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RADIOACTIVE AIR EFFLUENT EMISSION MEASUREMENTS AT TWO RESEARCH REACTORS *

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ABSTRACT

Sandia National Laboratories operates two reactors which fall under U. S. Environmental Protection Agency regulations for emission of radionuclides to the ambient air. These reactors are: 1) the Annular Core Research Reactor, a pool-type reactor and 2) the Sandia Pulsed Reactor III, a Godiva-type reactor. The annual radioactive air emissions from these two reactors had been estimated based on engineering calculations and used in the facility Safety Analysis Report. The calculated release rates had never been confirmed through measurements. The purpose of this work was to obtain confirmatory radioactive gas and aerosol concentration measurements for radionuclides in exhaust stacks of these reactors during normal operation; however, the measured production rate of argon-41 was significantly different from the engineering calculations for both reactors. The resolution of this difference is discussed.

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DISCLAIMER

1. Background Information

1.1 National Emission Standards for Hazardous Air Pollutants (NESHAP) Requirements

The U. S. Environmental Protection Agency (EPA) regulations 40 CFR 61, Subpart H[1] require that emissions of radionuclides to the ambient air from U. S. Department of Energy (DOE) facilities shall not exceed those amounts that would cause any member of the public to receive in any year an effective dose equivalent (EDE) of 10 millirem/year (mrem/yr). Continuous stack monitoring is required for sources contributing greater than 0.1 mrem/yr to the nearest off site public receptor, and periodic confirmatory monitoring is required for sources contributing less than 0.1 mrem/yr. Sandia National Laboratories, operates two research reactors for the DOE, the Annular Core Research Reactor (ACRR) and the Sandia Pulsed Reactor (SPR III). The annual radioactive air emissions from these two reactors had been estimated based on engineering calculations presented in the facility Safety Analysis Report (SAR).[2,3] The calculated release rates had never been confirmed through measurements.

1.2 Description of Facilities

The ACRR is 2-megawatt (MW) pool-type reactor capable of pulse, steady-state, and tailored transient rod withdrawal operations. The annular-shaped core is designed in a close-packed lattice of BeO-UO_2 fuel elements. Their longitudinal axes are aligned vertically and arranged in a hexagonal grid around the central cavity. The complete core assembly consists of fuel elements, six fuel-followed control rods, two fuel-followed safety rods, and three void-followed transient rods. The number of fuel elements in the core depends on experiment requirements and is normally 200–250. The facility provides a large central irradiation cavity (23 centimeters (cm) diameter), a neutron radiography facility, and two large interchangeable fuel ringed external cavities (FREC I and II) of 38 and 51 cm diameter respectively. The ACRR is authorized to operate at a maximum reactor production rate of 1.06×10^7 megajoules (MJ) per year; however, the average operation over eight years has been 3.4×10^5 MJ. The primary radionuclide effluent generated by the operation of the ACRR is argon-41 (^{41}Ar) produced by the neutron activation of stable argon-40 (^{40}Ar), a normal trace constituent of the air present in the central cavity.

The SPR III is a Godiva-type reactor that operates in a fast-burst mode; however, steady-state operations are also common. The reactor uses an unmoderated cylindrical assembly of bare, solid uranium metal enriched to 93 percent (%) uranium-235, with 10% molybdenum by weight to ensure phase stabilization of the fuel material. The total mass of the core is about 258 kilograms (kg). The SPR III is housed in a hemispherical, dome-shaped building, often referred to as the Kiva. The walls are constructed of steel-reinforced concrete, 122 cm thick. Outside diameter of the building is 12 meters (m). The internal volume of the

Kiva is about 210 m^3. The DOE has authorized the SPR III to operate at a maximum annual production rate of 2.08×10^4 MJ/yr. The average production rate over the last eight years has been 2.1×10^3 MJ. Similar to the ACRR, ^{41}Ar is the primary effluent produced by neutron activation of the stable, inert ^{40}Ar within the Kiva.

Information on the release parameters for each reactor ventilation stack is listed in Table 1.

Table 1. Release Parameters for ACRR and SPR III.

Facility	Stack Height (m)	Diameter at top (m)	Flow Velocity (m/s)	Exit Temp. (°C)	Volumetric Flow Rate (m^3/s)
ACRR	16.5	0.2	11.2	21	0.35
SPR III	8.2	0.5	17.8	21	3.5

2. Methodology

The assumption was made that ^{41}Ar was the major contributor to the annual dose, greater than 90% of the EDE to the nearest off site receptor. Based on this assumption the methodology was developed to confirm the activity concentration of ^{41}Ar in the effluent of both facilities and confirm the production rate of ^{41}Ar in curies/megajoule (Ci/MJ). Confirmatory measurements for ^{41}Ar releases at the ACRR and SPR III reactors were performed by capturing and compressing a volume of air (100 pounds per square inch, gauge) taken from the stack into a stainless steel cylinder. Activity of the captured gas was determined by gamma spectral analysis using a hyper-pure germanium gamma (HPGe) spectrometer. Triplicate air samples were taken at each facility. The ACRR stack was sampled during operations at 2 MW, 1 MW and 500 kilowatts (kW). The SPR III stack was sampled during steady-state operations of 10 kW.

Although it was assumed that the particulate contribution to the total dose would be insignificant, less than 10% of EDE, a methodology was developed to quantify the particulate contribution. Particulate samples were extracted from the air stream using a 47-millimeter diameter in-line glass fiber filter on a continuous air monitor (CAM), Eberline Alpha 6 CAM. Filter samples were taken upstream and downstream of the HEPA filters at the ACRR during a 2-MW steady-state operation and upstream of the HEPA filter at the SPR III during both pulsed and steady-state operation.[4] A gas flow proportional counter was used to analyze the filter samples for gross alpha and beta activity. Gamma spectral analysis was performed using a HPGe detector. Activity concentration was normalized to megajoules of output during the sampling operation.

3. Measurement System Description

The compressed air samples were analyzed using a 19% efficient, high-resolution HPGe gamma-ray spectrometer in a close geometry (5 cm). The stainless steel gas cylinders were placed horizontally, so the vertically facing detector saw the center of the cylinder side wall. A 4-inch lead shield surrounds the detector and sample on five sides. Data collection was performed with a Canberra Series-90 multichannel analyzer system with 8192 energy channels. Samples were counted for a nominal data collection time of 1800 seconds. Analysis was performed using the vendor-provided software.[5] The code includes background subtraction, peak identification, and quantitative nuclide analysis.

Calibration of the measurement system was accomplished by counting a simulated gas standard in an identical geometry. The simulated gas standard consisted of a 1-liter stainless steel sample bottle, filled with a solid matrix solid foam (density 0.018 grams (g)/cm^3) spiked with a mixed gamma standard. A gamma-ray efficiency curve was generated over the energy range of 88–1836 kilo electron volts (keV), based on analysis identical to that used for the actual sample counting. Uncertainties for the gamma-ray standard are nominally 5% for each of the calibration peaks.[6]

The filter samples were analyzed on a Tennelec gas flow proportional counter with a 80-microgram (μg)/cm^2 mylar window. Plutonium-238 and strontium/yttrium-90 traceable standards were used to perform an efficiency calibration. The system efficiency for alpha and beta particulate was determined to be 36% and 54% respectively. The data was collected and analyzed using vendor-provided software.[7]

3.1 ACRR Sampling Results

The measured activity concentration of ^{41}Ar in the air sample extracted from the ACRR stack was 6.37 ± 0.26 x 10^{-4}, 3.26 ± 0.12 x 10^{-4}, and 1.76 ± 0.03 x10^{-4} microcuries/milliliter (μCi/mL), at 2.0 MW, 1.0 MW and 500 kW respectively. This corresponds to a production rate of 1.09 x 10^{-4} Ci/MJ at 2.0 MW. The gross alpha and beta analyses of the filters showed activity concentrations on the particulate samples slightly above the minimum detectable concentration for both the upstream and downstream samples. Gamma spectral analysis of the particulate samples showed no gamma emitting nuclides above the minimum detectable concentration. [8]

The yearly emission of ^{41}Ar from the ACRR stack was calculated to be 3.72 x 10^7 μCi /yr, using a production rate of 1.09 x 10^{-4} Ci/MJ and average yearly output of the reactor. The annual EDE to the nearest receptor was calculated using the CAP88-PC air dispersion code.[9] The calculations show the EDE to be 1.60 x 10^{-2} mrem/yr. This is below the threshold for the continuous air monitoring requirement set by EPA. The analysis confirmed that the dose due to particulates was insignificant, less than 10% of the EDE. Therefore, there is no requirement to monitor the particulate emissions.

While the measured production rate for ^{41}Ar met EPA EDE requirements, it was higher than the previous values that had been calculated for the SAR. This difference will be addressed in Section 4.

3.2 SPR III Sampling Results

The measured ^{41}Ar concentration in the air extracted from the SPR III stack was $3.6 \pm 0.18 \times 10^{-6}$ millicuries (mCi)/mL. This corresponds to a production of 1.26×10^{-3} Ci/MJ. The yearly emission of ^{41}Ar from the SPR III was calculated to be 2.59×10^{6} μCi/yr, using a production rate of 1.26×10^{-3} Ci/MJ and average yearly output of the reactor. The annual EDE to the nearest receptor was calculated using the CAP88-PC air dispersion code. The calculations show the EDE to be 3.2×10^{-4} mrem/yr. This EDE is also below the threshold for continuous air monitoring .[8]

The results of the analyses of the particulate samples from the SPR III stack show that fission products generated during operation reach the stack in measurable concentrations. The production rate (μCi/MJ) for these particulates was higher during the steady-state operation as compared to the pulsed operations. The pulsed operations were normalized to MJ for comparison purposes. The source term resulting from these measurements was used in CAP88-PC to determine the EDE to the nearest receptor. The particulate emission from the SPR III contributes less than 10% of the calculated dose; therefore, particulates are considered insignificant and there is no requirement to monitor the particulate emissions.

The measured ^{41}Ar production rates in the stack of the SPR III were higher than the previous estimates stated in the SAR, 1.26×10^{-3} Ci/MJ versus 2.94×10^{-4} Ci/MJ. This variance will be discussed in Section 4.

4. Resolution of the Discrepancies

The variation between the measured production rate and the engineering calculations in the SAR for each facility was greater than expected and the cause was investigated. Initially, all the quality assurance, sampling methods, and analytical procedures were reviewed, and it was determined that these were not contributing factors. Historical information involving the measurement and calculations on production rates for each facility was reviewed.

4.1 ACRR

The ACRR SAR[2] estimated the ^{41}Ar production to be 6.1×10^{-6} Ci/MJ, compared to the measured value of 1.09×10^{-5} Ci/MJ. There had been three previous calculations and/or experiments performed to estimate the production rate in the central cavity in

addition to the confirmatory measurements. Each of these used different parameters. 1) The calculated value in the SAR, 6.1 x 10^{-6} Ci/MJ, used the thermal neutron flux in the central cavity and thermal cross section for 2-MW steady-state operation. 2) Calculations using the MCNP Monte Carlo code estimated the production rate to be 4.09 x 10^{-5} Ci/MJ.[10] 3) An experiment performed in 1987 in which 250 mL of stable argon was irradiated in the central cavity estimated the production rate to be 1.9 x 10^{-5} Ci/MJ.[11] The samples extracted from the stack and analyzed during the confirmatory measurements included the production of ^{41}Ar in the central cavity along with the contributions from the FREC II external cavity and the radiography unit. The additional volume of activated air would account for a higher production rate than the previous calculations and experiment.

4.2 SPR III

The production rate of ^{41}Ar calculated from the confirmatory measurements was 1.26 x 10^{-3} Ci/MJ compared to 2.94 x 10^{-4} Ci/MJ, the value calculated for the SPR III SAR.[3] The production rate calculated in the SAR used only the thermal cross section and did not account for the change in the neutron spectrum at varying distances from the reactor, which is located in the center of the Kiva. More recent calculations estimated the ^{41}Ar production rate to be 5.03 x 10^{-4} Ci/MJ. These estimates were based on data using nickel and sulfur activation foils for neutrons, thermoluminescent dosimeters for gamma dose, and current cross section data.[12] The difference between the more recent calculations and the measured data is within the range of expected differences.

5. Conclusions

The measurements of the source term at each facility confirmed that the EDE to the nearest receptor was less than 0.1 mrem/year. Therefore, neither facility is required to have continuous air monitoring under current operating conditions. The facilities are required to make periodic measurements to confirm that the production rate at the facilities and the source term, ^{41}Ar, has not changed. The methodology of using compressed gas samples to meet the NESHAP confirmatory measurement requirements was an efficient and reliable method. That method will be easy to replicate for the required periodic measurements. The variations in measured to predicted values of the production rates was to be expected when the assumptions and parameters applied in establishing the predicted rates are examined. However, caution needs to be taken when comparing values in existing documentation to insure that the same operating conditions exist and the parameters used to estimate the values are the same.

6. References

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