DETERMINATION OF ^{90} Sr IN SOIL SAMPLES

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ABSTRACT

Radionuclide identification and measurement became essential in environmental samples (air particulate, water,soil and all foodstuff) after Chernobyl accident. In chemistry laboratory scientists have already substantial experience about radioisotope analysis and some radiochemical separation techniques. On the other hand quality control measures are necessary to show that the analytical results are reliable. For checking the accuracy of our analytical results, our laboratory has participated in the Technical Co-operation (TC) regional project entitled ''Quality Assurance and Quality Control of Nuclear Analytical Techniques'' For this purpose, IAEA Quality Control service has sent to our laboratory 4 soil samples for determination of ^{90}Sr activities.

Radiostrontium was extracted from the soil with 6M hidrochloric acid and then radiochemical separation and purification methods were applied. Solution was stored at 21 days for ^{90}Sr-^{90}Y equilibrium. After 21 days, Yttrium oxalate precipitation was obtained and it was counted by using Protean Instrument WPC 9550 Low Level Alpha/Beta Counter.

INTRODUCTION

^{89}Sr and ^{90}Sr/^{90}Y are fission products that are created in nuclear explosions and nuclear fuel consumption. Therefore, the main source of these radionuclides is nuclear weapons testing in the atmosphere and releases from the nuclear fuel cycle. Fallout from nuclear weapons testing is primarily responsible for the ^{89}Sr and ^{90}Sr/^{90}Y concentrations found in soil.

^{90}Sr is an important fission product because of its long physical (28.5 y) and biological (11 y) half life. It is produced with a fission yield of about 5.7 %, which is equivalent to 3.6x10^{15} Bq Sr^{90} per 4.1x10^{5} joule energy release.

Since the nuclear test ban treaty in 1963 to 1981 the total amount of ^{90}Sr deposited on the surface of the earth amounted to approximately 5x10^{17} Bq. After the Chernobyl nuclear power plant accident, an increase of the ^{90}Sr deposition took place.

Because of its chemical properties as an alkaline earth element, ^{90}Sr enters the food chain of animals and humans through the pathway soil- grass- cattle- milk- man. Mainly the bone tissue is exposed to the radiation of ^{90}Sr and ^{90}Y.Therefore, data on the ^{90}Sr content of soil, animal feed and human foodstuffs have radiological importance.

EXPERIMENTAL

Description of sample:

Matrix origin: Soil from China

Sample preparation: The samples (each containing 5g of soil) were spiked with known amounts of a certified standard solution containing a mixture of ^{90}Sr,^{238}Pu,$^{239+240}$Pu and ^{241}Am radionuclides. No attempt was made to homogenise the samples after spiking, therefore analysts were performed measurements on the entire sample which were transferred quantitatively from the bottle.

Materials

All reagents used were of analytical reagent grade. Standard solution and the blank sample were obtained from the IAEA.

Method

All procedures for the total analysis of radiostrontium are rather complicated and slow since they require the separation and purification of strontium from various inactive and radioactive elements present in complex sample matrix by chemical procedures exhibiting only limited selectivity .Exemplary is the standard method of analysis of radiostrontium , based on the precipitation of strontium with fuming HNO_3.This method which consist of nine different precipitation steps, requires 21 days. Although this method is time consuming, it is reliable and safe.

Radiochemical procedure

5g dry soil is treated directly without ashing.

- 25 ml of distilled water and 2 ml Sr-carrier (20mg/ml)were added and then 25 ml 11 M HCI was added and stirred. After 24 hours it was filtered. Soil was washed with distilled water and second extraction was carried with 1:1 diluted conc. HCI Filtrates were combined.

- After addition oxalic acid and ammonium acetate, solution's pH was adjusted to 4 with ammonium hydroxide and oxalate precipitation was obtained.

Note: Since there is no excess iron, brown precipitation was not observed.

Therefore further oxalic acid was not added

- Oxalate precipitate was transferred to a silica dish and dried in an oven and ignited in a muffle furnace at 700-800 ^{0}C for 30 minutes. After addition distilled water and conc. HNO_3 was boiled to remove CO_2. Solution was diluted and 6 M ammonium hydroxide added for aluminium and iron precipitation and then filtered.

- Solid ammonium carbonates was added to the filtrate until precipitation of calcium and strontium is complete. The mixed carbonates were dissolved in the minimum of 3.5 M HNO_3 and separation of Sr-Ca was carried with fuming HNO3 in the ice-bath. Residue of $Sr(NO_3)_2$

was dissolved with distilled water and then Ba- carrier and methyl red indicator were added. Solution was neutralised with 6 M ammonium hydroxide. After addition 1 ml of 6 M acetic acid and 2 ml of %25 ammonium acetate the solution was diluted and boiled. %30 sodium chromate was added and about 5 minutes boiled again.

- The solution was made alkaline with ammonium hydroxide and then solid ammonium carbonate was added for precipitation of $SrCO_3$ and then residue was dissolved with diluted HNO_3. The solution was boiled after addition 1 drop of H_2O_2 and Fe-carrier and heated and stirred to remove CO_2.The solution treated with ammonium hydroxide and $Fe(OH)_3$ precipitation was obtained in an alkaline media.

- It was centrifuged and filtered. HNO_3 was added to obtain acidic solution. After addition Y-carrier it was stored for ^{90}Sr-^{90}Y equilibrium for at least 14 days.

Experimental determination of counting efficiency

From the standard (equilibrium) ^{90}Sr-^{90}Y solution, a freshly separated strontium source of known ^{90}Sr activity (1000 pCi/ml) is prepared in the same chemical form and counted under the same conditions as the samples to be analysed and a pure yttrium source is also prepared and counted under identical conditions. Counting efficiency for ^{90}Sr is 0.4566.

Counting Conditions

The decay of ^{90}Sr is counted by Ultra Low Level Alpha/Beta Counting System (Protean Instrument Corporation (PIC)-WPC9550)

Beta background was 0.017±0.002 cps. Time of measurement was 6000 s.

By weighing the precipitates of strontium carbonate and yttrium oxalate the chemical yield were calculated.

^{90}Sr activity results of samples, standard and blank were summarised in Table1.

Table 1

Sample	Activity	Combined standard Uncertainty(±σ)	Limit of dedection	Chemical yield for Y (%)	Chemical yield for Sr (%)
Sample 1	0.11 Bq/s	0.02 Bq	0.04 Bq	87.05	33.00
Sample 2	0.09 Bq/s	0.02 Bq	0.04 Bq	93.97	33.00
Sample 3	0.13 Bq/s	0.02 Bq	0.03 Bq	86.09	38.85
Blank	0.004 Bq/g	0.001 Bq/g	0.004 Bq/g	99.35	34.74
Standard	9.13 Bq/g	0.07 Bq/g	0.007 Bq/g	83.03	100.00

Activity Concentration /sample

$$A_{Sr90}=\frac{R_2}{\%Sr.\%Y.E.M.e^{-\lambda_{Y90}t}}$$

R_2: Net count (cps)

%Sr : Chemical yield of Sr

%Y : Chemical yield of Y

Counting efficiency for ^{90}Y

t : time between Sr precipitation and middle of Sr measurement

M : Sample mass

Statistical counting error

$$S_{ASr90}=\frac{\sqrt{\frac{I_2}{(t_{M2})^2}}+\sqrt{\frac{I_{02}}{(t_{02})^2}}}{\%Sr.\%Y.E.M.e^{-\lambda_{Y90}t}}$$

I_2 : Counts of Y measurement in counting time t_{M2} in s

I_{02}: Background of Y measurement in t_{02} in s

t_{M2} : Counting time in s

t_{02} : Background time in s

Limit of detection

$$LD_{Sr90}=\frac{3.\sqrt{I_{02}\left(\frac{1}{(t_{M2})^2}+\frac{1}{(t_{02})^2}\right)}}{\%Sr.\%Y.E.M.e^{-\lambda_{Y90}t}}$$