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Validation of Rapid Radiochemical Method for Total Radiostrontium (Sr-90) in Brick Samples for Environmental Remediation Following Radiological Incidents

U.S. Environmental Protection Agency

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This report was prepared for the National Analytical Radiation Environmental Laboratory of the Office of Radiation and Indoor Air and the National Homeland Security Research Center of the Office of Research and Development, United States Environmental Protection Agency (EPA). It was prepared by Environmental Management Support, Inc., of Silver Spring, Maryland, under contract EP-W-13-016, Task Order 0014, managed by Dan Askren. This document has been reviewed in accordance with EPA policy and approved for publication. Note that approval does not signify that the contents necessarily reflect the views of the Agency. Mention of trade names, products, or services does not convey EPA approval, endorsement, or recommendation.

CONTENTS

Figure

Tables

Acronyms, Abbreviations, Units, and Symbols

AAL	analytical action level
ACS	American Chemical Society
APS	analytical protocol specification
Bq	becquerel
CL_{NC}	critical net concentration
CSU	combined standard uncertainty
Ci	curie
cpm	counts per minute
cps	counts per second
d	day
DL	discrimination level
dpm	disintegrations per minute
dps	disintegrations per second
DQO	data quality objective
DRP	discrete radioactive particle
E&Z	Eckert & Ziegler Analytics
EPA	U.S. Environmental Protection Agency
FRMAC	Federal Radiological Monitoring and Assessment Center
ft	foot
g	gram
gal	gallon
G-M	Geiger-Muller [counter or probe]
GPC	gas-flow proportional counter
Gy	gray
h	hour
ICP-AES	inductively coupled plasma – atomic emission spectrometry
ID	identifier/identification number
IND	improvised nuclear device
IUPAC	International Union of Pure and Applied Chemistry
kg	kilogram (10^3 gram)
L	liter
Lc	critical level
LCS	laboratory control sample
m	meter
M	molar
MARLAP	*Multi-Agency Radiological Laboratory Analytical Protocols Manual*
MDA	minimum detectable activity
MDC	minimum detectable concentration
MeV	mega electron volts (10^6 electron volts)
mg	milligram (10^{-3}grams)
min	minute
mL	milliliter (10^{-3} liter)
mm	millimeter (10^{-3} meter)
MQO	measurement quality objective
MS	matrix spike

MVRM	method validation reference material
µm	micrometer (10^{-6} m)
µCi	microcurie (10^{-6} curie)
NAREL	EPA's National Analytical Radiation Environmental Laboratory, Montgomery, AL
NHSRC	EPA's National Homeland Security Research Center, Cincinnati, OH
NIST	National Institute of Standards and Technology
ORD	EPA Office of Research and Development
ORIA	EPA Office of Radiation and Indoor Air
φ_{MR}	required relative method uncertainty
pCi	picocurie (10^{-12} curie)
PPE	personal protective equipment
ppm	parts per million
PT	proficiency test
QAPP	quality assurance project plan
R	Roentgen – unit of X and γ radiation exposure in air
rad	unit of radiation absorbed dose in any material
RDD	radiological dispersal device
rem	roentgen equivalent: man
s	second
SI	International System of Units
STS	sample test source
Sv	sievert
u_{MR}	required method uncertainty
WCS	working calibration source
wt%	percent by mass
y	year

Radiometric and General Unit Conversions

To Convert	To	Multiply by	To Convert	To	Multiply by
years (y)	seconds (s)	3.16×10^{7}	s	y	3.17×10^{-8}
	minutes (min)	5.26×10^{5}	min		1.90×10^{-6}
	hours (h)	8.77×10^{3}	h		1.14×10^{-4}
	days (d)	3.65×10^{2}	d		2.74×10^{-3}
disintegrations per second (dps)	becquerel (Bq)	1	Bq	dps	1
Bq	picocurie (pCi)	27.0	pCi	Bq	3.70×10^{-2}
Bq/kilogram (kg)	pCi/gram (g)	2.70×10^{-2}	pCi/g	Bq/kg	37.0
Bq/cubic meters (m^3)	pCi/g	2.70×10^{-2}	pCi/g	Bq/m^3	37.0
Bq/m^3	Bq/L	10^{-3}	Bq/L	Bq/m^3	10^{3}
microcuries per milliliter (μCi/mL)	pCi/g	10^{9}	pCi/g	μCi/mL	10^{-9}
disintegrations per minute (dpm)	μCi	4.50×10^{-7}	pCi	dpm	2.22
	pCi	4.50×10^{-1}			
cubic feet (ft^3)	m^3	2.83×10^{-2}	m^3	ft^3	35.3
gallons (gal)	liters (L)	3.78	L	gal	0.264
gray (Gy)	rad	10^{2}	rad	Gy	10^{-2}
roentgen equivalent: man (rem)	sievert (Sv)	10^{-2}	Sv	rem	10^{2}

NOTE: Traditional units are used throughout this document instead of the International System of Units (SI). Conversion to SI units will be aided by the unit conversions in this table.

Acknowledgments

The U.S. Environmental Protection Agency's (EPA's) Office of Radiation and Indoor Air's (ORIA) National Analytical Radiation Environmental Laboratory (NAREL), in conjunction with the EPA Office of Research and Development's National Homeland Security Research Center (NHSRC) developed this method validation report. Dr. John Griggs served as project lead. Several individuals provided valuable support and input to this document throughout its development. Special acknowledgment and appreciation are extended to Kathleen M. Hall, of NHSRC.

We also wish to acknowledge the valuable suggestions provided by the staff of NAREL, who conducted the method validation studies. Dr. Keith McCroan, of NAREL, provided significant assistance with the equations used to calculate minimum detectable concentrations and critical levels. Numerous other individuals, both inside and outside of EPA, provided comments and criticisms of this method, and their suggestions contributed greatly to the quality, consistency, and usefulness of the final method. Environmental Management Support, Inc. provided technical support.

1. Introduction

Rapid methods need to be developed and validated for processing samples taken in response to a radiological incident. In order to address this need, a project was initiated to develop rapid methods that can be used to prioritize environmental sample processing as well as provide quantitative results that meet measurement quality objectives (MQOs) that apply to the intermediate and recovery phases of an incident.[1] Similar to the rapid method project initiated in 2007 for other radionuclides in water samples (EPA 2008), this rapid method development project for a brick matrix addressed four different radionuclides in addition to ^{90}Sr: ^{241}Am, ^{nat}U, ^{226}Ra, and $^{239/240}$Pu. Each of these radionuclides will have separate method validation reports for a brick matrix.

The method validation plan developed for the rapid methods project follows the guidance in *Method Validation Guide for Qualifying Methods Used by Radiological Laboratories Participating in Incident Response Activities* (EPA 2009), *Validation and Peer Review of U.S. Environmental Protection Agency Radiochemical Methods of Analysis* (2006), and Chapter 6 of *Multi-Agency Radiological Laboratory Analytical Protocols Manual* (MARLAP) (EPA 2004). The method was evaluated according to MARLAP method validation “Level C.” The method formulated was preliminarily tested at a commercial laboratory and refinements to the method made according to the feedback from the laboratory and the quality of the generated results. For the method validation process, the laboratory analyzed several sets of blind proficiency test (PT) samples according to specifications that meet established MQOs and guidance outlined in *Radiological Sample Analysis Guide for Radiological Incidents – Radionuclides in Soil* (EPA 2012). The MQO specification for the required method uncertainty of 0.32 pCi/g was based on a ^{90}Sr brick concentration similar to the MQO for the soil matrix, i.e., at approximately 1×10^{-5} risk limit for a 50-year exposure of 2.4 pCi/g.

This report provides a summary of the results of the method validation process for a combination of two methods: *Rapid Method for Sodium Hydroxide Fusion of Concrete and Brick Matrices Prior to Americium, Plutonium, Strontium, Radium, and Uranium Analyses for Environmental Remediation Following Radiological Incidents* (Attachment II) and *Rapid Radiochemical Method for Total Radiostrontium (Sr-90) in Building Materials for Environmental Remediation Following Radiological Incidents* (Attachment III). In this document, the combined methods are referred to as “combined rapid ^{90}Sr - Brick method.” The method validation process is applied to the fusion dissolution of soil and building material using sodium hydroxide and the subsequent separation and quantitative analysis of ^{90}Sr using a calibrated gas proportional counter to detect the beta particles of ^{90}Sr (β_{mean} = 0.547 million electron volts [MeV] and ^{90}Y (β_{mean} = 0.934 MeV). The chemical yield of each sample processed was determined by gravimetric means of a $Sr(NO_3)_2$ precipitate. The laboratory’s complete report, including a case narrative and a compilation of the reported results for this study, can be obtained by contacting EPA’s National Analytical Radiation Environmental Laboratory (NAREL) (http://www.epa.gov/narel/contactus.html).

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[1] ORIA and the Office of Research and Development jointly undertook the rapid methods development projects. The MQOs were derived from Protective Action Guides determined by ORIA.

2. Radioanalytical Methods

The combined rapid ^{90}Sr - Brick method was written in a format consistent with EPA guidance and conventions. The rapid method was formulated to optimize analytical throughput for sample preparation, fusion process, chemical processing and radiation detection. The final step of the fusion method prepares the sample for separation and analysis by the *Rapid Radiochemical Method for Total Radiostrontium (Sr-90) in Building Materials for Environmental Remediation Following Radiological Incidents.* Specifications for sample processing were incorporated into the rapid method. These specifications are reflected in the scope and application and in the body of the methods. The specifications include the use of a gravimetric yield monitor and the required method uncertainty. Known interferences are addressed in Section 4 of the attached method (Attachment III). For this validation study, the laboratory used a 100-minute counting time for the three test level samples as well as for the required minimum detectable concentration (MDC) samples. A 1.5-g sample of the brick matrix was processed (see Attachment IV for the chemical composition of the brick matrix). A summary of the rapid method is presented in Section 8.1 prior to presenting the experimental results of the method validation analyses.

The final combined rapid ^{90}Sr - Brick method is included as Attachments II and III to this report. Although this final method is a departure from the originally tested method, the incorporated revisions are significant improvements and do not change the general methodology. The validation process was performed using the final combined method in the attachments.

3. Method Validation Process Summary

The method validation plan for the combined rapid ^{90}Sr - Brick method follows the guidance provided in *Method Validation Guide for Qualifying Methods Used by Radiological Laboratories Participating in Incident Response Activities* (EPA 2009), *Validation and Peer Review of U.S. Environmental Protection Agency Radiochemical Methods of Analysis* (EPA 2006), and Chapter 6 of MARLAP (2004). This method validation process was conducted under the generic *Quality Assurance Project Plan Validation of Rapid Radiochemical Methods for Radionuclides Listed in EPA's Standardized Analytical Methods (SAM) for Use During Homeland Security Events* (EPA 2011). The combined rapid ^{90}Sr - Brick method is considered a "modified application" of an existing method (EPA 2004, Section 6.6.3.5). Therefore, the method was evaluated according to MARLAP method validation "Level C." More specifically, the method was validated against acceptance criteria for the required method uncertainty (u_{MR}) at a specified analytical action level (AAL) concentration and the required MDC. In addition, analytical results were evaluated for statistical bias, absolute bias for blank samples and relative bias at each of the three test level radionuclide activities. The gravimetric yield of the method was also evaluated as a characteristic of method ruggedness.

The method validation process was divided into four phases:

1. Phase I
    a. Laboratory familiarization with the method concept.
    b. Set-up of the laboratory and acquisition of reagents, standards and preparation of in-house performance test (PT) samples.

    c. Perform preliminary tests of the rapid fusion method.
    d. Make changes to improve the method based on the preliminary tests.
2. Phase II
    a. Conduct blank sample analyses to assess the method critical level concentration.
    b. Conduct method validation for required method uncertainty.
3. Phase III
    a. Conduct verification of the required MDC.
4. Phase IV
    a. Report results.
    b. Laboratory writes report to describe the process and narratives on the method.
    c. Review and comment on method.
    d. Environmental Management Support, Inc., writes method validation report, which is reviewed by laboratory.

During Phases I, II, and III, the laboratory processed and evaluated batch quality control samples according to their laboratory quality manual, including an analytical reagent blank, laboratory control sample (LCS) and a sample duplicate.

The objective of the first (preliminary) phase was to familiarize the laboratory with the formulated rapid method and then gain hands-on experience in using the rapid method to identify areas that might require optimization. During this phase, the laboratory processed samples of blank brick material, and blank brick that was spiked in-house with ^{90}Sr – activities consistent with evaluating the required method uncertainty at the AAL and the required MDC (see "Sr-90 Method Validation Test Concentrations and Results," Table 1). The analysis of the blank and laboratory spiked brick samples used in Phase I provided insight into the feasibility of the proposed method. Based on information and experience gained during Phase I practice runs, the combined rapid ^{90}Sr - Brick method was optimized without compromising data collected during the validation process in Phases II and III. The method was not subjected to a "formal" method validation evaluation process in Phase I.

During Phases II and III of the method validation process, the laboratory analyzed PT samples (consisting of method validation reference materials [MVRM]) provided by an external, National Institute of Standards and Technology (NIST)-traceable source manufacturer (Eckert & Ziegler Analytics (E&Z), Atlanta, GA). The MVRM was pulverized brick prepared by E&Z. The macro-analysis of brick material is provided in Attachment IV. The laboratory was instructed to analyze specific blind PT samples having concentration levels consistent with validation test levels for the required method uncertainty and the required MDC. The test levels of the PT samples are listed in Tables 1 and 2. Following completion of the method validation studies, comments from the laboratory were evaluated, and the method revised to conform to the documented "as-tested" conditions in Phases II and III. Thus the validation data presented in this report reflect the combined final method included in the attachments to this document.

4. Participating Laboratory

NAREL validated the combined rapid ^{90}Sr - Brick method using chemically characterized brick samples spiked with NIST-traceable ^{90}Sr source. NAREL has demonstrated satisfactory performance in national radioanalytical performance proficiency programs for radionuclides in water and other matrices.

5. Measurement Quality Objectives

The combined rapid ^{90}Sr - Brick method was developed to meet pre-established MQOs for the rapid methods project. The selected MQOs included the radionuclide concentration range, the required method uncertainty at a specified radionuclide concentration (e.g., AAL), and the required MDC. The required relative method uncertainty (φ_{MR}) for the combined rapid ^{90}Sr - Brick method was set at 13%[2] at a targeted brick AAL equal to 2.41 pCi/g, which is approximately the 1×10^{-5} risk concentration for a 50-year exposure period for a soil matrix. This soil concentration value is based on guidance found in the Federal Radiological Monitoring and Assessment Center (FRMAC).[3] The target MDC for the combined rapid ^{90}Sr - Brick method for the brick matrix was 0.40 pCi/g (~17% of the AAL). However, the PT sample supplier generated method validation and MDC samples having ^{90}Sr concentrations slightly different than the targeted values. Table 1 summarizes the targeted MQOs for the ^{90}Sr method validation process and the study test values based on the actual spiked concentrations of the PT samples. It should be noted that the method was validated for a brick matrix having a typical chemical composition and four additional radionuclides in concentrations corresponding to a 10^{-5} risk AAL for soil. The brick concentrations for the four other radionuclides were ^{241}Am (1.570 pCi/g), ^{238}U (12.35 pCi/), ^{226}Ra (4.755 pCi/g), and ^{239}Pu (1.890 pCi/g). The PT sample supplier provided test data for 10, 1-gram samples that documents the spread in the spike in the samples as a 1.59% standard deviation of the distribution of results.

Table 1 – Sr-90 Method Validation Test Concentrations and Results

	Target Value	**Known Value, pCi/g (*k* = 1)**	**Average Measured Value**	**Required Method Uncertainty, u_{MR}**	**Standard Deviation [2]**
MDC	0.40	0.4040 ±0.0093	0.51	—	0.19
½ × AAL	1.21	1.210 ± 0.028	1.160	0.32	0.073
AAL	2.41	2.440 ± 0.063	2.17	0.32	0.23
3 × AAL	7.23	7.28 ± 0.16	6.83	0.95 [1]	0.43

[1] The value of 0.95 pCi/g is the absolute value for the required method uncertainty and represents 13% of 7.28 pCi/g.

[2] Calculated standard deviation of the 10 and 5 measurement results for the MDC and Test Level samples, respectively.

6. Method Validation Plan

The combined rapid ^{90}Sr - Brick method was evaluated for the six important performance characteristics for radioanalytical methods specified in *Quality Assurance Project Plan*

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[2] Type I and II decision error rates were set at $z_{1-\alpha}= 0.01$ and $z_{1-\beta}= 0.05$. The required method uncertainty is calculated using the formula, $u_{MR} = (\text{AAL-DL})/[z_{1-\alpha} + z_{1-\beta}]$ where the analytical action level (AAL) is as noted above and the discrimination level (DL) is ½ the AAL.

[3] Federal Radiological Monitoring and Assessment Center. Appendix C of the FRMAC Manual (FRMAC 2010) or calculated using Turbo. FRMAC 2010 available from Sandia National Laboratory.

Validation of Rapid Radiochemical Methods for Radionuclides Listed in EPA's Standardized Analytical Methods (SAM) for Use During Homeland Security Events (EPA 2011). These characteristics include method uncertainty, detection capability, bias, analyte activity range, method ruggedness, and method specificity. A summary of the manner in which these performance characteristics were evaluated is presented below. The chemical yield of the method, an important characteristic for method ruggedness, was also evaluated.

6.1 Method Uncertainty

The method uncertainty of the combined rapid ^{90}Sr - Brick method was evaluated at the AAL concentration (2.44 pCi/g known value) specified in the MQOs presented in Table 1. In accordance with MARLAP method validation "Level C," this is a new application and was evaluated at each of three test concentration levels, one of which was the AAL equivalent activity concentration to approximately 1×10^{-5} risk for a soil matrix. The laboratory analyzed five replicate external MVRM PT samples containing ^{90}Sr activities at approximately one-half the AAL, the AAL and three times the AAL. The method was evaluated against the required method uncertainty, u_{MR} = 0.32 pCi/g, at and below the AAL, and against the relative required method uncertainty, φ_{MR} = 13% of the known test value, above the AAL. The test level concentrations analyzed are listed in Table 1 "Known Value."

6.2 Detection Capability

The detection capability of the combined rapid ^{90}Sr - Brick method was evaluated to meet a required MDC of 0.4040 pCi/g as specified in Table 2. In accordance with the guidance provided in *Method Validation Guide for Radiological Laboratories Participating in Incident Response Activities* (EPA 2009), the laboratory estimated the critical net concentration based on the results of seven reagent blank samples. Results from 10 replicate samples having an "as tested" concentration at the required MDC were compared to the critical net concentration to determine method detection capability.

Table 2 – Sample Identification and Test Concentration Level for Evaluating the Required Minimum Detectable Concentration

Test Sample Designation	Number of Samples Prepared	Nuclide	MDC Known Value (pCi/g)	Mean Measured Concentration (pCi/g) [1]
MA1-MA10 (Brick MDC samples)	10	^{90}Sr	0.4040 ± 0.0093 (k = 1)	0.51 ± 0.19 (k = 1)
BMDC1-BMDC7 (Brick[2] matrix blanks)	7	^{90}Sr	—	0.119 ± 0.077

[1] Mean and standard deviation of 10 spiked samples and 7 blanks. The stated combined standard uncertainty (CSU) includes the uncertainty in the ^{90}Sr reference standard used to prepare the samples. The concentration of ^{90}Sr in the blank brick sample matrix was not statistically different than zero. No limit for detection was provided.

[2] Blank brick matrix supplied by Eckert & Ziegler Analytics, Atlanta, Georgia.

6.3 Method Bias

Two types of method bias were evaluated, absolute and relative.

Absolute Bias

Absolute bias was determined as a method performance parameter. The results from the seven blank brick samples for the required MDC evaluation were assessed for absolute bias according to the protocol and equation presented in the *Method Validation Guide for Qualifying Methods Used by Radiological Laboratories Participating in Incident Response Activities* (EPA 2009). There was no acceptance limit for bias established for the method in the validation process.

The following protocol was used to test the combined rapid ^{90}Sr - Brick method for absolute bias:

1. Calculate the mean ($\overline{X}$) and estimated standard deviation (s_x) for “N” (at least seven) blank sample net results.

2. Use the equation below to calculate the |T| value:

$$\left|T\right| = \frac{\left|\overline{X}\right|}{s_X / \sqrt{N}} \qquad (1)$$

   1. An absolute bias in the measurement process is indicated if

$$\left|T\right| > t_{1-\alpha/2}(N-1) \qquad (2)$$

where $t_{1-\alpha/2}\ (N-1)$ represents the (1 – α/2)-quantile of the *t*-distribution with *N*–1 degrees of freedom. For seven blanks, an absolute bias is identified at a significance level of 0.05, when |T| > 2.447.

Relative Bias

The results from the five external PT brick samples for each of the three test levels were evaluated for relative bias according to the protocol and equation presented in the *Method Validation Requirements for Qualifying Methods Used by Radioanalytical Laboratories Participating in Incident Response Activities*. No acceptable relative bias limit was specified for this method validation process.

The following protocol was used to test the combined rapid ^{90}Sr - Brick method for relative bias:

1. Calculate the mean ($\overline{X}$) and estimated standard deviation (s_X) of the replicate results for each method validation test level.

2. Use the equation below to calculate the |T| value

$$\left|T\right| = \frac{\left|\overline{X} - K\right|}{\sqrt{s_X^2 / N + u^2(K)}} \qquad (3)$$

where:

$\overline{X}$ is the average measured value
s_X is the experimental standard deviation of the measured values
N is the number of replicates
K is the reference value
$u(K)$ is the standard uncertainty of the reference value

A relative bias in the measurement process is indicated if

$$\left|T\right| > t_{1-\alpha/2(\nu_{eff})} \tag{3a}$$

The number of *effective degrees of freedom* for the T statistic is calculated as follows:

$$\nu_{\text{eff}} = (N-1)\left(1+\frac{u^2(K)}{s_X^2/N}\right)^2 \tag{4}$$

ν_{eff} as calculated by the equation generally is not an integer so ν_{eff} should be truncated (rounded down) to an integer. Then, given the significance level, 0.05, the critical value for "|T|" is defined to be $t_{1-\alpha/2(\nu\text{eff})}$, the $(1 - \alpha/2)$-quantile of the *t*-distribution with ν_{eff} degrees of freedom (see MARLAP Appendix G, Table G.2).

6.4 Analyte Concentration Range

The combined rapid ^{90}Sr - Brick method was evaluated for the required method uncertainty at three test level activities. The five replicate PT samples from each test level concentration were analyzed. The proposed (target) and "as tested" (known) test level activities are presented in Table 1. Note that the final test concentration values for the PT samples varied from the proposed test levels but that these values were well within the sample preparation specifications provided to the PT sample provider.

6.5 Method Specificity

The brick sample is fused with sodium hydroxide in zirconium crucibles for ~15 minutes at 600 °C in a furnace. The fused material is dissolved using water and transferred to a centrifuge tube. A preconcentration step with calcium phosphate enhanced with iron hydroxide is used to remove the Sr from the alkaline matrix. The precipitate is dissolved in dilute acid and a calcium fluoride precipitation is performed to further remove brick matrix components such as silicates that can adversely affect column flow. The precipitate is dissolved in strong nitric acid with boric acid and aluminum present and the solution is passed through a Sr Resin extraction chromatography cartridges that selectively retains strontium while allowing most interfering radionuclides and matrix constituents to pass through to waste. Strontium is eluted from the column with 0.05 M HNO_3 and taken to dryness in a tared, stainless steel planchet. The planchet containing the strontium nitrate precipitate is weighed to determine the strontium yield.

6.6 Method Ruggedness

The sodium hydroxide fusion has been used successfully on U.S. Department of Energy's Mixed Analyte Performance Evaluation Program soil samples containing refractory actinides and strontium. The method is rapid and simple yet very rugged. The calcium fluoride step with HF present removes silicates, which tend to clog the resin cartridges. The sample size was limited to 1.5 g of brick to minimize any undissolved solids in the column load solutions. A calcium phosphate co-precipitation method was used instead of calcium carbonate since complete dissolution of the final precipitate in the column load solution could not be achieved with calcium carbonate.

The method validation reference material samples contained other beta emitting radionuclides (U and ^{226}Ra decay products).

7. Techniques Used to Evaluate the Measurement Quality Objectives for the Rapid Methods Development Project

A general description of the specifications and techniques used to evaluate the required method uncertainty, required MDC and bias was presented in Section 6. The detailed method evaluation process for each MQO, the bias and the radiochemical yield is presented in this section.

7.1 Required Method Uncertainty

The combined rapid ^{90}Sr - Brick method was evaluated following the guidance presented for "Level C Method Validation: Adapted, Newly Developed Methods, Including Rapid Methods" in *Method Validation Guide for Qualifying Methods Used by Radiological Laboratories Participating in Incident Response Activities* (EPA 2009) and Chapter 6 of *Multi-Agency Radiological Laboratory Analytical Protocols Manual* (EPA 2004).

MARLAP "Level C" method validation requires the laboratory to conduct a method validation study wherein five replicate samples from each of the three concentration levels are analyzed according to the method. The concentration test levels analyzed are listed in Table 1. For validation "Level C," externally prepared PT samples consisting of NIST-traceable ^{90}Sr were used to spike MVRM. In order to determine if the proposed method met the rapid methods development project MQO requirements for the required method uncertainty (u_{MR} = 0.32 pCi/g), each external PT sample result was compared with the method uncertainty acceptance criteria listed in the table below. The acceptance criteria stated in Table 3 for "Level C" validation stipulate that, for each test sample analyzed, the measured value had to be within ± 2.9 u_{MR} (required method uncertainty) for test level activities at or less than the AAL, or ± 2.9 φ_{MR} (required relative method uncertainty) for test level activities above the AAL.

Table 3 – MARLAP Level C Acceptance Criteria

MARLAP Validation Level	Application	Sample Type [1]	Acceptance Criteria [2]	Number of Test Levels	Number of Replicates	Total Number of Analyses
C	New Application	Method Validation Reference Materials	Measured value within ± 2.9 u_{MR} or ± 2.9 φ_{MR} of validation value	3	5	15

[1] "Method Validation Reference Materials" is not a requirement of MARLAP for these test levels. However, in order to assure laboratory independence in the method validation process, a NIST-traceable source manufacturer was contracted to produce the testing materials for Phases II and III of the project.

[2] The measured value must be within ± 2.9 u_{MR} for test level concentrations at or less than the AAL and within ± 2.9 φ_{MR} for a test level concentration above the AAL. It was assumed that the uncertainty of a test sample concentration will be negligible compared to the method uncertainty acceptance criteria and was not incorporated in the acceptance criteria.

7.2 Required Minimum Detectable Concentration

The analytical results reported for the PT samples having a ^{90}Sr concentration at the tested MDC of 0.4040 pCi/g were evaluated according to Sections 5.5.1 and 5.5.2 of Testing for the Required MDC in *Method Validation Guide for Qualifying Methods Used by Radiological Laboratories Participating in Incident Response Activities* (EPA 2009). NAREL analyzed the external PT samples in accordance with the proposed rapid method.

Critical Net Concentration

In order to evaluate whether the combined method can meet the required MDC (0.4040 pCi/g), the critical net concentration, as determined from the results of analytical blanks, must be calculated. The critical net concentration with a Type I error probability of α = 0.05, was calculated using the following equation (consistent with MARLAP, Chapter 20, Equation 20.35):

$$CL_{\mathrm{NC}}(\mathrm{pCi}) = \mathrm{t}_{1-\alpha}(\mathrm{n}-1) \times \mathrm{s}_{\mathrm{Blanks}} \tag{5}$$

where s_{Blanks} is the standard deviation of the *n* blank-sample net results (corrected for instrument background) in radionuclide concentration units of pCi/g, and $t_{1-\alpha}(n-1)$ is the (1 – α)-quantile of the *t*-distribution with *n*–1 degrees of freedom (see MARLAP Table G.2 in Appendix G). For this method validation study a Type I error rate of 0.05 was chosen.

For seven (minimum) blank results (six degrees of freedom) and a Type I error probability of 0.05, the previous equation reduces to:

$$CL_{\mathrm{NC}}(\mathrm{pCi}) = 1.94 \times \mathrm{s}_{\mathrm{Blanks}} \tag{6}$$

The use of the above equations assumes that the method being evaluated has no bias.

Verification of Required MDC

Each of the 10 analytical results reported for the PT samples having a concentration at the required MDC for ^{90}Sr (0.4040 pCi/g) was compared to the estimated critical net concentration for the method. The following protocol was used to verify a method's capability to meet the required method MDC for a radionuclide-matrix combination:

I. Analyze a minimum of seven matrix blank samples for the radionuclide.

II. From the blank sample net results, calculate the estimated *Critical Net Concentration*, CL_{NC}.

III. Analyze 10 replicate samples spiked at the required MDC.

IV. From the results of the 10 replicate samples spiked at the required MDC, determine the number (Y) of sample results at or below the estimated CL_{NC}.

V. If $Y \leq 2$, the method evaluated at the required MDC passes the test for the required MDC specification.

VI. If $Y > 2$, the method evaluated at the required MDC fails the test for the required MDC specification.

8. Evaluation of Experimental Results

Only the experimental results for Phases II and III of the method validation process are reported and evaluated in this study. Information presented in this section will include results for Sections 6 and 7. The ^{90}Sr analytical results were evaluated for the required method uncertainty, required MDC and bias. In addition, the mean radiochemical yield for the method for Phases II and III is reported to provide the method user the expected mean and range of this method performance characteristic.

8.1 Summary of the Combined Rapid ^{90}Sr - Brick Method

The brick sample is fused with sodium hydroxide, dissolved in water and preconcentrated using calcium phosphate and calcium fluoride precipitations. The brick samples are analyzed for ^{90}Sr using the method for building materials (*Rapid Radiochemical Method for Total Radiostrontium (Sr-90) in Building Materials for Environmental Remediation Following Radiological Incidents*, Attachment III).

8.2 Required Method Uncertainty

Table 4A summarizes the ^{90}Sr results for a 1.5-g sample and the acceptability of each result compared to the acceptance criteria presented in Section 7.1. The final sample test sources were a $SrNO_3$ precipitate counted on a gas proportional counting system for 100 minutes that was capable of meeting a required method uncertainty of 0.32 pCi/g at and below the AAL of 2.440 pCi/g.

Table 4A – Sr-90 Analytical Results for Required Method Uncertainty Evaluation

Nuclide: Sr-90 Matrix: Brick AAL Tested: 2.440 pCi/g
Proposed Method: Combined Rapid ^{90}Sr - Brick Method
Required Method Validation Level: MARLAP “C”
Required Method Uncertainty, u_{MR}: 0.32 pCi/g at and below AAL; 13% above AAL
Acceptance Criteria:
Test Levels 1 and 2: 2.9 × u_{MR} = ± 0.92 pCi/g of quoted known value of sample in test level
Test Level 3: 2.9 × φ_{MR} = ± 37.7% of quoted known value of sample in test level (7.28 pCi/g)

Test Level 1

Sample (Analytics Batch SO-U1)	pCi/g Known	Uncertainty [1] (pCi/g)	pCi/g Measured	CSU [2] (pCi/g)	Allowable Range (pCi/g)	Acceptable Y/N
S01	1.210	0.028	1.26	0.14	0.29 – 2.1	Y
S02			1.14	0.14		Y
S03			1.19	0.15		Y
S04			1.15	0.14		Y
S05			1.06	0.14		Y

Test Level 2

Sample (Analytics Batch SO-U2)	pCi/g Known	Uncertainty [1] (pCi/g)	pCi/g Measured	CSU [2] (pCi/g)	Allowable Range (pCi/g)	Acceptable Y/N
SR6	**2.440**	**0.063**	**1.98**	**0.15**	**1.5- 3.4**	**Y**
SR7			**2.13**	**0.16**		**Y**
SR8			**1.94**	**0.15**		**Y**
SR9			**2.48**	**0.18**		**Y**
SR10			**2.33**	**0.17**		**Y**

Test Level 3						
Sample (Analytics Batch SO-U3)	**pCi/g Known**	**Uncertainty [1] (pCi/g)**	**pCi/g Measured**	**CSU [2] (pCi/g)**	**Allowable Range (pCi/g)**	**Acceptable Y/N**
S14	7.28	0.16	7.20	0.27	4.5 – 10	Y
S15			6.81	0.26		Y
S16			6.24	0.24		Y
S17			6.61	0.25		Y
S18			7.30	0.27		Y

[1] Quoted uncertainty (*k* = 1) by the radioactive source manufacturer.
[2] Combined standard uncertainty (CSU), coverage factor *k*=1.

As a measure of the expected variability of results for a test level, the calculated standard deviation of the five measurements of each test level is provided in Table 4B. The standard deviation of the analytical results for a test level was much smaller than the required method uncertainty.

Table 4B – Experimental Standard Deviation of the Five PT Samples by Test Level

Test Level	**Mean Concentration Measured (pCi/g)**	**Standard Deviation of Measurements (pCi/g)**	**Required Method Uncertainty (pCi/g)**
1	1.160	0.073	0.32
2 (AAL)	2.17	0.23	0.32
3	6.83	0.43 (6.3%)	0.95 [1] (13%)

[1] This figure represents the absolute value of the required method uncertainty, calculated by multiplying the known value of Test Level 3 (7.28 pCi/g) by the required relative method uncertainty.

8.3 Required Minimum Detectable Concentration

The combined rapid ^{90}Sr - Brick method was validated for the required MDC using a 1.5 g sample and a gas proportional counting time of 100 minutes.

Tables 5 and 6 summarize the ^{90}Sr results and the acceptability of the method's performance specified in Section 7.2 to meet the tested required MDC of 0.4040 pCi/g.

Table 5 – Reported ^{90}Sr Concentration Blank Brick Samples

Sample ID	Concentration (pCi/g)	CSU [1] (pCi/g)
S41	0.24	0.11
S42	0.11	0.10
S43	0.069	0.088
S44	0.058	0.097
S45	0.21	0.10
S46	0.106	0.085
S47	0.042	0.084
Mean [2]	**0.119**	
Standard Deviation	**0.077**	
Critical Net Concentration (pCi/g)	**0.15**	

[1] Combined standard uncertainty (CSU), *k*=1 or coverage factor of 1.

[2] Mean and standard deviation were calculated before rounding.

Critical Net Concentration

The critical net concentration for the method under evaluation was calculated using Equation 6 from Section 7.2. Based on the results of the seven blanks (Table 5), the critical net concentration for the combined method was determined to be 0.15 pCi/g.

Required MDC

A summary of the reported results for samples containing ^{90}Sr at the required MDC is presented in Table 6. The mean measured value and standard deviation of the ten ^{90}Sr MDC test samples were calculated as 0.51± 0.19 pCi/g (*k*=1). Each result was compared to the critical net concentration of 0.15 pCi/g. If the result was at or below the critical net concentration, a “Y” qualifier was applied to the sample result; otherwise an “N” qualifier was applied. As presented in the table, the number of Y qualifiers is ≤ 2, so the combined rapid ^{90}Sr - Brick method evaluated passes the test for the required MDC specification (see *Method Validation Guide for Qualifying Methods Used by Radiological Laboratories Participating in Incident Response Activities* [EPA 2009] for a description of the test).

Table 6 – Reported Results for Samples Containing ^{90}Sr at the As-Tested MDC Value (0.4040 pCi/g)

Sample ID	Concentration (pCi/g)	CSU [1] (pCi/g)	Test Result ≤ CL_{NC} [3]
S30	0.38	0.11	N
S31	0.99	0.14	N
S32	0.34	0.11	N
S33	0.45	0.11	N
S34	0.55	0.11	N
S35	0.48	0.12	N
S36	0.44	0.12	N
S37	0.37	0.10	N
S38	0.44	0.11	N
S39	0.61	0.11	N
Mean [2]	**0.51**		—
Standard Deviation of Results	**0.19**		
	CL_{NC}	**0.15 pCi/g**	—
	Acceptable maximum values ≤ CL_{NC} (Y)	2	**—**
	Number of results > CL_{NC}	—	10
	Number of results ≤ CL_{NC}	—	0
		Evaluation	**PASS**

[1] Combined standard uncertainty (CSU), coverage factor *k*=1.
[2] Mean and standard deviation were calculated before rounding.
[3] Critical net concentration.

Based on the validation study results, it may be concluded that the combined rapid ^{90}Sr - Brick method for 1.5 g sample and a 100-minute counting time is capable of meeting a required MDC for ^{90}Sr of 0.4040 pCi/g (the known value of the MDC PT sample).

8.4 Evaluation of the Absolute and Relative Bias

The ^{90}Sr results for the seven blank brick samples (Table 5), 10 MDC samples (Table 6) and the five replicate PT samples on the three test levels (Table 4A) were evaluated for absolute and relative bias according to the equations presented in Section 6.3. The results and interpretation of the evaluation are presented below in Table 7.

Table 7 – Absolute and Relative Bias Evaluation of the Combined Rapid ^{90}Sr - Brick Method

Type of Bias	Test Level	Known Value ± CSU (k=1) (pCi/g) [1]	Mean of Measurements ± Standard Deviation [2] (pCi/g)	Difference from Known	Number of Measurements/ Degrees of Freedom	|T|	t_{df}	Bias Yes / No
Absolute	Blanks	0.0000	0.119 ± 0.077	0.1193	7/6	4.11	2.45	Y
Relative	MDC	0.4040 ± 0.0093	0.51 ± 0.19	0.11	10/9	1.67	2.26	N
Relative	1	1.210 ± 0.028	1.160 ± 0.073	-0.050	5/12	1.16	2.18	N
Relative	2 – AAL	2.440 ± 0.063	2.17 ± 0.23	-0.27	5/7	2.22	2.36	N
Relative	3	7.28 ± 0.16	6.83 ± 0.43	-0.45	5/11	1.77	2.20	N

[1] The stated CSU includes the uncertainty in the ^{90}Sr reference standard used to prepare the samples and the standard deviation of the spiked test samples.

[2] Standard deviation of the measurements.

The results for the seven blank samples had a mean and standard deviation of 0.1193 ± 0.077 pCi/g. A statistical analysis of the data indicated that there was an absolute bias for the blank samples.

No relative bias was noted for the measurements performed on the 10 MDC or any of the method uncertainty test levels. The mean concentration of 0.51 pCi/g for the 10 MDC test samples falls within 0.106 pCi/g (or 26%) of the known value of 0.4040 pCi/g.

As determined by the paired-*t* test described in section 7, no relative bias was indicated for any of the method uncertainty test levels. The relative percent difference for each test level was:

- Test Level 1: -4.1%.
- Test Level 2: -11%.
- Test Level 3: -6.2%.

8.5 Method Ruggedness and Specificity

The results summarized in Table 8 represent the gravimetric yields for all three test levels, blanks, laboratory control samples and MDC samples that were processed in accordance with the final methods in Attachments II and III.

Table 8 – Summary of Sr-90 Gravimetric % Yield Results for Test and Quality Control Samples

Number of Samples	48
Mean Gravimetric Yield	85.2
Standard Deviation of Distribution (1σ)	6.8
Median	83.5
Minimum Value	73.8
5th Percentile	77.1
95th Percentile	98.9
Maximum Value	99.9

The yields for samples evaluated using this method are shown on Figure 1. The gravimetric yields were very good, with an average yield of 85.2% ± 6.8% (1 standard deviation). The lowest chemical yield was ~73.8% while 90% of the yields were between 77.1 and 98.9%. The high chemical yields are an indication of the ruggedness of the combined ^{90}Sr rapid method.

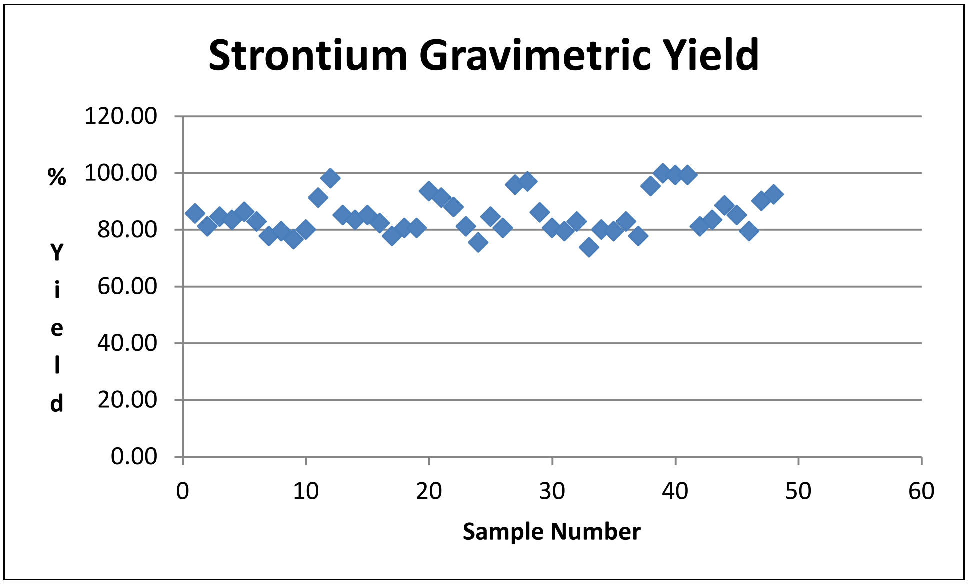

Figure 1 – Sr Yields for Method Based on Measurement of Sr(NO_3)$_2$

9. Timeline to Complete a Batch of Samples

NAREL kept a timeline log on processing a batch of samples and associated internal quality control samples. The total time to process a batch of samples, including counting of the samples and data review/analysis, was approximately 7.5 to 8.5 hours, assuming a 1 to 2-hour count time. NAREL's breakdown of the timeline by method-process step is presented in Attachment I (this information is also presented in more detail in the method flow chart in Attachment III, Section 17.5).

10. Reported Modifications and Recommendations

NAREL performed the method validation and made one significant modification to the method prior to analyzing samples for Phases II and III of the project. The selected modification provided by NAREL is listed below.

The primary modification was to employ a calcium phosphate precipitation instead of calcium carbonate since the calcium phosphate approach allowed the analysis of 1.5-g sample aliquants with less dissolution problems in the column load solution.

The following changes were made in the procedure *Rapid Method for Sodium Hydroxide Fusion of Concrete and Brick Matrices Prior to Americium, Plutonium, Strontium, Radium, and*

Uranium Analyses for Environmental Remediation Following Radiological Incidents (Attachment II).

NOTE: The step numbers below may have changed in the post-validation method in Attachment II.

1.1. Preconcentration of ^{90}Sr from Hydroxide Matrix (Brick)

NOTE: The preconcentration steps for ^{90}Sr in this section, using calcium phosphate instead of calcium carbonate, can also be applied to concrete samples but this will have to be validated by the laboratory. See Section 11.3 for steps validated for ^{90}Sr in concrete samples.

1.1.1. Transfer each fused sample to a 225-mL centrifuge tube, rinse crucibles well with water, and transfer rinses to each tube.

1.1.2. Dilute to approximately 150 mL with water.

1.1.3. Pipet 2 mL 1.25M $Ca(NO_3)_2$ into each tube.

1.1.4. Add 1 mL 50-mg/mL iron carrier into each tube.

1.1.5. Add 5 mL 3.2M $(NH_4)_2HPO_4$ to each tube.

1.1.6. Cap tubes and mix well.

1.1.7. Centrifuge tubes for 5 minutes at 3500 rpm.

1.1.8. Pour off the supernate and discard to waste.

1.1.9. Add 1.5M HCl to each tube to redissolve each sample in a total volume of ~60 mL.

1.1.10. Cap and shake each tube to dissolve solids as well as possible.

1.1.11. Dilute each tube to ~170 mL with 0.01M HCl. Cap and mix.

1.1.12. Add 22 mL of concentrated HF into each tube. Cap and mix well.

1.1.13. Place tubes to set in an ice bath for ~10 minutes to get the tubes very cold.

1.1.14. Centrifuge for ~6 minutes at 3500 rpm.

1.1.15. Pour off supernate and discard to waste.

1.1.16. Pipet 5 mL of concentrated HNO_3 and 5 mL of 3-M HNO_3 – 0.25M boric acid into each 225 mL tube to dissolve precipitate.

1.1.17. Cap and mix well. Transfer contents of the tube into a labeled 50-mL centrifuge tube.

1.1.18. Pipet 5 mL of 3M HNO_3 and 5 mL of 2M aluminum nitrate into each tube, cap tube and mix.

1.1.19. Transfer rinse solutions to 50 mL centrifuge tubes and mix well (shake or use vortex stirrer).

1.1.20. Centrifuge the 50 mL tubes at 3500 rpm for 5 minutes to remove any traces of solids.

1.1.21. Transfer solutions to labeled beakers or new 50 mL tubes for further processing.

1.1.22. If solids remain, add 5 mL 3M HNO_3 to each tube, cap and mix well, centrifuge for 5 minutes and add the supernate to the sample solution. Discard any residual solids.

1.1.23. Set aside for ^{90}Sr analysis using *Rapid Radiochemical Method for Total Radiostrontium (Sr-90) In Building Materials for Environmental Remediation Following Radiological Incidents* (Reference 16.4)

11. Summary and Conclusions

The combined rapid ^{90}Sr - Brick method was successfully validated according to *Method Validation Requirements for Qualifying Methods Used by Radioanalytical Laboratories Participating in Incident Response Activities* and Chapter 6 of *Multi-Agency Radiological Laboratory Analytical Protocols Manual* (EPA 2004) for a typical brick matrix containing ^{241}Am, isotopic uranium, ^{226}Ra, and ^{239}Pu in similar concentrations corresponding to a 10^{-5} risk for a soil exposure pathway. The method was evaluated using well-characterized brick analyzed for its macro-constituents by an independent laboratory[4] and for its radiological constituents (Attachment IV) using the combined rapid ^{90} Sr - brick method by NAREL, The radiochemical results provided by NAREL for the brick blank samples indicated that that there was some detectable ^{90}Sr in the blank material (0.119 ± 0.029 standard error, pCi/g). It is unknown whether the measured ^{90}Sr was from the blank brick material or from the reagents used in the method.

The pulverized brick samples were spiked with three low-level ^{90}Sr concentrations consistent with a concentration range that incorporated the 10^{-5} exposure risk contaminant level in soil (1.210 pCi/g, 2.440 pCi/g, and 7.28 pCi/g) in the presence of low-level concentrations of ^{241}Am, ^{239}Pu, ^{226}Ra, and uranium (Table 1). The combined rapid ^{90}Sr method met MARLAP Validation Level “C” requirements for required method uncertainty (0.32 pCi/g) at and below the AAL, and for a required relative method uncertainty of (13%) above the AAL concentration of 2.440 pCi/g. A sample size of 1.5 grams and a counting time of 100 minutes were used in the method validation process.

Based on the results of the seven blank brick samples (Table 5), the critical net concentration for the combined method was determined to be 0.15 pCi/g for a 1.5 g sample and a 100-minute counting time. The results for the seven blank samples had a mean and standard deviation of −0.119 ± 0.077 pCi/g. A statistical analysis of the data indicated there was an absolute bias (difference from the expected zero concentration) for the blank brick samples.

The mean measured value and standard deviation of the 10 ^{90}Sr in the MDC test samples were calculated as 0.51 ± 0.19 pCi/g (k=1). Each result was compared to the critical net concentration of 0.15 pCi/g. All 10 measurements had a result greater than the critical net concentration, thus verifying that the method can meet a required MDC of 0.4040 pCi/g.

Predicated on the statistical tests provided in the *Method Validation Requirements for Qualifying Methods Used by Radioanalytical Laboratories Participating in Incident Response Activities*, the combined method was found not to have a relative bias for the three test levels. The mean

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[4] Wyoming Analytical Laboratories, Inc. of Golden, Colorado, performed the macro analysis.

relative difference from the known for the low, AAL and high test levels was −4.1%, −11.1%, and −6.28%. The average bias versus reference values at the 3 test levels, as well as the MDC study, indicates this rapid method is a very robust, reliable method to determine ^{90}Sr in brick samples.

Although radionuclide and chemical interferences (^{241}Am, uranium, ^{239}Pu, ^{226}Ra and typical constituents in the blank brick) were in the test samples, the method specificity was adequate under conditions as tested. Additionally, high and reproducible chemical yield results (mean yield = 85.2 ± 6.8%) was observed for the 48 analyses evaluated. The method is rapid and the validation study indicates it can be used with confidence after a radiological incident for the analysis of emergency brick samples.

12. References

Multi-Agency Radiological Laboratory Analytical Protocols Manual (MARLAP). 2004. EPA 402-B-1304 04-001A, July. Volume I, Chapters 6, 7, 20, Glossary; Volume II and Volume III, Appendix G. Available at www.epa.gov/radiation/marlap/.

U.S. Environmental Protection Agency (EPA). 2006. *Validation and Peer Review of U.S. Environmental Protection Agency Radiochemical Methods of Analysis*. FEM Document Number 2006-01, September 29. Available at: www.epa.gov/fem/agency_methods.htm.

U.S. Environmental Protection Agency (EPA) 2008. Radiological Laboratory Sample Analysis Guide for Incidents of National Significance – Radionuclides in Water, Office of Air and Radiation, Washington, DC, EPA 402-R-07-007, January 2008. Available at: http://nepis.epa.gov/Adobe/PDF/60000LAW.PDF.

U.S. Environmental Protection Agency (EPA). 2009. *Method Validation Guide for Qualifying Methods Used by Radiological Laboratories Participating in Incident Response Activities*. Revision 0. Office of Air and Radiation, Washington, DC. EPA 402-R-09-006, June. Available at: www.epa.gov/narel.

U.S. Environmental Protection Agency (EPA). 2011. *Quality Assurance Project Plan Validation of Rapid Radiochemical Methods For Radionuclides Listed in EPA's Standardized Analytical Methods (SAM) For Use During Homeland Security Events*. July, Revision 2. Office of Air and Radiation, National Analytical Radiation Environmental Laboratory.

U.S. Environmental Protection Agency (EPA). 2012. *Radiological Sample Analysis Guide for Incident Response — Radionuclides in Soil*. Revision 0. Office of Air and Radiation, Washington, DC. EPA 402-R-12-006, September 2012.

Attachment I:

Estimated Elapsed Times
Combined Rapid ^{90}Sr - Brick Method

Step	Elapsed Time (hours)*
Rapid Fusion	2.5
Vacuum Box Setup	2.75
Load Sample to Sr Resin	4.25
Elute Sr from resin	5.0
Planchet Mounting	6.5
Count sample test source (1–2 hours)	7.5–8.5

* These estimates depend on the number of samples that can be processed simultaneously.

Attachment II:

Rapid Method for Sodium Hydroxide Fusion of Concrete[5] and Brick Matrices Prior to Americium, Plutonium, Strontium, Radium, and Uranium Analyses for Environmental Remediation Following Radiological Incidents

1. Scope and Application
   1.1. The method is applicable to the sodium hydroxide fusion of concrete and brick samples, prior to the chemical separation procedures described in the following procedures:
      1.1.1. Rapid Radiochemical Method for Americium-241 in Building Materials for Environmental Remediation Following Radiological Incidents (Reference 16.1).
      1.1.2. Rapid Radiochemical Method for Plutonium-238 and Plutonium-239/240 in Building Materials for Environmental Remediation Following Radiological Incidents (Reference 16.2).
      1.1.3. Rapid Radiochemical Method for Radium-226 in Building Materials for Environmental Remediation Following Radiological Incidents (Reference 16.3).
      1.1.4. Rapid Radiochemical Method for Total Radiostrontium (Sr-90) in Building Materials for Environmental Remediation Following Radiological Incidents (Reference 16.4).
      1.1.5. Rapid Radiochemical Method for Isotopic Uranium in Building Materials for Environmental Remediation Following Radiological Incidents (Reference 16.5).
   1.2. This general method for concrete and brick building material applies to samples collected following a radiological or nuclear incident. The concrete and brick samples may be received as core samples, pieces of various sizes, dust or particles (wet or dry) from scabbling, or powder samples.
   1.3. The fusion method is rapid and rigorous, effectively digesting refractory radionuclide particles that may be present.
   1.4. Concrete or brick samples should be ground to at least 50–100 mesh size prior to fusion, if possible.
   1.5. After a homogeneous, finely ground sample is obtained, the dissolution of concrete or brick matrices by this fusion method is expected to take approximately 1 hour per batch of 20 samples. This method assumes the laboratory starts with a representative, finely ground, 1–1.5-g aliquant of sample and employs simultaneous heating in multiple furnaces. The preconcentration steps to eliminate the alkaline fusion matrix and collect the radionuclides are expected to take approximately 1 hour.
   1.6. As this method is a sample digestion and pretreatment technique, to be used prior to other separation and analysis methods, the user should refer to those individual methods

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[5] U.S. Environmental Protection Agency (EPA). 2014. *Rapid Radiochemical Method for Plutonium-238 and Plutonium-239/240 in Building Materials for Environmental Remediation Following Radiological Incidents*, Office of Air and Radiation, Washington, DC, EPA 402-R-07-007, April 2014. Unpublished.

and any project-specific requirements for the determination of applicable measurement quality objectives (MQOs).

1.7. Application of this method by any laboratory should be validated by the laboratory using the protocols provided in *Method Validation Guide for Qualifying Methods Used by Radioanalytical Laboratories Participating in Incident Response Activities* (Reference 16.6), or the protocols published by a recognized standards organization for method validation.

1.7.1. In the absence of project-specific guidance, MQOs for concrete or brick samples may be based on the Analytical Action Levels (AALs), the Required Method Uncertainty (u_{MR}) and the Required Relative Method Uncertainty (φ_{MR}) found in the *Radiological Laboratory Sample Analysis Guide for Incident Response — Radionuclides in Soil* (Reference 16.7).

2. Summary of Method

2.1. The method is based on the rapid fusion of a representative, finely ground 1–1.5-g aliquant using rapid sodium hydroxide fusion at 600 °C.

2.2. Pu, U, and Am are separated from the alkaline matrix using an iron/titanium hydroxide precipitation (enhanced with calcium phosphate precipitation) followed by a lanthanum fluoride matrix removal step.

2.3. Sr is separated from the alkaline matrix using a carbonate precipitation, followed by a calcium fluoride precipitation to remove silicates.

2.4. Ra is separated from the alkaline matrix using a carbonate precipitation.

3. Definitions, Abbreviations and Acronyms

3.1. Discrete Radioactive Particles (DRPs or “hot particles”). Particulate matter in a sample of any matrix where a high concentration of radioactive material is present as a tiny particle (µm range).

3.2. *Multi-Agency Radiological Analytical Laboratory Protocols (MARLAP) Manual* (Reference 16.8).

3.3. The use of the term concrete or brick throughout this method is not intended to be limiting or prescriptive, and the method described herein refers to all concrete or masonry-related materials. In cases where the distinction is important, the specific issues related to a particular sample type will be discussed.

4. Interferences and Limitations

NOTE: Large amounts of extraneous debris (pebbles larger than ¼", non-soil related debris) are not generally considered to be part of a concrete or brick matrix. When consistent with data quality objectives (DQOs), materials should be removed from the sample prior to drying. It is recommended this step be verified with Incident Command before discarding any materials.

4.1. Concrete or brick samples with larger particle size may require a longer fusion time during Step 11.1.8.

4.2. As much information regarding the elemental composition of the sample should be obtained as possible. For example some concrete or brick may have native concentrations of uranium, radium, thorium, strontium or barium, all of which may have an effect on the chemical separations used following the fusion of the sample. In some cases (e.g., radium or strontium analysis), elemental analysis of the digest prior to chemical separations may be necessary to determine native concentrations of carrier elements present in the sample.

NOTE: In those samples where native constituents are present that could interfere with the determination of the chemical yield (e.g., strontium for ^{90}Sr analysis) or with the creation of a sample test source (e.g., Ba for ^{226}Ra analysis by alpha spectrometry), it may be necessary to determine the concentration of these native constituents in advance of chemical separation (using a separate aliquant of fused material) and make appropriate adjustments to the yield calculations or amount of carrier added.

4.3. Matrix blanks for these matrices may not be practical to obtain. Efforts should be made to obtain independent, analyte-free materials that have similar composition as the samples to be analyzed. These blanks will serve as process monitors for the fusion, and as potential monitors for cross contamination during batch processing.

4.4. Uncontaminated concrete or brick material may be acceptable blank material for Pu, Am, and Sr analyses, but these materials will typically contain background levels of U and Ra isotopes.

4.4.1. If analyte-free blank material is not available and an empty crucible is used to generate a reagent blank sample, it is recommended that 100–125 milligram (mg) calcium (Ca) per gram of samples be added as calcium nitrate to the empty crucible as blank simulant. This step facilitates Sr/Ra carbonate precipitations from the alkaline fusion matrix.

4.4.2. Tracer yields may be slightly lower for reagent blank matrices, since the concrete and brick matrix components typically enhance recoveries across the precipitation steps.

4.5. Samples with elevated activity or samples that require multiple analyses from a single concrete or brick sample may need to be split after dissolution. In these cases the initial digestate and the split fractions should be carefully measured to ensure that the sample aliquant for analysis is accurately determined.

4.5.1. Tracer or carrier amounts (added for yield determination) may be increased where the split allows for the normal added amount to be present in the subsequent aliquant. For very high activity samples, the addition of the tracer or carrier may need to be postponed until following the split, in which case special care must be taken to ensure that the process is quantitative until isotopic exchange with the yield monitor is achieved. This deviation from the method should be thoroughly documented and reported in the case narrative.

4.5.2. When this method is employed and the entire volume of fused sample is processed in the subsequent chemical separation method, the original sample size and units are used in all calculations, with the final results reported in the units requested by the project manager.

4.5.3. In cases where the sample digestate is split prior to analysis, the fractional aliquant of the sample is used to determine the sample size. The calculation of the appropriate sample size used for analysis is described in Section 12, below.

4.6. In the preparation of blank samples, laboratory control samples (LCSs) and duplicates, care should be taken to create these quality control samples as early in the process as possible, and to follow the same tracer/carrier additions, digestion process, and sample splitting used for the field samples. In the case of this method, quality control samples should be initiated at the point samples are aliquanted into crucibles for the fusion.

4.7. Although this method is applicable to a variety of subsequent chemical separation procedures, it is not appropriate where the analysis of volatile constituents such as iodine or polonium is required. The user of this method must ensure that analysis is not required for any radionuclide that may be volatile under these sample preparation conditions, prior to performing this procedure.

4.8. Zirconium crucibles used in the fusion process may be reused.

4.8.1. It is very important that the laboratory have a process for cleaning and residual contamination assessment of the reused zirconium crucibles. The crucibles should be cleaned very well using soap and water, followed by warm nitric acid and then water. Blank measurements should be monitored to ensure effective cleaning.

4.8.2. Segregation of crucibles used for low and high activity samples is recommended to minimize the risk of cross-contamination while maximizing the efficient use of crucibles.

4.9. Centrifuge speed of 3500 rpm is prescribed but lower rpm speeds (>2500 rpm) may be used if 3500 rpm is not available.

4.10. Titanium chloride ($TiCl_3$) reductant is used during the co-precipitation step with iron hydroxide for actinides to ensure tracer equilibrium and reduce uranium from U^{+6} to U^{+4} to enhance chemical yields. This method adds 5 mL 10 percent by mass (wt%) $TiCl_3$ along with the Fe. Adding up to 10 mL of 10 wt% $TiCl_3$ may increase uranium chemical yields, but this will need to be validated by the laboratory.

4.11. Trace levels of ^{226}Ra may be present in Na_2CO_3 used in the ^{226}Ra pre-concentration step used in this method. Adding less 2M Na_2CO_3 (<25 mL used in this method) may reduce ^{226}Ra reagent blank levels, while still effectively pre-concentrating ^{226}Ra from the fusion matrix. This will need to be validated by the laboratory.

4.12. La is used to pre-concentrate actinides along with LaF_3 in this method to eliminate matrix interferences, including silica, which can cause column flow problems. La follows Am in subsequent column separations and must be removed. Less La (2 mg) was used for brick samples to minimize the chance of La interference on alpha spectrometry peaks. While this may also be effective for concrete samples, this will have to be validated by the laboratory.

5. Safety

   5.1. General

      5.1.1. Refer to your laboratory safety manual for concerns of contamination control, personal exposure monitoring and radiation dose monitoring.

      5.1.2. Refer to your laboratory's chemical hygiene plan (or equivalent) for general safety rules regarding chemicals in the workplace.

   5.2. Radiological

      5.2.1. Discrete Radioactive Particles (DRPs or "hot particles")

         5.2.1.1. Hot particles will be small, on the order of 1 millimeter (mm) or less. DRPs are typically not evenly distributed in the media and their radiation emissions are not uniform in all directions (anisotropic).

         5.2.1.2. Concrete/brick media should be individually surveyed using a thickness of the solid sample that is appropriate for detection of the radionuclide decay particles.

         NOTE: The information regarding DRPs should accompany the samples during processing as well as be described in the case narrative that accompanies the sample results.

   5.3. Procedure-Specific Non-Radiological Hazards:

      5.3.1. The sodium hydroxide fusion is performed in a furnace at 600 °C. The operator should exercise extreme care when using the furnace and when handling the hot crucibles. Long tongs are recommended. Thermal protection gloves are also recommended when performing this part of the procedure. The fusion furnace should be used in a ventilated area (hood, trunk exhaust, etc.).

      5.3.2. Particular attention should be paid to the use of hydrofluoric acid (HF). HF is an extremely dangerous chemical used in the preparation of some of the reagents and in the microprecipitation procedure. Appropriate personal protective equipment (PPE) must be used in strict accordance with the laboratory safety program specification.

6. Equipment and Supplies

   6.1. Adjustable temperature laboratory hotplates.

   6.2. Balance, top loading or analytical, readout display of at least ± 0.01 g.

   6.3. Beakers, 100 mL, 150 mL capacity.

   6.4. Centrifuge able to accommodate 225 mL tubes.

   6.5. Centrifuge tubes, plastic, 50 mL and 225 mL capacity.

   6.6. Crucibles, 250 mL, zirconium, with lids.

   6.7. 100 µL, 200 µL, 500 µL, and 1 mL pipets or equivalent and appropriate plastic tips.

   6.8. 1-10 mL electronic/manual pipet(s).

   6.9. Drill with masonry bit (¼-inch carbide bit recommended).

6.10. Hot water bath or dry bath equivalent.

6.11. Ice bath.

6.12. Muffle furnace capable of reaching at least 600 °C.

6.13. Tongs for handling crucibles (small and long tongs).

6.14. Tweezers or forceps.

6.15. Sample size reduction equipment (ball mill, paint shaker, etc.) and screens. The necessary equipment will be based on a laboratory's specific method for the process of producing a uniformly ground sample from which to procure an aliquant.

NOTE: See appendix for a method for ball-milling and homogenization of concrete or brick.

6.16. Vortex stirrer.

7. Reagents and Standards

NOTES:

Unless otherwise indicated, all references to water should be understood to mean Type I reagent water (ASTM D1193; Reference 16.9).

All reagents are American Chemical Society (ACS)-grade or equivalent unless otherwise specified.

7.1. Type I reagent water as defined in ASTM Standard D1193 (Reference 16.9).

7.2. Aluminum nitrate ($Al(NO_3)_3 \cdot 9H_2O$)

7.2.1. Aluminum nitrate solution (2M): Add 750 g of aluminum nitrate ($Al(NO_3)_3 \cdot 9H_2O$) to ~700 mL of water and dilute to 1 L with water. Low-levels of uranium are typically present in $Al(NO_3)_3$ solution.

NOTE: Aluminum nitrate reagent typically contains trace levels of uranium concentration. To achieve the lowest possible blanks for isotopic uranium measurements, some labs have removed the trace uranium by passing ~250 mL of the 2M aluminum nitrate reagent through ~7 mL TRU® Resin or UTEVA® Resin (Eichrom Technologies), but this will have to be tested and validated by the laboratory.

7.3. Ammonium hydrogen phosphate (3.2M): Dissolve 106 g of $(NH_4)_2HPO_4$ in 200 mL of water, heat on low to medium heat on a hot plate to dissolve and dilute to 250 mL with water.

7.4. Boric Acid, H_3BO_3.

7.5. Calcium nitrate (1.25M): Dissolve 147 g of calcium nitrate tetrahydrate ($Ca(NO_3)_2 \cdot 4H_2O$) in 300 mL of water and dilute to 500 mL with water.

7.6. Iron carrier (50 mg/mL): Dissolve 181 g of ferric nitrate ($Fe(NO_3)_3 \cdot 9H_2O$) in 300 mL water and dilute to 500 mL with water.

7.7. Hydrochloric acid (12M): Concentrated HCl, available commercially.

7.6.1. Hydrochloric acid (0.01M): Add 0.83 mL of concentrated HCl to 800 mL of water and dilute with water to 1 L.

7.6.2. Hydrochloric acid (1.5M): Add 125 mL of concentrated HCl to 800 mL of water and dilute with water to 1 L.

7.8. Hydrofluoric acid (28M): Concentrated HF, available commercially.

7.9. Lanthanum carrier (1.0 mg La^{3+}/mL): Add 1.56 g lanthanum (III) nitrate hexahydrate [$La(NO_3)_3 \cdot 6H_2O$] in 300 mL water, diluted to 500 mL with water.

7.10. Nitric acid (16M): Concentrated HNO_3, available commercially.

7.10.1. Nitric acid (3M): Add 191 mL of concentrated HNO_3 to 700 mL of water and dilute to 1 L with water.

7.10.2. Nitric acid–boric acid solution (3M-0.25M): Add 15.4 g of boric acid and 190 mL of concentrated HNO_3 to 500 mL of water, heat to dissolve, and dilute to 1 liter with water.

7.10.3. Nitric acid (7M): Add 443 mL of concentrated HNO_3 to 400 mL of water and dilute to 1 L with water.

7.10.4. Nitric acid (8M): Add 506 mL of concentrated HNO_3 to 400 mL of water and dilute to 1 L with water.

7.11. Sodium carbonate (2M): Dissolve 212 g anhydrous Na_2CO_3 in 800 mL of water, then dilute to 1 L with water.

7.12. Sodium hydroxide pellets.

7.13. Titanium (III) chloride solution ($TiCl_3$), 10 wt% solution in 20–30 wt% hydrochloric acid.

7.14. Radioactive tracers/carriers (used as yield monitors) and spiking solutions. A radiotracer is a radioactive isotope of the analyte that is added to the sample to measure any losses of the analyte. A carrier is a stable isotope form of a radionuclide (usually the analyte) added to increase the total amount of that element so that a measureable mass of the element is present. A carrier can be used to determine the yield of the chemical process and/or to carry the analyte or radiotracer through the chemical process. Refer to the chemical separation method(s) to be employed upon completion of this dissolution technique. Tracers/carriers that are used to monitor radiochemical/chemical yield should be added at the beginning of this procedure. This timing allows for monitoring and correction of chemical losses in the combined digestion process, as well as in the chemical separation method. Carriers used to prepare sample test sources but not used for chemical yield determination (e.g., cerium added for microprecipitation of plutonium or uranium), should be added where indicated.

8. Sample Collection, Preservation, and Storage

Not Applicable.

9. Quality Control

9.1. Where the subsequent chemical separation technique requires the addition of carriers and radioactive tracers for chemical yield determinations, these are to be added prior to beginning the fusion procedure, unless there is good technical justification for doing otherwise.

9.2. Batch quality control results shall be evaluated and meet applicable analytical protocol specifications (APS) prior to release of unqualified data. In the absence of project-defined APS or a project-specific quality assurance project plan (QAPP), the quality control sample acceptance criteria defined in the laboratory's Quality Manual and procedures shall be used to determine acceptable performance for this method.

9.2.1. An exception to this approach may need to be taken for samples of exceptionally high activity where human safety may be involved.

9.3. Quality control samples are generally specified in the laboratory's Quality Manual or in a project's APS. At the very minimum the following are suggested:

9.3.1. A laboratory control sample (LCS), which consists solely of the reagents used in this procedure and a known quantity of radionuclide spiking solution, shall be run with each batch of samples. The concentration of the LCS should be at or near the action level or level of interest for the project

9.3.2. One reagent blank shall be run with each batch of samples. The blank should consist solely of the reagents used in this procedure (including tracer or carrier from the analytical method added prior to the fusion process).

9.3.3. A sample duplicate that is equal in size to the original aliquant should be analyzed with each batch of samples. This approach provides assurance that the laboratory's sample size reduction and sub-sampling processes are reproducible.

10. Calibration and Standardization

10.1. Refer to the individual chemical separation and analysis methods for calibration and standardization protocols.

11. Procedure

11.1. Fusion

11.1.1. In accordance with the DQOs and sample processing requirements stated in the project plan documents, remove extraneous materials from the concrete or brick sample using a clean forceps or tweezers.

11.1.2. Weigh out a representative, finely ground 1-g aliquant of sample into a labeled crucible (1.5-g aliquants for ^{90}Sr analysis).

NOTES:

It is anticipated that concrete or brick powder sample material will be dry enough to aliquant without a preliminary drying step. In the event samples are received that contain moisture, the samples may be dried in a drying oven at 105 °C prior to taking the aliquant.

For Sr and Ra analyses, a reagent blank of 100–150 mg calcium per gram of sample (prepared by evaporating 2.5 mL of 1.25M calcium nitrate, $Ca(NO_3)_2$, for radium and 3 mL of 1.25M $Ca(NO_3)_2$ for strontium) should be added to the crucible as a blank simulant to ensure the blank behaves like the concrete or brick samples during the precipitation steps.

11.1.3. Add the proper amount of tracer or carrier appropriate for the method being used and the number of aliquants needed.

11.1.4. Place crucibles on a hot plate and heat to dryness on medium heat.

NOTE: Heat on medium heat to dry quickly but not so high as to cause splattering.

11.1.5. Remove crucibles from hot plate and allow to cool.

11.1.6. Add the following amounts of sodium hydroxide based on the aliquant size/analysis required.

1 g for Pu, Am, U:	15 g NaOH
1.5 g for Sr:	15 g NaOH
1 g for Ra:	10 g NaOH

11.1.7. Place the crucibles with lids in the 600 °C furnace using tongs.

11.1.8. Fuse samples in the crucibles for ~15 minutes.

NOTE: Longer times may be needed for larger particles.

11.1.9. Remove hot crucibles from furnace very carefully using tongs, and transfer to hood.

11.1.10. Add ~25-50 mL of water to each crucible ~8 to 10 minutes (or longer) after removing crucibles from furnace, and heat on hotplate to loosen/dissolve solids.

11.1.11. If necessary for dissolution, add more water, and warm as needed on a hotplate.

11.1.12. Proceed to Section 11.2 for the actinide preconcentration procedure, 11.3 or 11.4 for Sr preconcentration, or 11.5 for Ra preconcentration steps.

11.2. Preconcentration of Actinides (Pu, U, or Am) from Hydroxide Matrix

11.2.1. Pipet 2.5 mL of iron carrier (50 mg/mL) into a labeled 225-mL centrifuge tube for each sample.

11.2.2. Add La carrier to each 225-mL tube as follows:

Concrete: 5 mL of 1 mg La/mL for Pu, Am, U
Brick: 5 mL of 1 mg La/mL for Pu, and U; 2 mL 1 mg La/mL for Am

11.2.3. Transfer each fused sample to a labeled 225 mL centrifuge tube, rinse.

11.2.4. crucibles well with water, and transfer rinses to each tube.

11.2.5. Dilute each sample to approximately 180 mL with water.

11.2.6. Cool the 225 mL centrifuge tubes in an ice bath to approximately room temperature as needed.

11.2.7. Pipet 1.25M $Ca(NO_3)_2$ and 3.2M $(NH_4)_2HPO_4$ into each tube as follows:

Pu, Am:	2 mL 1.25M $Ca(NO_3)_2$ and 3 mL 3.2M $(NH_4)_2HPO_4$
U:	3 mL 1.25M $Ca(NO_3)_2$ and 5 mL 3.2M $(NH_4)_2HPO_4$

11.2.8. Cap tubes and mix well.

11.2.9. Pipet 5 mL of 10 wt% $TiCl_3$ into each tube, and cap and mix immediately.

11.2.10. Cool the 225 mL centrifuge tubes in an ice bath for ~10 minutes.

11.2.11. Centrifuge tubes for 6 minutes at 3500 rpm.

11.2.12. Pour off the supernate, and discard to waste.

11.2.13. Add 1.5M HCl to each tube to redissolve each sample in a total volume of ~60 mL.

11.2.14. Cap and shake each tube to dissolve solids as well as possible.

NOTE: There will typically be undissolved solids, which is acceptable.

11.2.15. Dilute each tube to ~170 mL with 0.01M HCl. Cap and mix.

11.2.16. Pipet 1 mL of 1.0 mg La/mL into each tube.

11.2.17. Pipet 3 mL of 10 wt% $TiCl_3$ into each tube. Cap and mix.

11.2.18. Add 22 mL of concentrated HF into each tube. Cap and mix well.

11.2.19. Place tubes to set in an ice bath for ~10 minutes to get the tubes very cold.

11.2.20. Centrifuge for ~10 minutes at 3000 rpm or more or as needed.

11.2.21. Pour off supernate, and discard to waste.

11.2.22. Pipet 5 mL of 3M HNO_3 - 0.25M boric acid into each tube.

11.2.23. Cap, mix and transfer contents of the tube into a labeled 50 mL centrifuge tube.

11.2.24. Pipet 6 mL of 7M HNO_3 and 7 mL of 2M aluminum nitrate into each tube, cap and mix (shake or use a vortex stirrer), and transfer rinse to 50-mL centrifuge tube.

11.2.25. Pipet 3 ml of 3M HNO_3 directly into the 50 mL centrifuge tube.

11.2.26. Warm each 50 mL centrifuge tube in a hot water bath for a few minutes, swirling to dissolve.

11.2.27. Remove each 50 mL centrifuge tube from the water bath and allow to cool to room temperature

11.2.28. Centrifuge the 50 ml tubes at 3500 rpm for 5 minutes to remove any traces of solids (may not be visible prior to centrifuging), and transfer solutions to labeled beakers or tubes for further processing. Discard any solids.

11.2.29. Proceed directly to any of those methods listed in Sections 1.1.1, 1.1.2, or 1.1.5 (for Pu, U, or Am).

11.3. Preconcentration of ^{90}Sr from Hydroxide Matrix (Concrete)

NOTE: The preconcentration steps for ^{90}Sr in this section can also be applied to brick samples, but this will have to be validated by the laboratory. See Section 11.4 for steps validated for ^{90}Sr in brick samples.

11.3.1. Transfer each fused sample to a 225-mL centrifuge tube, rinse crucibles well with water, and transfer rinses to each tube.

11.3.2. Dilute to approximately 150 mL with water.

11.3.3. Add 15-mL concentrated HCl to each tube.

11.3.4. Cap and mix solution in each tube.

11.3.5. Pipet 1-mL of 1.25M $Ca(NO_3)_2$ into each tube.

11.3.6. Add 2-mL of 50-mg/mL iron carrier into each tube.

11.3.7. Add 25-mL of 2M Na_2CO_3 to each tube.

11.3.8. Cap tubes and mix well.

11.3.9. Cool the 225-mL centrifuge tubes in an ice bath for ~10 minutes.

11.3.10. Centrifuge tubes for 5 minutes at 3500 rpm.

11.3.11. Pour off the supernate, and discard to waste.

11.3.12. Add 1.5M HCl to each tube to redissolve each sample in a total volume of ~50 mL.

11.3.13. Cap and shake each tube to dissolve solids as well as possible.

11.3.14. Dilute each tube to ~170 mL with 0.01M HCl. Cap and mix.

11.3.15. Add 22 mL of concentrated HF into each tube. Cap and mix well.

11.3.16. Place tubes to set in an ice bath for ~10 minutes to get the tubes very cold.

11.3.17. Centrifuge for ~6 minutes at 3500 rpm.

11.3.18. Pour off supernate, and discard to waste.

11.3.19. Pipet 5 mL of concentrated HNO_3 and 5 mL of 3M HNO_3 - 0.25M boric acid into each 225 mL tube to dissolve precipitate.

11.3.20. Cap and mix well. Transfer contents of the tube into a labeled 50-mL centrifuge tube.

11.3.21. Pipet 5 mL of 3M HNO_3 and 5 mL of 2M aluminum nitrate into each tube, cap tube and mix.

11.3.22. Transfer rinse solutions to labeled 50-mL centrifuge tubes and mix well (shake or use vortex stirrer).

11.3.23. Centrifuge the 50 mL tubes at 3500 rpm for 5 minutes to remove any traces of solids.

11.3.24. Transfer solutions to labeled beakers or new 50 mL tubes for further processing.

11.3.25. If solids remain in the original 50 mL tubes (step 11.3.23), add 5 mL of 3M HNO3 to each tube containing solids, cap, and mix well, Centrifuge for 5 minutes and add the supernate to the sample solution from step 11.3.24. Discard any remaining solids.

11.3.26. Set aside for ^{90}Sr analysis using *Rapid Radiochemical Method for Total Radiostrontium (Sr-90) In Building Materials for Environmental Remediation Following Radiological Incidents* (Reference 16.4).

11.4. Preconcentration of ^{90}Sr from Hydroxide Matrix (Brick)

NOTE: The preconcentration steps for ^{90}Sr in this section, using calcium phosphate instead of calcium carbonate, can also be applied to concrete samples but this will have to be validated by the laboratory. See Section 11.3 for steps validated for ^{90}Sr in concrete samples.

11.4.1. Transfer each fused sample to a labeled 225-mL centrifuge tube, rinse crucibles well with water, and transfer rinses to each tube.

11.4.2. Dilute to approximately 150 mL with water.

11.4.3. Pipet 2 mL 1.25M $Ca(NO_3)_2$ into each tube.

11.4.4. Add 1 mL 50-mg/mL iron carrier into each tube.

11.4.5. Add 5 mL 3.2M $(NH_4)_2HPO_4$ to each tube.

11.4.6. Cap tubes and mix well.

11.4.7. Centrifuge tubes for 5 minutes at 3500 rpm.

11.4.8. Pour off the supernate and discard to waste.

11.4.9. Add 1.5M HCl to each tube to redissolve each sample in a total volume of ~60 mL.

11.4.10. Cap and shake each tube to dissolve solids as well as possible.

11.4.11. Dilute each tube to ~170 mL with 0.01M HCl. Cap and mix.

11.4.12. Add 22 mL of concentrated HF into each tube. Cap and mix well.

11.4.13. Place tubes to set in an ice bath for ~10 minutes to get the tubes very cold.

11.4.14. Centrifuge for ~6 minutes at 3500 rpm.

11.4.15. Pour off supernate and discard to waste.

11.4.16. Pipet 5 mL of concentrated HNO_3 and 5 mL of 3M HNO_3 – 0.25M boric acid into each 225 mL tube to dissolve precipitate.

11.4.17. Cap and mix well. Transfer contents of the tube into a labeled 50-mL centrifuge tube.

11.4.18. Pipet 5 mL of 3M HNO_3 and 5 mL of 2M aluminum nitrate into each tube, cap tube and mix.

11.4.19. Transfer rinse solutions to labeled 50 mL centrifuge tubes and mix well (shake or use vortex stirrer).

11.4.20. Centrifuge the 50 mL tubes at 3500 rpm for 5 minutes to remove any traces of solids.

11.4.21. Transfer solutions to labeled beakers or new 50 mL tubes for further processing.

11.4.22. If solids remain in the original 50 mL tubes (step 11.4.20), add 5 mL of 3M HNO3 to each tube containing solids, cap, and mix well, Centrifuge for 5 minutes and add the supernate to the sample solution from step 11.4.21. Discard any remaining solids.

11.4.23. Set aside for ^{90}Sr analysis using *Rapid Radiochemical Method for Total Radiostrontium (Sr-90) In Building Materials for Environmental Remediation Following Radiological Incidents* (Reference 16.4).

11.5. Preconcentration of ^{226}Ra from Hydroxide Matrix

11.5.1. Transfer each sample to a labeled 225 mL centrifuge tube, rinse crucibles well with water, and transfer rinses to each tube.

11.5.2. Dilute to approximately 150 mL with water.

11.5.3. Add 10 mL of concentrated HCl to each tube.

11.5.4. Cap and mix each tube well.

11.5.5. Pipet 0.5 mL of 1.25M $Ca(NO_3)_2$ into each tube.

11.5.6. Add 25 mL of 2M Na_2CO_3 to each tube.

11.5.7. Cap tubes and mix.

11.5.8. Cool the 225-mL centrifuge tubes in an ice bath for ~5–10 minutes.

11.5.9. Centrifuge tubes for 6 minutes at 3500 rpm.

11.5.10. Pour off the supernate, and discard to waste.

11.5.11. Pipet 10 mL 1.5M HCl into each tube to dissolve precipitate. Cap and mix.

11.5.12. Transfer sample solution to a labeled 50-mL centrifuge tube.

11.5.13. Pipet 10 mL 1.5M HCl into each 225-mL tube to rinse. Cap and rinse well.

11.5.14. Transfer rinse solution to 50 mL-tube and mix well.

NOTE: Typically the HCl added to dissolve the carbonate precipitate is sufficient to acidify the sample. If the precipitate was unusually large and milky suspended solids remain, indicating additional acid is needed, the pH can be checked to verify it is pH 1 or less. To acidify the pH <1, 1 or 2 mL of concentrated hydrochloric acid may be added to acidify the solution further and get it to clear. Undissolved solids may be more likely to occur with brick samples. Tubes may be warmed in a water bath to help dissolve samples.

11.5.15. If solids remain in the original 225 mL tubes, add 5 mL of 1.5M HCl to each tube containing solids, cap, and mix well. Centrifuge for 5 minutes and add the supernate to the sample solution from step 11.5.14. Discard any remaining solids.

11.5.16. Set aside for ^{226}Ra analysis using *Rapid Radiochemical Method for Radium-226 in Building Materials for Environmental Remediation Following Radiological Incidents* (Reference 16.3).

12. Data Analysis and Calculations

12.1. Equations for determination of final result, combined standard uncertainty, and radiochemical yield (if required) are found in the corresponding chemical separation and analysis methods, with the project manager providing the units.

12.2. In cases where samples have elevated activity, smaller initial sample aliquants may be taken from the original sample. Alternately, smaller aliquant volumes may be taken from the final sample volume containing the dissolved precipitate (digestate). Aliquants should be removed carefully and accurately from this final sample volume.

NOTE: Small aliquants taken from the final sample digestate for Sr and Ra analysis may be used in the respective analytical procedures as is. Smaller aliquants for actinide analysis should be diluted to a 15 mL total volume with 3M HNO_3 so that load solution acidity is maintained when valence adjustment reagents are added.

For a single split, the effective size of sample is calculated:

$$W_a = W_s \frac{D_a}{D_s} \qquad (1)$$

Where:

W_s = original sample size, in the units designated by the project manager (e.g., 1 g, etc.)

D_s = mass or volume of the entire final digestate, (e.g., 20 mL, etc.).

D_a = mass or volume of the aliquant of digestate used for the individual analyses, (e.g., 5.0 mL, etc.). Note that the values for D_a must be in the same units used in D_s.

W_a = sample aliquant size, used for analysis, in the units designated by the project manager (e.g., kg, g, etc.).

NOTE: For higher activity samples, additional dilution may be needed. In such cases, Equation 1 should be modified to reflect the number of splits and dilutions performed. It is also important to measure the masses or volumes, used for aliquanting or dilution, to enough significant figures so that their uncertainties have an insignificant impact on the final uncertainty budget. In cases where the sample will not be split prior to analysis, the sample aliquant size is simply equal to the original sample size, in the same units requested by the project manager.

13. Method Performance

13.1. Report method validation results.

13.2. The method performance data for the analysis of concrete and brick by this dissolution method may be found in the attached appendices.

13.3. Expected turnaround time per sample

13.3.1. For a representative, finely ground 1-g aliquant of sample, the fusion should add approximately 2 hours per batch to the time specified in the individual chemical separation methods.

13.3.2. The preconcentration steps should add approximately 2 to 2.5 hours per batch.

NOTE**: Processing times for the subsequent chemical separation methods are given in those methods for batch preparations.**

14. Pollution Prevention

This method inherently produces no significant pollutants. The sample and fusion reagents are retained in the final product and are carried into the ensuing chemical separation techniques, which marginally increases the salt content of the effluent waste. It is noted that if the sampled particulates include radionuclides that may be volatile under the fusion conditions, these constituents will be exhausted through the fume hood system.

15. Waste Management

15.1. Refer to the appropriate chemical separation methods for waste disposal information.

16. References

Cited References

16.1. U.S. Environmental Protection Agency (EPA). 2013. Rapid Radiochemical Method for Americium-241 in Building Materials for Environmental Remediation Following Radiological Incidents. Revision 0, EPA 402-R14-007. Office of Air and Radiation, Washington, DC. Available at: www.epa.gov/narel.

16.2. U.S. Environmental Protection Agency (EPA). 2013. Rapid Radiochemical Method for Pu-238 and Pu-239/240 in Building Materials for Environmental Remediation Following Radiological Incidents. Revision 0, EPA 402-R14-006. Office of Air and Radiation, Washington, DC. Available at: www.epa.gov/narel.

16.3. U.S. Environmental Protection Agency (EPA). 2013. Rapid Radiochemical Method for Radium-226 in Building Materials for Environmental Remediation Following Radiological Incidents. Revision 0, EPA 402-R14-002. Office of Air and Radiation, Washington, DC. Available at: www.epa.gov/narel.

16.4. U.S. Environmental Protection Agency (EPA). 2013. Rapid Radiochemical Method for Total Radiostrontium (Sr-90) in Building Materials for Environmental Remediation Following Radiological Incidents. Revision 0, EPA 402-R14-001. Office of Air and Radiation, Washington, DC. Available at: www.epa.gov/narel.

16.5. U.S. Environmental Protection Agency (EPA). 2013. Rapid Radiochemical Method for Isotopic Uranium in Building Materials for Environmental Remediation Following Radiological Incidents. Revision 0, EPA 402-R14-005. Office of Air and Radiation, Washington, DC. Available at: www.epa.gov/narel.

16.6. U.S. Environmental Protection Agency (EPA). 2009. *Method Validation Guide for Qualifying Methods Used by Radiological Laboratories Participating in Incident Response Activities*. Revision 0. Office of Air and Radiation, Washington, DC. EPA 402-R-09-006, June. Available at: www.epa.gov/narel.

16.7. *U.S. Environmental Protection Ag*ency (EPA). 2012. *Radiological Laboratory Sample Analysis Guide for Incident Response — Radionuclides in Soil*. Revision 0. Office of Air and Radiation, Washington, DC. EPA 402-R-12-006, September 2012. Available at: www.epa.gov/narel.

16.8. MARLAP. *Multi-Agency Radiological Laboratory Analytical Protocols Manual*. 2004. Volumes 1 – 3. Washington, DC: EPA 402-B-04-001A-C, NUREG 1576, NTIS PB2004-105421, July. Available at: www.epa.gov/radiation/marlap.

16.9. ASTM D1193, "Standard Specification for Reagent Water" ASTM Book of Standards 11.01, current version, ASTM International, West Conshohocken, PA.

Other References

16.10. Maxwell, S., Culligan, B. and Noyes, G. 2010. Rapid method for actinides in emergency soil samples, *Radiochimica Acta*. 98(12): 793-800.

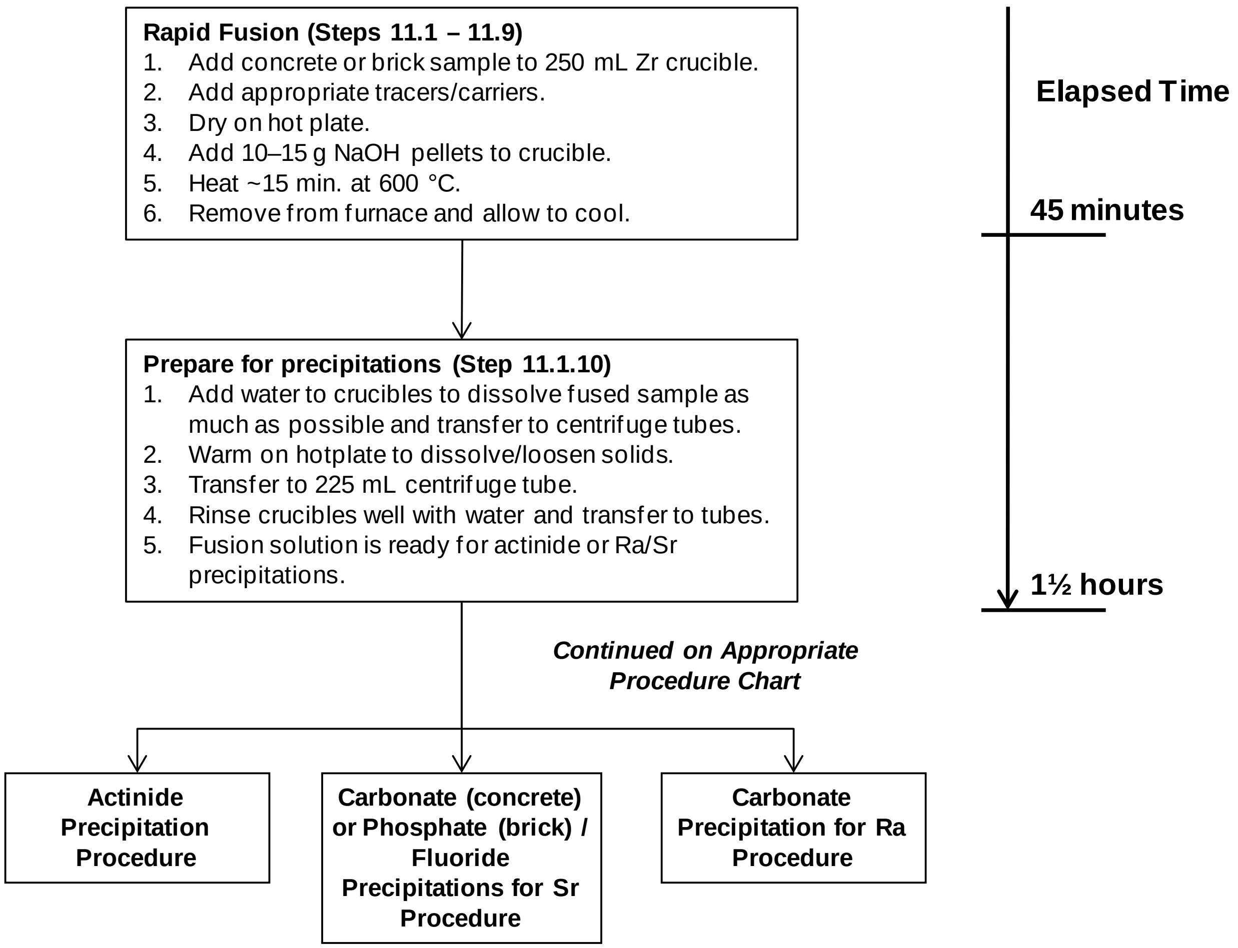
16.11. Maxwell, S., Culligan, B., Kelsey-Wall, A. and Shaw, P. 2011. "Rapid Radiochemical Method for Actinides in Emergency Concrete and Brick Samples," *Analytica Chimica Acta*. 701(1): 112-8.

16.12. U.S. Environmental Protection Agency (EPA). 2010. *Rapid Radiochemical Methods for Selected Radionuclides in Water for Environmental Restoration Following Homeland Security Events*, Office of Air and Radiation. EPA 402-R-10-001, February. Revision 0.1 of rapid methods issued October 2011. Available at: www.epa.gov/narel/.

17. Tables, Diagrams, and Flow Charts

17.1. Fusion Flow Chart

Timeline for Rapid Fusion and Preparation of Building Materials Samples for Precipitation and Analysis



Rapid Fusion (Steps 11.1 – 11.9)

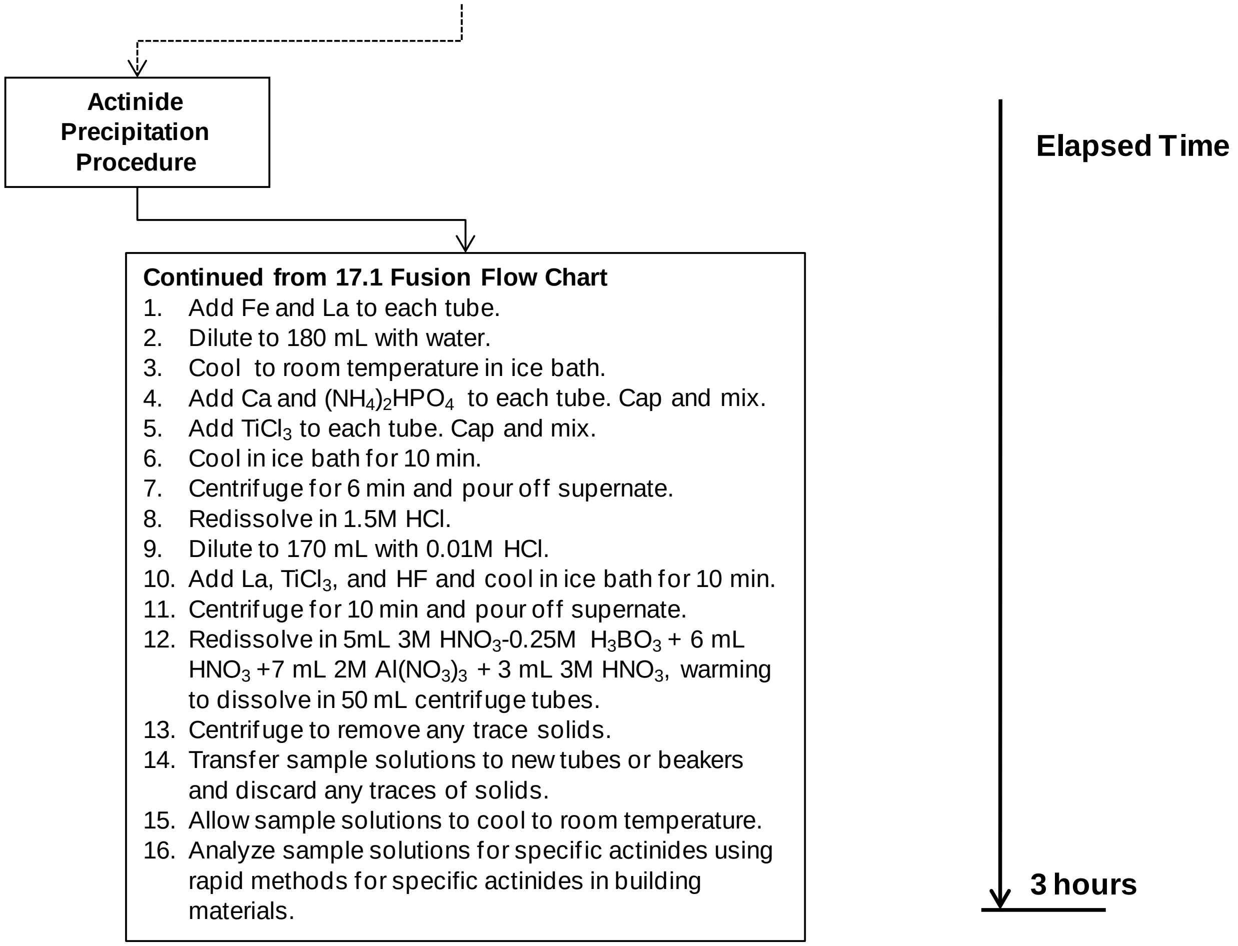
1. Add concrete or brick sample to 250 mL Zr crucible.
2. Add appropriate tracers/carriers.
3. Dry on hot plate.
4. Add 10–15 g NaOH pellets to crucible.
5. Heat ~15 min. at 600 °C.
6. Remove from furnace and allow to cool.

Prepare for precipitations (Step 11.1.10)

1. Add water to crucibles to dissolve fused sample as much as possible and transfer to centrifuge tubes.
2. Warm on hotplate to dissolve/loosen solids.
3. Transfer to 225 mL centrifuge tube.
4. Rinse crucibles well with water and transfer to tubes.
5. Fusion solution is ready for actinide or Ra/Sr precipitations.

17.2. Actinide Precipitation Flow Chart

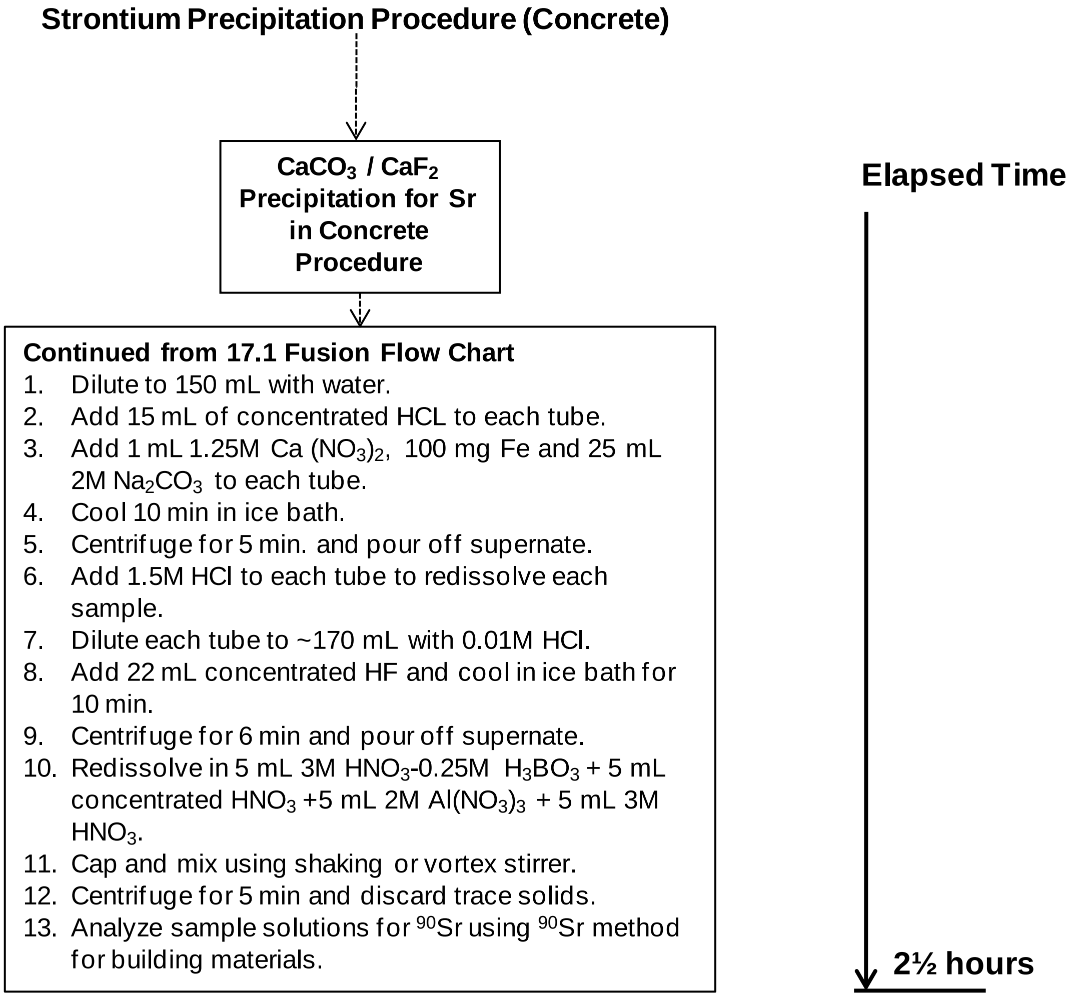
Actinide Precipitation Procedure



Continued from 17.1 Fusion Flow Chart

1. Add Fe and La to each tube.
2. Dilute to 180 mL with water.
3. Cool to room temperature in ice bath.
4. Add Ca and $(NH_4)_2HPO_4$ to each tube. Cap and mix.
5. Add $TiCl_3$ to each tube. Cap and mix.
6. Cool in ice bath for 10 min.
7. Centrifuge for 6 min and pour off supernate.
8. Redissolve in 1.5M HCl.
9. Dilute to 170 mL with 0.01M HCl.
10. Add La, $TiCl_3$, and HF and cool in ice bath for 10 min.
11. Centrifuge for 10 min and pour off supernate.
12. Redissolve in 5mL 3M HNO_3-0.25M H_3BO_3 + 6 mL HNO_3 +7 mL 2M $Al(NO_3)_3$ + 3 mL 3M HNO_3, warming to dissolve in 50 mL centrifuge tubes.
13. Centrifuge to remove any trace solids.
14. Transfer sample solutions to new tubes or beakers and discard any traces of solids.
15. Allow sample solutions to cool to room temperature.
16. Analyze sample solutions for specific actinides using rapid methods for specific actinides in building materials.

17.3. Strontium Precipitation Flow Chart



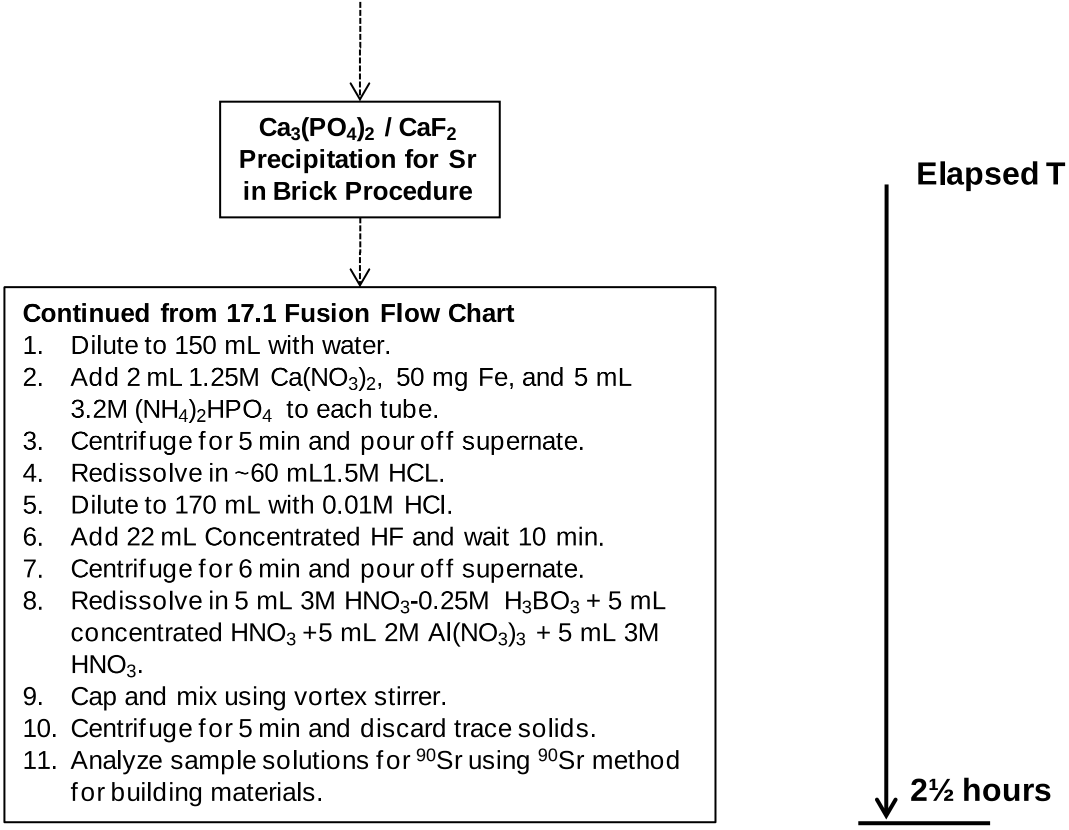
Strontium Precipitation Procedure (Concrete)

$CaCO_3$ / CaF_2 Precipitation for Sr in Concrete Procedure

Continued from 17.1 Fusion Flow Chart

1. Dilute to 150 mL with water.
2. Add 15 mL of concentrated HCL to each tube.
3. Add 1 mL 1.25M Ca $(NO_3)_2$, 100 mg Fe and 25 mL 2M Na_2CO_3 to each tube.
4. Cool 10 min in ice bath.
5. Centrifuge for 5 min. and pour off supernate.
6. Add 1.5M HCl to each tube to redissolve each sample.
7. Dilute each tube to ~170 mL with 0.01M HCl.
8. Add 22 mL concentrated HF and cool in ice bath for 10 min.
9. Centrifuge for 6 min and pour off supernate.
10. Redissolve in 5 mL 3M HNO_3-0.25M H_3BO_3 + 5 mL concentrated HNO_3 +5 mL 2M $Al(NO_3)_3$ + 5 mL 3M HNO_3.
11. Cap and mix using shaking or vortex stirrer.
12. Centrifuge for 5 min and discard trace solids.
13. Analyze sample solutions for ^{90}Sr using ^{90}Sr method for building materials.

Strontium Precipitation Procedure (Brick)

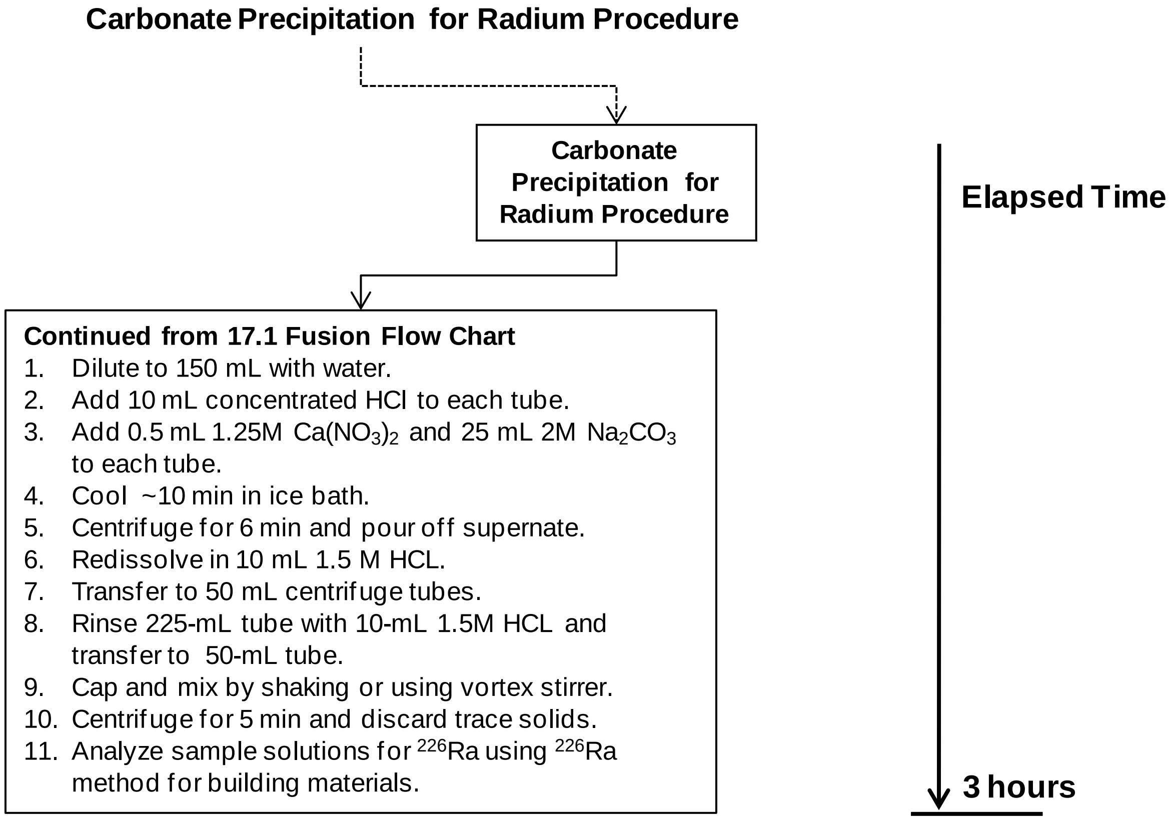


$Ca_3(PO_4)_2$ / CaF_2 Precipitation for Sr in Brick Procedure

Continued from 17.1 Fusion Flow Chart

1. Dilute to 150 mL with water.
2. Add 2 mL 1.25M $Ca(NO_3)_2$, 50 mg Fe, and 5 mL 3.2M $(NH_4)_2HPO_4$ to each tube.
3. Centrifuge for 5 min and pour off supernate.
4. Redissolve in ~60 mL1.5M HCL.
5. Dilute to 170 mL with 0.01M HCl.
6. Add 22 mL Concentrated HF and wait 10 min.
7. Centrifuge for 6 min and pour off supernate.
8. Redissolve in 5 mL 3M HNO_3-0.25M H_3BO_3 + 5 mL concentrated HNO_3 +5 mL 2M $Al(NO_3)_3$ + 5 mL 3M HNO_3.
9. Cap and mix using vortex stirrer.
10. Centrifuge for 5 min and discard trace solids.
11. Analyze sample solutions for ^{90}Sr using ^{90}Sr method for building materials.

17.4. Radium Precipitation Flow Chart



Continued from 17.1 Fusion Flow Chart

1. Dilute to 150 mL with water.
2. Add 10 mL concentrated HCl to each tube.
3. Add 0.5 mL 1.25M $Ca(NO_3)_2$ and 25 mL 2M Na_2CO_3 to each tube.
4. Cool ~10 min in ice bath.
5. Centrifuge for 6 min and pour off supernate.
6. Redissolve in 10 mL 1.5 M HCL.
7. Transfer to 50 mL centrifuge tubes.
8. Rinse 225-mL tube with 10-mL 1.5M HCL and transfer to 50-mL tube.
9. Cap and mix by shaking or using vortex stirrer.
10. Centrifuge for 5 min and discard trace solids.
11. Analyze sample solutions for ^{226}Ra using ^{226}Ra method for building materials.

Appendix:

Rapid Technique for Milling and Homogenizing Concrete and Brick Samples

A1. Scope and Application

A1.1. Concrete or brick samples may be received as powder, core samples or other size pieces or chunks. The goal is to obtain representative sample aliquants from homogeneous amounts of sample.

A1.2. The ball mill method describes one approach for the rapid, gross preparation of concrete or brick samples to yield representative 1–2-g aliquant for radiochemical analysis of non-volatile radionuclides. The method addresses steps for splitting, drying, and milling of 50–2,000 g concrete or brick samples. The concrete or brick sample must be reduced to pieces or fragments less than ~25 mm in diameter prior to using the ball mill. This can be done with a hydraulic press or mallet.

A1.3. The method is designed to be used as a preparatory step for the attached methods for fusion of concrete or brick for ^{241}Am, $^{239/240}$Pu, U, ^{90}Sr, and ^{226}Ra. It may also be applied to other matrices whose physical form is amenable to pulverization in the ball mill.

A1.4. If the levels of activity in the sample are low enough to permit safe radiological operations, up to 2 kg of concrete or brick can be processed.

A1.5. For smaller amounts of concrete or brick samples, a drill with masonry bit can be used in a lab hood inside a plastic bag to collect the powder that results.

A2. Summary of Methods

A2.1. This method uses only disposable equipment to contact the sample, minimizing the risk of contamination and cross-contamination and eliminating concerns about adequate cleaning of equipment.

A2.2. Extraneous material, such as rocks or debris, may be removed prior to processing the sample unless the project requires that they be processed as part of the sample.

NOTE: The sample mass is generally used for measuring the size of solid samples. The initial process of acquiring a representative aliquant uses the volume of the sample, as the total sample size is generally based on a certain volume of concrete or brick (e.g., 500 mL).

A2.3. The entire sample as received (after reducing fragment size to less than ~25 mm diameter) is split by coning and quartering until 75-150 mL of concrete or brick are available for subsequent processing. If less than 450 mL of concrete or brick is received, the entire sample is processed.

A2.4. The concrete or brick is transferred to a paint can or equivalent. Percent solids are determined, if required, by drying in a drying oven. A mallet and plastic bag or hydraulic press may be needed to break up larger pieces.

A2.5. Grinding media (stainless steel or ceramic balls or rods) are added, and the sample is milled to produce a finely-ground, well-homogenized, powder with predominant particle size less than 250 micrometers (µm).

NOTE: A mortar and pestle may also be used as needed to grind the sample further.

A2.6. If the sample may contain discreet radioactive particles (DRPs), particles larger than a nominal size of 150 µm are screened for radioactivity, and further milled, or processed with another appropriate method to ensure that they will be chemically available for subsequent processing.

A2.7. The resulting milled sample is stored in, and aliquanted directly from, the container used for pulverization.

A2.8. The drill bit method involves drilling into the sample using a drill bit. The operation is performed inside a disposable plastic bag in a hood so that the drilled out sample is caught within the plastic bag (this approach also minimizes the spread of contamination). A drill bit such as a ¼-inch carbide bit is recommended. The holes should be drilled in such a way as to obtain representative powdered samples. The drill bit should be cleaned between uses on different samples using soap and water.

A3. Definitions, Abbreviations, and Acronyms

A3.1. Discrete Radioactive Particles (DRPs or “hot particles”). Particulate matter in a sample of any matrix where a high concentration of radioactive material is contained in a tiny particle (µm range).

A3.2. *Multi-Agency Radiological Analytical Laboratory Protocols (MARLAP) Manual* (Reference A16.3).

A3.3. *ASTM C999 Standard Practice for Soil Sample Preparation for the Determination of Radionuclides* (Reference A16.4).

A4. Interferences

A4.1. Radiological Interferences

A4.1.1. Coning and quartering provides a mechanism for rapidly decreasing the overall size of the sample that must be processed while optimizing the representativeness of the subsampling process. By decreasing the time and effort needed to prepare the sample for subsequent processing, sample throughput can be significantly improved. Openly handling large amounts of highly contaminated materials, however, even within the containment provided by a fume hood, may pose an unacceptable risk of inhalation of airborne contamination and exposure to laboratory personnel from radioactive or other hazardous materials. Similarly, it may unacceptably increase the risk of contamination of the laboratory.

A4.1.2. In such cases, the coning and quartering process may be eliminated in lieu of processing the entire sample. The time needed to dry the sample will increase significantly, and the container size and the number and size of grinding media used will need to be adjusted to optimize the milling process. See *ASTM C999* for an approach for homogenization and milling of larger soil samples.

A4.1.3. The precise particle size of the milled sample is not critical to subsequent processes. However, milling the sample to smaller particle sizes, and thorough mixing, both facilitate representative sub-sampling by minimizing the amount of sample that is not pulverized to fine mesh and must be discarded. Additionally, subsequent fusion and digestion processes are more effective when performed on more finely milled samples.

A4.1.4. This method assumes that radioactivity in the sample is primarily adsorbed onto the surface of particles, as opposed to being present as a hot particle (see discussion of DRPs below). Thus, nearly all of the activity in a sample will be associated with sample fines. By visually comparing the sample to a qualitative standard of 50–100 mesh size particles, it is possible to rapidly determine whether the sample is fine enough to facilitate the subsequent fusion or digestion. This method assumes that when greater than 95% of the sample is as fine or finer than the 50–100 mesh sample, bias imparted from losses of larger particles will be minimal.

A4.1.5. If the sample was collected near the epicenter of a radiological dispersal device (RDD) or improvised nuclear device (IND) explosion, it may contain millimeter- to micrometer-sized particles of contaminant referred to as "discrete radioactive particles" or DRPs. DRPs may consist of small pieces of the original radioactive source and thus may have very high specific activity. They may also consist of chemically intractable material and present special challenges in the analytical process. Even when the size is reduced to less than 50-100 mesh, these particles may resist fusion or digestion of the solids into ionic form that can be subjected to chemical separations.

A4.1.6. When DRPs may be present, this method isolates larger particles by passing the sample through a disposable 50-mesh screen after which they can be reliably checked for radioactivity. DRPs may reliably be identified by their very high specific activity, which is readily detectable, since they show high count rates using hand-held survey equipment such as a thin-window Geiger-Muller (G-M) probe.

A4.1.7. When present, DRPs may be further milled and then recombined with the original sample. Alternatively, the particles, or the entire sample may need to be processed using a different method capable of completely solubilizing the contaminants such that the radionuclides they contain are available for subsequent chemical separation.

A5. Safety

A5.1. General

A5.1.1. Refer to your safety manual for concerns of contamination control, personal exposure monitoring, and radiation dose monitoring.

A5.1.2. Refer to your laboratory's chemical hygiene plan (or equivalent) for general safety rules regarding chemicals in the workplace.

A5.2. Radiological

A5.2.1. Refer to your radiation safety manual for direction on working with known or suspected radioactive materials.

A5.2.2. This method has the potential to generate airborne radioactive contamination. The process should be carefully evaluated to ensure that airborne contamination is maintained at acceptable levels. This should take into account the activity level, and physical and chemical form of contaminants possibly present, as well as other engineering and administrative controls available.

A5.2.3. Hot Particles (DRPs)

A5.2.3.1. Hot particles will usually be small, on the order of 1 mm or less. Typically, DRPs are not evenly distributed in the media, and their radiation emissions are not uniform in all directions (anisotropic). Filtration using a 0.45 μm or smaller filter may be needed following subsequent fusion to identify the presence of smaller DRPs.

A5.2.3.2. Care should be taken to provide suitable containment for filter media used in the pretreatment of samples that may have DRPs, because the particles become highly statically charged as they dry out and will "jump" to other surfaces potentially creating contamination-control issues.

A5.3. Method-Specific Non-Radiological Hazards

A5.3.1. This method employs a mechanical shaker and should be evaluated for personnel hazards associated with the high kinetic energy associated with the milling process.

A5.3.2. This method employs a mechanical shaker and involves vigorous agitation of steel or ceramic balls inside steel cans. The process should be evaluated to determine whether hearing protection is needed to protect the hearing of personnel present in the area in which the apparatus is operated.

A6. Equipment and supplies

A6.1. Balance, top-loading, range to accommodate sample size encountered, readability to ±1%.

A6.2. Drying oven, at 110 ± 10 ºC.

A6.3. Steel paint cans and lids (pint, quart, 2-quart, 1-gallon, as needed).

A6.4. Steel or ceramic grinding balls or rods for ball milling, ~15–25 mm diameter. The size and number of grinding media used should be optimized to suit the types of concrete or brick, the size of the can, and the volume of sample processed.

A6.5. Disposable wire cloth – nominal 48 mesh size (~300 μm).

A6.6. Disposable sieves, U.S. Series No. 50 (300 µm or 48 mesh) and U.S. Series No. 100 (150 µm or 100 mesh).

A6.7. Red Devil 5400 mechanical paint shaker or equivalent.

A6.8. Disposable scoop, scraper, tongue depressor or equivalent.

A7. Reagents and Standards

No reagents needed.

A8. Sample Collection, Preservation and Storage

A8.1. Samples should be collected in appropriately sized plastic, metal or glass containers.

A8.2. No sample preservation is required. If samples are to be held for an extended period of time, refrigeration may help minimize bacterial growth in the sample.

A8.3. Default sample collection protocols generally provide solid sample volumes equivalent to approximately 500 mL of sample. Such samples will require two splits to obtain a ~100 mL sample.

A9. Quality Control

A9.1. Batch quality control results shall be evaluated and meet applicable Analytical Protocol Specifications (APS) prior to release of unqualified data. In the absence of project-defined APS or a project-specific quality assurance project plan (QAPP), the quality control sample acceptance criteria defined in the laboratory quality manual and procedures shall be used to determine acceptable performance for this method.

A9.2. Quality control samples should be initiated as early in the process as possible. Since the risk of cross-contamination using this process is relatively low, initiating blanks and laboratory control samples at the start of the chemical separation process is acceptable. If sufficient sample is available, a duplicate sample should be prepared from the two discarded quarters of the final split of the coning and quartering procedure.

A10. Procedure

NOTE: This method ensures that only disposable equipment comes in contact with sample materials to greatly minimize the risk of sample cross-contamination and concerns about adequate cleaning of equipment. Under certain circumstances (disposable sieves are not available, for example), careful, thorough cleaning of the sieves with water and the ethanol may be an option.

A10.1. If necessary, reduce the concrete or brick particle diameter to less than ~25 mm using a hydraulic press, mallet, or alternate equipment capable or reducing the fragment size.

A10.2. Estimate the total volume of sample, as received.

NOTE: If the sample is dry, the risk of resuspension and inhalation of the solids may be determined to be unacceptable. In such cases, the entire sample may be processed in a larger can. The drying and milling time will be increased, and more grinding media will be required to obtain a satisfactory result.

NOTE: The next step uses absorbent paper in the reverse fashion for the normal use of this type of paper; it allows for a smooth division of the sample and control of contamination.

A10.2.1. Spread a large piece of plastic backed absorbent paper, plastic side *up* in a hood.

A10.2.2. If the sample volume is less than 450 mL, there is no benefit to coning and quartering.[6]

A10.2.2.1. Carefully pour the sample onto the paper.

A10.2.2.2. Remove extraneous material, such as rocks or debris, unless the project requires that such material be processed as part of the sample. Continue with Step A10.2.5.

A10.2.3. If the sample volume is greater than ~450 mL, carefully pour the entire sample into a cone onto the paper.

Remove extraneous material, such as rocks or debris unless the project requires that such material be processed as part of the sample.

A10.2.4. If levels of gross activity in the sample permit, the sample is split at least twice using the coning and quartering steps that follow.

NOTE: Unused quarters are considered representative of the original sample and may be reserved for additional testing. The process should be carried out expediently to minimize loss of volatile components in the sample, especially if volatile components or percent solids are to be determined.

A10.2.4.1. Spread the material into a flat circular cake of soil using a tongue depressor or other suitable disposable implement. Divide the cake radially and return two opposing quarters to the original sample container.

A10.2.4.2. Reshape the remaining two quarters into a smaller cone, and repeat Step A10.2.2.1 until the total volume of the remaining material is approximately 100-150 mL.

NOTE: Tare the can and lid together. Do not apply an adhesive label. Rather, la bel the can with permanent marker since the can will be placed in a drying oven. The lid should be labeled separately since it will be removed from the can during drying.

A10.2.5. Transfer the coned and quartered sample to a tared, labeled 1-pint paint can. If the total volume was less than ~450 mL, transfer the entire sample to a tared, labeled 1-quart paint can.

NOTE: **Constant mass may be determined by removing the container from the oven and weighing repeatedly until the mass remains constant with within 1% of the starting mass of the sample. This determination may also be achieved**

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[6] International Union of Pure and Applied Chemistry (IUPAC). 1997. Compendium 1675 of Chemical Terminology, 2nd ed. (the "Gold Book"). Compiled by A. D. (Reference A16.1).

operationally by observing the time needed to ensure that 99% of all samples will obtain constant mass.

A10.3. Place the can (without lid) in an oven at 110 ± 10 ºC and dry the concrete or brick to constant mass.

NOTE: **Concrete or brick samples may be dry enough such that heating prior to homogenizing the sample is not required.**

A10.4. Weigh the combined mass of the can, sample, and lid. If the percent solids are required see Section A12.1 calculations. Remove can from oven and allow to cool.

A10.5. Add five 1.5 cm stainless steel or ceramic balls or rods to the can. Replace the lid and seal well.

A10.6. Shake the can and contents for 5 minutes, or longer, as needed to produce a finely-milled, well-homogenized, sample.

NOTE: **Although the precise particle size of the milled sample is not critical, complete pulverization and fine particle size facilitates representative sub-sampling and subsequent fusion or digestion processes. A qualitative standard can be prepared by passing quartz sand or other milled material through a 50-mesh and then a 100-mesh screen. The portion of the sample retained in the 100 mesh screen can be used as a qualitative visual standard to determine if samples have been adequately pulverized.**

A10.7. Visually compare the resulting milled sample to a qualitative 50–100 mesh pulverized sample (~150–300 µm or 50–100 mesh using the Tyler screen scale). The process is complete once 95% of the sample (or greater) is as fine, or finer, than the qualitative standard. If, by visual estimation, more than ~5% of total volume of the particles in the sample appear to be larger than the particle size in the standard, return the sample to the shaker and continue milling until the process is complete.

A10.8. Following milling, a small fraction of residual larger particles may remain in the sample.

A10.8.1. If the sample was collected close to the epicenter of an RDD or IND explosion, it may also contain particles of contaminant referred to as "discrete radioactive particles" or DRPs. In such a case, the larger particles should be isolated by passing through a disposable 48 mesh screen and checked for radioactivity. DRPs are readily identified by their very high specific activity which is detectable using hand-held survey equipment such as a thin-window G-M probe held within an inch of the particles.

A10.8.1.1. If radioactivity is clearly detected, the sieved material is returned to the can and ball milled until the desired mesh is obtained. In some cases, these materials may be resistant to further pulverization and may need to be processed according to a method specially designed to address highly intractable solids.

A10.8.1.2. If the presence of DRPs is of no concern, the larger particles need not be included in subsequent subsamples taken for analysis. It may be possible to easily avoid including them during aliquanting with a disposable scoop. If not, however, they should be removed by sieving through a nominal 50 mesh screen (disposable) prior to further subsampling for subsequent analyses.

A10.9. Sample fines may be stored in, and aliquanted directly from, the container used for drying and pulverization.

A11. Calibration and Standardization

A11.1. Balances used shall be calibrated using National Institute of Standards and Technology (NIST)-traceable weights according to the process defined by the laboratory's quality manual.

A12. Data Analysis and Calculations

A12.1. The percent solids (dry-to-as-received mass ratio) for each sample is calculated from data obtained during the preparation of the sample as follows:

$$\%\ \text{Solids} = \frac{M_{\text{dry}} - M_{\text{tare}}}{M_{\text{as rec}} - M_{\text{tare}}} \times 100$$

Where:

M_{dry} = mass of dry sample + labeled can + lid (g)

M_{tare} = tare mass of labeled can + lid (g)

$M_{\text{as rec}}$ = mass of sample as received + labeled can + lid (g)

A12.2. If requested, convert the equivalent mass of sample, as received, to dry mass. Dry mass is calculated from a measurement of the total as received mass of the sample received as follows:

$$\text{Dry Sample Equivalent} = M_{\text{total-as rec.}} \times \frac{\%\ \text{Solids}}{100}$$

Where:

$M_{\text{total-as rec.}}$ = total mass of sample, as received (g)

A12.3. Results Reporting

A12.3.1. The result for percent solids and the approximate total mass of sample as received should generally be reported for each result.

A13. Method Performance

A13.1. Results of method validation performance are to be archived and available for reporting purposes.

A13.2. Expected turnaround time is about 3 hours for an individual sample and about 4 hours per batch.

A14. Pollution Prevention.

Not applicable

A15. Waste Management

A15.1. All radioactive and other regulated wastes shall be handled according to prevailing regulations.

A16. References

A16.1. International Union of Pure and Applied Chemistry (IUPAC). 1997. Compendium of Chemical Terminology, 2nd ed. (the “Gold Book”). Compiled by A. D. McNaught and A. Wilkinson. Blackwell Scientific Publications, Oxford. XML on-line corrected version: http://goldbook.iupac.org/C01265.html. (2006) created by M. Nic, J. Jirat, B. Kosata; updates compiled by A. Jenkins. Last update: 2010-12-22.

A16.2. ALS Laboratories, Fort Collins, SOP 736.

A16.3. MARLAP. *Multi-Agency Radiological Laboratory Analytical Protocols Manual.* 2004. Volumes 1 – 3. Washington, DC: EPA 402-B-04-001A-C, NUREG 1576, NTIS PB2004-105421, July. Available at: www.epa.gov/radiation/marlap.

A16.4. ASTM C 999-05, “Standard Practice for Soil Sample Preparation for the Determination of Radionuclides,” Volume 12.01, ASTM, 2005.

Attachment III:

Rapid Radiochemical Method for Total Radiostrontium (Sr-90) in Building Materials for Environmental Remediation Following Radiological Incidents

1. Scope and Application

   1.1. The method will be applicable to samples where the source of the contamination is either from known or unknown origins. The method provides a very rapid screen for total radiostrontium (^{89}Sr + ^{90}Sr) in building materials samples, such as concrete and brick.

   1.2. This method is specific for beta-emitting isotopes of strontium in building materials such as concrete and brick. It uses rapid digestion and precipitation steps to preconcentrate strontium isotopes, followed by final purification using Sr Resin (see footnote on next page) to remove interferences.

   1.3. This method uses rapid radiochemical separations techniques for the determination of beta-emitting strontium radioisotopes in concrete or brick samples following a nuclear or radiological incident.

   1.4. The method is capable of satisfying a required method uncertainty for ^{90}Sr (total as ^{90}Sr) of 0.32 pCi/g at an analytical action level (AAL) of 2.44 pCi/g, a required relative method uncertainty (φ_{MR}) of 13% above the AAL, and a MDC of 0.40 pCi/g. To attain the required method uncertainty at the AAL, a sample weight of 1.5 g and a count time of approximately 1.5 hours are recommended. The sample turnaround time and throughput may vary based on additional project measurement quality objectives (MQOs), the time for analysis of the final counting form, and initial sample volume. The method must be validated prior to use following the protocols provided in *Method Validation Guide for Qualifying Methods Used by Radiological Laboratories Participating in Incident Response Activities* (Reference 16.1).

   1.5. This method is intended to be used for building materials. The rapid ^{90}Sr method was initially validated for concrete building materials following the guidance presented for “Level E Method Validation: Adapted or Newly Developed Methods, Including Rapid Methods” in *Method Validation Guide for Qualifying Methods Used by Radiological Laboratories Participating in Incident Response Activities* (Reference 16.1) and Chapter 6 of *Multi-Agency Radiological Laboratory Analytical Protocols Manual* (EPA 2004, Reference 16.2). Subsequent building material matrices were validated at Level C (“Similar Matrix/New Application”).

   1.6. Other solid samples such as soil can be digested using the rapid sodium hydroxide fusion procedure as an alternative to other digestion techniques, but the laboratory will have to validate this procedure.

2. Summary of Method

   2.1. Strontium is collected and purified from the building materials sample matrix using sodium hydroxide fusion (Reference 16.3) and purified from potentially interfering radionuclides and matrix constituents using a strontium-specific, rapid chemical separation method. The sample is equilibrated with strontium carrier, and preconcentrated by Sr/$CaCO_3$ coprecipitation from the alkaline fusion matrix. The carbonate precipitate is dissolved in HCl and strontium is precipitated with calcium fluoride to remove silicates. The precipitate is dissolved in strong nitric acid and the solution is passed through a Sr Resin extraction chromatography column[7] that selectively retains strontium while allowing most interfering radionuclides and matrix constituents to pass through to waste. If present in the sample, residual plutonium and several interfering tetravalent radionuclides are stripped from the column using an oxalic/nitric acid rinse. Strontium is eluted from the column with 0.05M HNO_3 and taken to dryness in a tared, stainless steel planchet. The planchet containing the strontium nitrate precipitate is weighed to determine the strontium yield.

2.2. The sample test source is promptly counted on a gas flow proportional counter to determine the beta emission rate, which is used to calculate the total radiostrontium activity.

2.2.1. The same prepared sample test source can be recounted after ~10 days to attempt to differentiate ^{89}Sr from ^{90}Sr. If the initial and second counts agree (based on the expected ingrowth of ^{90}Y), this is an indication that ^{89}Sr is not present in significant amounts relative to ^{90}Sr (within the uncertainty of the measurement).

2.2.2. Computational methods are available for resolving the concentration of ^{89}Sr and ^{90}Sr from two sequential counts of the sample. An example of an approach that has been used successfully at a number of laboratories is presented in Appendix B to this method. It is the responsibility of the laboratory, however, to validate this approach prior to its use.

2.2.3. It is also possible to determine ^{89}Sr more rapidly using Cerenkov counting if significant amounts of ^{89}Sr are suspected; this method must be validated independently. The minimum detectable activity (MDA) levels with Cerenkov counting, however, will be higher than of determination with gas proportional counting and may or may not meet measurement quality objectives.

3. Definitions, Abbreviations, and Acronyms

3.1. Analytical Protocol Specification (APS). The output of a directed planning process that contains the project's analytical data needs and requirements in an organized, concise form.

3.2. Analytical Action Level (AAL). The term analytical action level is used to denote the value of a quantity that will cause the decision-maker to choose one of the alternative actions.

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[7] Sr-Resin™ is a proprietary extraction chromatography resin consisting of octanol solution of 4,4'(5')-bis (t-butyl-cyclohexanol)-18-crown-6-sorbed on an inert polymeric support. The resin can be employed in a traditional chromatography column configuration (gravity or vacuum) or in a flow cartridge configuration designed for use with vacuum box technology. Sr-Resin™ is available from Eichrom Technologies, Lisle, IL. Throughout the remainder of the method, the terms "Sr-Resin" or "Sr-cartridge" will be used for Sr-Resin™.

3.3. Discrete Radioactive Particles (DRPs or “hot particles”). Particulate matter in a sample of any matrix where a high concentration of radioactive material is contained in a tiny particle (µm range).

3.4. Laboratory Control Sample (LCS). A standard material of known composition or an artificial sample (created by fortification of a clean material similar in nature to the sample), which is prepared and analyzed in the same manner as the sample. In an ideal situation, the result of an analysis of the laboratory control sample should be equivalent to (give 100 percent of) the target analyte concentration or activity known to be present in the fortified sample or standard material. The result normally is expressed as percent recovery.

3.5. Matrix Spike (MS). An aliquant of a sample prepared by adding a known quantity of target analytes to specified amount of matrix and subjected to the entire analytical procedure to establish if the method or procedure is appropriate for the analysis of the particular matrix.

3.6. *Multi-Agency Radiological Analytical Laboratory Protocols (MARLAP) Manual* provides guidance for the planning, implementation, and assessment phases of those projects that require the laboratory analysis of radionuclides (Reference 16.2).

3.7. Measurement Quality Objective (MQO). MQOs are the analytical data requirements of the data quality objectives and are project- or program-specific. They can be quantitative or qualitative. MQOs serve as measurement performance criteria or objectives of the analytical process.

3.8. Radiological Dispersal Device (RDD), i.e., a “dirty bomb.” This device is an unconventional weapon constructed to distribute radioactive material(s) into the environment either by incorporating them into a conventional bomb or by using sprays, canisters, or manual dispersal.

3.9. Required Method Uncertainty (u_{MR}). The required method uncertainty is a target value for the individual measurement uncertainties and is an estimate of uncertainty (of measurement) before the sample is actually measured. The required method uncertainty is applicable below an AAL.

3.10. Relative Required Method Uncertainty (φ_{MR}). The relative required method uncertainty is the u_{MR} divided by the AAL and is typically expressed as a percentage. It is applicable above the AAL.

3.11. Sample Test Source (STS). This is the final form of the sample that is used for nuclear counting. This form is usually specific for the nuclear counting technique in the method, such as a solid deposited on a filter for alpha spectrometry analysis.

3.12. Total Radiostrontium (also called Total Strontium): A radiological measurement that does not differentiate between ^{89}Sr and ^{90}Sr. The assumption is that all of the strontium is in the form of ^{90}Sr. When it is certain that no ^{89}Sr is present, the total radiostrontium activity is equal to the ^{90}Sr activity and may be reported as such.

3.13. Working Calibration Source (WCS): A prepared source, made from a certified reference material (standard), including those diluted or prepared by chemical procedure, for the purpose of calibrating an instrument.

4. Interferences

4.1. Radiological

4.1.1. Count results should be monitored for detectable alpha activity and appropriate corrective actions taken when observed. Failure to address the presence of alpha emitters in the sample test source may lead to high result bias due to alpha-to-beta crosstalk.

4.1.1.1. Elevated levels of radioisotopes of tetravalent plutonium, neptunium, cerium, and ruthenium in the sample may hold up on the column and co-elute with strontium. The method employs an oxalic acid rinse that should address low to moderate levels of these interferences in samples.

The resin has a higher affinity for polonium than strontium at low nitric acid concentrations but only minimal retention in 8M HNO_3. If there were any residual Po (IV) retained, it would likely be removed using the 3M HNO_3-0.05M oxalic acid rinse.

4.1.2. Significant levels of ^{89}Sr in the sample will interfere with the total radiostrontium analysis.

4.1.2.1. The absence of higher activities of interfering ^{89}Sr may be detected by counting the sample test source quickly after initial separation (minimizing ingrowth of ^{90}Y), and then recounting the sample test source after 1–21 days to verify that the calculated activity (based on the expected ingrowth of ^{90}Y) does not change significantly. The presence of only ^{89}Sr may be indicated when the calculated activity of the second count is less than that of the first count by an amount greater than that which can be attributed to statistical variation in the two analyses. However, the second count activity is a complex function of the amount of the ^{89}Sr present, the ingrowth of the ^{90}Y from ^{90}Sr and the time between the first and second counts.

4.1.2.2. Alternatively, Appendix B provides a numerical approach for the isotopic determination ^{89}Sr and ^{90}Sr from two sequential counts of the sample, one immediately following separation, and one after a delay to allow for ingrowth of ^{90}Y and decay of ^{89}Sr.

4.1.3. High levels of ^{210}Pb may interfere with low-level strontium analysis due to ingrowth of short-lived ^{210}Bi during chemical separations, where Pb is retained by Sr Resin but is not eluted. If ^{210}Pb is known to be present in samples, minimizing the time between the final rinse and the elution of strontium to less than 15 minutes will maintain levels of interfering ^{210}Bi to less than 0.1% of the ^{210}Pb activity present. The presence or absence of interfering ^{210}Bi may be determined by recounting the sample test source to verify the half-life of the nuclide present. Bi-214 also can grow in during the elution step if ^{214}Pb (half life-26.8 minutes) makes it to the Sr Resin. In this case, holding the samples until the ^{214}Bi decays (~2 hours) may be advisable.

4.1.4. High levels of ^{228}Th or its decay progeny ^{224}Ra and ^{212}Pb may interfere with low-level strontium determinations due to ingrowth of short-lived decay products during chemical separations. Monitoring count data for alpha activity may provide indications of interferences. Minimizing the time between the final rinse and the elution of strontium from the column to 5 minutes should maintain levels of interfering ^{212}Pb and ^{208}Tl to less than 2% of the parent nuclide activity. The presence or absence of ^{212}Pb may be determined by recounting the sample test source to verify the half-life of the nuclide present.

4.1.5. Levels of radioactive cesium or cobalt in excess of approximately 10^3 times the activity of strontium being measured may not be completely removed and may interfere with final results. Column rinsing to remove interferences may be increased to minimize interference if high levels of cesium or cobalt are known to be present. Changing cartridge connector tips and/or column reservoirs prior to final elution of strontium from Sr Resin can facilitate removal of sample matrix interferences. It may also be possible to increase the nitric acid in the eluted Sr fraction to 8M HNO_3 and reprocess the sample through the column separation method again.

4.2. Non-Radiological

4.2.1. Stable strontium present in the concrete, brick or other solid sample at levels that are significant relative to the stable Sr carrier added will increase the apparent gravimetric yield and cause a negative bias in the final results. If the quantity of native strontium in the sample aliquant exceeds ~5% of the expected strontium carrier mass, chemical yield measurements will be affected unless the native strontium is accounted for in the yield calculations.

4.2.2. The native strontium content in the sample may be determined by an independent spectrometric measurement (such as inductively coupled plasma - atomic emission spectroscopy [ICP-AES], etc.) or by taking an aliquant and processing the sample without the addition of strontium carrier to obtain an estimate of the native strontium content of the sample.

4.2.3. Sr Resin has a greater affinity for lead than for strontium. Lead will quantitatively displace strontium from the column when the two are present in combined amounts approaching or exceeding the capacity of the column. If the combined quantity of lead and strontium carrier in the sample exceeds the capacity of the column, decreased strontium yields will be observed. High lead levels are not typically seen in building materials samples. However, decreasing the sample size will help address samples with elevated levels of lead.

4.2.4. High levels of calcium, barium or potassium may compete slightly with strontium for uptake on the resin, possibly leading to low chemical yield. If these interfering matrix constituents are present in the final sample test source, yield results will overestimate the true strontium yield and cause a low result bias.

4.2.5. Rinsing Sr Resin with 8M HNO_3 minimizes retention of Ba, K ions which have more retention at lower nitric acid levels to optimize removal of interferences.

4.2.6. The final solids on the planchets containing strontium nitrate should be white to very light brown. A significant brown color could indicate formation of iron oxide solids from the stainless steel planchets. This can cause a positive bias in the gravimetric chemical yields. Annealing the planchets properly minimizes the formation of iron oxide solids (Step 6.8).

5. Safety

5.1. General

5.1.1. Refer to your safety manual for concerns of contamination control, personal exposure monitoring and radiation dose monitoring.

5.1.2. Refer to your laboratory's chemical hygiene plan for general chemical safety rules.

5.2. Radiological

5.2.1. Hot Particles (DRPs)

5.2.1.1. Hot particles, also termed "discrete radioactive particles" (DRPs), will be small, on the order of 1 mm or less. Typically, DRPs are not evenly distributed in the media and their radiation emissions are not uniform in all directions (anisotropic).

5.2.2. For samples with detectable activity concentrations of these radionuclides, labware should be used only once due to potential for cross contamination.

5.3. Procedure-Specific Non-Radiological Hazards

None noted.

6. Equipment and supplies

6.1. Analytical balance with 10^{-4} g readability or better.

6.2. Centrifuge able to accommodate 225 mL and 50 mL centrifuge tubes.

6.3. Centrifuge tubes, 50 mL and 225 mL.

6.4. Hot plate.

6.5. Low-background gas flow proportional counter.

6.6. 100 µL, 200 µL, 500 µL, and 1 mL pipets or equivalent and appropriate plastic tips.

6.7. 1–10 mL electronic pipet.

6.8. Stainless steel planchets or other sample mounts: ~2-inch diameter, annealed at 530-550 °C or higher in a furnace for ~3.5 to 4 hours with a volume of ~5 mL. Planchets annealed properly will typically have a bronze/brown color. Do not overheat as the planchets will become more susceptible to acid degradation and iron oxide formation.

6.9. Tips, white inner, Eichrom part number AC-1000-IT, or PFA 5/32" × ¼" heavy-wall tubing connectors, natural, Ref P/N 00070EE, cut to 1 inch, Cole Parmer, or equivalent.

6.10. Tips, yellow outer, Eichrom part number AC-1000-OT, or equivalent.

6.11. Vacuum box, such as Eichrom part number AC-24-BOX, or equivalent.

6.12. Vacuum pump or laboratory vacuum system.

7. Reagents and Standards

NOTES:

All reagents are American Chemical Society (ACS) reagent grade or equivalent unless otherwise specified.

Unless otherwise indicated, all references to water should be understood to mean Type I reagent water (ASTM D1193, Reference 16.5). All solutions used in microprecipitation should be prepared with water filtered through a 0.45 µm (or better) filter.

7.1. Type I reagent water as defined in ASTM Standard D1193 (Reference 16.5).

7.2. Aluminum nitrate ($Al(NO_3)_3 \cdot 9H_2O$).

7.2.1. Aluminum nitrate solution, 2M ($Al(NO_3)_3$): Add 750 g of aluminum nitrate ($Al(NO_3)_3 \cdot 9H_2O$) to ~700 mL of water and dilute to 1 L with water.

7.3. Ethanol, reagent (C_2H_5OH): Available commercially (or mix 95 mL 100% ethanol and 5 mL water).

7.4. Nitric Acid, HNO_3 (15.8M), concentrated, available commercially.

7.4.1. Nitric acid (8M): Add 506 mL of concentrated HNO_3 to 400 mL of water and dilute to 1 L with water.

7.4.2. Nitric acid (3M): Add 190 mL of concentrated HNO_3 to 800 mL of water and dilute to 1 L with water.

7.4.3. Nitric acid (0.1M): Add 6.4 mL of concentrated HNO_3 to 900 mL water. Dilute to 1 L with water.

7.4.4. Nitric acid (0.05M): Add 3.2 mL of concentrated HNO_3 to 900 mL water. Dilute to 1 L with water.

7.5. Nitric acid (3M)/oxalic acid solution (0.05M): Add 190 mL of concentrated HNO_3 (7.3) and 6.3 g of oxalic acid dihydrate ($C_2H_2O_4 \cdot 2H_2O$), to 800 mL of demineralized water and dilute to 1 L with de-ionized water.

7.6. Sr Resin columns, ~1.00 g resin, 3 mL, small particle size (50–100 µm), in appropriately sized column or stacked 2 mL+ 1 mL pre-packed cartridges. (Available from Eichrom Technologies, Inc., Lisle IL.)

7.7. Strontium carrier solution, 7 mg/mL in 0.1M HNO_3, traceable to a national standards body such as NIST or standardized at the laboratory by comparison to independent standards.

7.7.1. Option 1: Dilute elemental strontium standard to a concentration of 7.00 mg/mL (or mg/g) in 0.1M HNO_3. Verify per Step 7.7.3.

7.7.2. Option 2: To ~200 mL de-ionized water, add 6.3 mL HNO_3 and approximately 16.90 g of strontium nitrate ($Sr(NO_3)_2$ dried to constant mass and the mass being determined to at least 0.001 g). Dilute to 1000 mL with water. Calculate the amount of strontium nitrate/mL actually present and verify per Step 7.7.3.

7.7.3. Prior to use, verify the strontium carrier solution concentration by transferring at least five 1.00-mL portions of the carrier to tared stainless steel planchets. Evaporate to dryness on a medium heat on a hotplate using the same technique as that used for samples (Heat 5 minutes after dryness is reached to ensure complete dryness). Allow to cool and weigh as the nitrate to the nearest 0.1 mg. The relative standard deviation for replicates should be less than 5% and the average residue mass within 5% of the expected value.

7.8. ^{90}Sr standard solution (carrier free), traceable to a national standards body such as NIST, in 0.5M HNO_3 solution.

8. Sample Collection, Preservation, and Storage
Not Applicable.

9. Quality Control

9.1. Batch quality control results shall be evaluated and meet applicable Analytical Protocol Specifications (APS) prior to release of unqualified data. In the absence of project-defined APS or a project-specific quality assurance project plan (QAPP), the quality control sample acceptance criteria defined in the laboratory quality manual and procedures shall be used to determine acceptable performance for this method.

9.1.1. A laboratory control sample (LCS) shall be run with each batch of samples. The concentration of the LCS should be at or near the action level or a level of interest for the project.

9.1.2. One method blank shall be run with each batch of samples fused using the procedure *Rapid Method for Sodium Hydroxide Fusion of Concrete and Brick Matrices Prior to Americium, Plutonium, Strontium, Radium, and Uranium Analyses* (Reference 16.3). If analyte-free blank material is not available and an empty crucible is used to generate a reagent blank sample, it is recommended that100 mg calcium be added as calcium nitrate to the empty crucible as blank simulant. This addition facilitates strontium carbonate precipitations from the alkaline fusion matrix.

9.1.3. One laboratory duplicate shall be run with each batch of samples. The laboratory duplicate is prepared by removing an aliquant from the original sample container.

9.1.4. A matrix spike sample may be included as a batch quality control sample if there is concern that matrix interferences, such as the presence of elemental strontium in the sample, may compromise chemical yield measurements, or overall data quality.

10. Calibration and Standardization

10.1. The effective detection efficiency for total radiostrontium (referenced to ^{90}Sr) is calculated as the weighted sum of the ^{90}Sr and ^{90}Y efficiencies that reflects the relative proportions of ^{90}Y and ^{90}Sr based on the ^{90}Y ingrowth after ^{90}Sr separation.

10.2. Set up, operate, and perform quality control for gas-flow proportional counters (GPC) in accordance with the laboratory's quality manual and standard operating procedures, and consistent with ASTM Standard Practice D7282, Sections 7–13 (Reference 16.4).

10.3. See Appendix A for details on calibration/standardization of the GPC specific to ^{90}Sr and ^{90}Y.

11. Procedure

11.1. Initial Sample Preparation for ^{89}Sr + ^{90}Sr

11.1.1. ^{89,90}Sr may be preconcentrated from building material samples using the separate procedure (Reference 16.3), which fuses the samples using rapid NaOH fusion followed by carbonate and fluoride precipitations to preconcentrate ^{89,90}Sr from the hydroxide matrix.

NOTE: The fusion procedure provides a column load solution for each sample (consisting of 20 mL of 8M HNO_3–0.5M $Al(NO_3)_3$), ready for column separation on Sr Resin.

11.1.2. This separation can be used with other solid sample matrices if the initial sample preparation steps result in a column load solution containing ~8M HNO_3- 0.5M $Al(NO_3)_3$ is used.

11.1.3. A smaller volume of the total load solution may be taken and analyzed as needed for very high activity samples, with appropriate dilution factor calculations applied.

11.2. Rapid Sr Separation using Sr Resin

11.2.1. Set up vacuum box

11.2.1.1. Place the inner tube rack (supplied with vacuum box) into the vacuum box with the centrifuge tubes in the rack. Place the lid onto the vacuum box system.

11.2.1.2. Place the yellow outer tips into all 24 openings of the lid of the vacuum box. Fit in the inner white tip into each yellow tip.

11.2.1.3. For each sample solution, place the Sr Resin cartridges (2 mL+1 mL cartridges) on to the inner white tip.

11.2.1.4. Place reservoirs on the top end of the Sr Resin cartridge.

11.2.1.5. Turn the vacuum on (building vacuum or pump) and ensure proper fitting of the lid.

IMPORTANT: The unused openings on the vacuum box should be sealed. Yellow caps (included with the vacuum box) can be used to plug unused white tips to achieve a good seal during the separation. Alternately, plastic tape can be used to seal the unused lid holes as well.

11.2.1.6. Add 5 mL of 8M HNO_3 to the column reservoir to precondition the Sr Resin cartridges.

11.2.1.7. Adjust the vacuum to achieve a flow-rate of ~1 mL/min.

NOTE: Unless otherwise specified in the procedure, use a flow rate of ~1 mL/min for load and strip solutions and ~2-3 mL/min for rinse solutions.

11.2.2. Sr Resin Separation

11.2.2.1. Transfer each sample solution from the fusion procedure (Reference 16.3) into the appropriate reservoir. Allow solution to pass through the Sr Resin cartridge at a flow rate of ~1 mL/min.

11.2.2.2. Add 5 mL of 8M HNO_3 to each beaker/tube (from Step 11.2.2.1) as a rinse and transfer each solution into the appropriate reservoir (the flow rate can be adjusted to ~2 mL/min).

11.2.2.3. Add 15 mL of 8M HNO_3 into each reservoir as second column rinse (flow rate ~3–4 mL/min).

11.2.2.4. Turn off vacuum and discard rinse solutions.

11.2.2.5. Add~5 mL 3M HNO_3 - 0.05M oxalic acid solution to each column (flow rate ~1–2 mL/min).

11.2.2.6. Add 5 mL of 8M HNO_3 into each reservoir as second column rinse (flow rate ~3 mL/min).

11.2.2.7. Discard column rinses.

11.2.2.8. Record time and date of the end of last rinse to the nearest 15 minutes as t_1, “time of strontium separation.”

11.2.2.9. Place clean 50 mL centrifuge tubes beneath the columns to catch the strontium eluate before proceeding to the next step.

11.2.2.10. Elute strontium from the columns by adding 15 mL of 0.05M HNO_3 at ~1 mL/min.

11.2.2.11. Discard Sr Resin cartridges.

11.2.3. Preparation of the STS and determination of chemical yield

11.2.3.1. Clean and label a stainless steel planchet for each STS.

11.2.3.2. Weigh and record the tare mass of each planchet to the nearest 0.1 mg.

11.2.3.3. Transfer the strontium eluate from Step 11.2.2.10 to the planchet and take to dryness on a hotplate (medium heat) to produce a uniformly distributed residue across the bottom of the planchet.

NOTE: A few mL at a time typically is added to the planchet during evaporation. Do not evaporate all the way to dryness to prevent splattering. After adding all 15 mL, take the planchet all the way to dryness.

11.2.3.4. Rinse tubes with ~2 mL 0.05M HNO_3 and add to the planchet.

11.2.3.5. Heat on hot plate for ~5–10 minutes after initial dryness is reached.

11.2.3.6. Allow planchets to cool.

11.2.3.7. Weigh and record the gross mass of each planchet to the nearest 0.1 mg.

NOTE: If gravimetric yields are unusually high with the possibility of moisture present, additional heating and reweighing should be performed.

11.2.3.8. Calculate the chemical yield as presented in Step 12 of this method.

11.3. Counting the Sample Test Source

11.3.1. On a calibrated gas-flow proportional detector that has passed all required daily performance and background checks, count the STS for a period as needed to satisfy MQOs.

11.3.1.1. If the presence of ^{89}Sr cannot be excluded, and total radiostrontium is being determined as a screen for the presence of ^{89}Sr or ^{90}Sr, count the STS as soon as practicable after preparation to minimize the ingrowth of ^{90}Y into the STS.

11.3.1.2. If the presence of ^{89}Sr can be excluded, total radiostrontium will provide isotopic ^{90}Sr results and the STS may be counted at any time after preparation (taking into account the appropriate increase in activity due to ^{90}Y ingrowth).

11.3.2. Calculate the total radiostrontium (^{90}Sr) sample results using calculations presented in Step 12.

11.3.3. Hold planchets for recounting as needed.

12. Data Analysis and Calculations

12.1. Calculation of Total Radiostrontium

12.1.1. When a sample is analyzed for total radiostrontium (equivalent ^{90}Sr), the effective efficiency is calculated as follows:

$$\varepsilon_{\text{Total Sr}} = \varepsilon_{\text{Sr90}} + \left(1 - \mathrm{e}^{-\lambda_{Y90}(t_2 - t_1)}\right) \times \varepsilon_{\text{Y90}} \qquad (1)$$

where

$\varepsilon_{\text{Total Sr}}$ = effective detection efficiency for total radiostrontium referenced to ^{90}Sr
$\varepsilon_{\text{Sr90}}$ = final ^{90}Sr detection efficiency
ε_{Y90} = final ^{90}Y detection efficiency
λ_{Y90} = decay constant for ^{90}Y, 3.005×10^{-6} seconds $(\text{s})^{-1}$
t_1 = date and time of the Sr/Y separation (s)
t_2 = date and time of the midpoint of the count (s)

NOTE: The elapsed time between the sample count and the reference date must be calculated using the same time units as the decay constant.

12.1.2. The standard uncertainty of the effective efficiency is calculated as follows:

$$u(\varepsilon_{\text{Total Sr}}) = \sqrt{u^2(\varepsilon_{\text{Sr90}}) + \left(1 - \mathrm{e}^{-\lambda_{Y90}(t_2 - t_1)}\right)^2 u^2(\varepsilon_{\text{Y90}}) + 2\left(1 - \mathrm{e}^{-\lambda_{Y90}(t_2 - t_1)}\right) u(\varepsilon_{\text{Sr90}}, \varepsilon_{\text{Y90}})} \qquad (2)$$

where

$$u(\varepsilon_{\text{Sr90}}, \varepsilon_{\text{Y90}}) = r(\varepsilon_{\text{Sr90}}, \varepsilon_{\text{Y90}})\, u(\varepsilon_{\text{Sr90}})\, u(\varepsilon_{\text{Y90}})$$

NOTE: The terms $u(\varepsilon_{\text{Sr90}}, \varepsilon_{\text{Y90}})$ and $r(\varepsilon_{\text{Sr90}}, \varepsilon_{\text{Y90}})$ are derived during calibrations as shown in Appendix A, Step A4.

12.1.3. The total radiostrontium activity concentration ($AC_{\text{Total Sr}}$) equivalent to ^{90}Sr is calculated as follows:

$$AC_{\text{Total Sr}} = \frac{R_{\text{a}} - R_{\text{b}}}{2.22 \times \varepsilon_{\text{Total Sr}} \times Y \times W \times DF} \qquad (3)$$

where

$$DF = \mathrm{e}^{-\lambda_{\text{Sr90}}(t_1 - t_0)}$$

and where

R_{a} = beta gross count rate for the sample (counts per minute [cpm])
R_{b} = beta background count rate (cpm)
$\varepsilon_{\text{Total Sr}}$ = effective efficiency of the detector for total strontium referenced to ^{90}Sr
Y = fractional chemical yield for strontium
W = weight of the sample aliquant (g)
DF = correction factor for decay of the sample from its reference date until the midpoint of the total strontium count
λ_{Sr90} = decay constant for ^{90}Sr, 7.642×10^{-10} s^{-1}
t_0 = reference date and time for the sample (s)
t_1 = date and time of the Sr/Y separation (s)

NOTE: The elapsed time between the sample count and the reference date must be calculated using the same time units as the decay constant

12.1.4. The standard counting uncertainty of the total radiostrontium activity concentration, $u_{\text{cC}}(AC_{\text{Total Sr}})$ is calculated as follows:

$$u_{\text{cC}}(AC_{\text{Total Sr}}) = \frac{\sqrt{\dfrac{R_{\text{a}}}{t_{\text{a}}} + \dfrac{R_{\text{b}}}{t_{\text{b}}}}}{2.22 \times \varepsilon_{\text{Total Sr}} \times Y \times W \times DF} \qquad (4)$$

where:

t_a = Duration of the sample count (min)
t_b = Duration of the background subtraction count (min)

12.1.5. The combined standard uncertainty (CSU) for the total radiostrontium activity concentration, $u_c(AC_{\text{Total Sr}})$, is calculated as follows:

$$u_{\mathrm{c}}(AC_{\text{Total Sr}}) = \sqrt{u_{\mathrm{cC}}^{2}(AC_{\text{Total Sr}}) + AC_{\text{Total Sr}}^{2}\left(\frac{u^{2}(\varepsilon_{\text{Total Sr}})}{\varepsilon_{\text{Total Sr}}^{2}} + \frac{u^{2}(Y)}{Y^{2}} + \frac{u^{2}(W)}{W^{2}}\right)} \tag{5}$$

where:

u(Y) = standard uncertainty of fractional chemical yield for strontium
u(W) = standard uncertainty of the weight of the sample aliquant (g)

12.1.6. If the critical level concentration (L_c) or the minimum detectable concentration (MDC) are requested (at an error rate of 5%), they can be calculated using the following equations:[8]

$$L_{\mathrm{C}} = \frac{\left[0.4\times\left(\frac{t_s}{t_b}-1\right)+0.677\times\left(1+\frac{t_s}{t_b}\right)+1.645\times\sqrt{\left(R_b\,t_b+0.4\right)\times\frac{t_s}{t_b}\times\left(1+\frac{t_s}{t_b}\right)}\right]}{t_s\times 2.22\times\varepsilon_{\text{Total Sr}}\times Y\times V\times DF} \tag{6}$$

$$MDC = \frac{\left[2.71\times\left(1+\frac{t_{\mathrm{s}}}{t_{\mathrm{b}}}\right)+3.29\times\sqrt{R_{\mathrm{b}}\,t_{\mathrm{s}}\times\left(1+\frac{t_{\mathrm{s}}}{t_{\mathrm{b}}}\right)}\right]}{t_{\mathrm{s}}\times 2.22\times\varepsilon_{\text{Total Sr}}\times Y\times W\times DF} \tag{7}$$

12.2. Chemical Yield for Strontium

12.2.1. Calculate the chemical yield for strontium using the gravimetric data collected in Step 11.2.3:

$$Y = \frac{m_{\mathrm{s}}F_{\mathrm{Sr(NO_3)_2}}}{c_{\mathrm{c}}V_{\mathrm{c}} + c_{\mathrm{n}}W} \tag{8}$$

where:

Y = strontium yield, expressed as a fraction
m_s = mass of $Sr(NO_3)_2$ recovered from the sample (mg)
$F_{\mathrm{Sr(NO_3)_2}}$ = gravimetric factor for strontium weighed as the nitrate, 0.414
c_c = Sr mass concentration in the strontium carrier solution (mg/mL)
V_c = volume of strontium carrier added to the sample (mL)

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[8] The formulations for the critical level and minimum detectable concentrations are based on the Stapleton Approximation as recommended in MARLAP Section 20A.2.2, Equations 20.54 and 20A.3.2, and Equation 20.74, respectively. The formulations presented assume $\alpha = 0.05$, $\beta = 0.05$ (with $z_{1-\alpha} = z_{1-\beta} = 1.645$), and $d = 0.4$.

c_n = Sr mass concentration native to the sample – if determined (mg/g)
W = weight of sample aliquant (g)

12.2.2. Calculate the standard uncertainty of the yield as follows:

$$u(Y) = Y \times \sqrt{u_\text{r}^2(m_\text{s}) + \frac{u^2(c_\text{c})V_\text{c}^2 + c_\text{c}^2\,u^2(V_\text{c}) + u^2(c_\text{n})V^2 + c_\text{n}^2\,u^2(W)}{(c_\text{c}V_\text{c} + c_\text{n}W)^2}} \quad (9)$$

where
$u(\cdot)$ = standard uncertainty of the quantity in parentheses,
$u_\text{r}(\cdot)$ = relative standard uncertainty of the quantity in parentheses.

12.3. Results Reporting

12.3.1. Unless otherwise specified in the APS, the following items should be reported for each result:

12.3.1.1. Result for total radiostrontium in scientific notation ±1 combined standard uncertainty.

12.3.1.2. Weight of sample aliquant and any dilutions used.

12.3.1.3. Yield of tracer and its uncertainty.

12.3.1.4. Case narrative.

13. Method Performance

13.1. Results of method validation performance are to be archived and available for reporting purposes.

13.2. Expected turnaround time per sample or per batch (See Step 17.5 for typical processing times (assumes samples are not from RDD).

13.2.1. Preparation and chemical separations for a batch of 20 samples can be performed by using a vacuum box system (24 ports each) simultaneously, assuming 24 detectors are available. For an analysis of a 1-g sample aliquant, sample preparation and digestion should take ~2.5 h.

13.2.2. Purification and separation of the strontium fraction using cartridges and vacuum box system should take ~2.5 h.

13.2.3. Sample test source preparation takes ~1.5 h.

13.2.4. A 60–90-minute counting time is sufficient to meet the MQO in Step 9.2, assuming 1.5-g aliquant, a background of 1 cpm, detector efficiency of 0.4–0.5, and radiochemical yield of at least 0.5.

13.3. Total radiostrontium (^{89}Sr+ ^{90}Sr) data reduction should be achievable between 5 and 8.5 hours after the beginning of the analysis, depending on batch size and count time.

13.4. The sample may be recounted following a delay of 10–21 days to differentiate the ^{89}Sr and ^{90}Sr activities. If the source contains pure ^{90}Sr, the total radiostrontium activity calculated from the two counts should agree within the uncertainty of the measurements. Minimizing the time between the chemical separation of Sr and the initial count, longer count times, and increasing the delay between the two counts, will minimize the overall uncertainty of the data and provide more sensitive and reliable measures of the radiochemical purity of the STS.

NOTE: The ^{89}Sr and ^{90}Sr may be determined from two consecutive counts of the source – calculations are presented in Appendix B. This approach must be validated prior to use.

14. Pollution Prevention

14.1. The use of Sr Resin reduces the amount of acids and hazardous metals that would otherwise be needed to co-precipitate and purify the sample and prepare the final counting form.

15. Waste Management

15.1. Nitric acid and hydrochloric acid wastes should be neutralized before disposal and then disposed in accordance with prevailing laboratory, local, state and federal requirements.

15.2. Final precipitated materials may contain radiostrontium and should be treated as radioactive waste and disposed in accordance with the restrictions provided in the facility's radioactive materials license and any prevailing government restrictions.

15.3. Used resins and columns should be considered radioactive waste and disposed of in accordance with restriction provided in the facility's radioactive materials license and any prevailing government restrictions.

16. References

Cited References

16.1. U.S. Environmental Protection Agency (EPA). 2009. *Method Validation Guide for Radiological Laboratories Participating in Incident Response Activities*. Revision 0. Office of Air and Radiation, Washington, DC. EPA 402-R-09-006, June. Available at: www.epa.gov/narel.

16.2. *Multi-Agency Radiological Laboratory Analytical Protocols Manual* (MARLAP). 2004. EPA 402-B-1304 04-001A, July. Volume I, Chapters 6, 7, 20, Glossary; Volume II and Volume III, Appendix G. Available at: www.epa.gov/radiation/marlap.

16.3. U.S. Environmental Protection Agency (EPA). 2014. *Rapid Method for Sodium Hydroxide Fusion of Concrete and Brick Matrices Prior to Americium, Plutonium, Strontium, Radium, and Uranium Analyses*. Revision 0, EPA 402-R-14-004. Office of Air and Radiation, Washington, DC. Available at: www.epa.gov/narel.

16.4. ASTM D7282 "Standard Practice for Set-Up, Calibration, and Quality Control of Instruments Used for Radioactivity Measurements," ASTM Book of Standards 11.02, current version, ASTM International, West Conshohocken, PA.

16.5. ASTM D1193, "Standard Specification for Reagent Water," ASTM Book of Standards 11.02, current version, ASTM International, West Conshohocken, PA.

Other References

16.6. Maxwell, S. 2006. “Rapid Column Extraction Method for Actinides and $^{89/90}$Sr in Water Samples,” *J. Radioanalytical and Nuclear Chemistry*. 267(3): 537-543.

16.7. Maxwell, S., Culligan, B. and Noyes, G. 2010. Rapid method for actinides in emergency soil samples, *Radiochimica Acta*. 98(12): 793-800.

16.8. Maxwell, S., Culligan, B., Kelsey-Wall, A. and Shaw, P. 2011. “Rapid Radiochemical Method for Actinides in Emergency Concrete and Brick Samples,” *Analytica Chimica Acta*. 701(1): 112-8.

16.9. SR-04, “Radiochemical Determination of Radiostrontium in Water, Sea Water, and Other Aqueous Media,” Eastern Environmental Radiation Facility (EERF) Radiochemistry Procedures Manual, Montgomery, AL, EPA 520/5-84-006 (August 1984).

16.10. SRW04-11, “Strontium 89, 90 in Water,” Eichrom Technologies, Inc., Lisle, Illinois (February 2003).

16.11. Nuclear data from NUDAT 2.3 and the National Nuclear Data Center at Brookhaven National Laboratory; available at: www.nndc.bnl.gov/nudat2/indx_dec.jsp, database version of 6/30/2009.

16.12. U.S. Environmental Protection Agency (EPA). 2014. Rapid Radiochemical Method for Americium-241 in Building Materials for Environmental Remediation Following Radiological Incidents. Revision 0, EPA 402-R-14-007. Office of Air and Radiation, Washington, DC. Available at: www.epa.gov/narel.

16.13. U.S. Environmental Protection Agency (EPA). 2014. Rapid Radiochemical Method for Pu-238 and Pu-239/240 in Building Materials for Environmental Remediation Following Radiological Incidents. Revision 0, EPA 402-R-14-006. Office of Air and Radiation, Washington, DC. Available at: www.epa.gov/narel.

16.14. U.S. Environmental Protection Agency (EPA). 2014. Rapid Radiochemical Method for Radium-226 in Building Materials for Environmental Remediation Following Radiological Incidents. Revision 0, EPA 402-R-14-002. Office of Air and Radiation, Washington, DC. Available at: www.epa.gov/narel.

16.15. U.S. Environmental Protection Agency (EPA). 2014. Rapid Radiochemical Method for Total Radiostrontium (Sr-90) in Building Materials for Environmental Remediation Following Radiological Incidents. Revision 0, EPA 402-R-14-001. Office of Air and Radiation, Washington, DC. Available at: www.epa.gov/narel.

16.16. U.S. Environmental Protection Agency (EPA). 2014. Rapid Radiochemical Method for Isotopic Uranium in Building Materials for Environmental Remediation Following Radiological Incidents. Revision 0, EPA 402-R-14-005. Office of Air and Radiation, Washington, DC. Available at: www.epa.gov/narel.

17. Tables, Diagrams, Flow Charts and Validation Data

17.1. Validation Data

This section intentionally left blank.

17.2. Nuclide Decay and Radiation Data

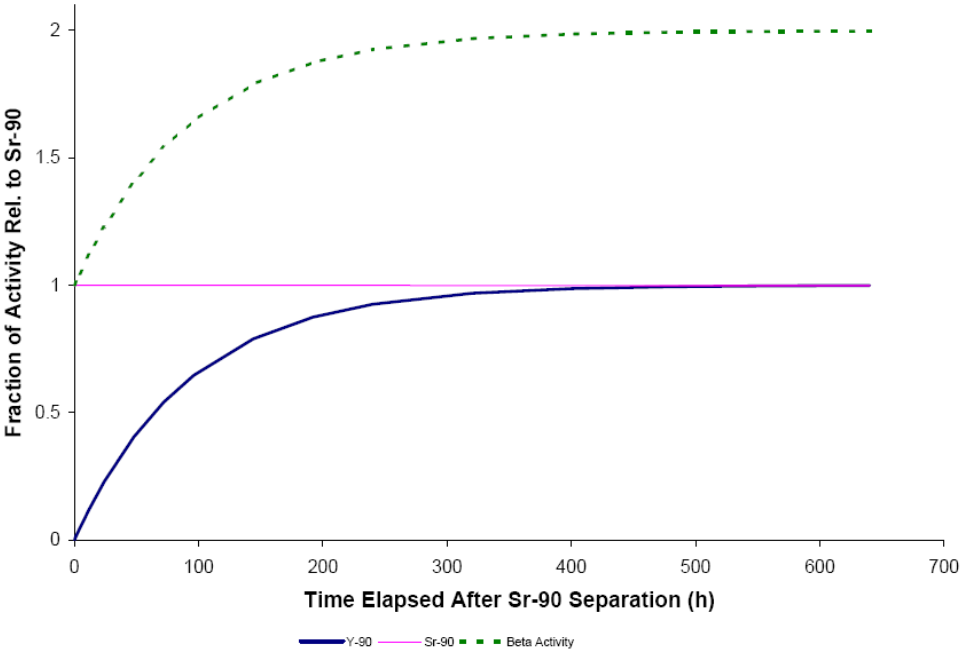
Table 17.2 – Decay and Radiation Data

Nuclide	Half-life (days)	λ (s^{-1})	Abundance	β_{max} (MeV[1])	β_{avg} (MeV)
^{90}Sr	1.052E+04	7.642×10^{-10}	1.00	0.546	0.196
^{90}Y	2.6667	3.005×10^{-6}	1.00	2.280	0.934
^{89}Sr	50.53	1.587×10^{-7}	1.00	1.495	0.585

[1] MeV – mega electron volts

17.3. Ingrowth and Decay Curves and Factors

In-Growth Curve for ^{90}Y in ^{90}Sr

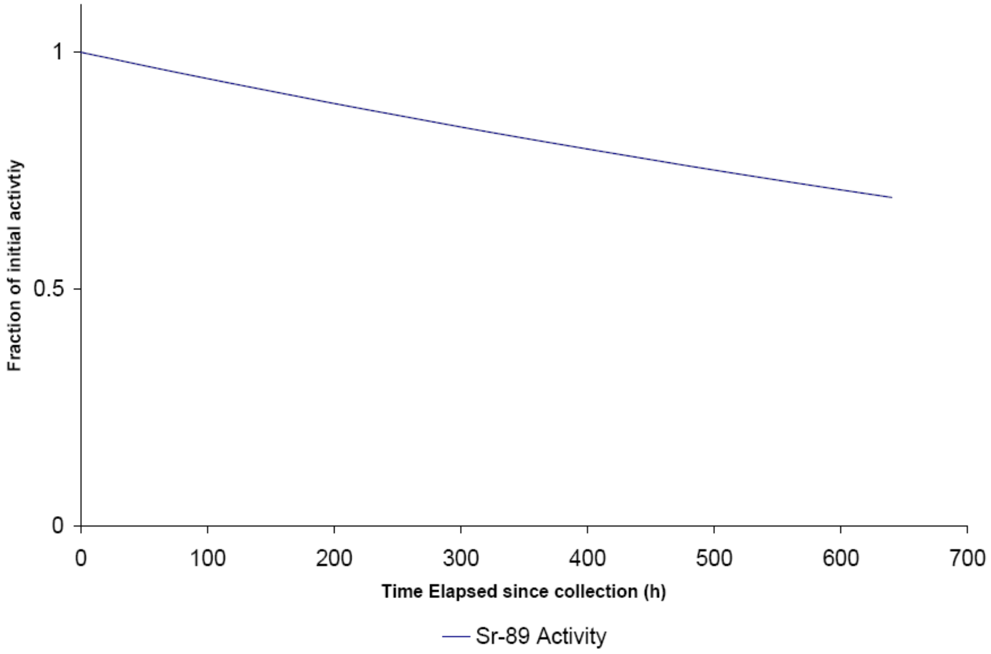


Total Beta Activity Ingrowth Factors for ^{90}Y in ^{90}Sr

Ingrowth time elapsed (hours)	0.25	2	4	12	24	48	72	96
Factor	0.003	0.021	0.042	0.122	0.229	0.405	0.541	0.646
Ingrowth time elapsed (hours)	144	192	240	320	400	480	560	640
Factor	0.790	0.875	0.926	0.969	0.987	0.994	0.998	0.999

Factor = (^{90}Y activity/^{90}Sr activity at zero hours of ingrowth)

Decay Curve for ^{89}Sr

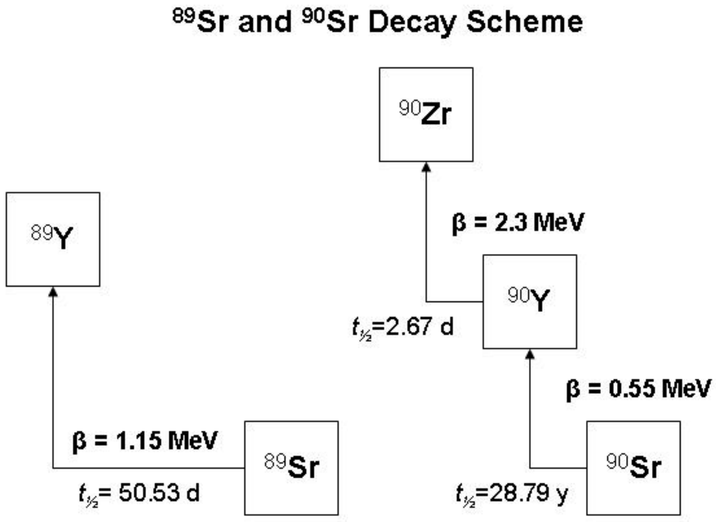


Decay Factors for ^{89}Sr

Decay time elapsed (hours)	0.25	2	4	12	24	48	72	96
Factor	1.000	0.999	0.998	0.993	0.986	0.973	0.960	0.947
Decay time elapsed (hours)	144	192	240	320	400	480	560	640
Factor	0.921	0.896	0.872	0.833	0.796	0.760	0.726	0.694

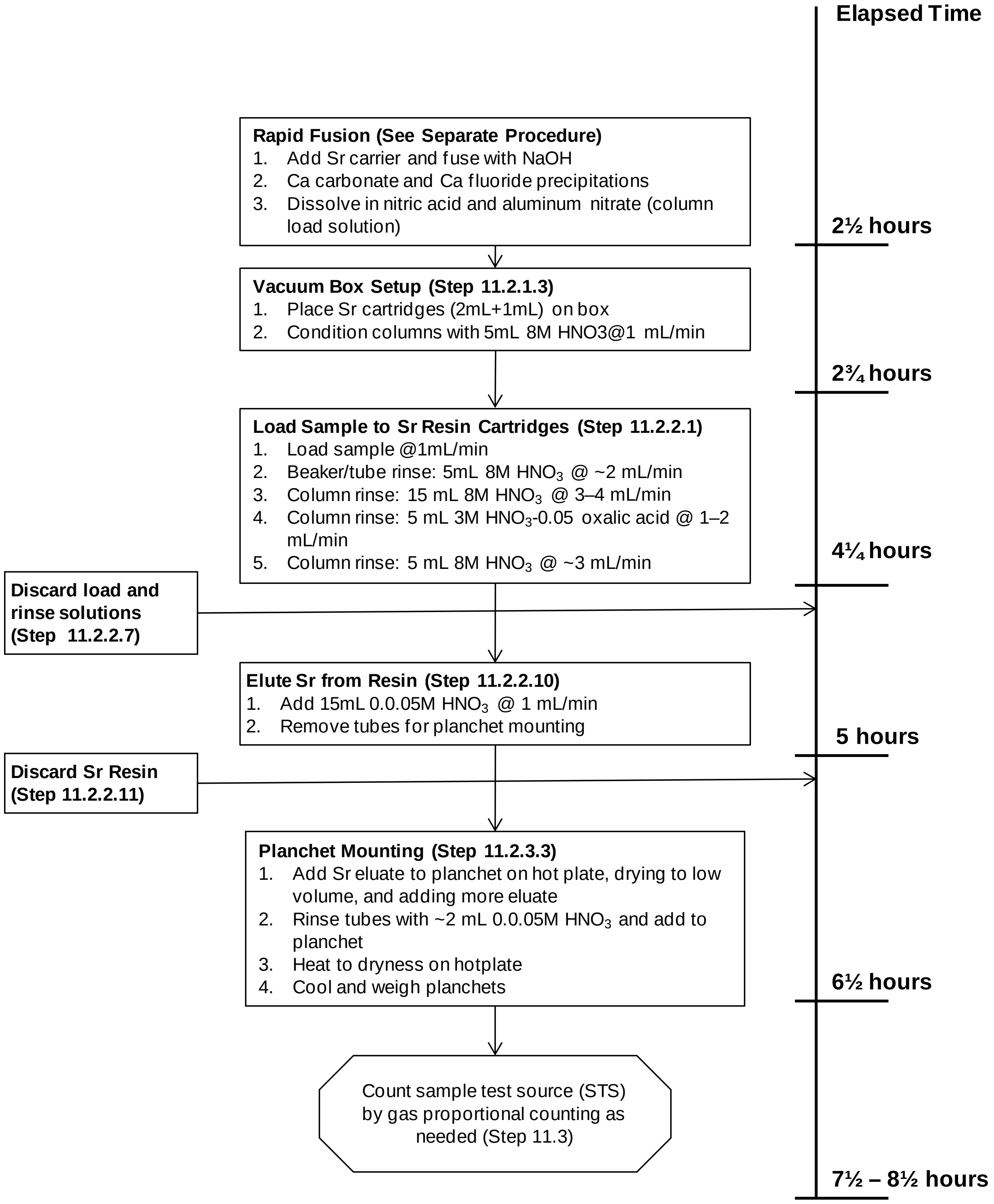
Factor = (^{89}Sr activity/^{89}Sr activity at zero hours of ingrowth)

17.4. Decay Schemes for ^{89}Sr and ^{90}Sr



17.5. Process Flow with Typical Processing Times

Separation Scheme and Timeline for Determination of Strontium Isotopes in Building Materials Samples



Appendix A:

Method and Calculations for Detector Calibration

A1. The effective detection efficiency for total radiostrontium (referenced to ^{90}Sr) is calculated as the weighted sum of the ^{90}Sr and ^{90}Y efficiencies that reflects the relative proportions of ^{90}Y and ^{90}Sr based on the ^{90}Y ingrowth after strontium separation.

NOTE: While ^{89}Sr efficiency calibration is not needed unless ^{89}Sr analysis will be performed, instructions for preparation are provided to support the two count approach should this option be desired.

A1.1. Due to the low mass of carrier used for this method, self-absorption effects may be assumed to be constant. Calibrate each detector used to count samples according to ASTM Standard Practice D7282, Section 16, "Single Point Efficiency or Constant Test Mass for a Specific Radionuclide" and the instructions below.

A1.2. Prepare a blank and at least three working calibration sources (WCS) for ^{90}Sr and ^{90}Y, and ^{89}Sr (if needed) as follows:

A1.2.1. The ^{90}Sr and ^{89}Sr radioactive standard solutions used to prepare WCSs shall be traceable to a national standards body such as NIST and shall originate from a standards supplier (or lot) different from standards used for calibration verification and batch quality controls. The standards should be diluted in nitric acid.

A1.2.2. The planchets used for the sources shall be of the same size, materials and type as those used for the analysis of STSs.

A1.2.3. Preparation of ^{89}Sr WCSs (if needed): ^{89}Sr standard solution (in 0.5M HNO_3) is evaporated to dryness in a stainless steel planchet as follows:

A1.2.3.1. For each ^{89}Sr WCS to be prepared, and for the associated blank, add strontium carrier to 15 mL of 0.05M HNO_3 in a disposable 50 mL centrifuge tube. The amount of carrier should be adjusted to approximate the amount expected to be recovered from routine samples.

NOTES:

If the average recovery has not been determined, the laboratory may assume 85% chemical yield for determining the amount of carrier to use in Step A1.2.3.1.

If the ^{89}Sr standard contains residual chloride, it will attack the surface of the planchet and compromise the quality of the calibration standard. In such cases, convert the aliquant of standard solution to a nitrate system by adding 1 mL concentrated HNO_3 and taking to dryness 2 times prior to quantitatively transferring the solution to the planchet.

A1.2.3.2. For each WCS, add a precisely known amount of traceable ^{89}Sr solution to a 50 mL centrifuge tube. Sufficient activity must be present at the point of the count to permit accumulation of greater than 10,000 net counts in a counting period deemed to be reasonable by the laboratory. The minimum activity used, however, should produce WCS count rates at least 20 times the background signal but not greater than 5000 counts per second (cps).

A1.2.3.3. Mix the solution and quantitatively transfer each WCS and the blank to respective clean stainless steel counting planchets using three rinses of 0.05M HNO_3.

A1.2.3.4. Evaporate to dryness using the same techniques used for sample test sources.

A1.2.3.5. For each detector to be calibrated, count three ^{89}Sr WCSs for sufficient time to accumulate at least 10,000 net counts.

A1.3. Preparation of ^{90}Sr and ^{90}Y WCSs: Separate WCSs for ^{90}Sr and ^{90}Y are prepared by chemically separating ^{90}Y from a standard solution of ^{90}Sr.

A1.3.1. For each ^{90}Sr WCS to be prepared, and for the associated blank, add 1 mL of 7 mg/mL strontium carrier to a disposable 50-mL centrifuge tube containing 10 mL 8M HNO_3. The amount of carrier added should correspond to that expected to be recovered from a routine sample.

NOTE: If the average recovery has not been determined, the laboratory may assume 85% chemical yield for determining the amount of carrier to use for Step A1.3.1. 3M HNO_3 may be used instead of 8M HNO_3, however Sr yields may be slightly less.

A1.3.2. For each ^{90}Sr WCS, add a precisely known amount of traceable ^{90}Sr solution to a 50-mL centrifuge tube containing 10 mL of 8M HNO_3. Sufficient activity should be present at the point of the count to permit accumulation of greater than 10,000 ^{90}Sr and 10,000 ^{90}Y net counts in the respective sources in a counting period deemed to be reasonable by the laboratory. The minimum activity used, however should produce WCS count rates at least 20 times the background signal but not greater than 5000 cps.

A1.3.3. Set up one (2+1 mL) Sr Resin column for each ^{90}Sr WCS and for the associated blank. Condition each column with 5 mL of 8M HNO_3. Column effluents are discarded to waste.

A1.3.4. Place a clean centrifuge tube under each column to catch all combined ^{90}Y effluents.

NOTE: Unless otherwise specified in the procedure, use a flow rate of ~1 mL/min for load and strip solutions and ~3 mL/min for rinse solutions.

A1.3.5. Load the ^{90}Sr solution onto the column at 1 drop/second or less (~1 mL/min). The load solution effluent containing ^{90}Y is retained.

A1.3.6. Rinse the centrifuge tube with three successive 2-mL portions of 8M HNO_3 adding each of the rinses to the column after the previous rinse has reached the upper surface of the resin. These effluents also contain ^{90}Y and are retained.

A1.3.7. Rinse the column with 10 mL of 8M HNO_3 and retain the column effluents containing ^{90}Y. Record the date and time that the final rinse solution leaves the column to the nearest 5 minutes as t_1, "Time of ^{90}Y Separation." Remove the centrifuge tube that has the combined ^{90}Y effluents. Place a clean tube under the column to catch the strontium eluate in subsequent steps.

NOTE: From this point, ^{90}Sr must be eluted, and the ^{90}Sr WCS must be prepared and counted as expeditiously as possible to minimize ^{90}Y ingrowth and necessary corrections to the efficiency. Counting of the ^{90}Sr WCS should be completed, if possible, within 3–5 hours but no longer than 10 hours from the time of ^{90}Y separation. If processing or counting capacity is limited, concentrate resources on ^{90}Sr WCS and counting first. The ^{90}Y WCS are not compromised by ingrowth but must only be counted promptly enough to minimize decay and optimize counting statistics.

A1.3.8. Strip strontium from each column by adding 15 mL of 0.05M HNO_3 to each column, catching the effluents containing ^{90}Sr in the centrifuge tube.

A1.3.9. Quantitatively transfer ^{90}Sr and ^{90}Y fractions to respective tared planchets using three portions of 0.05M HNO_3.

A1.3.10. Evaporate to dryness using the same techniques used for sample test sources, with the same heat time applied after dryness is reached.

NOTE: Gravimetric measurements may be performed following the counting to minimize elapsed time between separation and counting.

A1.4. Weigh the ^{90}Sr and ^{90}Y WCS sources and calculate the net residue mass.

A1.4.1. The net mass of the strontium nitrate precipitate shall indicate near quantitative yield of strontium of 95–103%. If strontium yield falls outside this range, determine and address the cause for the losses and repeat the process. The known activity of ^{90}Sr in the standard is corrected for losses based on the measured chemical yields of the strontium carrier.

Note that no correction shall be applied for values greater than 100% because this will produce a negative bias in the calibrated efficiency.

A1.4.2. The net residue mass of the ^{90}Y should be low. Higher residue mass may indicate the breakthrough of strontium and will result in high bias in the ^{90}Y efficiency, but it may simply be the result of corrosion of the stainless planchet during evaporation of 8M HNO_3, forming a small amount of iron oxide. Lower Sr carrier yields on the Sr planchet would indicate the present of Sr breakthrough.

NOTE: Formation of a small amount of iron oxide on the planchet during evaporation may result in a slight mass on the ^{90}Y planchet (~1 mg) but does not affect the ^{90}Y counting significantly.

A1.4.3. Count three ^{90}Sr WCS on each detector to be calibrated, for sufficient time to accumulate at least 10,000 net counts.

A1.4.4. Count three ^{90}Y WCS on each detector to be calibrated, for sufficient time to accumulate at least 10,000 net counts.

A1.4.5. Count the associated blanks as a gross contamination check on the process. If indications of contamination are noted, take appropriate corrective actions to minimize spread and prevent cross-contamination of other samples in the laboratory.

A1.5. Verify the calibration of each detector according to ASTM Standard Practice D7282, Section 16, and the laboratory quality manual and standard operating procedures.

A1.6. Calculations and data reduction for ^{90}Sr and ^{90}Y calibrations and calibration verifications are presented in Sections A2, A3, and A4. Calculations for total radiostrontium are in Section 12 of the method.

A2. Calculation of Detection Efficiency for ^{90}Sr

A2.1. Calculate the following decay and ingrowth factors for each WCS:

$$DF_{\mathrm{s}} = \mathrm{e}^{-\lambda_{\mathrm{Sr90}}(t_1 - t_0)} \tag{A1}$$

$$IF_{\mathrm{Y90}} = 1 - \mathrm{e}^{-\lambda_{\mathrm{Y90}}(t_2 - t_1)} \tag{A2}$$

where

DF_{s} = decay factor for decay of the ^{90}Sr standard from its reference date until the ^{90}Sr/^{90}Y separation

IF_{Y90} = ingrowth factor for ingrowth of ^{90}Y after the ^{90}Sr/^{90}Y separation

λ_{Sr90} = decay constant for ^{90}Sr, 7.642×10^{-10} s^{-1}

λ_{Y90} = decay constant for ^{90}Y, 3.005×10^{-6} s^{-1}

t_0 = reference date and time for the ^{90}Sr standard (s)

t_1 = date and time of the Sr/Y separation (s)

t_2 = date and time of the midpoint of the ^{90}Sr count (s)

NOTE: The elapsed time between the sample count and the reference date must be calculated using the same time units as the decay constant

A2.2. Calculate the ^{90}Sr detection efficiency for each WCS:

$$\varepsilon_{\mathrm{Sr90},i} = \frac{R_{s,i} - R_b}{AC_{\mathrm{Sr90\ std}} \times V_{\mathrm{s},i} \times DF_{\mathrm{s},i}} - IF_{\mathrm{Y90},i} \times \bar{\varepsilon}_{\mathrm{Y90}} = \frac{R_{n,i}}{AC_{\mathrm{Sr90\ std}} \times V_{\mathrm{s},i} \times DF_{\mathrm{s},i}} - IF_{\mathrm{Y90},i} \times \bar{\varepsilon}_{\mathrm{Y90}} \tag{A3}$$

where

$\varepsilon_{\mathrm{Sr90},i}$ = ^{90}Sr detection efficiency for the i^{th} WCS

$\bar{\varepsilon}_{\mathrm{Y90}}$ = average ^{90}Y detection efficiency (from Step A3.2)

$R_{\mathrm{s},i}$ = beta gross count rate for the i^{th} WCS (cpm)

R_{b} = background count rate, in cpm

$R_{\mathrm{n},i}$ = beta net count rate for the i^{th} WCS (cpm)

$AC_{\mathrm{Sr90\ std}}$ = activity concentration of the ^{90}Sr standard solution on its reference date (dpm/mL or dpm/g)

$V_{\mathrm{s,i}}$ = amount (volume or mass) of the standard solution added to the i^{th} WCS (mL or g)

A2.3. Average the efficiencies determined in Step A2.2 for all the WCSs to obtain the final detection efficiency for ^{90}Sr.

$$\varepsilon_{\mathrm{Sr90}} = \bar{\varepsilon}_{\mathrm{Sr90}} = \frac{1}{n}\sum_{i=1}^{n} \varepsilon_{\mathrm{Sr90},i} \tag{A4}$$

where

$\varepsilon_{\mathrm{Sr90},i}$ = ^{90}Sr detection efficiency determined for the i^{th} WCS in A2.2,
n = number of WCSs prepared and counted.

A2.4. Calculate the standard uncertainty of the average ^{90}Sr detection efficiency as follows:

$$u(\bar{\varepsilon}_{\mathrm{Sr90}}) = \sqrt{\left[\frac{1}{n^2}\sum_{i=1}^{n}\frac{u^2(R_{n,i}) + R_{n,i}^2 u_{\mathrm{r}}^2(V_{\mathrm{s},i})}{AC_{\mathrm{Sr90\,std}}^2 V_{\mathrm{s},i}^2 DF_{\mathrm{s},i}^2}\right] + \left(u^2(\bar{\varepsilon}_{\mathrm{Y90}}) - \bar{\varepsilon}_{\mathrm{Y90}}^2 u_{\mathrm{r}}^2(AC_{\mathrm{Sr90\,std}})\right)\overline{IF}_{\mathrm{Y90}}^{2} + \bar{\varepsilon}_{\mathrm{Sr90}}^2 u_{\mathrm{r}}^2(AC_{\mathrm{Sr90\,std}})}$$

(A5)

where

$$\overline{IF}_{\mathrm{Y90}} = \frac{1}{n}\sum_{i=1}^{n} IF_{\mathrm{Y90},i} = \text{average value of } {}^{90}\text{Y ingrowth factors} \qquad \text{(A6)}$$

and

$u(\cdot)$ = standard uncertainty of the value in parentheses,
$u_{\mathrm{r}}(\cdot)$ = relative standard uncertainty of the value in parentheses.

A3. Detection Efficiency for ^{90}Y

A3.1. Calculate the ^{90}Y detection efficiency, $\varepsilon_{\mathrm{Y90},i}$, for each WCS,

$$\varepsilon_{\mathrm{Y90},i} = \frac{R_{\mathrm{s},i} - R_{\mathrm{b}}}{AC_{\mathrm{Sr90\,std}} V_{\mathrm{s},i}\, DF_{\mathrm{s},i}} = \frac{R_{\mathrm{n},i}}{AC_{\mathrm{Sr90\,std}} V_{\mathrm{s},i}\, DF_{\mathrm{s},i}} \qquad \text{(A7)}$$

where

$$DF_{\mathrm{s},i} = \mathrm{e}^{-\lambda_{\mathrm{Sr90}}(t_1 - t_0)}\, \mathrm{e}^{-\lambda_{\mathrm{Y90}}(t_2 - t_1)} \qquad \text{(A8)}$$

and

$\varepsilon_{\mathrm{Y90},i}$ = ^{90}Y detection efficiency determined for the WCS
$R_{\mathrm{s},i}$ = beta gross count rate for the i^{th} WCS (cpm)
R_{b} = background count rate, in cpm
$R_{\mathrm{n},i}$ = beta net count rate for the i^{th} WCS (cpm)
$AC_{\mathrm{Sr90\,std}}$ = activity concentration of the ^{90}Sr standard solution on its reference date (disintegrations per minute per mL [dpm]/mL or dpm/g)
$V_{\mathrm{s},i}$ = amount of the standard solution added to the i^{th} WCS (mL or g)
$DF_{\mathrm{s},i}$ = combined correction factor for decay of the ^{90}Sr standard in the i^{th} WCS from its reference date until ^{90}Y separation, and for the decay of ^{90}Y from its separation until the midpoint of the count
λ_{Sr90} = decay constant for ^{90}Sr, 7.642×10^{-10} s^{-1}
λ_{Y90} = decay constant for ^{90}Y, 3.005×10^{-6} s^{-1}
t_0 = reference date and time for the ^{90}Sr standard (s)
t_1 = date and time of the ^{90}Y separation (s)
t_2 = date and time at the midpoint of the ^{90}Y count (s)

NOTE: The elapsed time between the sample count and the reference date must be calculated using the same time units as the decay constant

A3.2. Average the efficiencies determined in Step A3.1 to obtain the final detection efficiency for ^{90}Y.

$$\varepsilon_{\mathrm{Y90}} = \bar{\varepsilon}_{\mathrm{Y90}} = \frac{1}{n}\sum_{i=1}^{n} \varepsilon_{\mathrm{Y90},i} \qquad \text{(A9)}$$

where

$\varepsilon_{\mathrm{Y90},i}$ = ^{90}Y detection efficiency determined for the i^{th} WCS in Step A3.1
n = number of WCS prepared and counted

A3.3. The combined standard uncertainty of the average efficiency for ^{90}Y including uncertainty associated with the preparation of the calibration standards is calculated as follows:

$$u(\bar{\varepsilon}_{\mathrm{Y90}}) = \sqrt{\left[\frac{1}{n^2}\sum_{i=1}^{n}\frac{u^2(R_{n,i}) + R_{n,i}^2 u_{\mathrm{r}}^2(V_{\mathrm{s},i})}{AC_{\mathrm{Sr90\,std}}^2 V_{\mathrm{s},i}^2 DF_{\mathrm{s},i}^2}\right] + \bar{\varepsilon}_{\mathrm{Y90}}^2 u_{\mathrm{r}}^2(AC_{\mathrm{Sr90\,std}})} \qquad \text{(A10)}$$

where

$u(\cdot)$ = standard uncertainty of the value in parentheses,
$u_{\mathrm{r}}(\cdot)$ = relative standard uncertainty of the value in parentheses.

NOTE: The uncertainty of the net count rate, $u_{Rn,i}$, includes the uncertainty of the background or $u^2(R_{n,i}) = u^2(R_s) + u^2(R_b) = (R_s/t_s) + (R_b/t_b)$

A4. Calculate the covariance and correlation coefficient for the ^{90}Sr efficiency and the ^{90}Y efficiency:

$$u(\bar{\varepsilon}_{\mathrm{Sr90}}, \bar{\varepsilon}_{\mathrm{Y90}}) = \bar{\varepsilon}_{\mathrm{Sr90}}\bar{\varepsilon}_{\mathrm{Y90}} u_{\mathrm{r}}^2(AC_{\mathrm{Sr90\,std}}) - \left(u^2(\bar{\varepsilon}_{\mathrm{Y90}}) - \bar{\varepsilon}_{\mathrm{Y90}}^2 u_{\mathrm{r}}^2(AC_{\mathrm{Sr90\,std}})\right)\overline{IF}_{\mathrm{Y90}} \qquad \text{(A11)}$$

and

$$r(\bar{\varepsilon}_{\mathrm{Sr90}}, \bar{\varepsilon}_{\mathrm{Y90}}) = \frac{u(\bar{\varepsilon}_{\mathrm{Sr90}}, \bar{\varepsilon}_{\mathrm{Y90}})}{u(\bar{\varepsilon}_{\mathrm{Sr90}})\,u(\bar{\varepsilon}_{\mathrm{Y90}})} \qquad \text{(A12)}$$

where

u(·,·) = estimated covariance of the two quantities in parentheses,
r(·,·) = estimated correlation coefficient of the two quantities in parentheses,
u(·) = standard uncertainty of the quantity in parentheses,
u$u_r(\cdot)$ = relative standard uncertainty of the quantity in parentheses.

A5. Detection Efficiency for ^{89}Sr (if needed for Appendix B Calculations)

A5.1. Calculate the detection efficiency, $\varepsilon_{\mathrm{Sr89},i}$, for each WCS as follows:

$$\varepsilon_{\mathrm{Sr89},i} = \frac{R_{\mathrm{s},i} - R_{\mathrm{b}}}{AC_{\mathrm{Sr89\,std}}\,V_{\mathrm{s},i}\,DF_{\mathrm{s},i}} = \frac{R_{\mathrm{n},i}}{AC_{\mathrm{Sr89\,std}}\,V_{\mathrm{s},i}\,DF_{\mathrm{s},i}} \qquad \text{(A13)}$$

where

$$DF_{\mathrm{s},i} = \mathrm{e}^{-\lambda_{\mathrm{Sr89}}(t_1 - t_0)} \qquad \text{(A14)}$$

and

$\varepsilon_{\mathrm{Sr89},i}$ = ^{89}Sr detection efficiency for the i^{th} WCS

$R_{\mathrm{s},i}$ = beta gross count rate for the i^{th} WCS (cpm)
R_{b} = background count rate, in cpm
$AC_{\text{Sr89 std}}$ = activity concentration of the ^{89}Sr standard solution on the reference date (dpm/mL or dpm/g)
$V_{\mathrm{s},i}$ = amount (volume or mass) of the standard solution added to the i^{th} WCS (mL or g)
$DF_{\mathrm{s},i}$ = correction factor for decay of the ^{89}Sr standard for the i^{th} WCS from its reference date until the midpoint of the sample count
λ_{Sr89} = decay constant for ^{89}Sr, 1.587×10^{-7} s^{-1}
t_0 = reference date and time for the ^{89}Sr standard (s)
t_1 = date and time at the midpoint of the ^{89}Sr count (s)

A5.1.1. Average the efficiencies determined in Step A5.1 to obtain the final detection efficiency for ^{89}Sr.

$$\varepsilon_{\text{Sr89}} = \bar{\varepsilon}_{\text{Sr89}} = \frac{1}{n}\sum_{i=1}^{n} \varepsilon_{\text{Sr89},i} \qquad \text{(A15)}$$

where

$\varepsilon_{\text{Sr89},i}$ = ^{89}Sr detection efficiency determined for the i^{th} WCS in Step A5.1,
n = number of WCSs prepared and counted.

A5.1.2. The combined standard uncertainty of the average efficiency for ^{89}Sr including uncertainty associated with the preparation of the calibration standards is calculated as follows:

$$u(\bar{\varepsilon}_{\text{Sr89}}) = \sqrt{\left[\frac{1}{n^2}\sum_{i=1}^{n}\frac{u^2(R_{n,i}) + R_{n,i}^2 u_{\mathrm{r}}^2(V_{\mathrm{s},i})}{AC_{\text{Sr89 std}}^2 V_{\mathrm{s},i}^2 DF_{\mathrm{s},i}^2}\right] + \bar{\varepsilon}_{\text{Sr89}}^2 u_{\mathrm{r}}^2(AC_{\text{Sr89 std}})} \qquad \text{(A16)}$$

where

$u(\cdot)$ = standard uncertainty of the value in parentheses,
$u_{\mathrm{r}}(\cdot)$ = relative standard uncertainty of the value in parentheses.

Appendix B:

Calculations for Isotopic ^{89}Sr and ^{90}Sr Results

A numerical approach for determining ^{89}Sr and ^{90}Sr activity from a single sample is performed by a number of laboratories. This presentation, however, allows a more rigorous evaluation of uncertainties than commonly employed. Lacking this treatment, many labs have found that the traditional approach (evaluating counting uncertainty for a single count only) has led to overestimation of the quality of results, and to poor decisions regarding the presence or absence of low activities of one radioisotope of strontium in the presence of elevated activities of the second.

These calculations may be valuable to laboratories who wish to determine isotopic ^{89}Sr and ^{90}Sr in a large number of samples with a minimum of additional effort beyond the initial preparation and counting of total radiostrontium. Specifically, it involves performing a second count of the same radiostrontium sample test source (STS) and mathematically resolving the activity of the two isotopes. Although the STS may be recounted as soon as 1–2 days after the initial count, resolution is optimized if the two counts span as large a range of the ^{90}Y ingrowth as practicable. The time elapsed between the chemical separation and the first count should be minimized, while the second count should optimally proceed as ^{90}Y approaches secular equilibrium with ^{90}Sr but before significant decay of ^{89}Sr has occurred, for example, after 3–5 half-lives of ^{90}Y have elapsed (1–2 weeks).

This section may not be employed without complete validation of the approach by the laboratory, including testing with samples containing ratios of ^{90}Sr relative to ^{89}Sr varying from pure ^{90}Sr to pure ^{89}Sr.

B1. The equations in this section are used to calculate the ^{90}Sr and ^{89}Sr activity of a sample from data generated from two successive counts of the same radiostrontium sample test source.

B1.1. For each of the two counting measurements (i = 1, 2), calculate the following decay and ingrowth factors:

$$DF_{\mathrm{Sr89},i} = \mathrm{e}^{-\lambda_{\mathrm{Sr89}}(t_i - t_0)} \tag{B1}$$

$$DF_{\mathrm{Sr90},i} = \mathrm{e}^{-\lambda_{\mathrm{Sr90}}(t_i - t_0)} \tag{B2}$$

$$F_{\mathrm{Y90},i} = \mathrm{e}^{-\lambda_{\mathrm{Sr90}}(t_{\mathrm{sep}} - t_0)}\left(1 - \mathrm{e}^{-\lambda_{\mathrm{Y90}}(t_i - t_{\mathrm{sep}})}\right) \tag{B3}$$

where:

$DF_{\mathrm{Sr89},i}$ = decay factor for decay of ^{89}Sr from the collection date to the midpoint of the i^{th} count of the STS

$DF_{\mathrm{Sr90},i}$ = decay factor for decay of ^{90}Sr from the collection date to the midpoint of the i^{th} count of the STS

$F_{\mathrm{Sr90},i}$ = combined decay and ingrowth factor for decay of ^{90}Sr from the collection date to the Sr/Y separation and ingrowth of ^{90}Y from the separation to the midpoint of the i^{th} count of the STS

λ_{Sr89} = decay constant for ^{89}Sr = $1.587\times10^{-7}\ \mathrm{s}^{-1}$
λ_{Sr90} = decay constant for ^{90}Sr = $7.642\times10^{-10}\ \mathrm{s}^{-1}$
t_0 = collection date and time for the sample (s)
t_{sep} = date and time of the Sr/Y separation (s)
t_i = date and time of the midpoint of the i^{th} count of the STS (s)

NOTE: The elapsed time between the sample count and the reference date must be calculated using the same time units as the decay constant

B1.2. For i = 1, 2, use the results from Section A5.1 in Appendix A to calculate the following sensitivity factors:

$$a_i = DF_{\mathrm{Sr89},i}\varepsilon_{\mathrm{Sr89},i} \tag{B4}$$

$$b_i = DF_{\mathrm{Sr90},i}\varepsilon_{\mathrm{Sr90},i} + F_{\mathrm{Y90},i}\varepsilon_{\mathrm{Y90},i} \tag{B5}$$

where

a_i = sensitivity of the count rate in the i^{th} measurement to ^{89}Sr activity
b_i = sensitivity of the count rate in the i^{th} measurement to ^{90}Sr activity
$\varepsilon_{\mathrm{Y90},i}$ = ^{90}Y efficiency of the detector for the i^{th} count of the STS
$\varepsilon_{\mathrm{Sr90},i}$ = ^{90}Sr efficiency of the detector for the i^{th} count of the STS

B1.3. Calculate the standard uncertainties of the sensitivity factors using the equations:

$$u(a_i) = DF_{\mathrm{Sr89},i}\, u(\varepsilon_{\mathrm{Sr89},i}) \tag{B6}$$

$$u(b_i) = \sqrt{DF^2_{\mathrm{Sr90},i}\, u^2(\varepsilon_{\mathrm{Sr90},i}) + F^2_{\mathrm{Y90},i}\, u^2(\varepsilon_{\mathrm{Y90},i}) + 2DF_{\mathrm{Sr90},i}F_{\mathrm{Y90},i}\, u(\varepsilon_{\mathrm{Sr90},i},\varepsilon_{\mathrm{Y90},i})} \tag{B7}$$

where the estimated covariance of the ^{90}Sr and ^{90}Y efficiencies is calculated as follows:

$$u(\varepsilon_{\mathrm{Sr90},i},\varepsilon_{\mathrm{Y90},i}) = r(\varepsilon_{\mathrm{Sr90},i},\varepsilon_{\mathrm{Y90},i})\,u(\varepsilon_{\mathrm{Sr90},i})\,u(\varepsilon_{\mathrm{Y90},i}) \tag{B8}$$

and where the estimated correlation coefficient $r(\varepsilon_{\mathrm{Sr90},i}, \varepsilon_{\mathrm{Y90},i})$ was determined during the calibration.

B1.4. Calculate the covariances $u(a_1,a_2)$ and $u(b_1,b_2)$ as follows:

$$u(a_1,a_2) = \begin{cases} u(a_1)\,u(a_2), & \text{if only one detector is used} \\ a_1 a_2\, u_{\mathrm{r}}^2(AC_{\mathrm{Sr89\ std}}), & \text{if two detectors are used} \end{cases} \tag{B9}$$

$$u(b_1,b_2)=\begin{cases}(DF_{\mathrm{Sr90,1}}F_{\mathrm{Y90,2}}+DF_{\mathrm{Sr90,2}}F_{\mathrm{Y\text{-}90,1}})\,u(\varepsilon_{\mathrm{Sr90,1}},\varepsilon_{\mathrm{Y90,1}}) \\ \quad +DF_{\mathrm{Sr90,1}}DF_{\mathrm{Sr90,2}}\,u^2(\varepsilon_{\mathrm{Sr90,1}})+F_{\mathrm{Y90,1}}F_{\mathrm{Y90,2}}\,u^2(\varepsilon_{\mathrm{Y90,1}}), & \text{using only one detector} \\ b_1b_2\,u_{\mathrm{r}}^2(AC_{\mathrm{Sr90\ std}}), & \text{using two detectors}\end{cases} \tag{B10}$$

where

$AC_{\mathrm{Sr89\ std}}$ = activity concentration of the ^{89}Sr standard used for calibration
$AC_{\mathrm{Sr90\ std}}$ = activity concentration of the ^{90}Sr standard used for calibration
$u_{\mathrm{r}}(\cdot)$ = relative standard uncertainty of the quantity in parentheses

B1.5. For i = 1, 2, calculate the net beta count rates, $R_{\mathrm{n},i}$, and their standard uncertainties:

$$R_{\mathrm{n},i}=R_{\mathrm{a},i}-R_{\mathrm{b},i} \tag{B11}$$

$$u(R_{\mathrm{n},i})=\sqrt{\frac{R_{\mathrm{a},i}}{t_{\mathrm{a},i}}+\frac{R_{\mathrm{b},i}}{t_{\mathrm{b},i}}} \tag{B12}$$

where:

$R_{\mathrm{n},i}$ = net beta count rate for the i^{th} count of the STS (cpm)
$R_{\mathrm{a},i}$ = beta gross count rate for the i^{th} count of the STS (cpm)
$R_{\mathrm{b},i}$ = beta background count rate for the i^{th} count of the STS (cpm)
$t_{\mathrm{a},i}$ = sample count time for the i^{th} count of the STS (min)
$t_{\mathrm{b},i}$ = background count time for the i^{th} count of the STS (min)

B1.6. Using the values calculated in A5.1 – A5.5, calculate the ^{89}Sr and ^{90}Sr activity concentrations:

$$AC_{\mathrm{Sr89}}=\frac{b_2R_{\mathrm{n,1}}-b_1R_{\mathrm{n,2}}}{2.22\times X\times V\times Y} \tag{B13}$$

$$AC_{\mathrm{Sr90}}=\frac{a_1R_{\mathrm{n,2}}-a_2R_{\mathrm{n,1}}}{2.22\times X\times V\times Y} \tag{B14}$$

where:

$$X=a_1b_2-a_2b_1 \tag{B15}$$

and where:

2.22 = conversion factor from dpm to pCi
Y = chemical yield for strontium
V = sample weight (g)

B2. The standard counting uncertainties for ^{89}Sr ($u_{\mathrm{cC}}(AC_{\mathrm{Sr89}})$) and ^{90}Sr ($u_{\mathrm{cC}}(AC_{\mathrm{Sr90}})$) are calculated in units of pCi/g as follows:

$$u_{\mathrm{cC}}(AC_{\mathrm{Sr89}})=\frac{\sqrt{b_2^2u^2(R_{\mathrm{n,1}})+b_1^2u^2(R_{\mathrm{n,2}})}}{2.22\times X\times W\times Y} \tag{B16}$$

$$u_{\mathrm{cC}}(AC_{\mathrm{Sr90}}) = \frac{\sqrt{a_1^2 u^2(R_{\mathrm{n},2}) + a_2^2 u^2(R_{\mathrm{n},1})}}{2.22 \times X \times W \times Y} \tag{B17}$$

B3. The combined standard uncertainties (CSU) for ^{89}Sr and ^{90}Sr are calculated as follows:

$$\begin{aligned} u_{\mathrm{c}}(AC_{\mathrm{Sr89}}) = \Bigg[& u_{\mathrm{cC}}^2(AC_{\mathrm{Sr89}}) \\ & + AC_{\mathrm{Sr89}}^2 \left(\frac{u^2(W)}{W^2} + \frac{u^2(Y)}{Y^2} + \frac{b_2^2 u^2(a_1) + b_1^2 u^2(a_2) - 2b_1 b_2\, u(a_1, a_2)}{X^2} \right) \\ & + AC_{\mathrm{Sr90}}^2 \, \frac{b_2^2 u^2(b_1) + b_1^2 u^2(b_2) - 2b_1 b_2\, u(b_1, b_2)}{X^2} \Bigg]^{1/2} \end{aligned} \tag{B18}$$

Attachment IV:

Composition of Brick for Spiking in this Study

Metals by ICP-AES [4]	Concentration (ppm) [1]
Silicon Dioxide	721,700
Aluminum	78,700
Barium	400
Calcium	1,600
Iron	40,000
Magnesium	4,600
Potassium	15,300
Sodium	1,500
Titanium	4,400
Manganese	600
Strontium	100
Uranium	<30
Thorium	<30
Non-Metals	
Chloride	—
Sulfur	5,600
Phosphorus	1,500
Radionuclide	**Concentration (pCi/g) [2, 3]**
Uranium 238, 234	1.054 ± 0.020, 1.102 ± 0.021
Plutonium 239/240	-0.0003 ± 0.0041
Americium 241	0.048 ± 0.039
Strontium 90	0.119 ± 0.077
Radium 226	1.025 ± 0.027

NOTE: Analyses conducted by an independent laboratory.

[1] Values below the reporting level are presented as less than (<) values. No measurement uncertainty was reported with the elemental analysis values. Parts per million (ppm).

[2] Reported values represent the average value of seven blank samples analyzed except for ^{226}Ra and U by NAREL. Ten blank brick samples were analyzed for ^{226}Ra. Sixteen blank brick samples were analyzed for the uranium isotopes.

[3] Reported uncertainty is the standard deviation of the results (k=1).

[4] ICP-AES=Inductively Coupled Plasma – Atomic Emission Spectrometry