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Technology Survey for Real-Time Monitoring of Plutonium in a Vitrifier Off-Gas System

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TECHNOLOGY SURVEY FOR REAL-TIME MONITORING OF PLUTONIUM IN A VITRIFIER OFF-GAS SYSTEM

by

John M. Berg and D. Kirk Veirs

ABSTRACT

We surveyed several promising measurement technologies for the real-time monitoring of plutonium in a vitrifier off-gas system. The vitrifier is being developed by Westinghouse Savannah River Corp. and will be used to demonstrate vitrification of plutonium dissolved in nitric acid for fissile material disposition. The risk of developing a criticality hazard in the off-gas processing equipment can be managed by using available measurement technologies. We identified several potential technologies and methods for detecting plutonium that are sensitive enough to detect the accumulation of a mass sufficient to form a criticality hazard. We recommend gross alpha-monitoring technologies as the most promising option for Westinghouse Savannah River Corp. to consider because that option appears to require the least additional development. We also recommend further consideration for several other technologies because they offer specific advantages and because gross alpha-monitoring could prove unsuitable when tested for this specific application.

INTRODUCTION

Westinghouse Savannah River Corp. plans to install a vitrification system in an F-canyon hot cell at the Savannah River Site (SRS) in FY 1997. The corporation intends to use the system in FY 1998 to vitrify a solution containing americium (Am) and curium (Cm) so that the actinide material can be shipped to Oak Ridge National Laboratory. Later the same year, the vitrification system is to be used for demonstrating plutonium (Pu) vitrification for disposition. One of the design criteria for the Pu vitrification states that all Pu-handling components must be criticality-safe or must be prevented from receiving enough fissile material to form a critical mass. The current design includes an off-gas collection system with one tank that is not criticality safe. The design must be modified either to make all tankage criticality safe or to ensure that problem quantities of fissile material do not reach the non-criticality-safe tankage. Los Alamos National Laboratory is tasked with recommending options for the latter type of design change by incorporating one or more monitoring systems for fissile material. This report summarizes the Los Alamos team’s survey of available technologies and provides our recommendations based on that survey.

We were guided primarily by the design information that we obtained in a series of meetings at SRS on August 10, 1995. At that time the system design was not final, and some details were not established. Thus some recommendations depend on the correctness of our assumptions about the final design and operating parameters. In some cases we tried to present several monitoring options, the applicability of which will depend on the final design details.

This report discusses some specific implementations of each type of monitoring technology identified for consideration. However, the background research for this report did not include a comprehensive, critical survey of all available manufacturers or developers. Thus, this report is not an exclusive endorsement of the implementations discussed here.

PRESENT SYSTEM DESIGN

The current conceptual design calls for the components shown in Fig. 1. However, there may not be adequate protection against the accumulation of fissile material in the tank labeled 17.2. Such an accumulation presents a criticality risk because tank 17.2 is not designed for safely holding quantities capable of forming a critical mass. The risk exists only for tank 17.2 because the other components have geometries that prevent formation of a critical mass.

Several options to deal with this design problem are under consideration. The first option is to dilute any entrained Pu so that its concentration in solution in tank 17.2 could never be high enough to form a critical mass. This option is incorporated into the schematic (Fig. 1) in the form of 443 added liters/hour of cooling water for diluting the Pu. Acidifying the added water may help keep Pu in solution. This option does not eliminate the problem, however, because Pu could precipitate and settle out of solution downstream of the settling tank. In this scenario a critical mass could assemble in the bottom of tank 17.2.

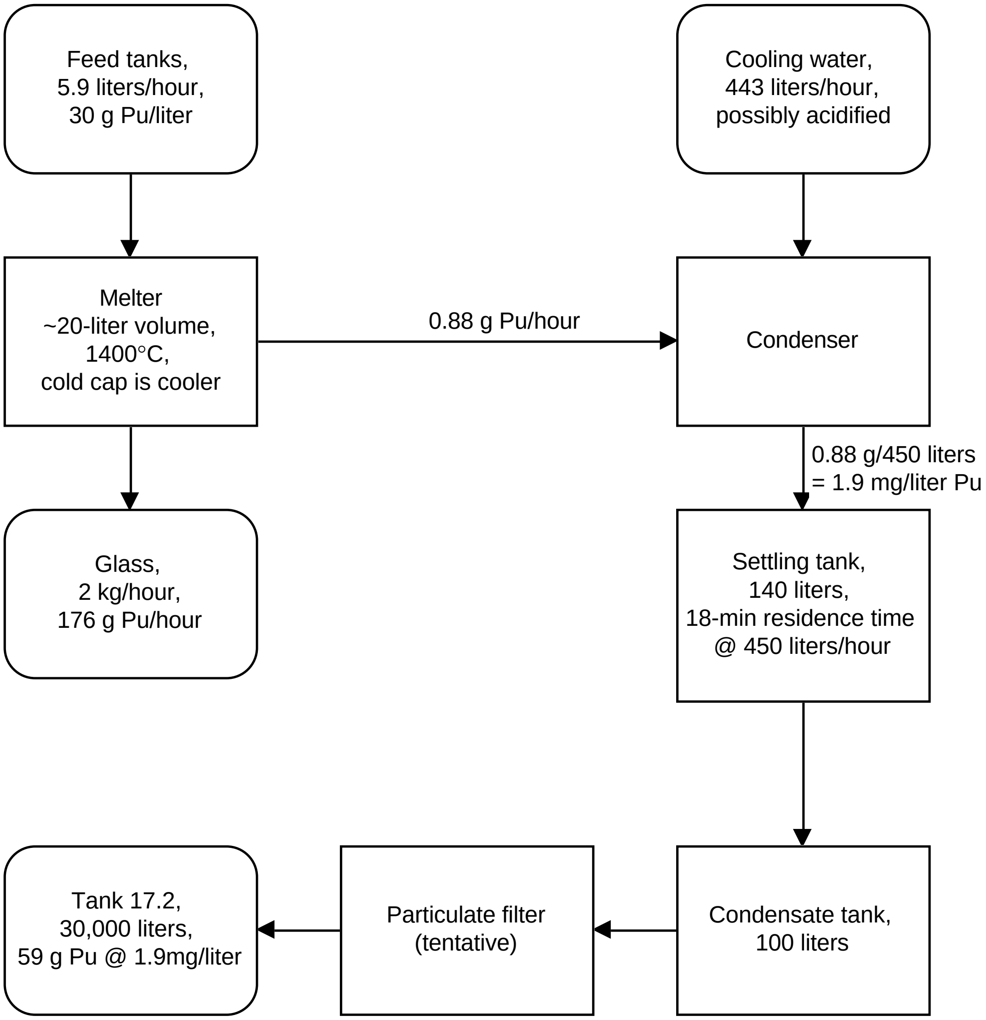

Fig. 1. Schematic of the planned vitrification system at the Savannah River Site.

A second option adds a filter upstream of tank 17.2 to catch particulates that get past the settling tank. The advantages of this option are that it is simple and passive, unless filter loading is high. One disadvantage is that a filter does not prevent dissolved Pu from reaching tank 17.2 and then precipitating. That scenario is possible because the residence time will be much longer in tank 17.2 than in the upstream components of the off-gas system, so kinetically slow precipitation might not occur until the solution reached that tank. Another disadvantage is that of filter maintenance, particularly if other solids such as silicates could form and clog the filter.

We were asked to evaluate technologies for a third option, that of monitoring the off-gas treatment streams for Pu so that the inventory of fissile material in tank 17.2 can be known at all times and the process can be altered if problem quantities begin building up. The additional advantage of this option is that system performance could be measured in real time, and operating parameters could be adjusted to reduce entrainment of problem components in the off gas.

SCENARIOS FOR PLUTONIUM TRANSPORT THROUGH OFF-GAS EQUIPMENT

We assume that plutonium leaves the melter and enters the off-gas system as particulates or liquid droplets entrained in the steam flow from the melter. The solids may dissolve again in the condensed and diluted aqueous solution into which the off gas flows. It is also possible that dissolved Pu may subsequently reprecipitate farther downstream. Figure 2 traces the possible transformations of Pu in the off-gas processing equipment and shows the resulting collection locations for the Pu. The boxes at the bottom of the chart indicate the four likely collection locations of the Pu that enters the off-gas system.

In one case Pu reaches tank 17.2 as suspended particulates and in a second case, as dissolved Pu. These two possibilities present potential criticality problems. Without precipitation, Pu in solution in tank 17.2 is not a criticality problem initially because the cooling water would dilute the condensate and thereby guarantee a Pu concentration too low for a criticality event to occur in a tank of any size or geometry. However, without some provision for monitoring and controlling the chemistry of Pu in tank 17.2, Pu could possibly precipitate some time after the Pu solution is introduced into the tank. Therefore, we assume that both cases—Pu as a suspended particulate and as dissolved Pu—are equally problematic.

The other two Pu collection locations identified in Fig. 2 are the results of Pu solids trapped upstream of tank 17.2. When Pu is trapped upstream, the inventory of Pu in tank 17.2 is reduced; however, both of these locations may later become sources of Pu flowing into tank 17.2 if the Pu becomes dislodged. While dislodging may be likely, we believe that such scenarios must be considered because of the risk associated with the failure to monitor a significant source of Pu that may enter tank 17.2.

The first point where Pu might collect and remain is the settling tank, which was incorporated in the current Savannah River Technical Center (SRTC) off-gas processing design in anticipation of entrained particulates in the off-gas stream remaining undissolved. The settling tank is a possible collection point for Pu unless the solids dissolve again or become resuspended. In that case they will contribute to the downstream migration of Pu to other collection points in the system. The second point where solids might collect is a particulate filter on the inflow into tank 17.2. The filter is not in the baseline off-gas processing design, but SRTC is considering its addition to reduce the total Pu flow into tank 17.2. Solids could dissolve after they are trapped on the filter, so we must consider the filter another possible delayed source of Pu flowing into tank 17.2.

MONITORING STRATEGIES AND PERFORMANCE CRITERIA

The total inventory of Pu in tank 17.2 must be monitored in order to assess the potential for a criticality hazard in that tank. The options, as we see them, are to monitor the contents of the tank, the inflow at the tank, or the off-gas system flow at one of several points upstream of the tank 17.2 inflow. Monitoring the tank contents may not be practical because the potential for the forming and settling of precipitates in the tank would make accurate sampling difficult. Monitoring the inflow to the tank would give the most reliable measure of the inventory because there is no potential for loss or gain of Pu between the monitoring point and tank 17.2; thus all the material in the tank would be sampled. Upstream monitoring is less attractive because Pu might be removed from the stream or added to the stream between the monitoring point and tank 17.2.

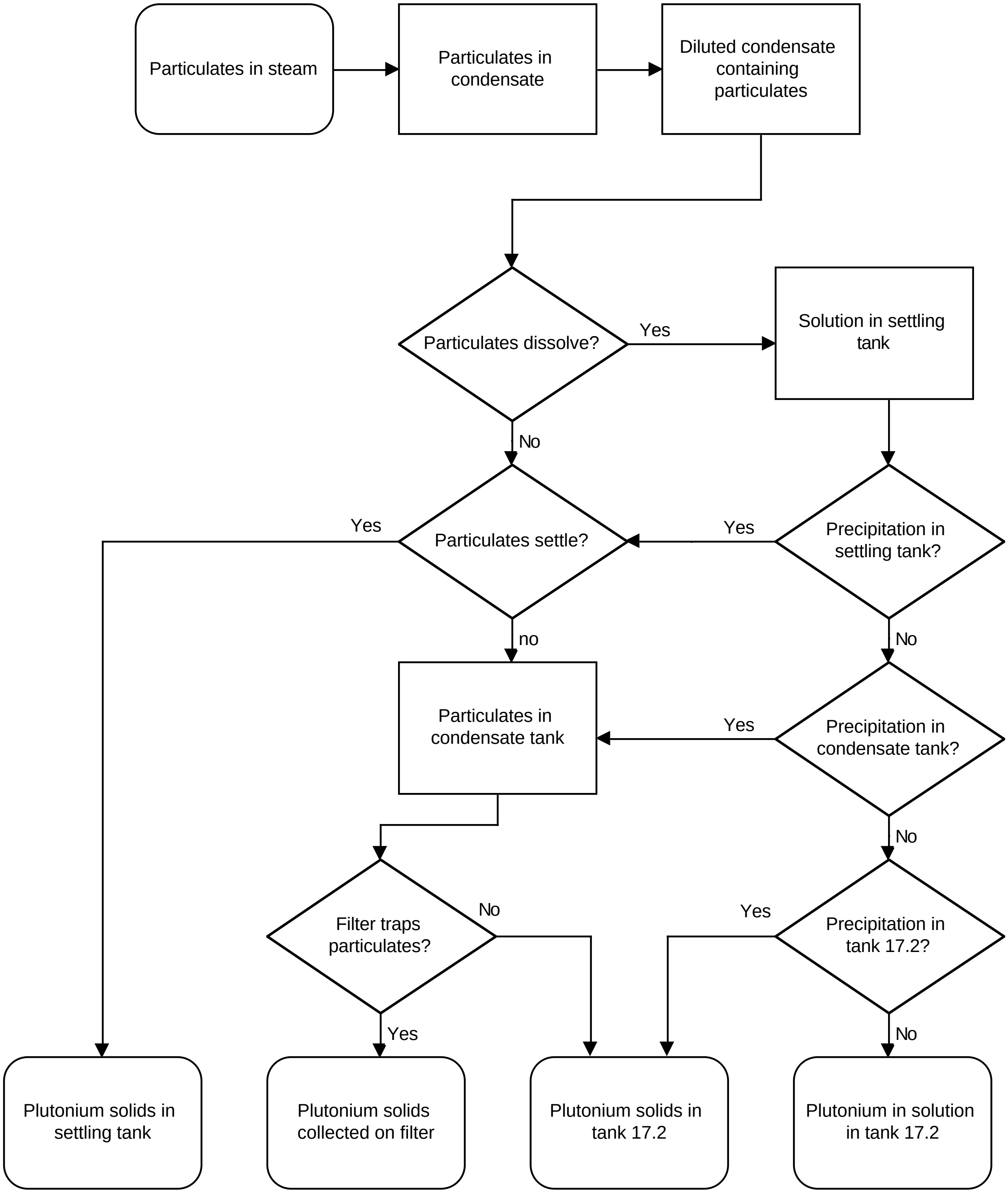

Fig. 2. Material flow paths of Pu through melter off gas. The boxes with rounded corners at the bottom of the chart represent the collection locations for Pu.

Upstream monitoring, however, offers some potential advantages. Information about total Pu entrainment in the off-gas stream could be obtained by monitoring upstream of any points at which Pu could be removed from the flow. Such monitoring should be done upstream of the settling tank. The information could be used for optimizing the overall system operating parameters in order to reduce Pu entrainment in the off-gas stream. A monitor in this position, in conjunction with a second monitor downstream of the settling tank, would enable operators to track the performance of the settling tank and its Pu inventory.

Monitoring positions downstream of the settling tank would all give equivalent results unless the proposed filter is installed between the condensate tank and tank 17.2. If a filter is present, monitoring upstream of the filter would give a maximum inventory of Pu reaching tank 17.2, or it could be used in conjunction with a monitor downstream of the filter to distinguish dissolved Pu from entrained particulates containing Pu and to monitor the performance of the filter.

There are also possible advantages to monitoring the off-gas vapor close to the melter. Sensitivity requirements might be easier to meet by monitoring before a large mass of cooling water dilutes the effluent stream, as is currently proposed. Also, some measurement techniques are more sensitive for vapor-phase samples. Monitoring the uncondensed steam from the vitrifier presents a practical difficulty: the monitoring system would have to tolerate acidic vapor at high temperature. The possibilities for overcoming this barrier are discussed in the section on individual monitoring technologies.

We recommend addressing the criticality concern by incorporating a monitor for total Pu directly at the inflow of tank 17.2, after any and all settling and filtration. Upstream monitoring above and below the settling tank should also be incorporated in order to track the performance and loading of the settling tank but should not be relied on for the criticality risk assessment of tank 17.2, except perhaps as a backup. Finally, vapor-phase monitoring of the off gas before condensation should be included in the design if cold testing of the melter suggests that entrainment may be at such low levels that it will not be possible to detect Pu after dilution of the condensate. These monitoring strategies depend on the availability of technologies capable of detecting Pu with sufficient sensitivity at each of these monitoring points.

The monitoring performance goal is the capability of measuring the Pu inventory in tank 17.2 accurately enough to ensure that a critical mass of Pu could not form in the tank under any conditions. To satisfy this goal, we set the conservative target of ensuring that the total inventory of Pu in tank 17.2 is less than 500 g. A 500-g mass of Pu cannot reach criticality even under optimum conditions of geometry, concentration and neutron moderation, and reflection by surrounding water.[1]

We assume that the vitrification system will operate in a batch mode and that tank 17.2 will accumulate off-gas processing material until it is full and then will be emptied completely. That is, no residue will remain to contribute to the Pu inventory in the tank during the next batch. In this case, monitoring the feed stream immediately above tank 17.2 and integrating over the batch will give the inventory of Pu in tank 17.2 at any time. If the assumption of complete emptying between batches is not valid, the monitoring schemes discussed in this report should be augmented by some form of Pu assay on the material removed from tank 17.2 at the end of each batch.

The sensitivity requirements are dictated by the need to measure Pu at the lowest concentration sufficient to accumulate 500 g in 30,000 liters, or about 16 mg/liter. The specific activity of ^{239}Pu is 63 mCi/g, so the activity of a 16-mg/liter solution is 1 mCi/liter. This is the required minimum detection capability for a condition of constant Pu concentration in the tank inflow. For a greatly varied flow, the dynamic range of the monitoring system would have to be great enough to measure accurately the highest concentrations in the inflow, and the sensitivity level would have to be at least as high as that required for the constant-feed-concentration scenario. A higher level of sensitivity would allow greater confidence at the required detection limit. We set a level of 0.5 mCi/liter as our minimum sensitivity requirement for monitoring the inflow of tank 17.2. This is half the activity level that would give 500 g of ^{239}Pu in 30,000 liters, allowing a safety margin of a factor of two in the measurements.

All the monitoring technologies discussed in this report measure concentration. For calculating the amount of Pu that passes the monitor, real-time flow measurements must be taken at the monitoring point. This report assumes that such flow-measurement capability will be installed and that it will sample at the same or greater frequency as that of the concentration-monitoring device(s) that is eventually chosen.

MONITORING TECHNOLOGIES AND TYPICAL IMPLEMENTATIONS

Alpha Detection

Alpha particle emission is the principal mode of decay of the ^{239}Pu nucleus. In the absence of other alpha-emitting radionuclides, gross alpha counting can give an accurate measure of the amount of Pu in a sample. The principal requirements for this application are sufficient sensitivity to Pu in solution and to entrained Pu particulates in water and an ability to operate reliably with little or no possibility of maintenance for a year or more.

In condensed phases, alpha particles must be detected close to their source because they are rapidly attenuated by any matter through which they pass. The mean free path of ^{239}Pu alpha emissions in water is only tens of microns. This is an important consideration because the best monitoring points, as identified in our analysis above, all require the detecting of alpha emissions in aqueous solution. On the other hand, alpha counting has an advantage: alpha particles that reach the detector can usually be detected with great efficiency because of their high energy and strong interaction with the detector material. For this report we investigated in detail two approaches to alpha counting in solution.

We also considered the technical feasibility of applying alpha counting to the off-gas stream before condensation. We were not able to identify any technologies that might work in the anticipated extreme environment of hot steam containing high HNO_3 concentrations, so options for that monitoring strategy are not analyzed in what follows. The barrier to such an application is survivability of the detector in the chemically hostile environment of the gas stream. Because the actual gas effluent may differ from what we surmised from our limited information about the design, we recommend revisiting the possibility of gas monitoring once the characteristics of this gas stream are better known.

Both the detection systems that we investigated for solution alpha detection use scintillation-based detection. One has recently been commercialized, and the other is in the pilot-scale testing stage of development. Both require some additional development before we can recommend their reliability for this monitoring application. They differ from each other in many technical aspects, but neither is unique in that each incorporates features that are available in part in other systems as well. We believe that each is close to the leading edge of technology for its type of system.

ZnS(Ag) Alpha Scintillator. At least two detectors using scintillation-based technology are commercially marketed. The commercialized system we investigated more thoroughly is manufactured by RIS Corp., Oak Ridge, Tennessee (model designation TUFF130AWM-H) and is based on scintillation in ZnS(Ag). The manufacturer of the system claims that it differs from other commercial detectors using that scintillator material in that its physical configuration minimizes attenuation of alpha particles by eliminating the protective window that is present in other sensors. Instead, RIS uses an epoxy coating to protect the sensor surface. RIS technical data sheets claim a sensitivity of 3.3×10^{-8} Ci/liter for ^{239}Pu, where the sensitivity is defined as that concentration that produces 10 counts per minute, given a detector background of <2 counts per minute.[2] This capability easily meets the sensitivity requirements we calculated for any of the liquid monitoring points.

Durability may be a concern, depending on the chemical composition of the off-gas stream. RIS product literature claims high chemical resistance, but the data supporting this assertion does not appear to be extensive. Chemical resistance down to pH 2.4 is claimed. RIS has not tested at lower pH or for specific resistance to nitric acid. Such testing would be necessary for evaluating the suitability of this detector for several of the monitoring points under consideration.

Despite the promising information about the RIS device, we recommend a thorough investigation of potential detectors from other manufacturers. Because the detection limit of the RIS device appears to be four orders of magnitude lower than that required for monitoring the tank-17.2 inflow, the advantage of the windowless design of the RIS device appears to be insignificant for this application. Personnel from SRS and Los Alamos have had experience with earlier detectors from other manufacturers. A similar detector was manufactured by IRT Corp. about 10 years ago. Our information is that the technology has since been sold to Eberline and may not be an actively manufactured product.[3]

Organic Polymer Scintillation Detectors. The operation principle for devices using organic polymer scintillators is the same as that described above. Alpha particles that reach the scintillating material are absorbed with great efficiency, and a portion of their energy dissipates as visible or ultraviolet photons emitted by the polymer. A portion of these photons are absorbed by the photocathode of a photomultiplier tube and are detected as voltage pulses, which are counted and recorded by detection electronics. The issues affecting sensitivity are the minimizing of alpha attenuation before the particles reach the scintillating material and the efficient collection and detection of the photons emitted. Interferences include scintillation that is due to cosmic rays and background beta, gamma, or neutron radiation, as well as thermally induced signals in the photomultiplier used to detect the photons.

Detection systems based on polymer scintillators have been in use for some time. Researchers at Los Alamos are currently pilot-testing a device that includes novel scintillator and photon detector geometries to increase detection efficiency and decrease background signal. The developers currently project a sensitivity of 1.2 nCi/liter for ^{239}Pu, where sensitivity is defined as the minimum alpha concentration that will produce a signal three standard deviations above the background signal.[4] Sensitivity to nonalpha scintillations is improved by efficient photon collection and the improved pulse height discrimination that high collection efficiency makes possible. Researchers have not yet tested the device for performance in gamma or neutron fields such as those that might occur in the vitrifier operating environment.

This device and the technical innovations that it incorporates are not yet commercialized, but further development to test its performance under the conditions projected in the off-gas system could take place in the near future. The device offers greater sensitivity than does the RIS device described above, but it will require higher development costs before it can be used for this application. This device also offers an alternative alpha-counting technology that should be considered because it may have better chemical compatibility or be more resistant to interferences from background radiation than other scintillator detectors.

Alpha Detection by Air Ionization. It is also possible to detect alpha decay in a liquid by measuring the ions produced in air by alpha particles escaping into a head space over the liquid surface. Researchers at Los Alamos have demonstrated this technique and are pilot-testing it to monitor a stirred tank in a liquid waste treatment facility.[5] The sensitivity appears to compare with that of the solid-scintillator techniques described above. One potential advantage lies in chemical durability since wetted surfaces could be made of a corrosion-resistant alloy. Although more testing would probably be necessary for this technology because it is not fully developed for liquid applications, it has already seen considerable use as a way to measure alpha contamination in soil and air.

Gamma Counting

In addition to alpha decay, ^{239}Pu has characteristic gamma emissions. These emissions can be detected and used not only for measuring Pu concentration, but also for discriminating Pu from other gamma emitters based on the gamma energy. Such a detection system has been useful for monitoring actinide separations processes.[6] In that application the sensitivity to Pu is only about 0.1 g/liter, which would not meet our monitoring requirements. However, since our investigation of the technique has not been extensive, it should not be completely ruled out at this point. Gamma counting may hold promise for monitoring upstream of the off-gas dilution because of the higher Pu concentrations there. It is advantageous that the sensor is not in physical contact with the liquid being monitored, so chemical compatibility would not be a problem.

Elemental Analysis by the Emission of Optical Photons

Atomic Emission from Laser-Generated Plasmas. Laser-induced plasma spectroscopy, laser-induced breakdown spectroscopy, and laser spark spectroscopy are all fundamentally the same analytical technique; they are based on the ability of a focused laser beam of moderate power to generate electric fields high enough to convert matter into a plasma in the laser’s focusing region. Elements in the plasma emit photons of characteristic energies as they recombine with plasma electrons and decay from highly excited electronic states to their ground states. By collecting these photons and analyzing them in a spectrometer, researchers can determine the elemental composition of the plasma. The system must be calibrated for the elements being analyzed, and their physical form must not deviate significantly from the form that was used for calibration. Both plasma generation and subsequent photon emission characteristics are sensitive to the form of the material being analyzed.

The applicability of the emission technique to this off-gas monitoring problem varies according to the monitoring strategy. For alpha monitoring, we sampled the liquid flow at the inlet of tank 17.2. Application of laser-generated plasma techniques at that potential sampling point is problematic. The sampling of bulk liquids with this technique showed low sensitivity for uranium. However, plasma generation at a liquid surface appears more promising, having displayed a sensitivity for uranium in solution of 0.1 g/liter (Ref. 7). The sensitivity to plutonium should be similar. This measurement does not meet the sensitivity requirement we established for detecting 500 g of Pu in 30,000 liters (0.017 g/liter), but it is included in this report because further investigation may show that increased sensitivity is achievable or that we have been overly conservative in estimating the sensitivity requirement. The application of this technique to the preferred sampling point would require that the feed to tank 17.2 pass through a sampling chamber with an air head space where the analysis could take place.

Laser-generated plasma emission spectroscopy may be better suited for sampling the melter's vapor-phase emissions before condensation than any of the other techniques that we examined for this report. Development of this suite of techniques for quantitative elemental analysis of aerosols has been going on for some time.[8]

Recently, Flower and colleagues at Sandia National Laboratories, in collaboration with researchers at Clemson University, bench-tested laser spark spectroscopy, as the technique is sometimes called, as a monitoring technique for quantitative elemental analysis of particulates entrained in a vitrification system off-gas stream.[9] They analyzed the melter effluent for metals regulated by the Clean Air Act, but the results should be similar for plutonium. Their sensitivity data ranged from 1 $\mu g/m^3$ for lead to 0.001 $\mu g/m^3$ for manganese.

Converting these sensitivities to the flow conditions from the proposed melter requires some assumptions on our part. First, we assume that the gas stream is composed entirely of steam and entrained particulates, that is, that no air or other gas is flowing through the system. Second, we assume a temperature of 200°C for the steam and a pressure of 1 atm. Superheated steam under these conditions has a specific volume of 2120 cm^3/g (33.9 ft^3/lb) (Ref. 10). With water feeding into the melter at the rate of 5.9 kg/hour, this gives a flow rate of 0.5 m^3/min of superheated steam. Assuming a minimum detectable Pu concentration of 1 $\mu g/m^3$, a Pu flow of 0.5 $\mu g/min$ (30 $\mu g/hour$) could be detected in the steam if performance equal to that seen by Flower et al. could be achieved in this application.

This flow rate is much lower than the design Pu loss of 0.88 g/hour. From another point of view, if Pu were to flow through the off-gas system at this estimated minimum detectable rate of 0.5 $\mu g/min$, it would take 1900 years of continuous operation for 500 g of Pu to accumulate in tank 17.2 in the case where no Pu is captured before it enters that tank. We conclude that if the total entrainment of Pu in the off gas could be monitored this way and shown to be low, criticality concerns could be answered without concern about where Pu collected in the off-gas stream. More realistically, if researchers could show that the Pu entrainment is well below 0.88 g/hour, they could significantly reduce the cooling-water dilution factor while keeping the maximum Pu concentration low in the off-gas liquid stream.

If this monitoring strategy is used, design differences between the system that Flower et al. tested and the one proposed for SRTC would necessitate significant further testing. This is the only system that we have evaluated that has meaningful potential for monitoring the off-gas vapor.

Inductively Coupled Plasma Emission or Mass Spectrometry. Another suite of analytical techniques converts the sample to a plasma and then analyzes its elemental composition by atomic emission (or absorption) in the plasma or by mass spectrometry sampling of the plasma. These techniques have sensitivities at the level of parts per billion or better and are frequently used to monitor for toxic elements in order to meet discharge or drinking water standards. Their sensitivities are many orders of magnitude better than the minimum requirements of this application. However, these technologies are not promising for this application because the plasma generation source requires frequent maintenance. We do not recommend that they be pursued.

Electronic Absorption Spectroscopy

Electronic absorption of plutonium is potentially a useful analytical technique because it can give information about the chemical form of plutonium in addition to quantitative information. However, we expect the sensitivity limit of electronic absorption spectroscopy, based on observation of Pu f-f transitions, to be 0.05 g/liter with conventional instrumentation. That level is not sufficiently sensitive to detect the accumulation of 500 g in tank 17.2 unless the accumulation occurs in a few large doses or unless the monitoring is done upstream of a dilution of the stream. Detection techniques other than measuring the attenuation of a light beam traversing the sample have been developed for reaching higher levels of sensitivity. These techniques include photoacoustic spectroscopy and photothermal lensing spectroscopy. At present, these are research tools, and significant development work would be required to use them in an on-line analytical application. Because they are optical techniques and are therefore amenable to remote implementation, they should not be dropped from further consideration. Rather, they should be considered for future generations of vitrifiers.

Technology Summary

Table I summarizes our assessments of the capabilities of each technology that we considered in preparing this report. Several techniques have more than sufficient sensitivity to meet our estimated requirements for monitoring at the inlet of tank 17.2.

TABLE I. Critical Comparison of Technologies Detailed in This Report

Detection Method	Estimated Sensitivity[a] to ^{239}Pu	Advantages	Concerns
Alpha-counting/ZnS(Ag) scintillator	0.5 μg/liter solution	Commercially available; simple	α contaminant interferences, β,γ background response; not element specific
Alpha-counting/polymer scintillator	0.02 μg/liter solution	High sensitivity; possibly better chemical resistance than ZnS(Ag) system	α contaminants, β,γ background response; not element specific; some development required
Alpha-counting/air ionization	0.2 μg/liter solution	Possibly better chemical compatibility than scintillators; simple and inexpensive—could install several as backups	α contaminants, β,γ background response; not element specific; some development required
Gamma detector outside of pipe	0.1 g/liter solution	Best chemical compatibility because the sensor is outside the pipe; elemental and isotopic information	γ background; relatively poor sensitivity
Laser-generated plasma emission, liquid sample	0.1 g/liter solution	Element specific; fiber optics allow remote instrumentation	Must sample the liquid surface; relatively low sensitivity
ICP/mass spectrometry	<0.001 μg/ liter solution	Element and isotope specific; very high sensitivity	High maintenance requirements; usually an off-line technique
ICP/atomic emission or atomic absorption	1 μg/liter solution	Element specific; high sensitivity	High maintenance requirements; usually an off-line technique
Laser-generated plasma emission, gas sample	4 μg/m^3 to 1000 μg/m^3 for metals in air at ambient pressure	Could be used on vapor-phase off gas if the optical interface with the stack is worked out; some vitrifier off-gas testing has already been done	Some development required; sensitivity to Pu has not been tested
Absorption spectroscopy of Pu electronic transitions	0.05 g/liter	Sensitive to Pu chemical form	Relatively insensitive to total Pu; calibration will be complicated if Pu chemistry varies

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[a] The sensitivity estimates are those of the authors. They are based on available literature and conversations with the technology developers.

INSTALLATION ISSUES

Historical uses of the equipment through which the off-gas stream will pass may affect the suitability of the monitoring strategies that we are recommending. In particular, internal contamination of the sampling volume may compromise the ability of ionizing radiation detectors to reach the desired sensitivity levels. The planned use of this equipment for an Am/Cm vitrification campaign before the Pu vitrification may necessitate designing sampling flow paths that will not be used until the Pu runs. They would be isolated from Am/Cm processing flows for their protection from background-producing contamination.

Background gamma radiation also can produce spurious signals in alpha detectors. Savannah River personnel should estimate the level of gamma background to be seen in the final installation after the Am/Cm runs, and the candidate detection systems should be tested in comparable radiation fields in order to characterize their behavior. This work should be done as soon as possible so that any problems can be addressed while there is still time to change monitoring strategies if necessary.

In this report we have considered detection system sensitivity, but we have not addressed the dynamic range requirements of the Pu detection system or the dynamic ranges of the technologies we have reviewed. If the dynamic range of the detection system is inadequate, high Pu concentrations could overload the monitoring system and cause inaccurate readings. This complication is particularly possible in counting-based techniques such as alpha counting, where signal pulses may interfere with each other at high count rates. Further Savannah River investigation of the recommended monitoring technologies should include a consideration of their dynamic range requirements. Because sensitivities of the alpha-counting techniques far exceed the requirements of this application, one solution would attenuate the alpha particles before they reach the detector so that the usable detection range is shifted to higher Pu concentration levels. Another solution would apply several detectors with different sensitivities to span the full range of possible Pu concentrations if no single detector could be shown to have adequate dynamic range.

Finally, we reiterate that real-time flow-measurement capability at the Pu-monitoring point is assumed. That capability is necessary to derive a total material throughput past the monitoring point.

RECOMMENDATIONS

Since several real-time analytical techniques have sufficient sensitivity to detect Pu at the levels required to ensure that a critical mass of ^{239}Pu does not form in tank 17.2, the choice depends on issues of reliability, robustness, and the relative usefulness of the information produced. We believe the best system to address the criticality issue alone is a liquid alpha-monitoring system on the pipe through which material must flow immediately before it enters tank 17.2. This monitoring system should be preceded by a particulate filter because its accuracy when measuring ^{239}Pu in suspended solids of unknown particle size will be difficult to ensure. Because commercial alpha detection systems seem to have sufficient sensitivity, we recommend them as the leading candidates for monitoring at this point. A thorough examination of all available commercial units should be done before the decision about what unit to purchase is made. We recommend purchasing at least two units for installation because they are not expensive (about $12,000) relative to the overall project and because loss of monitoring capability might be cause for ceasing operations.

If information about overall system performance is desired, several options are available. The simplest option is to monitor the stream into the settling tank with the same type of liquid alpha monitor that is used on the tank-17.2 inlet. In conjunction with the downstream monitor, this option will give information on the particulate loading and on the performance of the settling tank in removing those particulates. This option will also allow the operators to track the inventory of Pu in the settling tank. Serious consideration should also be given to the technique of laser-generated plasma emission for the optical monitoring of the vapor-phase before condensation. With this technique the nonradioactive components of the off gas could be monitored, and faster response to changing performance could be obtained.

Finally, we identified several alpha detection technologies under development and not yet commercialized. It is highly probable that the developers of these units could construct units for testing of materials compatibility and sensitivity to background radiation and that such testing could be completed during FY 1996. This avenue should be pursued because commercial units may prove unsuitable.

ACKNOWLEDGMENTS

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