STRONTIUM-90 LEVELS IN SEAWATER SAMPLES IN BRAZIL

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SUMMARY. A radiochemical procedure for low level strontium-90 analysis in seawater samples as well as the levels of this radionuclide in Brazilian samples are presented. The strontium yield is higher than 65%, and the yttrium yield is of 95%. The strontium-90 level in seawater is of (1.6 ± 0.3) Bq.m^{-3} and the detection limit is of 70 mBq.m^{-3}.

Key words: strontium-90, seawater, radiochemical analysis, radioactivity levels.

INTRODUCTION

In the last years, the environmental contamination from radionuclides originating from nuclear weapons tests, dumping operation and nuclear accidents have introduced artificial radionuclides to the environment, in particular the marine environment.

Strontium-90, due to its nuclear and chemistry characteristics is very important from the environmental point of view.

The estimates of input of the strontium-90 by fallout into the oceans are of 3.7×10^2 PBq. The inputs to the oceans in the southern hemisphere have been half of those in the northern hemisphere. The artificial radionuclides could affect marine organisms and its total activity is only a very rough guide to risks[1,2].

Determination of radionuclides in seawater generally requires their preconcentration from large volumes of sample and subsequent radiochemical analysis of the concentrate. The strontium determination in seawater presents serious difficulties because this matrix has significant contents of salts, such as magnesium and calcium, and these interfering ions must be removed from the strontium fraction.

In this work, the method of strontium-90 analysis consisted of strontium preconcentration from a large volume of sample as carbonate; elimination of magnesium and calcium by using ammonium chloride and sulphuric acid, respectively; removal of the other interferents by ferric hydroxide scavenger; ingrowth of the daughter; separation of strontium-90 from yttrium-90 and beta counting of the yttrium-90 in a low background Geiger-Muller detector.

The strontium yield was determined with strontium-85 tracer, and yttrium yield was determined gravimetricaly. The yttrium-90 purity was verified by decay curves.

MATERIALS AND METHODS

- Gamma ray spectrometer, high-purity Ge detector, type POP-TOP of EG & ORTEC, model 20190, connected to IBM-PC microcomputer, and eletronic associated;

- Low background (< 0.3 cpm) anticoincidence Geiger-Müller multicounter system GM-25-5, gas flow (99% argon - 1% methane), and high counting efficiency (28% for yttrium-90). (Denmark)

STRONTIUM-85 TRACER

About of 400 mg of strontium nitrate was irradiated in the IEA-R1 reactor of the Instituto de Pesquisas Energéticas e Nucleares (IPEN-CNEN/SP - Brazil), under a neutron flux of the 10^{12} $n.cm^{-2}.s^{-1}$, during 8 hours. After the irradiation it was dissolved in water, acidified with concentrated hydrochloric acid and diluted in water to 25 mL.

SEAWATER SAMPLING

Seawater sampling (300 L) was performed by the Oceanography Institute of the São Paulo University. Surface seawater samples were collected from the offshore of São Paulo State (45° longitude, 23° latitude), Atlantic Ocean. Samples were stored in polyethylene containers.

STRONTIUM-90 ANALYSIS

To determine strontium-90 in 100 liters of seawater, the analyse was carried out simultaneously in two portions of 50 liters each, and both precipitates obtained were mixed.

To 50 liters of seawater, 6.0 g of strontium carrier (strontium nitrate), strontium-85 tracer, 500 g of ammonium chloride and 875 g of sodium carbonate were added, in order to precipitate, strontium, calcium and only a little amount of magnesium carbonate.

The supernatant was retired by sucction and discarded, the strontium carbonate was dissolved with concentrated nitric acid. Further, for the elimination of the calcium interference, sulphuric acid (10% of solution volume) was added.

The strontium sulphate was filtered and converted to carbonate, by addition of 60 g of sodium carbonate. The supernatant was retired by sucction and discarded.

The strontium carbonate was dissolved with concentrated nitric acid. Iron carrier (10 mg of Fe) was added and the pH was increased up to 7 in order to precipitate ferric hydroxide. The retention of strontium in the hydroxide was lower than 15%.

To the strontium present in the supernatant 60 g of sodium carbonate were added, the strontium carbonate was stored by 14 days in order to attain the radioactive equilibrium. The separation of strontium-90 from yttrium-90 was carried out in several steps.

Strontium carbonate was dissolved with concentrated nitric acid and yttrium carrier (10 mg of Y) was added. The solution was heated by fifteen minutes. The yttrium was precipitated as hydroxide, by addition of concentrated ammonium hydroxide (pH 8). After filtering the precipitate was dissolved with concentrated hydrochloric acid, in the presence of 10 mg of strontium as hold back carrier. The yttrium was again precipitated as hydroxide, as here described for three times.

Finally, the yttrium hydroxide was dissolved with concentrated hydrochloric acid, and 3 mL of the solution 1 M oxalic acid (pH 1.5) were added. The yttrium oxalate was assayed by beta counting. The beta counting obtained for the yttrium-90 precipitate was corrected by building up and decay of yttrium-90, counter background, detector efficiency, strontium and yttrium yields and blank value in order to obtain the strontium-90 activity. Analysis of the blank were performed simultaneously to the seawater analysis.

RESULTS AND DISCUSSION

The yield of strontium was higher than 65% and of the yttrium was higher than 90%. The results obtained for the strontium-90 levels in the samples analysed are presented in Table 1.

TABLE 1 - Results obtained in three analysis of Brazilian coast seawater samples. (date of sampling: march/93)

ANALYSIS	Sr YIELD (%)	Y YIELD (%)	Sr-90 ACTIVITY($Bq.m^{-3}$)
1	69.0 ± 0.3	99 ± 6	1.3 ± 0.4
2	53.7 ± 0.2	100 ± 6	1.9 ± 0.5
3	66.4 ± 0.5	100 ± 6	1.7 ± 0.5
MEAN VALUE AND STANDARD DEVIATION	63 ± 8	100.0 ± 0.6	1.6 ± 0.3

The Figure 1 shows the scheme for the radiochemical analysis of strontium-90 in seawater samples. The yttrium purity was verified by decay curve, being obtained the value of (65 ± 6) hours for the half time of yttrium-90 (Figure 2).

STRONTIUM-90 ANALYSIS IN INTERCOMPARASION SAMPLE

The method developed in this work was applied to the intercomparasion water, "EMSL-LV Cross-Check" sample, from the USEPA and Dosimetry and Radioprotection Institute (Brazil). Data obtained are presented in Table 2.

TABLE 2 - Results obtained in the analysis of intercomparasion water.

ANALYSE	Sr YIELD (%)	Y YIELD (%)	Sr-90 ACTIVITY($mBq.L^{-1}$)
1	100.5 ± 0.4	88 ± 5	181 ± 64
2	94.9 ± 0.4	67 ± 4	117 ± 53
3	100.5 ± 0.4	79 ± 5	204 ± 64
4	94.9 ± 0.4	84 ± 5	127 ± 53
MEAN VALUE AND STANDARD DEVIATION	98 ± 3	80 ± 9	157 ± 42
REFERENCE VALUE (01/11/91)			187 ± 187

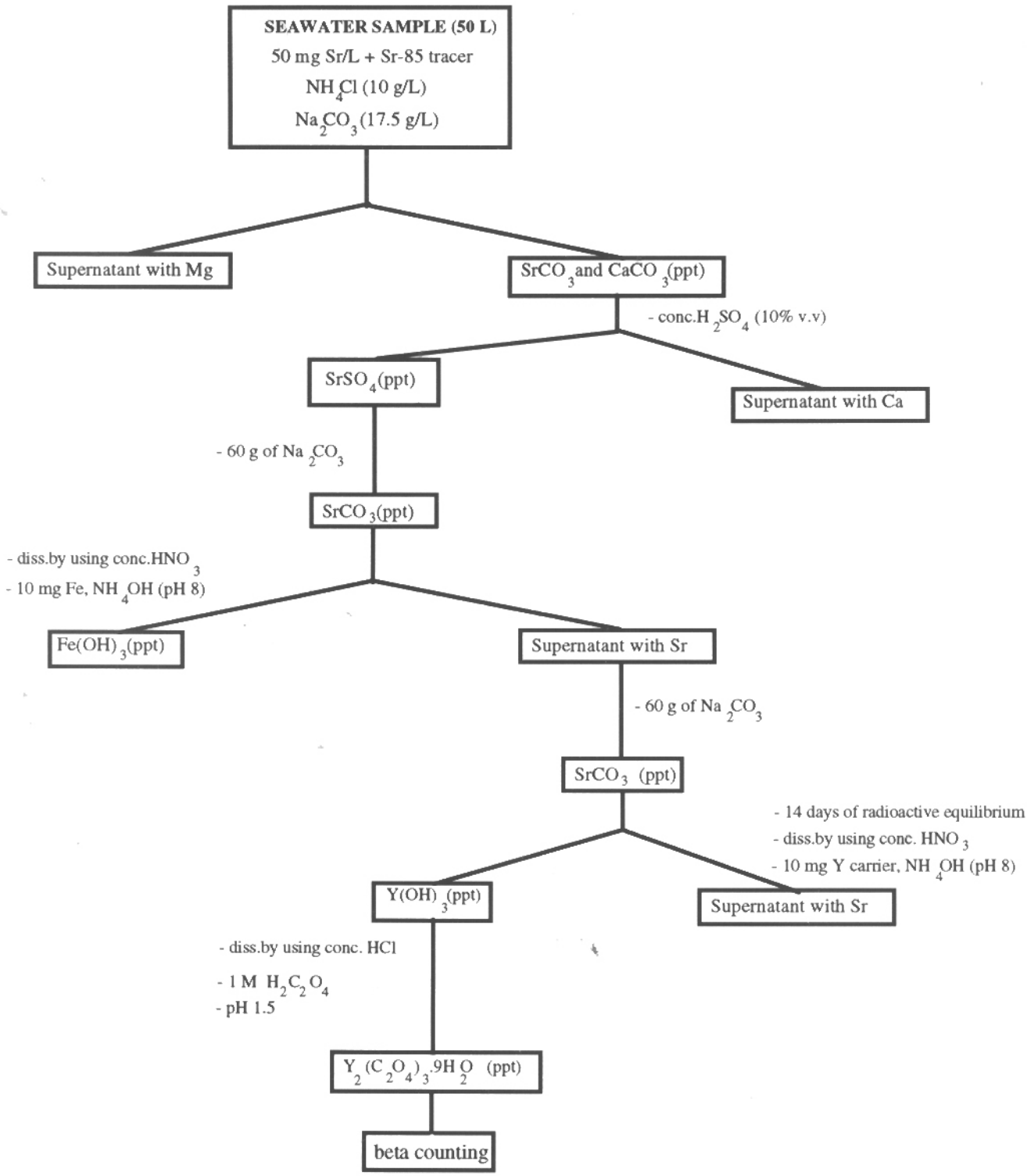

Figure 1 - Scheme for the radiochemical analysis of strontium-90 in seawater samples.

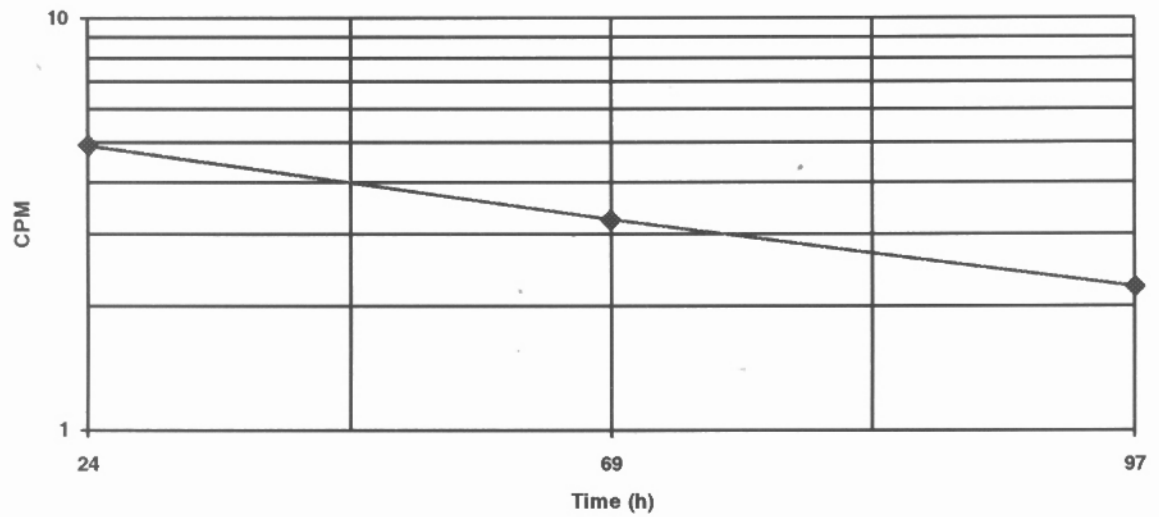

Figure 2 - Decay curve of yttrium-90 in seawater sample.

The results obtained for yttrium and strontium yields are in good agreement to the values published at the literature[2,3,4]. The strontium recuperation is of about 65%, due to the interferences of magnesium and calcium that are present in high concentration, as well as the strontium carbonate and strontium sulphate solubilities, 0.011 g/L and 0.11 g/L, respectively.

In this work, the elimination of the magnesium was made by using ammonium chloride, while the elimination of calcium was carried out by successive transformations of precipitate, carbonate to sulphate and sulphate to carbonate; the volume of sulphuric acid added is critical in order to minimize strontium sulphate losses by solubility. This method does not require fuming nitric acid, that is a reagent of high price for a monitoring programme and it is hazardous for the health.

The procedure of analysis here developed was applied to an intercomparasion sample. The results obtained showed the good precision and accuracy of the method established. It can be applied in a routine work where large volumes of sample are required.

Strontium-90 levels in Brazilian seawater are of (1.6 $\pm$ 0.3) $Bq.m^{-3}$. These concentrations are typical values due to fallout deposition in the southern hemisphere and are of the same order of magnitude of the regions, like North Pacific[1], 2.7-3.7 $Bq.m^{-3}$ and Atlantic North[6], 1.7-4.7 $Bq.m^{-3}$, but lower when compared to areas influenced by input from discharges of nuclear reprocessing plants or Chernobyl accident, such as Cap La Hague[7], 29 $Bq.m^{-3}$, Yellow Sea[8], 10 $Bq.m^{-3}$, and Baltic Sea[9], 17.6 $Bq.m^{-3}$.

Data obtained in this work are used as strontium-90 reference levels in Brazilian seawater samples and any alteration in this value can be attributed to an especific cause.

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