and Sr on Sr resin and separation from a number of elements by 6 mol/L HCl:3 mol/L HNO_3 and their mutual separation by eluting of Sr with 4 mol/L HCl is established. The method enables efficient simultaneous separation (with high recovery) of $^{89,90}\text{Sr}$ and ^{55}Fe and in combination with Čerenkov counting rapid determination of $^{89,90}\text{Sr}$.

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1. Introduction

As a rule, the determination of ^{55}Fe and strontium isotopes is complicated and time-consuming, especially the determination of $^{89,90}\text{Sr}$. Therefore, numerous methods have been developed for the analysis of radioactive strontium isolation (Horwitz et al., 1991, 1992; Mellado et al., 2005; Stamoulis et al., 2007; Mosqueda et al., 2007; Jäggi and Eikenberg, 2009; Günther et al., 2009; Heckel and Vogl, 2009; Ometakova et al., 2011) in various kinds of samples, but only few methods for ^{55}Fe determination mainly in liquid radioactive waste samples (Grahek and Rožmarić Mačefat, 2004; Hou et al., 2005; Mellado et al., 2005; Jäggi et al., 2008). One reason for this lies in the fact that strontium isotopes are highly radiotoxic fission products (especially ^{90}Sr with long half-life, 28.80 years), and as such they were an interesting subject for various kinds of investigations, from their distribution and behavior in natural systems to the influences on the human health. In distinction from strontium

isotopes, ^{55}Fe is an activation product, low energetic emitter with approximately 2.747 years half life. As such, it was not as interesting as strontium, but environmental concerns prompted the responsible institutions in the radiological monitoring program to include ^{55}Fe determinations in their procedures.

There are few published methods where $^{89,90}\text{Sr}$ and ^{55}Fe are separated in a single step, prior to measurement. The aim of this paper is to develop a method for the single step separation of ^{55}Fe and $^{89,90}\text{Sr}$ from liquid samples by using Sr specific resin with a mixture of nitric and hydrochloric acid as an eluant. It is well known that Sr specific resin is extremely selective and specific for Sr, enabling the separation of Sr from a range of elements on Sr resin column by using HNO_3 solution including Fe (Horwitz et al., 1992; Horwitz, 1993). However, Fe can also be bound on Sr specific resin if a mixture of HNO_3 and HCl is employed. As noted above, the Sr specific resin enables very efficient separation of Sr from many other elements, especially Ca and Pb, while Fe is closely studied. Therefore, this study will show how Fe and Sr can be bound and mutually separated on Sr resin and how this method can be used for their rapid determination in liquid samples. The binding strength and selectivity of Sr resin depend on the

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concentrations of HCl and HNO₃ and the separation of Fe from Sr can be achieved by altering the concentrations of these two acids. The outcome of this work is a method for rapid determination of ⁵⁵Fe and ^{89,90}Sr that has been tested using proficiency test samples and radioactive waste samples; these results are presented and discussed below.

2. Experimental

The data presented in this paper define the conditions under which the single step separation of Fe and Sr may be reliably achieved. The distribution coefficients and their variation under different acid concentrations are presented for Fe, Sr and a number of other cations on Sr specific resin. These data allow the optimization of the separation of Fe and Sr. This procedure was used for the determination of ⁵⁵Fe and ^{89,90}Sr in liquid samples and has been validated by the determination of these isotopes in crosscheck samples with known activity and in waste samples where iron and strontium isotopes were determined with classical methods.

2.1. Distribution coefficients and elution curve determination

The batch technique was used for the measurement of the distribution coefficients. To carry out this determination, 0.2 g of Sr resin were equilibrated for 24 h with 10 ml of HNO₃ and HCl solutions containing the metal cations being studied.

The distribution coefficient of the metal cations was calculated as follows:

$$K_d = \frac{C_0 - C}{C} \cdot \frac{V}{m}$$

where C_0 is the initial concentration of cations in solution, C is the concentration in solution after equilibrating, V is the solution volume (ml), and m is the mass of resin (g).

The behavior of Fe and Sr on the Sr columns was examined. Solutions of these elements in a mixture of nitric and hydrochloric acid were employed for the determination of the behavior of Fe and Sr on Sr specific resin and their separation from other cations. The sorption/desorption process was monitored by determining cation concentrations in the appropriate fractions by Atomic Absorption Spectrometry (AAS). This resulted in the basic procedure for isolation of Sr and Fe in one step as shown in Fig. 1.

2.2. Model and real samples of radioactive liquid waste

The model solution was obtained by mixing 1 l of deionised water with a 1 ml of strontium [5 mg l⁻¹] and iron [2.5 mg l⁻¹] carrier and low concentration of other cations and 1 ml of cross check samples of ⁵⁵Fe and ^{89,90}Sr obtained from Eckert and Ziegler Analytix (E&Z). The liquid cross check samples obtained from E&Z contained mixtures of isotopes (β- and γ-ray emitters) with a known activity of ^{89,90}Sr and ⁵⁵Fe, although the uncertainties were not specified. Real samples were obtained from NPP Krško and doped with ^{89,90}Sr.

2.3. Isolation of iron and strontium from model and real liquid radioactive waste samples

2.3.1. Procedure I

1 ml of Fe [2.5 mg l⁻¹] and Sr [5 mg l⁻¹] carriers were put into 1 l of liquid waste samples and evaporated almost to dryness and 30 ml of 6 mol/L HCl:3 mol/L HNO₃ were added. This solution was passed through the Sr column. If a combination of Sr and TRU

column is to be used, evaporated samples should be dissolved in 5 mol/L HNO₃.

2.3.2. Procedure II

1 ml of Fe [2.5 mg l⁻¹] and Sr [5 mg l⁻¹] carriers were added into 1 l of liquid waste with intense stirring. 2 g of cation exchanger IR-120 were added to this solution. This suspension was left for 2 h with vigorous stirring. After equilibration, the suspension was transferred to a glass column (1 cm in diameter) and the cations were eluted with 30 ml of 6 mol/L HCl:3 mol/L HNO₃. As in previous case, if a combination of Sr and TRU column is to be used, evaporated samples should be dissolved in 5 M mol/L NO₃.

2.3.3. New procedure for the separation of iron and strontium from Sr resin columns by HCl/HNO₃ elution

30 ml of 6 mol/L HCl:3 mol/L HNO₃ solution (from IR-120 column) were put onto a column filled with 1 g of Sr resin and passed through the column with 1 ml min⁻¹ flow rate. (The Sr column was preconditioned for separation by passing 20 ml of 6 mol/L HCl:3 mol/L HNO₃) After that, the column was washed with 20 ml of 6 mol/L HCl:3 mol/L HNO₃ (separation from other cations), strontium was eluted with 15 ml of 4 mol/L HCl (whole fraction should be kept for Čerenkov counting) and iron was eluted with 2 ml of 0.1 mol/L HCl. From both fractions 0.1 ml of solution was taken for the recovery determination. 0.1 ml of Fe and Sr were transferred in a 25 ml calibrated flask and their concentration was determined by AAS. Fe and Sr were also determined after isolation on TRU column and Sr column for the comparison of methods.

2.3.4. Separation of iron and strontium from TRU and Sr resin columns

30 ml of 5 M HNO₃ sample solution was passed through Sr resin column (2 g Sr resin in column, Ø=1 cm, 1 ml min⁻¹ flow rate). Column was rinsed with 30 ml 3 mol/L HNO₃, strontium was eluted with deionized water and strontium carbonate precipitation was followed. The Fe was passed through the Sr column and the Fe fraction was evaporated almost to dryness and after adding of 20 ml of 6 mol/L HCl Fe was bound on TRU column (2 g TRU resin in column, Ø=1 cm, 1 ml min⁻¹ flow rate). Column was rinsed with 40 ml of 4 M HCl solution and iron was eluted from the TRU column with 10 ml of 0.1 mol/L HCl. The phosphate precipitation was followed.

2.3.5. Determination of ⁵⁵Fe and ^{89,90}Sr by liquid scintillation counting

The determination of ⁵⁵Fe was by LSC. Recovery is determined by iron determination by AAS and single label liquid scintillation counting (LSC) was used for ⁵⁵Fe determination. In Fe fraction hydrogen phosphate was precipitated and dissolved in 2 ml of 2 mol/L HCl. 0.1 ml of this solution was transferred in calibrated flask and the concentration of Fe was determined by AAS. Mixture for counting contains 2 ml of sample and 15 ml Ultima Gold LLT scintillator in 25 ml plastic vial (Packard). ⁵⁵Fe activity concentration was obtained from the relation:

$$A = \frac{dpm}{60 \cdot V \cdot R}$$

where A is the activity of ⁵⁵Fe (Bq l⁻¹), dpm is the number of disintegrations per minute, R is the recovery, and V is the sample volume. Quench correction was used for the activity determination.

The determination of ^{89,90}Sr was carried out by LSC by Čerenkov counting in 4 M HCl. The sample was counted

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