

✓
070
✓ 8/15/83

LOAN COPY

THIS REPORT MAY BE RECALLED
AFTER CONSULTING THE SOURCE
RETURN PROMPTLY TO:

cy 1

INEL TECHNICAL LIBRARY

PROGRAM PLAN: EVALUATION, DEVELOPMENT AND VERIFICATION OF EFFLUENT MONITORING METHODS DURING NUCLEAR FUEL REPROCESSING AND WASTE SOLIDIFICATION

Public Reading Room
U. S. Department of Energy
Idaho Operations Office

March 1978 Public Reading Room
U. S. Department of Energy
Idaho Operations Office



Allied
Chemical

IDAHO CHEMICAL PROGRAMS



IDAHO NATIONAL ENGINEERING LABORATORY

DEPARTMENT OF ENERGY

070
✓ 8/1983

LOAN COPY

THIS REPORT MAY BE RECALLED
AFTER TWO WEEKS PLEASE
RETURN PROMPTLY TO:

ayl

INEL TECHNICAL LIBRARY

PROGRAM PLAN: EVALUATION, DEVELOPMENT
AND VERIFICATION OF
EFFLUENT MONITORING METHODS DURING
NUCLEAR FUEL REPROCESSING AND
WASTE SOLIDIFICATION



March 1978

Public Reading Room
U. S. Department of Energy
Idaho Operations Office



Allied
Chemical

IDAHO CHEMICAL PROGRAMS



IDAHO NATIONAL ENGINEERING LABORATORY

DEPARTMENT OF ENERGY

ACI-226, Rev. 1

ACI-226, Rev. 1

**PROGRAM PLAN: EVALUATION, DEVELOPMENT AND VERIFICATION
OF EFFLUENT MONITORING METHODS DURING NUCLEAR FUEL
REPROCESSING AND WASTE SOLIDIFICATION**

by

R. C. Girton

S. J. Fernandez

M. A. Wong

L. P. Murphy

Date Published - March 1978

**ALLIED CHEMICAL CORPORATION
IDAHO CHEMICAL PROGRAMS - OPERATIONS OFFICE**

**Work Performed for
DEPARTMENT OF ENERGY - IDAHO OPERATIONS OFFICE
UNDER CONTRACT EY-76-C-07-1540**

SUMMARY

This plan outlines seven effluent monitoring projects for study and investigation. These projects have been identified as significant in extending the state-of-the-art in effluent monitoring.

The objective of the plan is to evaluate, modify, and improve monitoring techniques at existing and future nuclear fuel reprocessing and waste solidification facilities. Innovative approaches to identified problems will be examined and, if feasible, demonstrated under operational conditions. Specific problems and needs in off-gas monitoring technology defined for investigation include the dioctylphthalate in-place test method for verification of HEPA filter efficiencies in a corrosive environment, monitoring of radionuclides with low-energy emissions, real time tritium monitoring, reliable analytical methodology for ^{14}C , ^{129}I and ^{85}Kr , on-line monitoring of alpha emissions, evaluation of sub-micrometre particle collection techniques, and particle size studies in plant off-gas streams.

Investigation of these monitoring needs will begin with laboratory and small-scale studies. The technology developed in the small-scale studies will be verified by in-plant sampling and measurements. Work on the combined projects will run from mid FY-1978 through FY-1981 and will require a total manpower effort of 15 man-years.

CONTENTS

SUMMARY	i
I. INTRODUCTION	1
II. EVALUATION OF THE HEPA FILTER IN-PLACE TEST METHOD	6
1. Problem Definition	6
2. Description of Test Method	7
3. Project Plan	7
4. Laboratory Apparatus	9
III. METHODS EVALUATION AND DEVELOPMENT FOR MONITORING OF RADIONUCLIDES WITH LOW ENERGY EMISSIONS	11
1. Problem Definition	11
2. Project Plan	11
IV. EVALUATION AND IMPROVEMENT OF TRITIUM MONITORING TECHNIQUES	11
1. Problem Definition	12
2. Project Plan	12
V. DEVELOPMENT OF RELIABLE ¹⁴ C, ¹²⁹ I, AND ⁸⁵ Kr ANALYTICAL METHODOLOGY	12
1. Problem Definition	12
2. Project Plan	13
VI. INVESTIGATION OF ALPHA MONITORING TECHNIQUES	14
1. Problem Definition	14
2. Project Plan	14
VII. EVALUATION OF SAMPLING METHODS FOR TOTAL COLLECTION OF SUB-MICROMETRE PARTICULATES	15
1. Problem Definition	15
2. Project Plan	15
VIII. PARTICLE SIZE STUDIES IN PLANT OFF-GAS STREAMS	15
1. Problem Definition	15
2. ICPP Plant Processes to be Investigated	16
3. Project Plan	19

CONTENTS (Continued):

IX. REFERENCES 20

FIGURES

**1. Time schedule for effluent monitoring methods evaluation
and development 3**

2. Overall manpower requirements and equipment costs 4

3. Critical resources 5

4. WCF off-gas decontamination system 8

5. HEPA filter test apparatus 10

**6. Proposed sampling locations for off-gas particulate
studies 17**

7. Isometric view of APS process off-gas area 18

I. INTRODUCTION

Experience in effluent monitoring at the Idaho Chemical Processing Plant (ICPP) resulted in the identification of specific problems and needs in off-gas monitoring technology at nuclear fuel reprocessing and waste solidification facilities. Seven major problem areas have been identified for investigation:

(1) Evaluation of the High Efficiency Particulate Air (HEPA) Filter In-Place Test Method in a Corrosive Off-Gas Environment

In-place testing of the Waste Calcining Facility (WCF) off-gas HEPA filters may be hindered by the combined effects of temperature, humidity and oxides of nitrogen. The effect of each parameter, individually and in combination with other parameters, on the standard test will be defined.

(2) Methods, Evaluation and Development for Monitoring of Radionuclides with Low-Energy Emissions

The reprocessing of defense and alternative nuclear fuels may release significant quantities of the long-lived low-energy radionuclides technetium-99 and selenium-79. An on-line continuous stack monitor system was designed and sensitivity limits will be established. In addition, a time integrated sampling technique will be developed for applications requiring greater sensitivity.

(3) Evaluation and Improvement of Tritium Monitoring Techniques

Tritium released from the ICPP is primarily in the form of tritiated water. Due to the active utilization of this species in plant and animal life and to the biologically long half-life, monitoring of tritium is a necessity. The upgrading of tritium monitoring to real time analysis would yield more detailed information on the relationship between plant processes and tritium released.

(4) Development of Reliable ^{14}C , ^{129}I and ^{85}Kr Analytical Methodology

The presently used molecular sieve ^{14}C sampler is inappropriate in process streams with high NO_x concentrations. The charcoal beds used for ^{129}I collection have yet to be verified during plant operations. Therefore, tests will be performed to verify the time integrated sampling technique for ^{14}C and ^{129}I . The project will also include improving the continuous ^{85}Kr monitor, and the development of continuous ^{14}C and ^{129}I monitors.

(5) Investigation of Alpha Monitoring Techniques

The quantitative analysis of low-level alpha-emitting nuclides is hampered by the presence of background interferences. A technique of measuring these alpha nuclides must be developed in anticipation of reprocessing defense and alternative reactor fuels.

(6) Evaluation of Sampling Methods for Total Collection of Sub-Micrometre Particulates

The study of radioactive particulates in the sub-micrometre range is important in regard to deposition within the human respiratory system. A more efficient sampling detection method is needed, particularly in the area of under 0.5 micrometre particle diameter. Collection systems will be investigated, evaluated and demonstrated.

(7) Particle Size Studies in Plant Off-Gas Streams

By determining the particulate characteristics of each process off-gas stream, stack release data can be related to a specific operation during multiple process off-gases so that data can be compared to present stack particulate results.

These monitoring problems need solutions for effective waste management operations of nuclear fuel reprocessing and waste solidification plants. An appraisal of present off-gas monitoring technology reveals a need to develop new methodology to solve problems such as the monitoring of radionuclides with low-energy emissions. Other problems, such as the development of reliable ^{14}C , ^{129}I , and ^{85}Kr monitors may require only improvement or upgrading in existing technology. Previous evaluation studies at the ICPP have, in general, confirmed that most of the existing monitoring methods and procedures have provided reliable and reasonably accurate data for the purposes intended. However, with the increasing growth and diversification of the nuclear industry, present monitoring techniques will not be adequate to meet the more stringent future requirements that are anticipated. Continuous measurement where feasible is needed for documentation in view of possible long-term legal problems.

The projects described seek to evaluate, develop or improve monitoring systems so that stack effluent releases can be measured and controlled in accordance with the requirements set forth in the ERDA Manual Ch. 0524. It is the responsibility of Allied Chemical Corporation, as a contractor for DOE, to use the available technical expertise, experience and facilities to continually upgrade existing monitoring methods and procedures.

The proposed timetable for the combined projects will run from FY-1978 through FY-1981. The time schedule for each project is illustrated in Figure 1. The total manpower effort will be 15 manyears, total operating costs will be \$915,000, and total capital equipment expenditures will be \$140,000 as shown in Figure 2. The critical resources for each project are shown in Figure 3.

Explanation of Time Schedule (Figure 1)

- 1.1 Laboratory testing.
- 1.2 Comparison testing with French and British methods.
- 1.3 In-plant demonstration and final report.

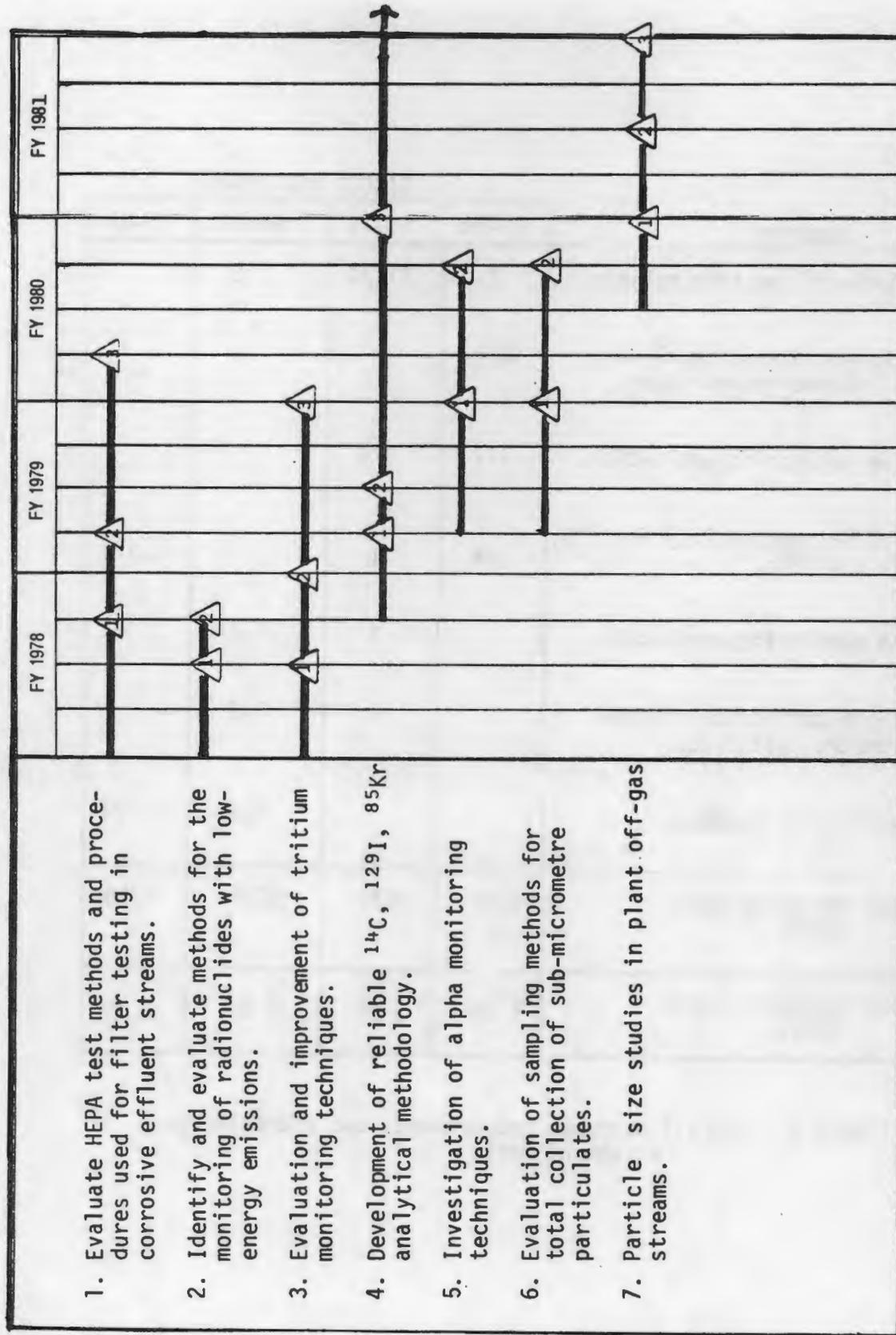


Figure 1. Time schedule for effluent monitoring methods evaluation and development

PROJECT	DIRECT MAN-YEARS			
	FY-78	FY-79	FY-80	FY-81
Evaluate filter test methods.	2	1-1/4	1/2	
Monitoring of radionuclides with low-energy emissions.	3/4			
Tritium monitoring evaluation.	1/2	3/4		
Analytical methodology for ¹⁴ C, ¹²⁹ I, and ⁸⁵ Kr.	3/4	1	1	1-3/4
Alpha monitoring evaluation.		1	1/2	
Particle collection of sub-micrometre particles.		1	1/2	
Particle size studies.			1/2	1-1/4
Total Operating Costs (\$000)	\$225	\$270	\$210	\$210
Total Equipment Costs (\$000)	\$ 30	\$ 50	\$ 30	\$ 30

Figure 2. Overall manpower requirements and operating and equipment costs

- 2.1 System design, laboratory tests, and instrumentation development using radioactive tracers.
- 2.2 Time integrated sampling technique site tested and final report.
- 3.1 Evaluate existing methods and issue report.
- 3.2 Complete experimental design and construct monitor.
- 3.3 Perform tests and final report.
- 4.1 Evaluate NRL continuous monitor for ^{129}I .
- 4.2 Evaluate existing methods for ^{129}I , ^{14}C and ^{85}Kr and issue report.
- 4.3 Perform tests to modify and improve existing time integrated methods for ^{129}I and ^{14}C and existing ^{85}Kr continuous monitor. Issue report.
- 5.1 Identify interferences from naturally occurring radioisotopes in various geographic areas and determine monitoring feasibility.
- 5.2 Demonstrate most acceptable method or instrumentation. Publish report or results.
- 6.1 Evaluate available systems such as cryogenic collection, electrostatic precipitation, scrubber cyclones, low-pressure cascade impactors.
- 6.2 Demonstration and installation at stack monitor. Publish report of results.
- 7.1 Identify sampling location and accessibility.
- 7.2 Evaluate commercial samplers and instrumentation.
- 7.3 Sampling, data evaluation and report.

II. EVALUATION OF THE HEPA FILTER IN-PLACE TEST METHOD

1. Problem Definition

The American National Standards Institute (ANSI) describes a procedure for in-place leak testing of high efficiency particulate air (HEPA) filters after filter replacement, or maintenance activity in the filter housing.¹ The method is also described in detail in Sec. 8.3.1 of the Nuclear Air Cleaning Handbook and in Sec. 3 of ANSI-N101.1.^{2,3} Erratic on-line test results of the HEPA filters at the Waste Calciner Facilities (WCF) have been recorded using the standard dioctylphthalate (DOP) test method.⁴ The WCF off-gas is at high temperature, high humidity and has a high NO_x concentration. This work will investigate the individual and combined effects of these parameters on the DOP test method. Filter test methods used in the U.K. and France will be evaluated under WCF conditions. The project will require a three-year plan, extending from mid FY-1977 to mid FY-1980 for a total of about four manyears of effort.

2. Description of Test Method

The WCF off-gas stream is cleaned by a system composed of a dry cyclone, a wet scrubber, silica gel adsorbers, and HEPA filters (Figure 4). At the filters, the off-gas has a temperature of 90°C, a relative humidity of 95-99% and an oxides of nitrogen concentration of 10,000 ppm. Filters in this off-gas stream are tested semiannually. A thermal DOP generator is used which has a generation capacity of 100 µg/L of 0.7 micrometre polydisperse DOP smoke. The smoke from the DOP generator is injected upstream of the HEPA filter bank to be tested. The DOP concentration in the filtered and unfiltered off-gas is measured with a photometer which detects the forward light scattering of the DOP aerosol. The procedure specifies a "fail" test if more than 0.03% of the DOP penetrates the filter bank. Each of the three parallel HEPA filter banks in the WCF off-gas system can be opened and closed for testing of individual banks. Each bank that is tested contains two filters in series, possibly causing poor test results due to agglomeration and evaporation between filters.⁵

High humidity can influence the DOP testing specifically, and nitrogen dioxide, a reddish-brown gas, may interfere with the operation of the photometer. At high humidity, nitric acid can form and will pit the photometer cell over long or constant exposures. The effect of all three factors in combination (humidity, temperature, and NO_x concentration) on the DOP test procedure is not known.

Experiments altering the temperature, humidity and nitrogen oxides conditions will be needed to investigate:

- (1) Combined effects of the parameters on the accuracy of results produced by photometers when using the DOP test method;
- (2) Reliability of the present DOP test method and possible modifications;
- (3) Comparison with results of other test methods.

3. Project Plan

The project is in three phases:

- (1) Evaluation of the present DOP test;
- (2) Comparison of the DOP test method with the British (sodium chloride) and French (uranine) methods;
- (3) In-place demonstration of the best method in the WCF off-gas system.

The DOP evaluation will be conducted according to a 2³ factorial experimental design. In a 2³ factorial experimental design, two levels (high and low) of the three parameters (temperature, humidity, NO_x concentrations) are selected for study. The high and low values for relative

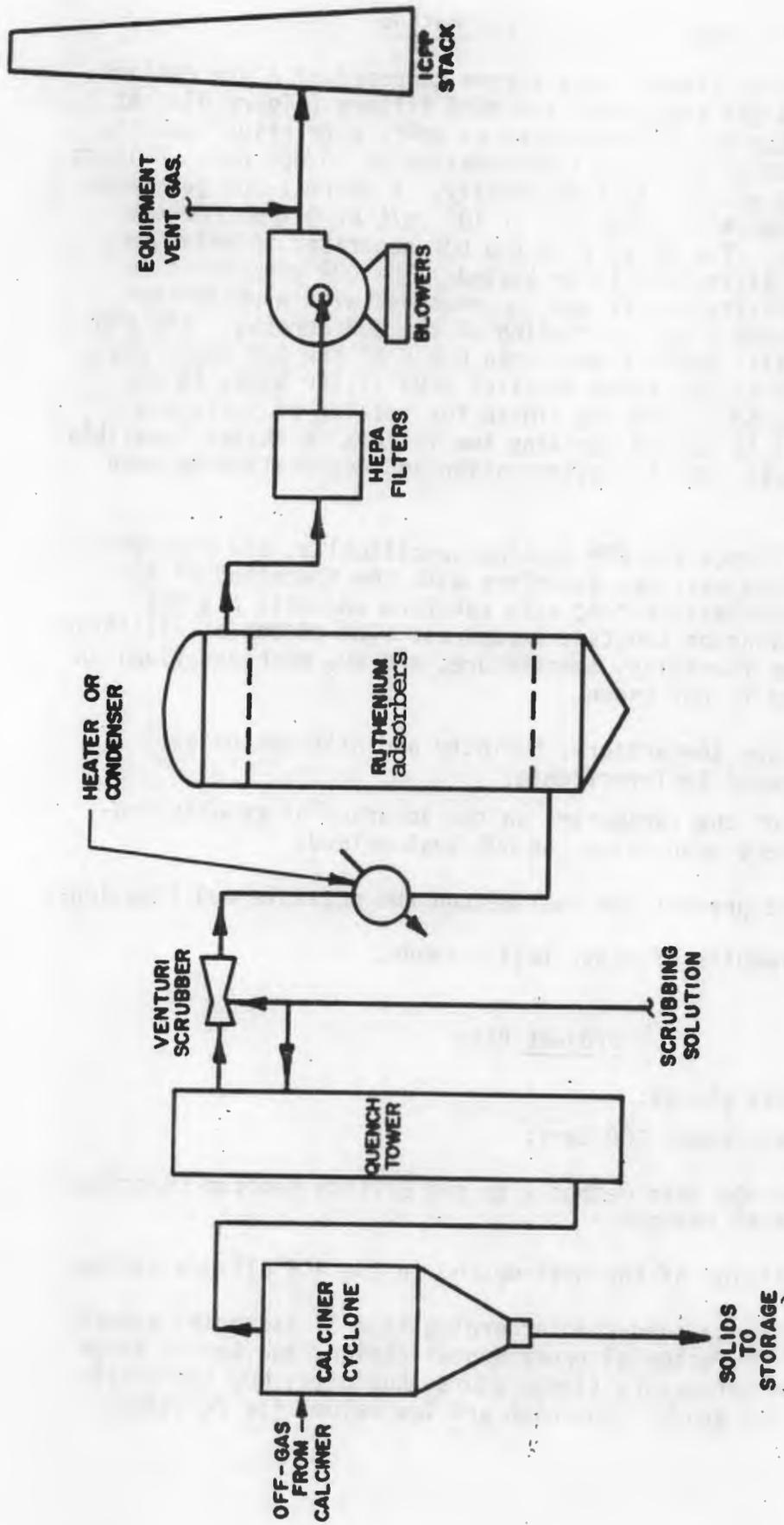


Figure 4. WCF Off-Gas Decontamination System

ACC-A-2760

humidity will be 50 and 95%, will be 500 and 10,000 ppm for NO_x concentrations, and will be 150 and 200°F for temperature. The combination of high and low values of the three parameters will be varied randomly and the filter efficiency for each combination calculated.

The advantage of this experimental design is that both the "main effect" and the "interactions" of each factor may be determined. Changes in the DOP procedure to minimize the effect of each parameter will be investigated.

The second phase of the project will involve a comparison of the DOP (U.S.), sodium chloride (British), and uranine (French) methods of testing HEPA filters. It is expected that the British and French methods can be adapted to the proposed DOP test apparatus with appropriate modifications.

The last phase will be the modification of the present DOP, sodium chloride, or uranine test equipment and/or procedures so that a more consistent and accurate HEPA filter efficiency can be measured. These changes will then be tested in the WCF off-gas HEPA banks to determine their effectiveness.

4. Laboratory Apparatus

The experimental apparatus is shown in Figure 5. The basic design will include an aerosol generator for DOP, injection points for air, steam, and NO, a reaction bomb for partial conversion of the NO to NO₂, two HEPA filters in series, and sample ports before each filter and after the filter bank.

Rotameters before and after the filter bank will indicate flow through the system, while pressure indicators will show the pressure drop across each filter. Humidity and nitrogen oxide concentration will be measured upstream of the filter system.

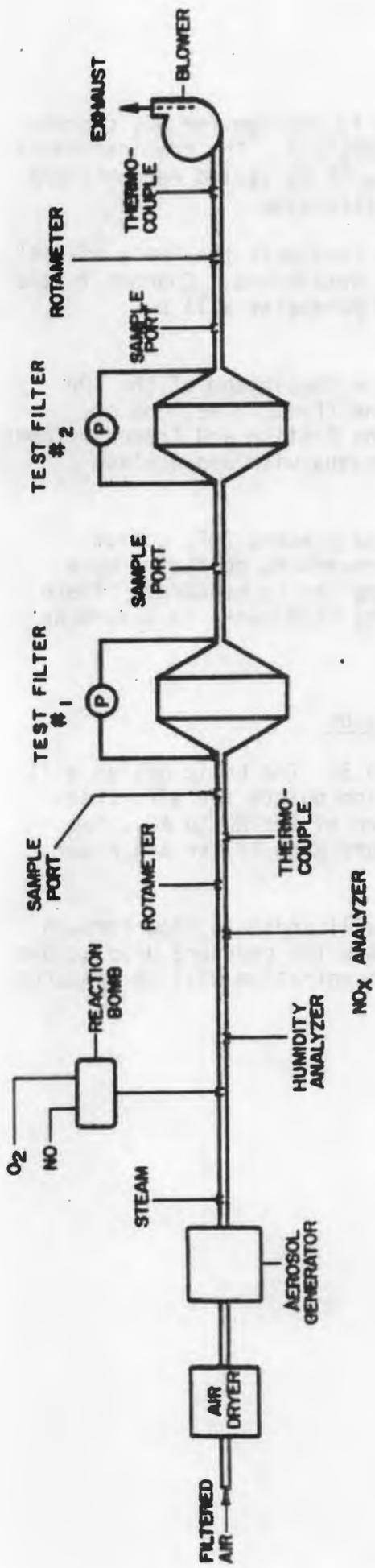


Figure 5. HEPA Filter Test Apparatus

ACC-A-2759

III. METHODS EVALUATION AND DEVELOPMENT FOR MONITORING OF RADIONUCLIDES WITH LOW ENERGY EMISSIONS

1. Problem Definition

Stack monitoring for low-energy beta and gamma emitting radionuclides has received little attention from the nuclear industry compared to the major effluent contaminants, cesium-137, ruthenium-106 and krypton-85. Monitoring or periodic sampling of all nuclides emitted to the atmosphere will eventually be required. In addition, the reprocessing of defense and alternative fuels will require analysis of long-lived nuclide concentrations. Complete knowledge of long-lived nuclides behavior in plant processes cannot precede the development of sensitive analytical techniques. In anticipation of these future monitoring requirements, two long-lived isotopes were selected for study, technetium-99 and selenium-79. These isotopes were selected on the basis of long half-life (>50,000 years), difficult-to-measure radiations ($\beta_{\max} < 0.3$ Mev, no γ), moderate toxicity,⁶ and significant presence in spent fuels.⁷

The project will be completed in late FY-1978 when a final report will be issued. About a one-manyear effort will be required.

2. Project Plan

A literature search to determine the feasibility of existing methods and instrumentation revealed that real time and time-integrated analysis was beyond present capabilities. The most likely approach to the real time analysis of these nuclides appears to be a rapid on-line chemical separation based on the relative volatility of Tc_2O_7 , SeO_2 , and H_2Se from acid media. The best approach to time-integrated sampling is the collection of the technetium and selenium on beds of silver zeolite and charcoal. Subsequent analysis would be by chemical purification followed by liquid scintillation counting.

The laboratory experiments will determine the design and operational parameters of the system, the sensitivity, specificity and reliability of the system.

The final phase of the project will be a demonstration of the prototype system.

IV. EVALUATION AND IMPROVEMENT OF TRITIUM MONITORING TECHNIQUES

1. Problem Definition

One of the most difficult monitoring problems at fuel reprocessing facilities has been real time monitoring of tritium. The nuclide is a primary contaminant in gaseous effluents. On-line separation techniques have not proven successful due to the difficulty of separating tritium from krypton-85, another major gaseous contaminant.

The project was started in early FY-1978 and will conclude with the issuance of a final report at the end of FY-1979. About a one-manyear effort will be required.

This project will investigate methods used to separate tritium from radioactive interferences that may occur in off-gases from reprocessing and waste solidification facilities. Present separation techniques include the automated collection of tritium as HTO on-silica gel or molecular sieves followed by stripping the tritium for counting. Present drawbacks to this approach are variable stripping efficiency, inadequate decontamination from interferences, and lack of detector specificity. A separation and analysis scheme based on the different permeabilities of krypton and HTO in polymer membranes appears promising.

2. Project Plan

A commercial permeable membrane separator will be evaluated in the laboratory for compatibility with stack gas humidity, temperature, and NO_x content. The catalytic conversion of all forms of tritium to HTO prior to separation will be documented. Development of an improved, state-of-the-art counter which will have a detection limit below 10^{-9} $\mu\text{Ci/cc}$ will be done in the laboratory phase. After the integrated system is proven on a laboratory scale, it will be demonstrated on-line at the ICPP stack.

V. DEVELOPMENT OF RELIABLE ^{14}C , ^{129}I , AND ^{85}Kr ANALYTICAL METHODOLOGY

1. Problem Definition

Among the most difficult nuclides to determine in effluent gas streams are ^{14}C , ^{129}I and ^{85}Kr . These nuclides exist as gases in the process streams and, along with tritium, are the most radiologically significant. Presently, ^{14}C is collected at the ICPP stack by oxidizing all chemical forms to $^{14}\text{CO}_2$ and collecting the formed $^{14}\text{CO}_2$ on beds of molecular sieve. Off-gas streams containing large amounts of NO_x cannot be accurately analyzed for ^{14}C with this system due to coadsorption of NO_2 on molecular sieve. Collection of radioiodine has been studied on a

laboratory scale. Presently ^{129}I is collected on an activated charcoal at the ICPP stack. Although charcoal has operated reliably in simulated off-gas streams, there is only a meager data base on its in-place performance. Krypton-85 is monitored continuously using a surface barrier detector. Other gaseous and particulate beta emitters are removed before counting by filters, charcoal beds, silver-exchanged zeolite beds, and silica gel beds. This monitor has occasionally produced anomolous results which could be produced by electronic interference, breakthrough of radiochemical interferences, or background variations. The proposed program plan will address each of these problems.

The project will begin at mid FY-1978 and conclude in FY-1981. A total of about four manyears effort will be required.

2. Project Plan

The project will examine the on-line behavior of the ^{14}C , ^{129}I and ^{85}Kr monitors. The molecular sieve beds will be replaced with two barium hydroxide impingers to collect ^{14}C . Among the parameters that will be determined on-line and in the laboratory will be nitrate formation (NO_x), optimization of liquid scintillation counting techniques, and the presence of radiochemical interferences in the BaCO_3 precipitate. The feasibility of a continuous on-line monitor will be examined. The behavior of the ^{129}I sampler will be evaluated by correlating the amounts of ^{129}I found in charcoal beds plumbed in series and in parallel. In addition, the iodine species emitted from various plant processes will be determined by sampling off-gas streams with a species selective sampler similar to that reported by Emel, et al.⁸ Once the iodine emissions are characterized, the feasibility of a continuous monitor will be examined. The ^{85}Kr monitor will be upgraded by the installation of isolation and line transformers to control electronic interferences. The efficacy of these transformers in controlling electrical noise will be documented.

The possibility of radiochemical breakthrough will be examined in laboratory tracer experiments and by analysis of all nuclides in the krypton fraction of the stack gas samples. The background variation will be documented by the use of matched surface barrier detectors, one detector to count the sample and the other detector to count background.

During the first quarter of FY-1979, a radioiodine detector based on laser-induced fluorescence will be evaluated. The instrument was developed at the Naval Research Laboratory (NRL); the prototype for evaluation is being built by Bendix Corporation. Evaluation at the ICPP will follow a testing program at NRL.

VI. INVESTIGATION OF ALPHA MONITORING TECHNIQUES

1. Problem Definition

Techniques do not exist for the real-time determination of alpha radiation stack emissions that will meet the low activity-high sensitivity measurement requirements needed after high efficiency cleanup systems in reprocessing and waste solidification facilities. If high burnup fuels are processed as expected, accurate alpha detection will be necessary as proof of meeting pollution control standards.

A major problem in the development of a satisfactory system will be background interferences, particularly at the ICPP where thorium deposits exist in the immediate vicinity. Using this plant as an extreme case for background interferences, the development of a monitoring system here for the detection of plant alpha releases should also provide satisfactory results when installed at any other existing or future plant. A system of high sensitivity capabilities would also be required, if it is desired to relate alpha emissions to specific plant processes.

This project would require at least 18 months of work, beginning in early FY 1979 and extending until late FY 1980. A 1-2 manyear effort would be needed.

2. Project Plan

Initially, all background interferences would have to be identified and separation or subtraction techniques developed. One possible solution to the interferences problem may require the use of matched detectors, i.e., one detector for the measurement of background alpha radiation and another detector to measure total activity. A computer would correct for background over a specified count time. Laboratory tracer studies may be necessary for complete system development and testing of instrumentation using the most sensitive detectors.

For satisfactory operation, the alpha monitor must be able to detect plutonium and uranium concentrations as low as 10^{-12} $\mu\text{Ci/cc}$. Ideally, this monitor should be capable of isotope differentiation since the maximum permissible concentration in air (MPCA) for the plutonium isotopes are much lower than the MPCA for uranium isotopes. One possibility for isotopic analysis is the measurement of the characteristic X-rays given off during alpha decay. The most sensitive and specific monitor will be installed, tested, and demonstrated using the ICPP stack effluent. The results will be documented in a final report.

VII. EVALUATION OF SAMPLING METHODS FOR TOTAL COLLECTION OF SUB-MICROMETRE PARTICULATES

1. Problem Definition

Particles in the sub-micrometre range will deposit in the lower portion of the human respiratory system so total particle collection should be of concern when developing a stack sampling system at a nuclear facility. When collecting particulates in an off-gas environment using sample filters of HEPA specifications, high collection efficiencies are obtained for particles of tenths of a micrometre diameter. Below about 0.05 micrometre diameter, high collection efficiency is questionable due to possible particle behavior as gases (Brownian movement). The project will investigate sub-micrometre collection efficiencies of various particle collection systems. Previous results at INEL-ICPP indicate that about 5% of the radioactivity associated with particulates escapes HEPA sampling filters.⁹ The Environmental Protection Agency method 5 was used.¹⁰ The project would require about 18 months to complete, beginning at mid FY-1979 and continuing through FY-1980. About 1-2 manyears of effort would be required.

2. Project Plan

The collection efficiency using various instrumentation and equipment will be defined in laboratory experiments using particle generators and various sub-micrometre particle size standards. Identified systems to be tested include cryogenic separators, electrostatic precipitators, scrubber cyclones, low-pressure cascade impactors, and the EPA sampling train.

On the basis of laboratory results, the most efficient collection system will be demonstrated using the ICPP stack effluent. The project goal is near 100% collection of particles of all size ranges, with particular interest directed toward the under 0.1 micrometre range.

VIII. PARTICLE SIZE STUDIES IN PLANT OFF-GAS STREAMS

1. Problem Definition

Work performed at the Idaho National Engineering Laboratory (INEL), using cascade impactors for particle size measurement of particulates at the stack, indicates that specific plant operations are related to stack particulate releases. This work has been reported in Allied Chemical Corporation - Idaho Operations monthly progress reports¹¹⁻¹⁵ and in a topical report.¹⁶

Preliminary measurements of stack effluents indicate the need for expanding the studies to obtain information at the point of release. At the ICPP, 90% of the stack effluents are plant ventilation air. This large dilution contributes significantly to errors in interpreting plant operation data. Another possible error would be due to changes in particulate size as particulate moves through transport lines from the point of release. It is proposed that particle size measurements be made close to the source of each gaseous effluent release. A better indication of particle characteristics in each stream would then be obtained and this information could be applied to the stack data for accurate and specific particle release information.

This project would result in providing the following particulate information for each major process off-gas stream: (1) particle size distribution, (2) radionuclides associated with particle size, (d) dust load, (4) element identification, and (5) emission concentration.

The project would require about 18 months to complete, beginning in mid FY-1979 and terminating at the end of FY-1980. However, the project would be dependent on ICPP and WCF plant operating schedules. About a 2-manyear effort would be needed.

2. ICPP Plant Processes to be Investigated

Because of the variety of plant processes, the ICPP is an ideal facility for studying particle characteristics in off-gas (OG) streams. The off-gases from the following processes will be investigated: (1) zirconium fuel dissolution (Zr-DOG), (2) aluminum fuel dissolution (CPM-DOG and C&D cell-DOG), (3) electrolytic process, (4) vessel off-gases (VOG), (5) Waste Calciner Facility off-gas (WCF-OG), and (6) ventilation air. Figure 6 shows these systems and the proposed sampling points.

The Zr-DOG (zirconium dissolver off-gas) cleanup system contains a caustic scrubber which removes hydrofluoric acid mist from off-gas having a maximum flow of 50 cfm. After the scrubber, the off-gas is cleaned by a demister, routed through a superheater, and filtered by a HEPA bank. This gas is further cleaned at the Atmospheric Protection System (APS) by a fiberglass-packed prefilter and another HEPA filter.¹⁷ The sample collection point will be downstream of the first HEPA, but before the APS prefilter.

During the dissolution of aluminum fuels, effluents (Al-DOG) can be released by either the C&D-DOG or the CPM-DOG. Off-gases from each are routed through a common cleanup system that consists of a demister, superheater, and HEPA filter. The flow rate of the gas is about 150 cfm. The off-gas is then combined with the cleaned WCF and vessel off-gases and the total gases are further decontaminated by the APS off-gas cleanup system (Figure 7). The best sampling station for the aluminum dissolver off-gas would be as close to the CPM-DOG HEPA filter as possible.

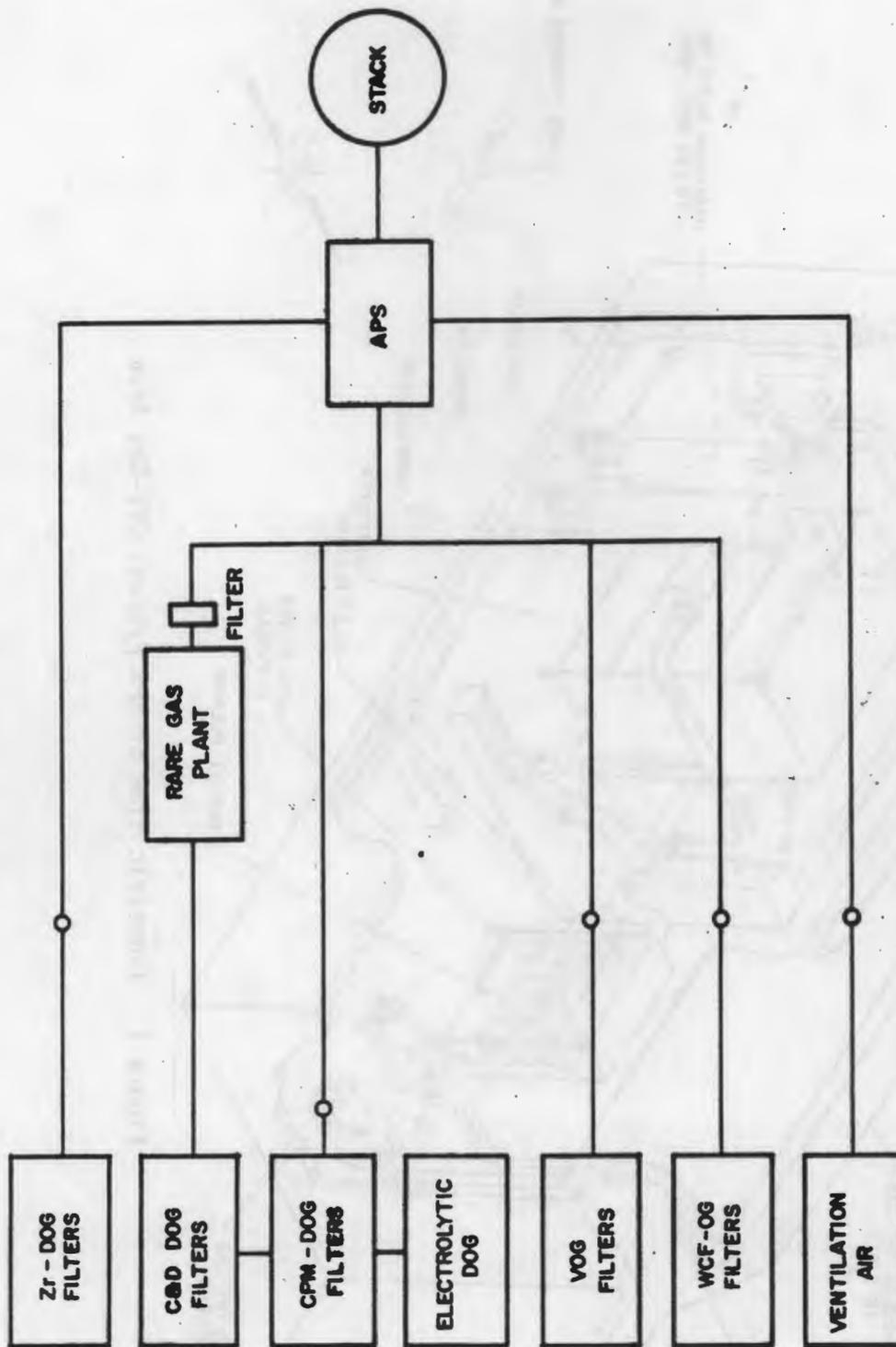


Figure 6. Proposed Sampling Locations for Off-Gas Particulate Studies (o)

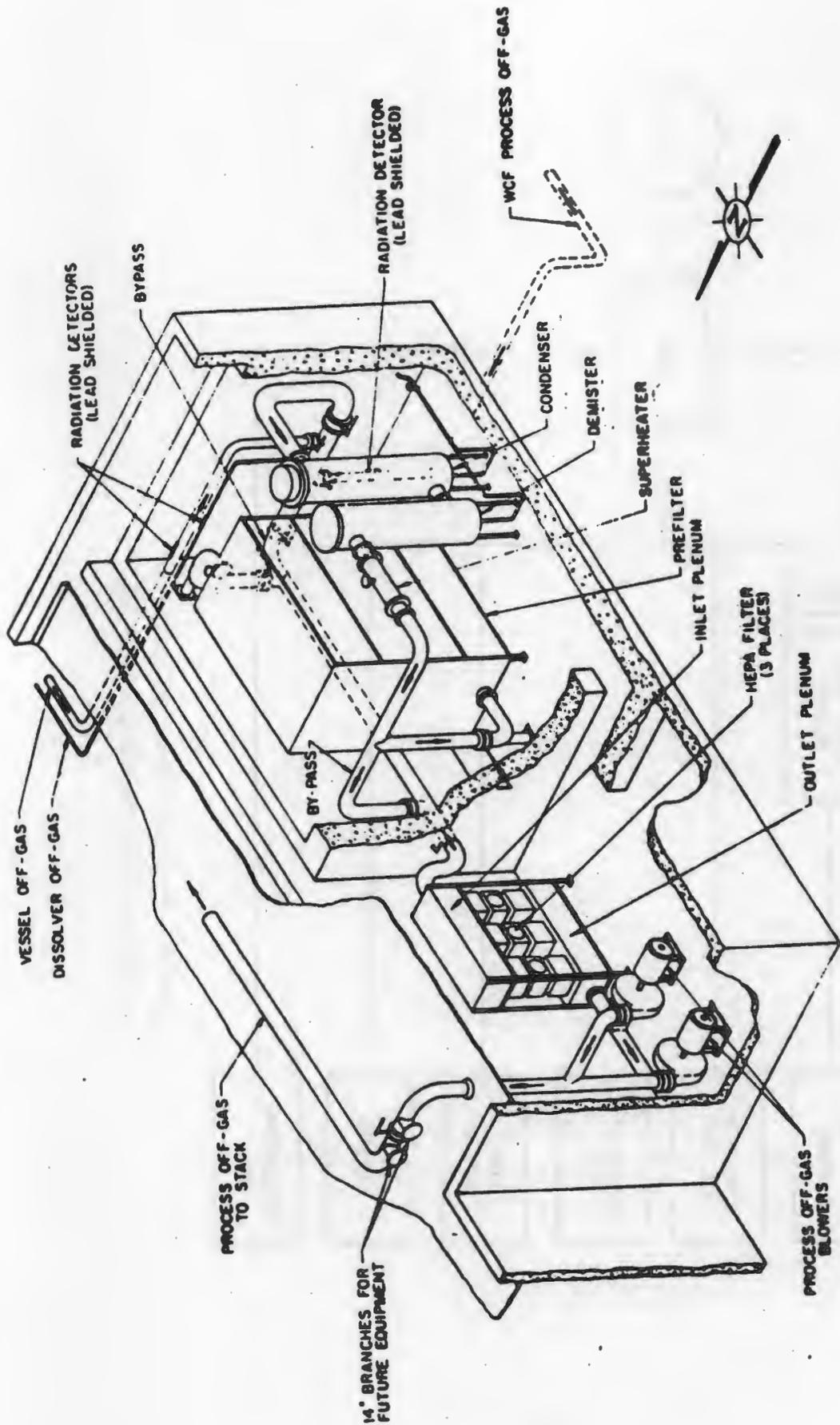


Figure 7. Isometric View of APS Process Off-Gas Area

Stainless steel clad fuels are dissolved in nitric acid by an electrolytic process. The dissolver off-gases are routed to the CPM-DOG at a flow rate of about 50 cfm. The sample point would be at the CPM-DOG sampling location.

The vessel off-gas system (VOG) vents all vessels containing radioactive solutions, except the dissolvers. Maximum flow from the VOG system is about 1000 cfm. The cleanup system is similar to the Zr-DOG and Al-DOG systems. The VOG is further cleaned at the APS after being combined with the Al dissolver and WCF off-gases. The particle sampling station will be immediately downstream of the VOG cleanup system.

The WCF exhausts about 2000 cfm of process off-gas containing NO_x at 90°C . The effluents pass through an extensive cleanup system consisting of cyclones, a scrubber, adsorbers, and HEPA filters. After combining with the vessel and Al dissolver off-gases, the WCF off-gases are further cleaned by the APS. Some particle sizes have been determined throughout the system, except after the HEPA filters. Sampling for this project would be close to the WCF HEPA exhaust and before the APS.

The ventilation air flows at a rate of about 90,000 cfm or 90% of the total stack flow and consists of room air from many of the ICPP buildings. This air is sometimes contaminated due to in-leakage from process areas, particularly at the WCF. Prefilters and HEPA filters are used to clean the ventilation air at the APS before it exhausts to the stack. The proposed sampling points would be before the APS.

3. Project Plan

This project will involve three phases. The first phase will determine the feasibility of installing the samplers at the previously discussed sample points.

The next phase will include an evaluation of all particle size samplers and instrumentation and the adaptability of each to the sampling conditions. This evaluation will include cascade impactors, light-scattering photometers, and laser spectrometers. For best statistical data, it is hoped that the same type of instrument can be used at each location. However, sampling accessibility and potential for instrument contamination will dictate the degree to which some equipment should be expended.

The last phase will involve formulating a sample program to include sampling frequency, coordinating of plant process operation and taking of samples, evaluation of data, and issuance of a final report.

IX. REFERENCES

1. ANSI-N510, Testing of Nuclear Air Cleaning Systems, American National Standards Institute, New York (1975).
2. C. A. Burchsted, A. B. Fuller and J. E. Kahn, Nuclear Air Cleaning Handbook, ERDA 76-21 (1976).
3. ANSI N1011-1972, Efficiency Testing of Air-Cleaning Systems Containing Devices for Removal of Particles, New York (1972).
4. Letter, G. E. Lohse to J. A. Buckham, Allied Chemical Corp. - Idaho Chemical Programs (March 18, 1976).
5. B. G. Shuster and D. J. Osetek, The Use of a Single Particle Intracavity Laser Particle Spectrometer for Measurements of HEPA Filters and Filter Systems, Proceedings of the 14th ERDA Air Cleaning Conf., ERDA Report CONF 760822 (February 1977).
6. IAEA, A Basic Toxicity Classification of Radionuclides, International Atomic Energy Agency, Vienna, Austria (1963).
7. J. O. Blomeke, C. W. Kee and J. P. Nickols, Projection of Radioactive Wastes to be Generated by the U.S. Nuclear Power Industry, ORNL-TM-3965 (1974).
8. W. A. Emel, D. C. Hetzer, C. A. Pelletier, et al., An Airborne Radioiodine Species Sampler and Its Application for Measuring Removal of Large Charcoal Adsorbers for Ventilation Exhaust Air, Proceedings of the 14th ERDA Air Cleaning Conference, ERDA Report CONF 760822 (February 1977).
9. R. C. Girton and D. T. Pence, Effluent Monitoring Associated with Fluidized-Bed Waste Calciner Facility Operations, Proceedings of the 2nd AEC Environmental Protection Conference, Wash-1332 (July 1974).
10. "Standards of Performance for New Stationary Sources," Environmental Protection Agency, Federal Register, vol. 36, No. 247, Part II (December 23, 1971).
11. Technical Progress Report, Allied Chemical Corporation - Idaho Chemical Programs (March 1973).
12. Technical Progress Report, Allied Chemical Corporation - Idaho Chemical Programs (October 1973).
13. Technical Progress Report, Allied Chemical Corporation - Idaho Chemical Programs (January 1974).

14. Technical Progress Report, Allied Chemical Corporation - Idaho Chemical Programs (February 1974).
15. Technical Progress Report, Allied Chemical Corporation - Idaho Chemical Programs (June 1974).
16. R. C. Girton, D. T. Pence and L. T. Lakey, The Stack Monitor System at the Idaho Chemical Processing Plant, ICP-1034 (September 1973).
17. P. I. Nelson, Final Safety Analysis Report for the Atmospheric Protection System, ICP-1082 (June 1976).