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INTERIM REPORT

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Progress Report on: "Characterization of Radionuclide Contamination Throughout Light Water Reactor Power Stations," April 11, 1980

To: Dr. Donald Solberg, USNRC, RES/SAFER

From: David E. Robertson, PNL

Findings to Date:

We will be initiating this program at two shut-down nuclear power plants--the Pathfinder Atomic Power Plant at Sioux Falls, South Dakota and at Indian Point Station, Unit 1 near Buchanan, New York. We felt that it was important to first examine the Pathfinder plant because of its lower contamination levels, its smaller size, the greater accessibility to a wide variety of contaminated materials, and the utilities great willingness to cooperate in every regard. We feel that this is a good place to initiate and perfect our sampling technology and on-site surveys. We are experiencing a slight delay in gaining access to Pathfinder because of licensing problems of the plant. The Northern States Power Company is in the process of applying for an amendment to the present license so that we can work there and remove samples for analysis. We are trying to expedite this as fast as possible. Northern States Power Company has estimated that entry into the plant will be possible sometime around the end of May, and I have set a tentative field trip for the second week in June. If we are not able to get into Pathfinder by this time, we will initiate our study at Indian Point Unit 1 and do Pathfinder later in the summer, as soon as the licensing amendment is approved. A detailed measurement plan for Pathfinder is enclosed. This plan has been submitted to Northern States Power Company and has formed the basis of the request for a licensing amendment.

We have assembled all of the necessary equipment for performing the field sampling and measurements. A state-of-the-art, portable intrinsic germanium gamma-ray spectrometer and related analyzer package has been purchased, assembled and is presently being tested and calibrated for a variety of geometries. This spectrometer can be hand carried and mounted in an omni-directional counting configuration to examine floors, walls, pipes, equipment, etc.

We have also assembled a beta proportional counter and a variety of radiation survey and dose rate meters to [illegible] in the field measurements program.

A concrete core drilling machine has been purchased, tested, and modified for use in contaminated environs. We will use this to collect 4" diameter x 6" deep concrete cores from contaminated floors and walls, and 1" diameter cores of the bioshield up to 10 feet long (the entire thickness of the bioshield concrete).

We have also purchased and tested a variety of portable metal cutting saws, including a portable band-saw, a portable circular-saw and a portable power hack-saw for obtaining sections of contaminated piping and metal components to ship back to PNL for comprehensive radionuclide analysis.

We have also been perfecting the radiochemical separation procedures for a number of difficult-to-analyze beta and alpha emitters (i.e., ^{63}Ni, ^{99}Tc, ^{129}I, Pu, Am, Cm) in a wide variety of matrix materials (soils, concrete, iron).

We have added to our research team an experienced and licensed health physics expert, Manford Leale, who will do our radiation monitoring at the reactor sites and be responsible for abiding by all regulations pertaining to handling and shipping of radioactive materials. He will spend about one-third of his time on this project.

Costs Incurred:

As of April 14, 1980 we have spend $114,502.00, broken down as follows:

Direct Labor Costs	$ 42,921
Procurements	7,890
Travel	3,463
Facilities and Equipment	12,594
Other Costs	1,262
Labor Overheads/Fee	42,358
Total Cost	4,014
TOTAL	$114,502

DETAILED SAMPLING AND ANALYSIS PLAN FOR PATHFINDER GENERATING PLANT

1. Purpose and Objectives

The purpose of this program is to identify the nature, distribution and inventory of residual radionuclide contamination in and around commercial light water nuclear power reactors to provide a data base for use in formulating policies and strategies for decommissioning retired nuclear power plants. The NRC and the operating utilities are charged with the responsibility of assuring the continued protection of the public from residual radioactivity or other potential hazards during and after decommissioning of the plants. In order to assure the safe and efficient decommissioning of nuclear power plants through appropriate regulations, guides and plans, measured data are needed to generically characterize the inventory and distribution of the radionuclide contamination of reactor systems and of the immediate surrounding environs. Such a knowledge will provide the predictive capability to permit a better assessment of the costs, radiation exposure and technology required for decontaminating and dismantling nuclear power plants, and restoring the reactor sites to an environmentally acceptable condition.

To acquire these data, a comprehensive measurements program is required. This measurement program will seek to characterize the residual radionuclide inventories in virtually all parts of the reactor sites which are known or suspected to contain radioactivity. This study will emphasize, but not be restricted to, the long-lived radionuclides ($t\ 1/2 > 1$ yr), including the transuranics and low energy beta and X-ray emitters. We plan to initiate this study at Pathfinder and Indian Point Unit 1 and then extend the measurements program to several operating plants.

The measurements plan will involve both an on-site examination of gamma and beta emitting radionuclides in all components at each reactor site, and the procurement of samples from the reactor sites for analyses at our laboratory in Richland, Washington.

Nondestructive On-site Surveys At Pathfinder

A portable collimated intrinsic germanium gamma-ray spectrometer and a beta detector will be utilized for nondestructive scanning for residual gamma and beta emitting radionuclides at Pathfinder. These instruments will be taken inside the Reactor Building, the Fuel Handling Building, the Turbine Building and auxiliary buildings to nondestructively measure residual radionuclide contamination on floors, walls, piping, equipment, stored scrap materials and any other materials containing contamination. Screening with a collimated G-M counter will first determine what areas will later be gamma-scanned using the intrinsic germanium detector. We will also examine the grounds immediately adjacent to the Pathfinder Generating Plant. It is not anticipated that we will require any maintenance assistance from NSP personnel during this phase of the study, except someone familiar with the plant who can escort us to all points of interest and explain the past operations. This portion of the work is anticipated to take about four days to complete.

Sample Procurement

Because of the potential difficulties of quantifying radionuclide concentrations within piping or on structural materials by the nondestructive scanning techniques, and to measure low energy beta emitters and the alpha-emitting radionuclides, samples of opportunity will be procured and shipped to our laboratory for comprehensive radionuclide analyses. These samples will consist of piping, structural materials, equipment, hardware and any other appropriate samples which would aid in establishing radionuclide inventories at Pathfinder.

We will also be prepared to obtain, when possible, four inch diameter concrete cores from highly contaminated floors and walls. Also, paint surfaces from contaminated areas will be sampled. Replicates of in-plant and environmental samples will simultaneously be collected for decontamination studies being conducted in another department at our laboratory and for archival purposes. The total radioactivity of all of the samples we would remove and ship to our laboratory is not expected to exceed one curie. Soil cores will also be taken around the grounds of the Pathfinder plant to determine if any small amounts of reactor produced radionuclides are present.

It is anticipated that this phase of the work will require about five days and would also require some assistance from NSP in the form of maintenance personnel to assist in pipe cutting and replacement etc. We will be prepared to do our own concrete coring and minor pipe cutting and coring. We are also prepared to do our own radiation monitoring and health physics. We have made arrangements with our radiation monitoring department to permanently assign one of their staff to this project. He will be responsible for assuring that all on-site work is carried out in compliance with safe and legal work practices. He will also arrange to obtain suitable shipping containers and assure that any shipments of materials outside containment to PNL are carried out in full compliance with DOT, NRC and state regulations. The sampling program, for convenience, can be organized on a building basis as described below.

Reactor Building

We would like to make as comprehensive an assessment of the radioactivity associated with all systems in the Reactor Building that is practically possible.

Two phases of sampling in the Reactor Building will be necessary. First, we will require samples of all contaminated materials outside of the bioshield to to determine the extent of residual radionuclide contamination which has been transported from the reactor vessel during plant operations. We would like to obtain sections of contaminated piping or at least four inch diameter cores from this piping. Also, any portions of contaminated pumps, valves and other hardware would be desirable to obtain for analysis. Concrete cores (four inch diameter by four inches deep) from contaminated floors and walls would also be collected from the most contaminated areas. Second, we would like to obtain samples of materials from the reactor vessel and bioshield which have been neutron activated. We will be prepared to take one or more one inch diameter cores through the entire concrete bioshield, hopefully at the midpoint of the reactor vessel. We would also like to obtain drill turnings of activated portions of the reactor vessel. The locations for taking the bioshield cores and reactor vessel steel samples will need to be discussed at greater length and perhaps we can make a preliminary site visit before the actual sampling and analysis work to determine this.

Fuel Handling Building

We would like to obtain samples of all contaminated piping, equipment, hardware and materials in all rooms associated with the Fuel Handling Building. Piping and tanks too large to cut into short segments for samples will be cored with a four inch diameter saw. We also would like to obtain four inch diameter by four inch deep concrete cores of contaminated floors. Since much of the contaminated piping, hardware, etc. from the dismantled portions of the plant have been stored in the fuel storage pool it will be necessary for us to obtain access to inside of the storage pool. Since the pool has been capped with a thick concrete cover it will be necessary for NSP to provide an entrance to the storage pool large enough for a person to enter. It would be convenient and speed the on-site work if this entrance could be provided before our on-site work begins. It is anticipated that the storage pool will have radiation levels too high to permit in-situ direct gamma-scanning so we intend to remove samples from this room for preliminary on-site gamma analysis at a low-background area of the plant.

Turbine Building

All contaminated material and equipment not being used in the fossil system which was removed and stored will be examined and sampled if possible. The "hot spots" in equipment and piping presently used in the fossil plant should be sampled, either by opening pipes and removing the crud or cutting out or coring small sections of the contaminated piping. (This can be optional if NSP does not want to disturb the operational facility.) Concrete cores would also be taken from contaminated floors (such as the sump floor).

Auxiliary Buildings

We would like to make several on-site gamma-spectrometric scans of the auxiliary buildings and structures at the site. We do not expect to find any significant contamination, but would like to verify that such is the case. Included in these buildings would be the Water Treatment Building, the cooling tower and any other structures that NSP would recommend. We would procure samples for comprehensive analysis only if we detected "significant" contamination.

Environmental Samples

We would like to take eight soil core samples in the eight compass directions surrounding the plant. The cores would be 5 cm deep and sectioned into 1 cm thick slabs of 200 cm^2 area. The cores will be collected about ten feet from the building in each direction.

We will make a survey of the grounds surrounding the plant with a G-M counter and also sample any "hot spots" that may be present in the soil. We will sample vegetation only if "significant" contamination is apparent.

Radionuclides To Be Measured

Using a portable intrinsic germanium gamma-ray spectrometer, on-site measurements of the gamma-emitters that are detectable by instrumental gamma-ray spectrometry will be made using nondestructive scanning techniques. A low-level beta counter will also be used in the nondestructive scanning measurements to document the beta activity at the same locations. We will key on the major gamma-emitting radionuclides to serve as indicators of contaminated areas. After the scanning is completed, measurements of selected hardware, structural materials and environmental samples will be performed on-site to allow more intelligent sampling. These rapid surveys will pinpoint the most desirable locations to conduct more intensive sampling and analysis, and will provide a great deal of information early in the program. The major, long-lived gamma-emitters that we expect to measure on-site by this method include ^{54}Mn, ^{65}Zn, ^{60}Co, ^{106}Ru, ^{110m}Ag, ^{134}Cs, ^{137}Cs, ^{144}Ce and possibly ^{125}Sb, ^{152}Eu and ^{154}Eu. Based on the on-site scanning results, extensive sampling will be conducted at all appropriate plant locations, and the instrumental gamma-ray spectrometric analyses of the bulk of these samples will be conducted at our laboratory in Richland. It is anticipated that approximately six to eight soil cores will be collected at each site for analyses at PNL, as well as a few samples of pond and trench sediments, cooling tower sludge and other appropriate environmental samples. Because the radiochemical analysis of the trace gamma-emitters, beta-emitters and the transuranic radionuclides are so time and manpower intensive, we will be very selective in choosing samples from each site for performing these additional difficult measurements. The actual selection of these samples will be guided by the results of the gamma spectrometric measurements. It is anticipated that only a few (no more than 8-10) in-plant samples at each site will be completely characterized to include the analysis of ^{108m}Ag, ^{94}Nb, ^{126}Sb-Sn, ^{55}Fe, ^{59}Fe, ^{59}Ni, ^{63}Ni, ^{90}Sr, ^{93}Zr-Nb, ^{99}Tc, ^{129}I, ^{147}Pm, and the transuranics ^{238}Pu, ^{239,240}Pu, ^{241}Am, ^{243}Am, ^{242}Cm and ^{244}Cm. Only the most contaminated soil and environmental samples (approximately 4-6) will be analyzed for these radionuclides. However, a few samples from areas considered to be background levels will also be included. If it becomes obvious that some anomalous contamination involving a specific radionuclide has occurred at a site, we will put special emphasis on delineating the extent of this contamination. Special attention will be given to the measurement of accurate isotopic ratios of various radionuclide pairs (e.g., ^{134}Cs/^{137}Cs, ^{238}Pu/$^{239-240}$Pu, ^{244}Cm/$^{239-240}$Pu, etc.) in the in-plant and environmental samples. These ratios can differ by several orders of magnitude when comparing reactor-generated radionuclides with those observed in weapons testing fallout. It is therefore possible to identify as little as a 2-3% contribution to the fallout radionuclide inventory from radionuclides of reactor origin. Since easily measurable fallout radionuclide concentrations will be present in the environmental samples, it will be important to document the fallout contribution relative to that coming from reactor contamination at each reactor site. These data will be greatly complemented by EPRI sponsored studies of the nature and distribution of radionuclides outside the boundaries of four nuclear power stations.

It is recognized that a number of intermediate-lived radionuclides will also be present in reactor contamination residues, including ^{46}Sc, ^{51}Cr, ^{57}Co, ^{58}Co, ^{59}Fe, ^{95}Zr-Nb, ^{103}Ru and ^{124}Sb. When these radionuclides are detectable by instrumental gamma-ray spectrometry they will be automatically measured by our counting equipment. However, we will make no special attempt at measuring them if their concentrations are below the instrumental (nondestructive) detection limits.

POTENTIAL RADIONUCLIDES TO BE MEASURED

Isotope	Half-Life	Decay Mode	Energy (KeV)
	Gamma-Emitters		
^{22}Na	2.6 y	β^{+} - γ	1275
^{54}Mn	312 d	EC - γ	834
^{60}Co	5.27 y	β - γ	1332
^{65}Zn	244 d	EC - β^{+} - γ	1115
^{94}Nb	2.0×10^{4} y	β - γ	871
$^{106}Ru\text{-}Rh$	36[illegible] d	β - γ	622
^{108m}Ag	130 y	EC - IT	434
^{110m}Ag	250 d	β - IT	658
^{125}Sb	2.77 y	β - γ	438
$^{126}Sb\text{-}Sn$	10^{5} y	β - γ	414
^{134}Cs	2.06 y	β - γ	796
^{137}Cs	30.17 y	β - γ	662
^{144}Ce	284 d	β - γ	134
^{152}Eu	13.6 yr	EC - β - γ	344
^{154}Eu	8.6 yr	β - γ	723
^{155}Eu	4.96 yr	β - γ	105
^{166m}Ho	1200 y	β - γ	184

POTENTIAL RADIONUCLIDES TO BE MEASURED

Isotope	Half-Life	Decay Mode	Energy (KeV)
	Beta and X-ray Emitters		
^{55}Fe	2.7 y	EC	5.9 X-ray
^{59}Ni	7.5×10^4 y	EC	6.9 X-ray
^{63}Ni	96 y	β	17 β^- avg.
^{90}Sr-Y	28.5 y	β	931 β^- avg. (from ^{90}Y)
^{93}Zr-Nb	9.5×10^5 y	β	19.6 β^- avg.
^{99}Tc	2.13×10^5 y	β	84 β^- avg.
^{129}I	1.57×10^7	β	29.8 X-ray
^{135}Cs	2.3×10^6	β	210 KeV
^{147}Pm	2.62	β	62 β^- avg.
^{151}Sm	90 y	β	19.6 β^- avg.
		Transuranics	
^{237}Np	2.14×10^6 y	α	4788
^{238}Pu	87.8 y	α	5499
$^{239-240}Pu$	2.44×10^4 y	α	5155
^{241}Pu	14.4 y	β	5.2 β^- avg.
^{241}Am	433 y	α	5485 (also 59.5 X-ray)
^{243}Am	7380 y	α	5275
^{242}Cm	163 d	α	6113
^{244}Cm	18.1 y	α	5805