A review on determination of $^{90}$Sr from alkaline waters using precipitation of Ca(OH)$_2$ and Ba(Ra)SO$_4$

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Abstract. Since the Chernobyl accident (1989) and the Fukushima Daiichi Nuclear Power Plant accident in 2011, concern about the contamination of $^{90}$Sr in the environment has increased considerably. It is important for public safety to monitor $^{90}$Sr in aquatic environments due to the radiotoxic of its biochemical similarities with calcium in living organisms. Several techniques for the separation of strontium from matrices have been reported such as solvent extraction, liquid membrane extraction, ion-exchange, and extraction chromatography using Sr-Resin. This review provided a summary on literature works to develop efficiency radiochemical separation for removing $^{90}$Sr from waters. Radiometric method normally requires a long time and complete separation of Sr from the matrix and other radionuclides. One of simple method based on Ca(OH)$_2$ precipitation in alkaline solution and purification of Yttrium using Ba(Ra)SO$_4$ precipitate was applied to the separation of Sr from seawater and underground water sample. This review is a mini-report for determination of $^{90}$Sr removal from contaminated waters.

1. Introduction
Radioactive strontium is one of the most biological hazardous radionuclides produced in nuclear fission processes and it can release into the environment due to fallout from nuclear weapon test, operation nuclear facilities and the accident of nuclear power plant. A major source of $^{89}$Sr and $^{90}$Sr released into the environment from global fallout has been estimated about $6.22 \times 10^{17}$ Bq during the period of nuclear weapon test [1]. The second major source is the accident of Chernobyl Nuclear Power Plant in 1986 and Fukushima Daiichi Nuclear Power Plant accident in 2011 due to high magnitude earthquake in Tohoku area. The approximate amount of $^{90}$Sr released during the Chernobyl NPP accident was $8 \times 10^{15}$ Bq [1][2] and around $10^{14}$–$10^{15}$ Bq was released from Fukushima Daiichi NPP accident into the Pacific Ocean due to a leakages of reactor coolant water [3][4]. As a result of these events, the long-term monitoring of radionuclides is necessary to assess the ongoing emissions from the nuclear facilities, dose risk, and the impact on the environment.

$^{90}$Sr is an important fission product because of its long physical and biological half-life and it can cause great external radiation doses to humans and other living things. On the other hand, $^{90}$Sr can be transferred into human body where it causes internal radiation doses through the inhalation or ingestion of contaminated food. Most of the $^{90}$Sr is accumulated in the bone skeleton resulting in...
chronic internal irradiation due to the similarity of its chemical characteristics to a calcium and it will incorporate in bones and teeth of humans. Furthermore, a long-term storage of $^{90}\text{Sr}$ deposited in the human body can lead to serious consequences, such as bone cancer or leukemia [5][6][7]. $^{89}\text{Sr}$ also share the same biological significance, but it has much shorter physical half-life and thus will not have a long-term environmental impact. However, the presence of both radioisotopes in environmental samples is of great interest. Since strontium can transported easily in waters is an important to determine the rapid and accurate analysis of radioactive strontium isotopes in waters in emergency or normal situations in order to protect the public from radiation hazards [8].

The analysis procedure for radioactive strontium is always complicated because $^{90}\text{Sr}$ and its daughter $^{90}\text{Y}$ are pure beta emitters, and prior to measurement they must be separated from the sample. A limited number of procedures have been described to determine $^{90}\text{Sr}$ in seawater and most of the analytical methods using $^{90}\text{Y}$. Therefore, accurately determining and monitoring of $^{90}\text{Sr}$ in the environment is important for long-term dose assessment. Moreover, $^{90}\text{Sr}$ have also served as useful tracers for the studies about dust transport, soil erosion, sediment chronology, seawater movement and so on [9][10][11][12]. Various analytical procedures for separating $^{90}\text{Sr}$ in environmental samples have been developed. For instance, the Sr can be separated from Ca by fuming nitric acid [13][14] and Ca hydroxide precipitation [15][16]. This report describes a new approach for the rapid determination of $^{90}\text{Sr}$ in waters using preconcentration by Ca(OH)$_2$ and Ba(Ra)SO$_4$ for purification of Yttrium methods. The resulting recommended procedure is designed to be of general use to a wide range of laboratories. It is expected that this rapid method for determining $^{90}\text{Sr}$ in waters will be useful and safer for routine environmental radioactivity monitoring.

2. Sample pretreatment

The quantitation of radiostrontium is based on three major considerations: sample pre-treatment (to bring the sample into a suitable matrix), isolation of radiostrontium by chemical treatment, and radiometric measurement [17]. Pretreatment of sample to analysis $^{90}\text{Sr}$ is relatively complicated, including digestion or co-precipitation techniques to release and concentrate $^{90}\text{Sr}$ from sample matrix to a small amount of liquid solution. The liquid environmental samples such as seawater, river water, and groundwater need to pretreatment included filtration, pH adjustment, co-precipitation or evaporation so that it can be applied to the next step or detected directly [7][18][19][20][21][22][23]. The purpose of pH adjustment is to prepare for the $^{90}\text{Sr}$ co-precipitation and remove major interferences so that $^{90}\text{Sr}$ can be concentrated from large volume liquid samples to a small volume of acid solution. Popov et al. [16] adjusted the pH of 40 l water samples with HNO$_3$ to acidify the solution at pH 3-4 and added by CaCl$_2$ solution in 12M HCl before removing carbonate precipitation by adjusted pH ~10 with NaOH and dry Na$_2$CO$_3$. The flowchart of the analytical procedures was schematically shown in Fig. 1.
3. Separation and purification of $^{90}$Sr from environmental samples

The environmental samples contain various $\beta$-ray emitting radionuclides and it is necessary requires specific radiochemical separation of Sr due to $\beta$-radiation is not monoenergetic and determination of spectral resolution of individual nuclides is difficult. Naturally occurring radionuclides has potentially interference with the determination of $^{90}$Sr, such as $^{40}$K and $^{238}$U exist predominantly in seawater and the decay chain of uranium and thorium series also emit beta particles [24]. Another significant interference for the analysis of $^{90}$Sr in environmental and biological samples is the Ca due to their similar chemical properties in the same group in period table. Moreover, concentrations of Ca in samples are much higher than those of $^{90}$Sr. Previous research has proven that K$^+$ had a definitely adverse effect on the recovery of $^{90}$Sr in resin which resulted in 7% loss of $^{90}$Sr when 30 mg K$^+$ was presented in the solution [25]. In order to remove the influence of matrix interferences, it is necessary to separate Sr from the sample matrix. Generally, four methods have been widely used in the $^{90}$Sr separation and purification such as extraction chromatography, liquid-liquid extraction, ion exchange chromatography, and co-precipitation.

Extraction chromatography was widely applied in $^{90}$Sr analytical methods, due to its higher purification efficiency especially the applications of DGA and Sr resins. Tazoe et al. [7] has been successfully applied to the analysis of $^{90}$Sr in large volume seawater samples through $^{90}$Y separated from Sr and other interferences. Characterized DGA resin having high distribution coefficients for different elements and the capability for separation with samples containing multiple elements. $^{90}$Y was completely separated from other nuclides such as Na, K, Ca, Mg, etc, that have a negative effect on $^{90}$Y $\beta$ measurements using the Fe(OH)$_3$ co-precipitation and DGA resin purification procedure with

![Figure 1. Summary of radiochemical procedure for analysis radiostrontium in water (Popov et al., 2006)](image-url)

**Figure 1.** Summary of radiochemical procedure for analysis radiostrontium in water (Popov et al., 2006)
decontamination factors of more than $10^6$ [26]. Sr resin is characterized as having high selectivity and a special adsorption capacity for Sr. Grahek et al. [19] has been described detail of Sr resin application conditions such as acid solution, adsorption coefficient, eluant selection, etc. Tazoe et al. [7] developed a simplified analytical method to determined $^{90}$Sr in 3 L seawater samples from direct separation of $^{90}$Y two weeks after getting equilibrium, with MDL of 1.5 mBq L$^{-1}$ after 20h of counting. For large volume freshwater samples, Tomita et al. [27] separated $^{90}$Y from $^{90}$Sr in 170 L water samples using multi-step co-precipitations, resin purification, and two weeks waiting to achieve secular equilibrium and counted with a gas-flow type $\beta$ counter to obtain a lower MDL of 0.1 mBq L$^{-1}$.

Another purification method that has been used is a liquid-liquid extraction using organic reagent to extract radionuclides from acid solution, which is a fast method for determination of $^{90}$Sr in environmental samples. Direct separation using Di-(2-ethylhexyl) phosphoric acid (HDEHP), tributyl phosphate (TBP), or t-octyl phosphine oxide (TOPO) have been commonly used for separation of $^{90}$Y from $^{90}$Sr. Applied HDEHP to selectively extract $^{90}$Sr from the acid solution was used by Clark, [28] in order to demonstrated effectiveness of the crown ether-based solid phase extraction from the calcium matrix containing Ca:Sr ratios exceeding 100. Aslan et al. [29] mixed HDEHP with toluene to selectively extract $^{90}$Sr into nitric acid solution. This method is easy and quick, and it offers high selectivity and recovery. However, organic solvents that must be used are usually toxic, volatile and expensive. Therefore, compared to other purification methods, liquid-liquid extraction for $^{90}$Sr analysis has not been widely applied [30].

Ion exchange chromatography was widely applied in determination $^{90}$Sr analytical methods due to its higher purification efficiency and convenient operation. Waples and Orlandini [31] developed a method combining the anion and cation exchange chromatography with coprecipitation using Fe hydroxide. This method included the measurement by beta counting of $^{90}$Y ($T_{1/2} = 64h$) as a daughter of $^{90}$Sr, which is in equilibrium state with $^{90}$Sr after letting samples equilibrate overnight. Isolation of Yttrium from other beta emitters such as Th was performed by passing the solution through an anion ion-exchange column Bio-Rad AG1-X8, 100-200 mesh. Ion exchange resins AG1-X8 and AG 50W-X8 have been the most commonly used to observation of the $^{90}$Sr-$^{90}$Y in the aquatic environment [3][4][32]. The cation exchange chromatography method is more complicated than the coprecipitation and DGA resin extraction chromatography method since the former requires multiple coprecipitation and resin purification steps. Though the cation exchange method can handle a large volume of sample, it has disadvantages of taking a long time, having a small exchange capacity, and having relatively complicated method procedures.

Much of the emphasis for assay of radiostrontium has been focused on separation chemistry. The classical purification method of co-precipitation is simple and offers easy separation of $^{90}$Sr from interferences, in which no required a complicated and expensive facilities. Chemically, strontium mimics calcium which makes separation difficult in environmental samples where, inevitably, calcium is abundant. Strontium is usually separated from the isotopes of barium by BaCrO$_4$ precipitation, but it is time consuming and difficult due to critical control of pH of the solution and removal of the excess of chromium [8]. Recently a simple method based on Ca(OH)$_2$ precipitation in alkaline solution and Ba(Ra)SO$_4$ precipitate was applied to the separation of Sr from sample matrices and interfering radionuclides. Chen et al. [15] developed a simple, cheap and safe procedure for the determination of $^{90}$Sr in water samples by separation of radiostrontium from calcium using precipitation of Ca(OH)$_2$ in alkaline solution instead of fuming HNO$_3$. The separation of Sr from Ca by Ca(OH)$_2$ precipitation from alkaline solution can remove $>99\%$ of Ca with 0.2 moll$^{-1}$ NaOH in one precipitation with Sr remaining $>95\%$ in the solution. The concentration of Ca in the solution does not significantly affect the separation of Sr from Ca in a solution containing $\leq$ 50 g of calcium.
Tomita et al. [27] developed analytical method to improved determining of low levels of radiostrontium in environmental freshwater samples. In this method, more than 60% of Ca was separated by Ca(OH)₂ precipitation, and the remaining Ca was removed by cation exchange. The strontium was separated by the cation exchange method for β counting after removal of most of the calcium (Ca) using Ca(OH)₂ precipitation. Through the procedure the Sr chemical yield was 88% on average with the detection limit of 90Sr activity approximately 0.1 mBq L⁻¹ in 170 L water samples. Popov et al. [16] developed another method based on the solubility difference of Sr(OH)₂ and Ca(OH)₂ to separated Sr from a large amount of Ca. In this analytical method proved that high radiochemical yield of strontium could be achieved at pH 4.5 with recovery value 83.6 % for strontium and 86.0 % for yttrium in water samples. The chemical yield of strontium is determined via gamma-spectrometric measurement of the activity of the radioactive tracer of 85Sr. The presented method is safer for the laboratory staff instead of using fuming nitric acid methods. Also, this procedure has advantages for good accuracy, high chemical yield and applicable for separated strontium from a large content of calcium if compared to previous analytical method.

4. Measurements of strontium
The isotopes of ⁸⁵Sr and ⁹⁰Sr/⁹⁰⁶Y emit only β-rays has been conventionally determined by radiometric methods usually measured by β counting, β spectrometry, or Cerenkov counting with proportional counters or liquid scintillation counters [9][16][33][34]. ⁹⁰Sr decays to ⁹⁰⁶Y before finally decaying to stable nuclide of ⁹⁰Zr. ⁹⁰⁶Y emits beta particles with energies Eₘ₅₆ = 2.28 MeV was higher than that ⁹⁰Sr with energy Eₘ₅₆ = 0.546 MeV, making it more suited for radiometric detection methods [35]. However, it takes up to three weeks to reach equilibrium, while the analysis itself can take as much as 1000 min [6][7]. Such time-consuming procedures cannot meet the need for quick response in nuclear emergency situations. However, ⁹⁰Sr is usually determined by measuring its short-lived daughter nuclide ⁹⁰⁶Y, and Y₂(SO₄)₃ is water soluble. Chen et al. [15] using the precipitation procedure of Ba(Ra)SO₄ to remove Ba and Ra from Y if only ⁹⁰⁶Y is to be determined. In this research shows the separation of Y from Ba, Ra and Sr by sulphate precipitation, more than 99% Sr and 99.5% of Ba and Ra can be removed by this method, while >98.5% Y still remained in the solution. Therefore, it is recommended that the procedure of Ba(Ra)SO₄ precipitation is used for the determination of ⁹⁰Sr. Measurement the radioactivity of ⁹⁰⁶Y using anti-coincident gas flow GM-counter with the low detection 5-10 mBg, or 0.02-0.05 Bq m⁻³ for 200 l samples and the analytical error at this level is >50%.

| Sample  | Purification               | Recovery % | MDL           | References               |
|---------|----------------------------|------------|---------------|--------------------------|
| Water   | Sr resin                   | 85-95      | 0.1-7 Bq      | Heckel and Vogl, 2009    |
| Water   | Sr resin                   | 86.2       | 140 mBq sample⁻¹ | Dai and Kramer-Tremblay, 2014 |
| Seawater| Co-precipitation + cation exchange | 86-93 | 0.1 mBq L⁻¹ | Tomita et al., 2015 |
| Seawater| AG 50W-X8 + Sr Resin       | 67 ± 22    | 1-3 mBq L⁻¹   | Karube et al., 2016      |
| Seawater| AG1-X8, AG 50W-X8          | 67-93      | 0.02-0.05 mBq L⁻¹ | Castrillejo et al., 2016 |
| Seawater| Co-precipitation + DGA     | 95.5 ± 2.3 | 1.5 mBq L⁻¹  | Tazoe et al., 2016       |
| Seawater| Sr resin + DGA resin       | 85.4 ± 3.3 | 1.7-3.5 Bq L⁻¹ | Tayeb et al., 2016       |
| Seawater| Co-precipitation + DGA     | 80         | 0.2 mBq L⁻¹   | Tazoe et al., 2017       |
| Water   | Sr resin                   | 97         | 14.5 Bq L⁻¹   | Kolacinska et al., 2017  |
| Seawater| Co-precipitation + Sr Disk | 84.2 ± 0.4 | 35 mBq L⁻¹    | Uesugi et al., 2018      |

Table 1. Analytical procedure for purification ⁹⁰Sr in environmental water
5. Conclusion
This mini-review summarized the literature works for developing selective Sr determination in high concentration of Ca using Ca(OH)$_2$ precipitation and purification of Y by Ba(Ra)SO$_4$ with focus in its use in alkaline waters. The result confirmed the capability of conventional procedure of co-precipitation method is effective for removing Sr and Y from waters. Co-precipitation method using Ca(OH)$_2$ and Ba(Ra)SO$_4$ can be separated is safer for the laboratory staff, environment and cheaper. For the determination of $^{90}$Sr, a more effective and specific separation of Ba and Ra can be achieved by Ba(Ra)SO$_4$ precipitation. This procedure can be used simultaneously for separation and further purification of $^{90}$Y from radiostrontium. The procedure of Ca(OH)$_2$ precipitation combined with Ba(Ra)SO$_4$ has been successfully applied for $^{90}$Sr analysis of 45l seawater and 200l drinking water samples.

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