METHOD OF EXPRESS DETERMINATION OF SR-90 CONTENT IN DIFFERENT OBJECTS WITH A HIGHLY SELECTIVE SORBENT

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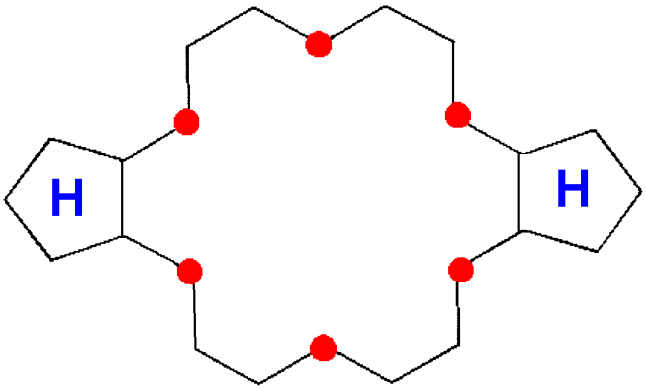
Abstract
The present method is worked out for ^{90}Sr determination in objects of different origin, especially in agricultural production, cattle production, and food. The method may be applied to determine the ^{90}Sr contamination of objects and territories, which were exposed to accidents at nuclear energy objects, buffer areas of NPP, and possible use of so called “dirty” bombs. The principle of the method is based on chromatographic extraction of strontium from nitrate solutions followed by radiometric determination of ^{90}Sr activity. A highly porous matrix of styrene-divinyl benzene in the form of beads ~1 mm in diameter is used as a substrate to support the extractant, dicyclohexyl-18-crown-6 (DCH18K6) in tetrachloroethane, which is in the liquid state deep in the pores of the matrix. Dicyclohexyl-18-crown-6 belongs to synthetic macrocyclic polyethers (so-called crown ethers) and has the ability to bind ^{90}Sr with high selectivity so that ^{90}Sr is extracted with 90 – 95 % efficiency. To register the equilibrium composition (^{90}Sr и ^{90}Y) Vavilov - Cherenkov effect or liquid scintillators are used. The ^{90}Sr activity is measured with allowance for the scintillating cell background and the conversion factor depending on the ^{90}Y accumulation time and the ^{90}Sr and ^{90}Y detection efficiency.

Introduction
The scientific bases of the most exact and express method of definition of the Sr-90 contents in the objects of environment were developed in GEOHI of the Russian Academy of Science (Moscow) under the direction of academician B.F.Mjasoedov at once after the accident on the Chernobyl NPP. This method was later adapted at the Institute of Physic NAS of Belarus on the β – radiometer “Beta-2”. In addition, after the investigation of the γ – emitting of ^{89}Sr the regime of making measurements was optimized at different stages. The present method is worked out for ^{90}Sr determination in objects of different origin, especially in agricultural production, cattle production, and food. The main advantage of the method is reduction of time needed for sample preparation and measurement (3-4 hours) in comparison with traditional radiochemical method (up to 2 weeks).

Methods
The key point of the method is Sr-90 extraction from the investigated sample by the high – selective sorbent of dicyclohexyl – 18 – crown – 6. It makes possible to exclude other background radioactive isotopes almost completely. The principle of the method is based on chromatographic extraction of strontium from nitrate solutions followed by radiometric determination of ^{90}Sr activity. A highly porous matrix of styrene-divinyl benzene in the form of beads ~1 mm in diameter is used as a substrate to support the extractant, dicyclohexyl-18-crown-6 (DCH18K6) in tetrachloroethane, which is in the liquid state deep in the pores of the matrix. Dicyclohexyl-18-crown-6 belongs to synthetic macrocyclic polyethers (so-called crown ethers**)** and has the ability to bind ^{90}Sr with high selectivity so that ^{90}Sr is extracted with 90 – 95 % efficiency. Illustration 1 depicts structural formula Dicyclohexyl-18-Crown-6. The first number in names of croun ethers indicates the size of a cycle, and the second number defines quantity of atoms of oxygen in a cycle. Illustration 1 depicts structural formula dicyclohexyl-18-crown-6

Illustration 1: Dicyclohexyl-18-Crown-6



Results

Institute of Physics of the Academy of Sciences of Belarus worked out two versions of this method. The first version is meant for Sr–90 determinations in soil, water, and milk. The method of ^{90}Sr determination on the basis of selective sorbent in the in the above objects includes three main stages:

1. Sample preparation. This stage incorporates double acid leaching of ashed samples by 7.5 M nitric acid, filtering, evaporation to wet salts, and final correction of the nitric acid concentration to 1.5 M of obtained solutions.
2. Preparation of selective sorbent, extract eluation trough a column. To prepare the selective sorbent, solid extragent (solex) balls are placed in the column and a 10% solution of the sorbent dicyclohexyl – 18 – grow – 6 (DCH18K6) in tetrachlorethane is poured. The nitrate extract of the sample is passed through the column (the time of completion of the sample passage is taken for the moment of separation of ^{90}Y and ^{90}Sr). Then the column is washed with 1.5 M nitric acid. Next the column is heated to 80 ^{0}C by a thermostat, hot water (90 ^{0}C) is poured, and strontium desorption from the column is carried out.
3. Radiometric measurement of the obtained precipitate. The ^{90}Sr – containing preparations of strontium carbonate and the accumulated ^{90}Y are placed in scintillating cells and measured on the “Beta – 2” apparatus. The ^{90}Sr activity is measured with allowance for the scintillating cell background and the conversion factor depending on the ^{90}Y accumulation time and the ^{90}Sr and ^{90}Y detection efficiency.

The second version is a later one. It is modernized. Use of the modernized methods considerably expands a range of researched objects. It is meant mainly for Sr–90 determination in objects of organic origin (cereals, leguminous plants, meat and milk production, food, etc.). Also will considerably decrease time of the analysis. At the step of sample preparation this version uses the wet combustion, which simplifies the sample preparation and allows realizing of radiometric measurement in DCH18K6 in tetrachlorethane.

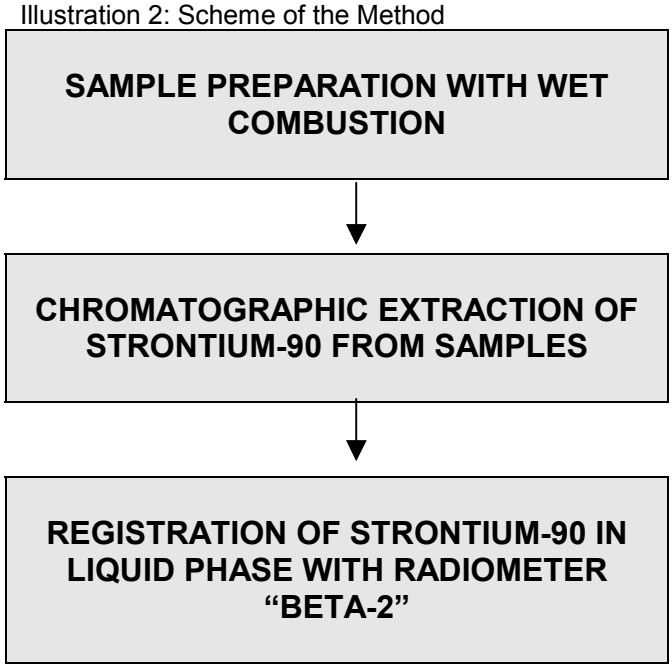
The second modernized version includes three main stages:

1. Sample preparation. The method of the wet combustion of the samples by aquafortis, filtering, evaporation, and final correction of the nitric acid concentration to 1.5 M of obtained solutions are used.
2. This stage does not change.
3. Radiometric measurement in the liquid phase by β-radiometer “Beta-2”. To register ^{90}Sr the moment the sample is passed through the column, liquid scintillators are used. The ^{90}Sr activity is measured with allowance for the scintillating cell background and the conversion factor depending on the ^{90}Y accumulation time and the ^{90}Sr and ^{90}Y detection efficiency.

Method «wet combustion» is based on combustion of dusty sample by the concentrated nitric acid at the presence of perchloric or sulfuric acid. Reacting with organic substance, nitric acid decays to water, nitrogen dioxide and oxygen. Oxygen in free state has huge reactivity and oxidizes hydrogen of organic substance up to water. Presence of sulfuric or perchloric acid considerably accelerates combustion.

Illustration 2 depicts scheme of the modernized method

Illustration 2: Scheme of the Method



The efficiency of strontium-90 desorption from the chromatographic column depends on the temperature of water and the crown ether concentration. Results of experiment are represented in the Table1.

Table1: Desorption of Strontium from the Chromatographic Column

Desorption temperature ^{0}C	60	70	80	90	60	70	80	90
Crown ether conc. %	5	5	5	5	10	10	10	10
Yield %	75	80	91	90	80	90	91	90

The basic technical parameters of the method are represented in Table 2.

Table 2: Technical Parameters

Characteristic		**Unit**	**Value**
Lower-range value		Bq	1
Upper-range value		Bq	1000
Time of realization of one analysis		Hour	3-4
Measurement error	1-3 Bq 3-10 Bq 10-1000 Bq	%	20 15 10

Discussion

The present method provides considerable reduction of time needed for measurement of a sample (3-4 hours) in comparison to traditional radiochemical method (up to 2 weeks). The present method is worked out for Sr-90 determination in objects of different origin, especially in food, agricultural production and cattle production. The method may be applied to determine the Sr-90 contamination of objects and territories, which were exposed to accidents at nuclear energy objects, buffer areas of NPP, and possible use of so called "dirty" bombs.

Conclusion

The stepwise investigation of the overall method showed that significant strontium losses occur only in acid leaching of the prepared soil samples and in desorption from the chromatographic column.

Losses are considerably reduced at use of a method " wet combustion ". Recovery at the leaching step is consistently near 90 %, so there are few benefits gained by reducing of losses at this step. The efficiency of strontium desorption from the chromatographic column with hot water is obviously temperature dependent. Nevertheless, consistent recovery of 90% can be obtained at the water temperature of 80°C. The results suggest that the desorption occurs in more consistent way when the crown ether concentration in the diluent is 10 %. The need for consistency in most environmental applications overrides the possible saving of using 5 % solution. Based on the results, re-use of the chromatographic column can be recommended only if it is known that the measured activities of all samples in the series are virtually equal.

Detection limits depend both on the chemical steps and the quality of the radiometer. With state-of-art equipment such as the "Beta-2" the method is characterized by the following relative measurement errors at the 95 % confidence level:

30% in the 1 to 3 Bq range,
20% in the 3 to 10 Bq range,
15% in the 10 to 1000 Bq range.

Slight modifications of the scintillation cells in the "Beta-2" are expected to permit measuring ^{9O}Sr activities of the order of 0.1 Bq

Further work to improve the method will include more thoroughly selection of the pore size in the polystyrene copolymer beads and replacing of tetrachloroethane by other types of diluents. Further work will probably use ultrasonic radiation and microwave radiation to decrease the time of sample preparation.

References

(1) S. Zablotsky, E. Rudak, L. Stopolyanskaya and J. Wiley “Improvents in the method of strontium-90 extraction from environmental samples with dicyclohexyl-18-crown-6” Kerntecnik, **V.60/5-6**, 262-264, (November 1995).