March 2019 Update: EPA has validated and published a rapid method for sodium hydroxide fusion of limestone matrices prior to americium, plutonium, strontium, radium and uranium analyses. The method is summarized and accessible through the link provided below.

Rapid Method for Sodium Hydroxide Fusion of Limestone Matrices Prior to Americium, Plutonium, Strontium, Radium, and Uranium Analyses for Environmental Remediation Following Radiological Incidents

Analyte(s)	CAS RN®
Americium-241	14596-10-2
Plutonium-238	13981-16-3
Plutonium-239	15117-48-3
Radium-226	13982-63-3
Strontium-90	10098-97-2
Uranium-234	13966-29-5
Uranium-235	15117-96-1
Uranium-238	7440-61-1

Analysis Purpose: Sample Preparation
Technique: Alpha spectrometry

Method Developed for: Americium-241 (^{241}Am), plutonium-238 (^{238}Pu), plutonium-239 (^{239}Pu), radium-226 (^{226}Ra), strontium-90 (^{90}Sr), uranium-234 (^{234}U), uranium-235 (^{235}U) and uranium-238 (^{238}U) in limestone samples.

Method Selected for: *Selected Analytical Methods for Environmental Remediation and Recovery* (SAM) now lists this method for qualitative analysis of americium-241 (^{241}Am), plutonium-238 (^{238}Pu), plutonium-239 (^{239}Pu), radium-226 (^{226}Ra), strontium-90 (^{90}Sr), uranium-234 (^{234}U), uranium-235 (^{235}U) and uranium-238 (^{238}U) in limestone samples.

Description of Method: This is a general method for limestone samples collected following a radiological or nuclear incident. The limestone samples may be received as core samples, crushed samples, or in pieces of various sizes. Limestone samples should be crushed and pulverized (milled) to achieve a particle size small enough that representative subsamples can be taken and representative aliquants analyzed. After a homogeneous subsample is obtained, the limestone sample aliquant is taken and fused to digest the limestone sample matrix, and matrix removal steps are employed to collect and preconcentrate the radionuclides from the alkaline fusion matrix. The method is based on the rapid dissolution of a representative, finely milled aliquants of approximately 1 g of limestone using sodium hydroxide fusion at 600 °C. Plutonium (Pu), uranium (U) and americium (Am) are separated from the alkaline matrix using an iron hydroxide/titanium hydroxide precipitation followed by a lanthanum fluoride matrix removal step. Strontium (Sr) is separated from the alkaline matrix using a phosphate precipitation followed by a calcium fluoride precipitation to remove silicates. Radium (Ra) is separated from the alkaline matrix using a carbonate precipitation. This method is a sample digestion and pretreatment technique to be used prior to the following separation and analysis methods:

- U.S. EPA. *Rapid Radiochemical Method for Americium-241 in Building Materials for Environmental Remediation Following Radiological Incidents* (EPA 402-R14-007, April 2014)
- U.S. EPA. *Rapid Radiochemical Method for Plutonium-238 and Plutonium-239/240 in Building Materials for Environmental Remediation Following Radiological Incidents* (EPA 402-R14-006, April 2014)
- U.S. EPA. *Improved Rapid Radiochemical Method for Radium-226 in Building Materials for Environmental Remediation Following Radiological Incidents* (Revision 1, EPA 402-S17-002, May 2017)
- U.S. EPA. *Rapid Radiochemical Method for Total Radiostrontium (Sr-90) in Building Materials for Environmental Remediation Following Radiological Incidents* (EPA 402-R14-00, April 2014)
- U.S. EPA. *Rapid Radiochemical Method for Isotopic Uranium in Building Materials for Environmental Remediation Following Radiological Incidents* (EPA 402-R14-005, April 2014)

Special Considerations:

Limestone samples with larger particle size may require a longer fusion time. Samples with elevated activity or samples that require multiple analyses from a single aliquant 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.

Information regarding the elemental composition of the sample may be helpful. For example, limestone may have native concentrations of uranium, radium, thorium (Th), strontium or barium (Ba), any of which may have an effect on the chemical separations used following the fusion of the sample. In some cases (e.g., strontium analysis), elemental analysis of the digestate prior to chemical separations may be necessary to determine native concentrations of carrier elements present in the sample. The amount of stable strontium added to the limestone samples is designed to minimize the impact from native stable strontium.

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 process, and as potential monitors for cross contamination during batch processing.

Uncontaminated limestone material may be acceptable blank material for Pu, Am and Sr analyses, but this material will contain background levels of naturally occurring U and Ra isotopes.

Additional information regarding potential interferences and procedures for addressing the interferences is provided in Section 4 of the sodium hydroxide fusion method listed below.

Source: U.S. EPA, National Air and Radiation Environmental Laboratory. "*Rapid Method for Sodium Hydroxide Fusion of Limestone Matrices Prior to Americium, Plutonium, Strontium, Radium, and Uranium Analyses for Environmental Remediation Following Radiological Incidents"* (EPA 402-R-18- 002, August 2018) https://www.epa.gov/sites/production/files/2018-08/documents/limestone_method_final.pdf