ROBERT J. EMERY. Retention and Subsequent Release of Radioactivity from the Incineration of Wastes Containing Microspheres. (Under the direction of Dr. James E. Watson, Jr. and Dr. Daniel D. Sprau.)

## ABSTRACT

Incineration has been chosen as the preferred method for disposing of animal carcasses containing radioactive microspheres at East Carolina University. Routine surveys of ash from successive non-radioactive burns have shown significant contamination from previously incinerated microspheres. Past studies on microsphere incineration quantified the amount of activity retained in ash, but did not address any subsequent releases. This topic was not considered in earlier studies because, in most cases, the carcasses were placed in some type of container to facilitate recovery of ash, preventing contamination of the incinerator refractory.

Five sets of controlled burns were performed to quantify the subsequent releases of the microsphere radioisotopes Ce-141, Sn-113, Ru-103, Nb-95, and Sc-46. Each set consisted of three successive burns. The first burn of each set incinerated a non-radioactive carcass, the second burn, a radioactive carcass, and the third, a non-radioactive carcass. In all of the burns, the carcasses were placed directly on the incinerator refractory

floor, as is the standard procedure during normal operations.

The data collected document that significant subsequent releases of activity from the refractory do occur. This results in contamination of ash from non-radioactive burns. The actual amount of activity released varies with each isotope. These findings have caused East Carolina University to re-evaluate the quantities of radioactivity which may be burned, and to initiate a program to monitor all incinerator ash for gamma emitting radionuclides.

TABLE OF CONTENTS

# LIST OF FIGURES

# LIST OF TABLES

# ACKNOWLEDGEMENTS

This report represents the culmination of a extended academic pursuit, so I am indebted to many individuals for their help along the way.

I am very thankful for the guidance provided by Professor James Watson, who served as my Advisor. His constant accessability for comment and helpful attitude made him a true pleasure to work with.

I would also like to thank Dr. Alvis Turner for his editorial and scientific comments as a reader of this report. He provided an important service by viewing this work with a critical eye, especially from the perspective of hazardous waste implications.

A special thanks goes to Dr. Daniel Sprau for his assistance with this project. He acted as an on-site advisor for this project and served as a reader of this report. He has enthusiastically supported all of my educational endeavors, and for this, I am eternally grateful.

The support offered by the East Carolina University Office of Radiation Safety, as well as the Administration of the East Carolina University School of Medicine is also greatly appreciated. Dr. Dean Hayek, Associate Dean, ECU School of Medicine, has been especially supportive of my efforts. Many other individuals, too numerous to mention here, gave assistance with technical problems, printing, graphics, and the like.

In addition, I would like to thank my good friend Ralton

Harris for his help. Ralton provided a critical review of this work in its early stages, and was always available to lend his expertise on the various environmental monitoring problems that I encountered.

Finally, I would like to thank my fiancee Maureen Triller, whose support and encouragement throughout the completion of this project has been unbelievable. I hope that some day I can be as helpful to her.

# I. INTRODUCTION

## A. Purpose of Project

Incineration has been chosen as the preferred method of disposal for animal carcasses containing radioactive microspheres at the East Carolina University School of Medicine. The routine monitoring of incinerator ash from non-radioactive burns has revealed significant contamination from previously incinerated microsphere isotopes. Several published studies have documented ash retention of radioactivity, but none have addressed these apparent subsequent releases.

This study will attempt to document the successive subsequent releases of radioactivity from the incineration of microspheres by performing a series of controlled burns using both radioactive and non-radioactive animal carcasses. This data will be used to develop a comprehensive program for the monitoring and disposal of incinerator ash for East Carolina University, and establish limits for the amounts of radioactive microspheres which may be burned.

## B. Literature Review

### 1. Microspheres and Their Applications

#### a. Historical Development

Solid foreign particles can be injected into the bloodstream and used to measure a variety of natural and induced phenomenon. This technique has been used since 1909 to evaluate such

parameters as the distribution of blood flow, cardiac output, and organ shunting.[1] The ability in recent years to accurately regulate the size of these particles and to incorporate radioactive isotope labels has created a marked increase in their popularity as a research tool.[2] Radioactive tracers, commonly called "microspheres" assist researchers in the detection and quantification of particle retention in various tissues, hence indicating flow patterns and destinations of different blood components.

The first radioactive foreign particles used for circulatory studies were produced by exposing small glass beads to a neutron flux which transformed the stable sodium in the glass into radioactive Na-24.[3] Intended to imitate red blood cells once injected in the animal, these miniature glass beads were too heavy and therefore did not accurately mimic the behavior of normal erythrocytes.[4] Microspheres made of ceramic were also produced, but these were also deemed inappropriate because of their tendency to quickly settle in a liquid medium.

---

[1]Heymann, M.A., Payne, B.D., Hoffman, J., and Rudolph, A.M., "Blood Flow Measurements With Radionuclide-labeled Particles", Progress in Cardiovascular Diseases, Vol. XX, No. 1, 1977, p. 55.

[2]Heymann, M.A., et. al., p. 55.

[3]Grim, E., Lindseth, F.O., "Distribution of Blood Flow to Tissues of the Small Intestine of the Dog.", University of Minnesota Medical Bulletin, No. 30, 1958, pp. 138-145.

[4]Heymann, M.A., et. al., p. 55.

A wide variety of plastic microspheres were subsequently developed. Early versions resulted in batches of microspheres with wide variations in particle size and the tendency to permit leaching of the radionuclide label.[5] Any results obtained from experiments with these tracers needed to be viewed with caution due to the large degree of error introduced by these characteristics.

Advances in manufacturing technology have resulted in microspheres of a constant particle size and in a form that prevents leaching. These current radioactive tracers are also available with different isotope labels so as to permit the use of several different microspheres in one experiment.

b. Physical Description

One of the most popular nuclide-labeled microspheres in use today is the insoluble carbonized plastic tracer microsphere produced by the DuPont NEN Research Products, Boston, Massachusetts. These tracers are used exclusively at the East Carolina University School of Medicine. Marketed under the name "NEN Trac Microspheres", these tracers are attractive for use in circulatory studies because:

---

[5]Rhodes, B.A., Zolle, I., Buchanan, J.W., et al, "Radioactive Albumin Microspheres for Studies of the Pulmonary Circulation.", Radiology, Vol. 92, 1969, pp. 1453-1460.

- The microspheres have a specific gravity of approximately 1.3, which is comparable to that of whole blood (1.05).[6]

- The microspheres are available in standard diameters ranging from 10±2 to 50±5 microns. This enables researchers to select a microsphere size which correlates with the blood constituent of interest.[7]

- Ten different gamma emitting isotopes are available, permitting the use of several different tracers with energies discrete enough for easy analytic detection and segregation (see Table 1). They are: Gd-153, Co-57, Ce-141, Cr-51, Sn-113, In-144m, Ru-103, Sr-85, Nb-95, and Sc-46. The half-lives and principal photon energies of each isotope are listed in Table 2. When used in combinations, the manufacturer recommends that a minimum of 100 keV energy separation be maintained between principal photon energy peaks.[8]

---

[6]NEN-Trac Microspheres Catalog, E. I. duPont De Nemours & Co. Biotechnology Systems Division, Boston, Mass. 1986.

[7]New England Nuclear Research Products Catalog, E. I. duPont De Nemours & Co., Boston, Mass., 1988. pp. 137-139.

[8]New England Nuclear Research Products Catalog, 1988, pp. 137-139.

Table 1

Half Lives and Principal Photon Energies of Gamma Emitting Radionuclides Used in Microspheres

| Radionuclide | Half Life | Photon Energy | Photon Abundance |
|---|---|---|---|
| Gd-153 | 242 days | 97-103 keV | 55% |
| | | (also 41 keV) | (100%) |
| Co-57 | 271 days | 122-136 keV | 98% |
| Ce-141 | 32.5 days | 145 keV | 48% |
| Cr-51 | 27.8 days | 320 kev | 9% |
| Sn-113 | 115 days | 393 keV | 64% |
| | | (also 255 keV) | (2%) |
| In-114m | 49.5 days | 192 keV | 17% |
| | | (also 558 keV | 3.5% |
| | | and 725 keV) | 3.5% |
| Ru-103 | 39.8 days | 497 keV | 88% |
| | | (also 610 keV) | (6%) |
| Nb-95 | 35 days | 765 keV | 100% |
| Sr-85 | 64.7 days | 514 keV | 100% |
| Sc-46 | 84 days | 889 keV | 100% |
| | | 1.120 MeV | 100% |

- The radionuclide tracer is incorporated into the plastic matrix of the sphere to prevent leaching. Quality control testing is performed at the factory to ensure tracer integrity.[9]

c. Quality Assurance

The microsphere shipments received at the East Carolina University School of Medicine are suspended in a 10% by volume polyoxethylene 80 sorbant monooleate (Tween 80).[10] Tween 80, a detergent, is added to prevent aggregation of the microspheres. (It should be noted that it is also possible to receive microspheres in a dry state.) After the standard check-in procedure by the Office of Radiation Safety, which is performed on all radioactive shipments, several other verifications are conducted by the actual user of the microspheres.

It is of paramount importance to confirm the actual size of the microspheres received. A standard accepted procedure established by Heymann, et. al. in 1977, calls for microscopic examination of several grab samples.[11] Diameter measurements are

---

[9] New England Nuclear Research Products Catalog, 1988, pp. 137-139.

[10] New England Nuclear Research Products Catalog, 1988, pp. 137-139.

[11] Heymann, M.A., Payne, B.D., Hoffman, J., and Rudolph, A.M., "Blood Flow Measurements With Radionuclide-labeled Particles", Progress in Cardiovascular Diseases, Vol. XX, No. 1. 1977, pp. 55-79.

made on 100 to 200 tracers, and their ranges plotted on a histogram. The acceptable range of diameter size is directly dependent upon the size of the microsphere ordered. The diameter range data provided by New England Nuclear Company through their Technical Services Division is listed in Table 2.

While under the microscope, a check is made for any clumping or aggregations of microspheres. Adhesions caused by bacterial growths in the solution can be broken up by vigorous shaking. Of greater concern is "bridging" that can occur between the spheres. Thin plastic strips sometimes still connect the spheres as a result of the manufacturing process. These bonds are not easily broken, and are cause for rejection of the batch.

A gamma spectral analysis is performed on each separate batch of microspheres to check for radionuclide specificity. Any cross-contamination or mislabeling will produce extraneous spectral peaks.

Soaking the spheres in a 0.5% Tween 80 isotonic saline solution for 24 hours is the technique used to check for isotope leaching. After the soaking period, the microspheres are filtered out, and the remaining solution counted for any residual radioactivity.

The final check is for specific activity. A sample of the microspheres is placed on a known field size, such as ordinary graph paper. A physical count is made of the number of spheres present in the field, and the total activity of the field is determined. By dividing the total activity by the number of

Table 2

NEN-Trac MICROSPHERE SIZES

| Nominal Size and Range | Approximate Number of Tracers/mg |
|---|---|
| 10±2 microns | 1,500,000 |
| 15±3 microns | 450,000 |
| 25±5 microns | 80,000 |
| 35±5 microns | 35,000 |
| 50±5 microns | 12,500 |

microspheres present, an activity per bead can be obtained.

Upon completion of these verifications, the microspheres are ready for use.

d. Experimental Applications

Since microspheres can be applied in a variety of experimental situations, it would be very difficult to outline all of the different applications. There are, however, some procedures which are common throughout most research applications.

Normally an animal is anesthetized and a catheter is inserted into the left atrium of the heart. The left atrium is the location of preference because of the high mixing of blood that occurs in this area. An initial injection of a well-vortexed 1 ml solution containing microspheres is made as a control. The choice of microsphere isotopes is randomized to ensure that no selective retention occurs over a series of experiments. The microsphere injection is followed by a heparinized saline flush. During, and immediately following the introduction of the microspheres, a series of ten 2-3 ml blood samples are drawn. These ten samples are counted using a NaI crystal connected to a multichannel analyzer to verify the injection and subsequent deposition of the microspheres into the tissue. Table 3 shows an example of ten blood samples taken at 12 second intervals depicting the introduction of radioactivity into the bloodstream and the total deposition of the radioactive material into the tissue in less than a minute. The data in this table was obtained from an actual

Table 3

**Blood Samples Withdrawn at 12 Second Intervals Immediately After Injection of Microspheres to Verify Tracer Deposition**

| time after microsphere injection (in seconds) | (in counts per minute) | | | | |
|---|---|---|---|---|---|
| | Ce-141 | Sn-113 | Ru-103 | Nb-95 | Sc-46 |
| 0 | 50 | 20 | 23 | 14 | 29 |
| 12 | 117 | 37 | 29 | 17 | 28 |
| 24 | 45063 | 8476 | 4050 | 6010 | 4943 |
| 36 | 14427 | 6017 | 2793 | 3207 | 1222 |
| 48 | 1360 | 634 | 429 | 378 | 156 |
| 60 | 334 | 159 | 113 | 83 | 53 |
| 72 | 533 | 127 | 60 | 53 | 64 |
| 84 | 271 | 86 | 44 | 38 | 81 |
| 96 | 420 | 102 | 48 | 35 | 73 |
| 108 | 397 | 124 | 45 | 40 | 63 |
| 120 | 284 | 108 | 48 | 44 | 70 |

experimental application of microspheres.

After the control injection is completed, any of a myriad of stimuli may be performed on the animal. During various stages of the experiment, different microsphere isotopes are injected, and the same procedure as detailed previously is followed. Using isotopes with different energies for each injection enables a researcher to compare tissue retention of blood components at various stages in the procedure.

At the end of the experiment, the animal is sacrificed, and the tissues of interest excised. The typical method of analyzing the tissue is counting on a multichannel analyzer system with a NaI(Tl) crystal, with the regions of interest on the counter set for the energies of the gamma rays expected.

Both the carcass and the excised tissue are contaminated with radioactive material and, as such, require special attention for their disposal.

e. Wastes Generated

At the East Carolina University School of Medicine, the animal carcasses and any excised tissues are brought to the Radiation Safety Laboratory upon completion of the experiment. Until their ultimate disposal is accomplished, the carcasses and tissues are temporarily stored in freezers. The Office maintains at least 10 freezers for the storage of these types of wastes at all times.

A small amount of solid waste is also generated in the course of the experiment. This consists mainly of contaminated gloves,

absorbent paper, tubing, etc.

Currently about 50-70 animal carcasses are being generated per year. It is estimated that the annual production of animal carcasses containing microspheres at East Carolina University will increase to 150-200 in the next fiscal year.

## 2. Review of Available Disposal Methods for Radioactive Carcasses

Three possible methods of disposal for these low-level radioactive biological wastes are currently available. The Office of Radiation Safety considered (1) burial at a low level radioactive waste disposal site, (2) storage for decay, or (3) incineration. Incineration was chosen as the preferred method of disposal for the following reasons:

- **Cost.** The present approximate cost for the burial for one 55 gallon drum of low-level radioactive biological waste is $400. Due to the stringent packaging requirements for biological wastes, approximately two animals (usually dogs or pigs) can be placed in each drum. At a generation rate of 200 animals per year, the annual cost for burial of microsphere-contaminated animals alone would be $40,000.

- **Availability of a Burial Site.** It is questionable whether radioactive waste generators in the State of North

Carolina will have uninterrupted access to any low-level radioactive waste disposal site in the future. The permanent closing of the Barnwell, S.C. facility is scheduled for 1992. Disagreements over the eventual siting of a new facility could delay any progress towards a long term solution to the low-level radioactive waste (LLRW) disposal problem.

- **Lack of Storage Space.** At most educational institutions, storage space (especially freezer storage space) is at a premium, and the East Carolina University School of Medicine is no exception. The space that would be needed to adequately store contaminated animal carcasses for a minimum of seven half-lives is simply not available.

- **Possible Packaging and Transportation Violations.** The regulations concerning packaging and shipment of LLRW are confusing at best, and subject to constant change. The possibility of violating some State, Federal, or contractors regulations during any phase of the packaging or shipment procedure can result in the loss of permission to continue shipping waste to a specific disposal site.

- **Volume Reduction.** The reported volume reduction of animal carcasses due to incineration is approximately 90-95%.

Incineration would greatly reduce the volume of waste to be shipped for burial, which in turn would reduce waste disposal costs.[12]

- **Other Hazardous Wastes.** Incineration is the preferred method of disposal for other types of wastes which may be generated in association with microspheres such as biohazardous, pathological, infectious, and chemical wastes. Regulations regarding the disposal of "mixed wastes", when completed, may impose further restrictions on generators.

- **Change of Waste Type Classification.** Incineration of animal carcasses transforms the classification of the waste from a biological to a solid. Solid radioactive wastes are subject to fewer packaging requirements.

- **Currently Available and Licensed Incinerator.** The Office of Radiation Safety has been using the incinerator at the East Carolina University School of Medicine since 1981 for the processing of low-level radioactive waste.

---

[12]Gregory, W.D., and Maillie, H.D., "Incineration of Animal Radioactive Waste: A Comparative Cost Analysis", Health Physics, Vol. 29, No. 9 (Sept.), 1975, pp. 389-392.

## 3. Previous Microsphere Incineration Studies

With the advent of the use of incineration as a technology for LLRW volume reduction, several researchers began to examine the possibility of burning microsphere-laden waste. When applying for a license to incinerate radioactive waste, it must be assumed that 100% of the activity is exhausted into the atmosphere for the purposes of calculating discharges of radioactivity. Preliminary results indicated that some of the activity in the form of microspheres was retained in the ash, so studies were performed in hopes of quantifying ash retention factors so that larger amounts of waste could be incinerated without exceeding established air concentration limits for various radionuclides.

Landholt, et. al. performed the first study published on the actual incineration of microsphere-ladened carcasses.[13] Eight animals containing Sc-46 labeled plastic tracers were individually placed directly into the incinerator and burned. The total ash from each of these burns was collected in a container and counted on a multichannel analyzer system with a 3" x 3" NaI(Tl) crystal. The mean percentage of retention of Sc-46 for the eight burns was 97.6$\pm$7.6%. Effluent monitoring was also performed, but no activity from the incineration of Sc-46 microspheres was reported. The total mass reduction ratio of the carcasses was reported to be 25:1. Contamination of ash from successive non-radioactive burns

---

[13]Landholt, R.R., Barton, T.P., Born, G.S., Morris, V.R., Vetter, R.J., Zimmerman, N.J., "Evaluation of a Small, Inexpensive Incinerator for Institutional Radioactive Waste", Health Physics, Vol. 44, No. 6, 1983, pp. 671-675.

was not addressed.

Brekke, et. al. monitored the stack effluent from the incineration of carcasses containing combinations of microspheres.[14] These carcasses were placed directly into the incinerator. No tray or container was utilized to collect the ash while the carcasses were burned. A modified EPA Method 5 stack sampling train was used to isokinetically remove a representative sample of the incinerator stack effluent. All of the radioactivity collected was retained on a particulate glass fiber filter located at the front of the sampling train. No significant activity was found in the HCl bubblers that followed the particulate filter. The mean percentage of released activity for the microspheres evaluated were: Sn-113, 12.3±4.9%; Gd-153, 5.0±2.5%; Nb-95, 4.8±1.1%; Co-57, 3.8±3.0%; and Ru-103, 16.5±5.0%. The Ru-103 release was assumed to be elevated due to the oxidation of the Ru metal to the volatile ruthenium tetroxide, $RuO_4$. No postulations were made concerning the apparent elevated release of tin. The radioactivity remaining in the incinerator ash was determined for only two nuclides. The mean ash retention for Nb-95 was 91±24%, and for Ru-103 was 58±8%. The ash retention for the other isotopes burned was not evaluated. Contamination of ash from subsequent burns was not discussed.

Finnegan, et. al. monitored both ash and effluent during the

---

[14]Brekke, D.D., Landholt, R.R., Zimmerman, N.J., "Measurement of Effluent Radioactivity During the Incineration of Carcasses Containing Radioactive Microspheres", Health Physics, Vol. 48, No. 3, 1985, pp. 339-341.

incineration of a mixture of isotopes.[15] The carcasses burned were placed on stainless steel trays to facilitate ash collection and removal. Only a gross range of 83-100% ash retention was reported for the isotopes Ce-141, Cr-51, Sn-113, Sr-85, and Sc-46. The stack effluent was sampled via a high volume air sampler with a particulate filter. No activity was reported to be found in the effluent by this method of stack sampling. The burns yielded a 95% reduction in weight. Ash from two subsequent non-radioactive burns was also collected and analyzed. Approximately 1 to 5% of the original activity burned was reported to be contained in these residues.

Classic, et. al. investigated solubility of radionuclides in residual ash resulting from the burning of radioactive carcasses.[16] During this experiment, microspheres containing various isotopes were burned while on stainless steel trays, and their percent activity retained in ash obtained. The results for these radioisotopes were: Sc-46, 95.4±1.1%; Co-57, 76.9±2.3%; Sr-85, 79.3±3.4%. This report also concluded that the ash-retained activity in an insoluble form.

---

[15]Finnegan, J.J., Miller,K.L., White, W.J., Bohner, K.R., "Incineration of Animal Carcasses Containing Gamma-Emitting Radioisotopes", Proceedings: 9th Biennial Conference of Campus Radiation Safety Officers, Columbia, Missouri, June 1983, pp. 20-22.

[16]Classic, K., Gross, G., Vetter, R.J., "Solubility of Radionuclides in Ash from the Incineration of Animals", Health Physics, Vol. 49, No. 6, 1985, pp. 1270-1271.

Van Swearingen evaluated the ash from three burns of Sc-46 and Sr-85 ladened microspheres.[17] Carcasses were placed on a bed of existing non-radioactive ash and burned. One gram ash samples were obtained from each burn and counted in a 2" X 2" NaI(Tl) detector connected to a single channel analyzer. The mean percentage of retention for Sc-46 was 79.7±12.9%; for Sr-85 86.3±32.8%. The study emphasized that problems exist in the exact determination of initial activity involved in the experiment.

In an attempt to better determine the exact amount of activity that was initially injected into the animal, Krueger and McLaughlin derived a syringe retention factor by counting several syringes while full of microspheres and then recounting after the expulsion of the contents.[18] The amount of activity in the full syringe minus the residual activity in the "empty" syringe was taken to be the true injected activity. Unfortunately, the actual syringe retention factor used was not included in the report. Krueger and McLaughlin also analyzed the remaining ash in a more detailed manner than in previous studies. Noting that the ash is in two distinct forms, powder and bone fragments, each constituent was analyzed separately. The bone samples did reveal some radioactivity, however the possibility of contamination from the

---

[17]Van Swearingen, F.L., "Incineration of Microspheres", Lecture Notes; Incineration of Low Level Wastes: 1985, Tucson, Arizona, March 21-23, 1985, pp. U-1 - U-5.

[18]Krueger, D.J., McLaughlin, J.E., "Residual Radioactivity in Ash from Incineration of Animal Carcasses Labeled With Radioactive Microspheres", Lecture Notes: Conference on the Incineration of Low Level Radioactive and Mixed Wastes, St. Charles, Ill., April 1987.

surrounding ash was not addressed. The results, published as Ash Retention Factors (ARF), were as follows: Gd-153, 67%; Co-57, 148%; Sn-113, 80%; Sr-85, 56%; and Sc-46, 92%. All carcasses were burned while resting on the floor of the incinerator. Although no subsequent releases of activity were investigated, the possibility of contamination of the refractory was mentioned. The unusually high retention factor for Co-57 was postulated to be caused by subsequent releases of activity which were trapped in the incinerator lining.

This report provides to-date, a comprehensive survey of the published data concerning microsphere incineration. Table 4 compiles all of the information available for ash retention values and stack effluent releases for the five isotopes burned at East Carolina University.

Table 4

Summary of Available Data:
Ash Retention and Effluent Concentrations (in Percent) for
Five Microspheres to be Incinerated at the
East Carolina University School of Medicine

| Study | | Microsphere Isotopes | | | | |
|---|---|---|---|---|---|---|
| | | Ce-141 | Sn-113 | Ru-103 | Nb-95 | Sc-46 |
| Landolt et. al., 1982 | in ash | | | | | 97.6±8% |
| | in air | | | | | |
| Brekke et. al., 1985 | in ash | | | 58±8% | 91±24% | |
| | in air | | 12.3±5% | 16.5±5% | 4.8±1% | |
| Finnegan, 1983 | in ash | 83-100% | 83-100% | | | 83-100% |
| | in air | | | | | |
| Classic et. al., 1984 | in ash | | | | | 95.4±1% |
| | in air | | | | | |
| Van Swearingen, 1985 | in ash | | | | | 79.7±13% |
| | in air | | | | | |
| Krueger et. al., 1986 | in ash | | 80% | | | 92% |
| | in air | | | | | |

## II. MATERIALS AND METHODS

The equipment used in this study included the East Carolina University School of Medicine incinerator, a modified isokinetic stack sampling train, a high efficiency particulate air (HEPA) filtered vacuum ash collection system, and a NaI(Tl) detector connected to a multichannel analyzer.

### A. Incinerator

The incinerator at the East Carolina University School of Medicine is an Environmental Control Products, Inc. Model 480E. This natural gas fired, double chamber, 350 pound per hour incinerator is manufactured primarily for the burning of Type IV wastes. (Type IV wastes consists of carcasses, organs, solid organic wastes containing up to 85% moisture and 5% non-combustible solids.) The specifications of the incinerator are listed in Table 5, and a schematic of the structure is shown in Figure 1. The incinerator was installed in 1980 on a 5 inches thick concrete floor of a room in the Utility Plant Building, located 500 feet from the main building of the Medical School. The primary and secondary chambers are both refractory lined. A 12 gauge galvanized steel rain hood was added after installation to protect the refractory lining of the secondary chamber.

The wastes to be incinerated are fed into the primary combustion chamber by a single ram, hydraulically operated feeder, and burned. The combustion products pass into the secondary chamber where they are mixed with a flame to complete the

## Table 5

## Incinerator Specifications

Model: Environmental Control Products, Inc. Model 480E

Cost: $60,000 (1980, estimate as part of total building)

Power: 6 HP, 280 Volts, 3 Phase

Fuel: Natural Gas

Primary Chamber Volume: 118 $ft^3$

Secondary Chamber Volume: 25 $ft^3$

Primary Chamber Temperature (maximum): 1800 $^oF$

Secondary Chamber Temperature (maximum): 1600 $^oF$

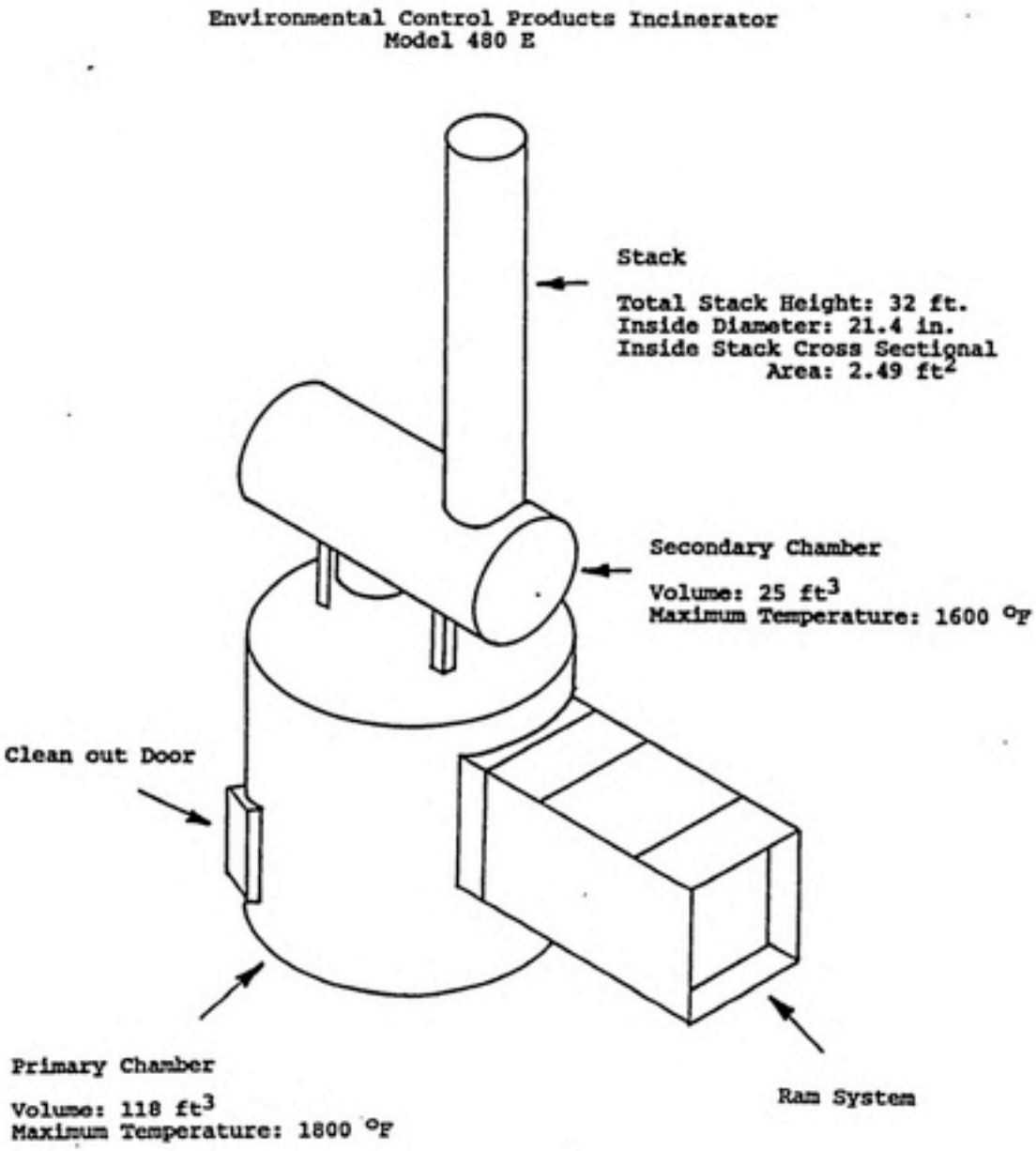
Stack Height: 32 feet

Inside Diameter of Stack: 21.4 inches

Total Area of Stack: 2.5 $ft^2$

# Figure 1

## Detail of Incinerator and Stack



combustion process. The resultant gases then pass through the refractory lined stack, which includes a spark arrestor, and into the atmosphere.Ashes collect on the floor of the refractory lined primary chamber and are removed manually. The remaining material is removed with a HEPA filtered vacuum system which directly deposits the ash into a 17H DOT 55 gallon drum. Access is gained to the refractory floor via a 16" by 16" door. The total height of the incinerator, including stack, is 32 feet, measured from the concrete floor.

The burners operate on natural gas. The primary burner has a maximum rating of $1.2 \times 10^6$ BTU per hour, while the secondary burner has a maximum rating of $8.0 \times 10^5$ BTU per hour. Both burners are provided with a timer control with a maximum setting of twelve hours.

### B. Stack Sampling System

The stack effluent sampling system used in this study isokinetically removed stack gases and passed the sample through a filter assembly. A description of the system is as follows:

**Probe and Nozzle:** A 3/8 inch buttonhook nozzle made of stainless steel was employed to remove the gas sample. This type of nozzle effectively removes a representative sample while causing minimal aerodynamic disturbance to the stream flow. The probe used to transport the sample to the filter assembly has an effective length of 5 feet and is constructed of inconel.

**Pitot Tube and Temperature Sensor:** Both of these components are located at the same position adjacent to the nozzle for the

purpose of measuring real time gas velocity and temperature. The pitot tube, a Stausscheibe type, was used to determine the exit gas velocity and pressure. The thermometer, a semiconductive resistance type, was used to measure stack gas temperature.

**Filter Assembly:** Stack gases that entered the sampler were passed to a filter holder containing a 7 cm diameter glass fiber filter produced by Fisher Products. This filter removed particulate matter with an efficiency of 99.97% for particles 3 microns in diameter or greater. The gases then passed through four bubblers. The first two bubblers were filled with water. The third one was left empty, and the fourth contained 6 mesh anhydrous $CaSO_4$, a desiccant. No other materials, such as acids, were used in the bubbler system because it was felt that no activity would be released in a gaseous state. The study by Brekke showed no activity in the gaseous releases when bubbled through HCl. In addition, all of the boiling points of the metals examined are in excess of 2700 $^{o}C$, which is far hotter than the temperatures experienced in the incinerator.

**Control Console:** After passing through the entire sample case, the stack gases are drawn via a hose connection into a Scientific Glass & Instruments, Inc. Model AP5500 Stac-o-lator control console. This unit contains a vacuum gauge, vacuum pump, dry gas meter, and dual manometer. This instrument was calibrated on July 19, 1988 by Nutech Corporation, Durham, N.C..

### C. Vacuum System

After the bulk of the residual ash is removed from the

incinerator using manual methods, a Hako Minuteman HEPA filtered vacuum system (Model Number C 80330-02, Hako Vacuum Systems, Addison, Ill.) is used to remove the remaining loose material. The vacuum system contains a high density impact filter and a HEPA filter which is 99.97% effective for trapping particulates as small as 0.3 microns. When drawn into the vacuum, the ash is directly deposited into a 55 gallon drum which is lined with a plastic bag.

### D. Multichannel Analyzer System

The detection system used to evaluate all of the samples was comprised of a Bicron 2" x 2" NaI(Tl) crystal connected to a Nucleus, Inc. Model 5010 scintillation amplifier and power supply. The spectrum was analyzed using a Nucleus, Inc. Personal Computer Analyzer card installed in an IBM XT personal computer. Ash samples were collected and counted in disposable 1 liter Marinelli beakers. Air filters and filter paper smears were placed in disposable plastic petri dishes for counting.

In order to determine a lower limit of detection (LLD) for the counting system in each of the counting configurations, the ordinary procedure is to count non-radioactive samples, and use the results as a background level, from which a LLD could be calculated. This technique could not be applied in this study because a combination of isotopes were being evaluated simultaneously. When using MCA's for analysis of mixed isotope samples, the background level in each region of interest is influenced by any activity recorded in the higher energy regions.

With this being the case, a LLD determined for a low energy gamma emission in a mixed isotope sample would not necessarily be what was calculated from the use of a non-radioactive sample. In recognition of this situation, an average background level in each region of interest for each counting configuration was determined by compiling the data from all of the samples collected. These "typical", or representative background values were then used to determine a LLD.[19]

Table 6 contains the typical background values used to calculate the LLD's. Also included in this table, for comparison, are the background values obtained from the counts of the non-radioactive samples.

The LLD's at a 95% Confidence Interval for all of the sample types were derived using the following equation:[20]

$$LLD = \frac{4.66\ (s_b)}{(E)\ \exp(-\lambda_i t_e)}$$

Where: $s_b = (N/t_b)^{1/2}$ standard deviation of background

$N$ = background count rate

$t_b$ = background count time

$E$ = efficiency (c/d)

---

[19]National Council on Radiation Protection and Measurements, 1985, A Handbook of Radioactivity Measurements Procedures, NCRP Publication 58, Washington, D.C., p. 307-311.

[20]Practical Statistics for Operational Health Physics, Tenth Annual Health Physics Society Summer School, Idaho State University, Pocatello, Idaho, July 1987

Table 6.

Background Levels for Counting Configurations

in Counts per Second

| Isotope | Ash Sample 12 Hour Count | | Air Filter 12 Hour Count | | Swipe Sample 2 Hour Count | |
|---|---|---|---|---|---|---|
| | Typical* Radioactive Sample | Non-Radioactive** Sample | Typical Radioactive Sample | Non-Radioactive Sample | Typical Radioactive Sample | Non-Radioactive Sample |
| Ce-141 | 350 | 1.14 | 1.4 | 1.06 | 0.9 | 0.6 |
| Sn-113 | 341 | 1.2 | 1.2 | 1.0 | 0.8 | 0.6 |
| Ru-103 | 331 | 1.07 | 1.2 | 0.94 | 0.82 | 0.53 |
| Nb-95 | 279 | 0.9 | 1.0 | 0.9 | 0.7 | 0.5 |
| Sc-46 | 241 | 0.7 | 0.8 | 0.7 | 0.6 | 0.45 |

*Typical Radioactive Sample background values are defined as the average of the background values from all of the actual experimental samples counted in this configuration.

**Non-Radioactive Sample background values are defined as the background counts of non-radioactive samples in the same counting configuration as the actual experimental samples.

$\lambda_i$ = decay constant of $i^{th}$ nuclide

$t_e$ = elapsed time

The LLD's for Ce-141, Sn-113, Ru-103, Nb-95,and Sc-46 are shown in Table 7.

To determine the detection efficiency for the ash counting configuration, a standard was prepared which closely simulated the actual ash samples to be analyzed. An ash sample was collected from the incinerator after a two month period of burning only non-radioactive wastes and carcasses. The ash sample was placed in a 1 liter Marinelli beaker, exactly like the beakers which were used for the counting of the actual samples. Known amounts of each of the five isotopes were incorporated into the ash with a syringe, using the same techniques employed during an actual experimental application. (A complete description of the mixing and injection of microspheres in given in Part I, Section d., Experimental Applications). The actual activity of each isotope injected was determined based on the microsphere batch assay information provided by the manufacturer, and a correction for decay. After all of the microspheres were injected, the ash was allowed to dry in a desiccant chamber for 48 hours and then sealed. Prior to counting, the beaker was tumbled to ensure uniform distribution of the microspheres.

A similar method was used for the filter paper counting standard. A filter paper was placed in a plastic petri dish and

Table 7.

Lower Limits of Detection

For Two Counting Configurations

| Isotope | 12 hour count Ash Sample (Bq) | 12 hour count Air Filter (Bq) | 2 hour count Swipe Sample (Bq) |
|---|---|---|---|
| Ce-141 | 17.5 | 0.2 | 0.4 |
| Sn-113 | 37.6 | 0.4 | 0.8 |
| Ru-103 | 40.7 | 0.5 | 0.9 |
| Nb-95 | 46.8 | 0.5 | 0.93 |
| Sc-46 | 87.0 | 1.1 | 2.2 |

known amounts of the five microspheres were added by syringe. The petri dish was placed in a desiccant chamber for 48 hours and then sealed.

The detection efficiencies for the five isotopes in each of the two counting configurations were determined by performing five 12-hour counts. The peaks produced by the five gamma ray emitters were marked and the net counts in each of the regions of interest were obtained. An average associated efficiency error of 5% was established using a worst case assumption. To obtain this error, each of the standards were counted ten times, and the detection efficiencies for each peak independently determined. The mean counting efficiency and standard deviation were calculated for each of the five isotopes. In the case of Sc-46, which has two gamma emissions, the peak of 1120 keV was always used for analysis. (The Sc-46 886 keV peak was interfered with by the 765 keV peak of Nb-95. This overlapping of peaks introduced error into the analysis of Nb-95, and is addressed in the section on Limiting Factors.) The highest error, that of the 1120 keV peak of Sc-46 in the Marinelli beaker configuration, was found to be 5%. This error was applied to all of the isotopes in each of the counting configurations as a conservative measure. The efficiencies and errors for the detection system used in this experiment are listed in Table 8.

The regions of interest for each gamma ray energy were established using the appropriate spiked standard prior to actual sample counting. The spiked sample was placed on the detector and

Table 8.

Detection Efficiencies (in percent) for Two Counting Configurations Using a 12 Hour Count

| Isotope | Ash in Marinelli Beaker | Filter in Petri Dish |
|---|---|---|
| Ce-141 | 2.4±0.12 | 12.2±0.6 |
| Sn-113 | 1.1±0.06 | 5.5±0.3 |
| Ru-103 | 1.0±0.05 | 4.6±0.2 |
| Nb-95 | 0.8±0.04 | 4.2±0.2 |
| Sc-46 | 0.4±0.02 | 1.7±0.09 |

counted for 30 minutes. This allowed an accumulation of a significant number counts so that the peaks could be easily distinguished from background. Regions of interest were marked on each side of the peaks, and stored in the computer memory. Energy calibration in keV/channel was also verified at this time. This short count was then erased, and the sample to be analyzed was counted.

At the end of the counting period, the regions of interest previously established were superimposed onto the sample spectrum. The method used by the computer to determine the net area of the peak is to draw a straight line from the average of the beginning region of interest channel contents and the three previous channels to the average of the final region of interest counts and the three following channels. All counts above the straight line are considered to be net area.[21]

The peak data for all of the regions of interest, including net counts, background counts, centroid value, and full width-half maximum (FWHM), are generated in a single summary report.

## E. Experimental Design

### 1. Objectives

The procedures developed for this project were specifically designed with the deficiencies of previous studies in mind. The primary objective of this study was to document the successive releases of radioactivity in non-radioactive burns which followed

---

[21]The Nucleus Inc., Personal Computer Analyzer Operation and Instruction Manual, 1986, Oak Ridge, Tn. pp. 39-40.

the incineration of microspheres. To accomplish this, the monitoring of five sets of burns was proposed. Each set would consist of three burns, the first containing non-radioactive carcasses, the second burn containing microspheres, and the third, another non-radioactive carcass. Each burn in the set would incinerate approximately the same volume of waste. This "series" approach would closely resemble the normal operating procedure which includes a microsphere burn, followed by set of burns comprised of non-radioactive wastes.

Along with attempting to perform test burns in the incinerator which reproduced actual standard operating procedures, all efforts were made to determine, as accurately as possible, the actual initial activity contained in the animal. The intention of monitoring the ash, refractory, and effluent was to perform a materials balance so as to determine the ultimate fate of the radioactivity in air, ash, or refractory.

**2. Limiting Factors**

Several parameters imposed limits on the design of the experimental burns. Because the incinerator is used for the disposal of large volumes of institutional wastes, exclusive access to this unit for research purposes was severely limited. To maximize the number of test burns that could be performed, a decision was made to utilize small volumes of waste which would reduce the total burn time required to totally incinerate each carcass. The manufacturers rated capacity for Type IV wastes (animal carcasses) in this incinerator is 350 lbs/hr. This value

could not be applied in this experimental design because it included long warm-up and burn-down periods. Past operating experience demonstrated this unit would totally ash 60-80 lbs of Type IV wastes in one hour with a 30 minute warm-up period.

Another constraint was imposed by the decision to burn only wastes which were currently being generated from research work. With the cooperation of the investigators, it was felt that an accurate determination of the injected radioactivity could be made without the need for generating additional wastes by "spiking" animal carcasses. Economics also played a role in this decision, since the microspheres themselves are very expensive (approximately $250 per 500 mg of each microsphere isotope). The decision to use currently generated wastes in this experiment effectively limited the amount of activity which would be incinerated in each burn. The typical amount of activity in each animal (13-27 kg dog or pig) is approximately 20 uCi of each of the five radioisotopes.

The stack sampling equipment produced another limiting factor. When incinerating wastes such as animal carcasses, the emission rate of effluents is not constant over the entire length of the burn. Therefore, it is important that the effluent be monitored over the entire burn period. The probe of the sample case is made of a metal which can only survive the effluent temperature of the stack for one hour. To properly monitor the effluent, burn times would need to be limited to no more than one hour.

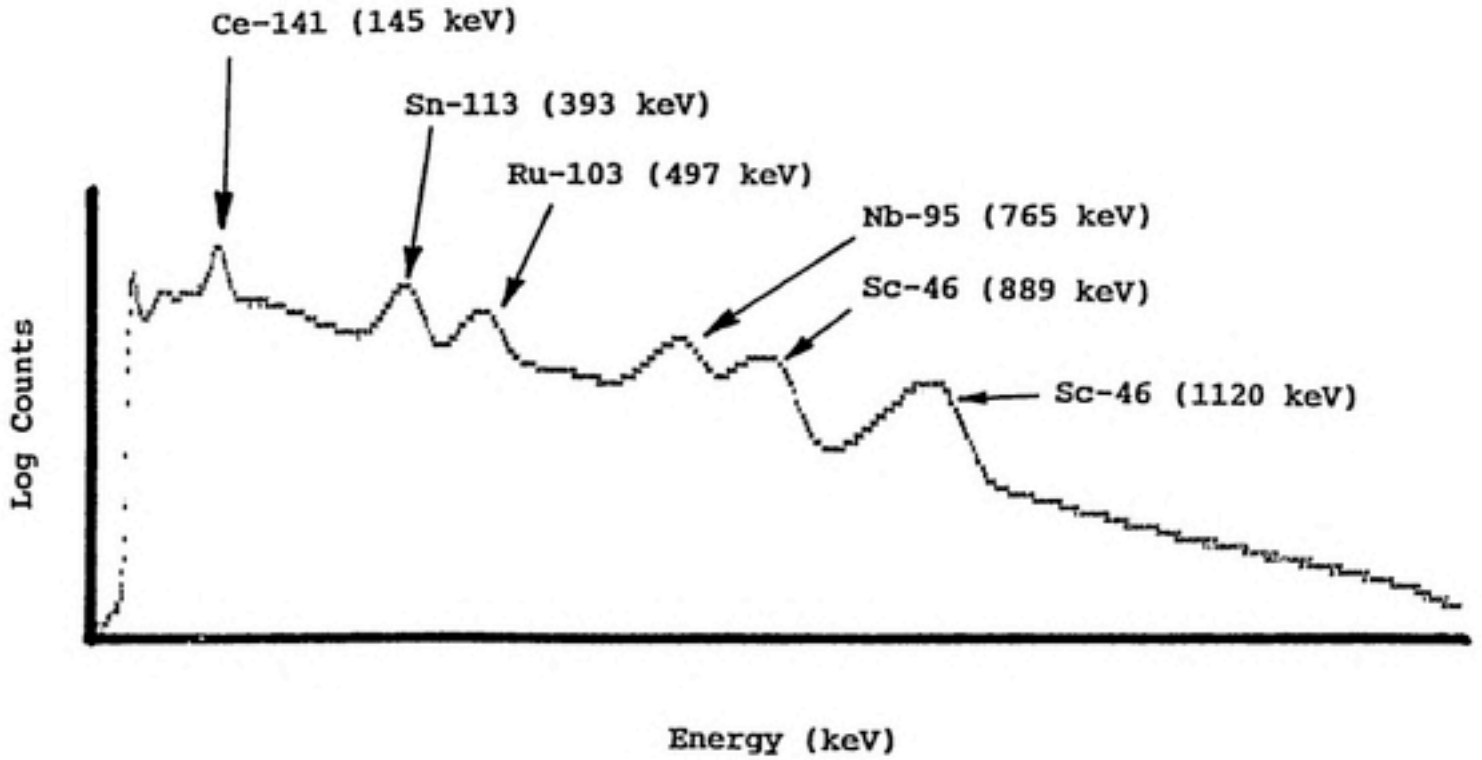
Considering all of these factors together, the decision was

made to perform each test burn with approximately 10-30 kg of animal carcass (one large animal) for a burn time of one hour. The incinerator would be allowed to warm up for 30 minutes prior to charging the chamber with waste. This schedule would permit all five sets of burns to take place with a minimum interruption to the normal waste processing at the facility.

It was recognized prior to the analysis of samples that the capabilities of the MCA system would also be a limiting factor in this experiment. Most of the microsphere isotopes produced gamma energy peaks which were easily resolved by the NaI(Tl) detector. The one exception was the overlapping of the Nb-95 peak at 765 keV with the 886 keV peak of Sc-46. Fortunately, Sc-46 emits two gamma energies, so the 1120 keV peak, which was unaffected by Nb-95, was used exclusively for spectral analysis. With the peak analysis capabilities available for this study, it was conceded that any Nb-95 results obtained should be viewed with this spectral resolution problem in mind. Figure 2 shows a typical spectrum obtained from a sample with a mixture of the five microsphere isotopes, and the overlap of the two peaks.

Figure 2

Multichannel Analyzer Spectrum
of Standard Containing Five Microsphere Isotopes
Showing Overlap of Nb-95 and Sc-46 Peaks



3. Determination of Actual Activity and Analysis of Error

A common problem encountered in all of the previous studies using institutionally generated waste was uncertainty associated with the initial activity placed in the incinerator. This uncertainty was due mainly to inaccurate recordkeeping by the researchers performing the actual microsphere applications. To aid in the determination of the actual amount of activity injected into the animals, assistance was solicited from one of the researchers who actually used the microspheres in experiments. All of the carcasses used in this study were obtained from this single researcher. Through this individual's diligent efforts, accurate records were maintained, including the date of injection, dilution of stock solutions, length of time vortexed and sonicated, and the actual volume of liquid suspension media injected. Fortunately, the experimental protocol used by this researcher called for the microsphere injection to be followed by a saline flush. This eliminated any possibility of microsphere retention in the syringe or associated tubing, as encountered in the study by Krueger and McLaughlin. The empty syringes were routinely monitored with a portable NaI(Tl) survey meter prior to disposal in a sharps container. No residual activity was ever detected. At the completion of the experiment, the animal carcass and any tissue that was excised for biological analysis was brought to the Office of Radiation Safety. In this way, all of the activity which was used in the experiment could be assembled for

incineration, without any losses due to missing tissue or blood samples.

Another source of uncertainty involved with the initial amount of radioactivity incinerated is the error associated with the actual radioactivity in each microsphere. Although each batch of microspheres is assayed by the manufacturer prior to shipment, this assay only indicates the measured specific activity of the beads, and not the associated standard deviation. Since the magnitude of this error is unknown, it would be prudent to make provisions for this error in any work incorporating the results of this study.

Since the Marinelli Beaker ash counting standard used to calibrate the counting system and the wastes were generated from the same batches of microspheres and were made using similar experimental methods, any uncertainty introduced by the activity per bead in the wastes was also introduced, in the same amount, into the ash counting standard. With this error in both the wastes and the counting standard, it was effectively eliminated, or cancelled out, with respect to the calculation of a percent retention. Although the uncertainty in the activity in the microspheres does not effect the determination of the percent retention, it would effect an estimate of the absolute amount of activity retained in the ash. Any estimate of the activity retained in the ash would include the uncertainty in the activity in the microspheres.

Appendix B provides a detailed explanation of the procedures and expressions used to obtain the percent retention values and a

sample calculation using actual data from one of the burns. This demonstrates that the value of the activity of the microspheres cancels when determining the percent retention factor.

4. Incineration Procedures

Prior to initiating each series of burns, arrangements were made with the University Physical Plant to have uninterrupted use of the incinerator for a minimum of 96 hours. This time period usually occurred on weekends or holidays.

Before starting the series of burns, the incinerator was manually cleaned out and the refractory vacuumed. The incinerator was then started and allowed to warm up for 30 minutes to reach operating temperatures. The animal carcass was then weighed, and in the case of radioactive carcasses, the activity information recorded. The animal was placed in the automatic ram feed system and charged into the incinerator at the end of the warm up period, and burned for one hour.

At the end of each burn period, the incinerator was turned off and allowed to cool for 4-6 hours before ash removal.

5. Stack Monitoring

Two different techniques were used to monitor the stack effluent. For the first two sets of burns (6 individual burns) the sample probe was placed at eight different sampling points on two transects as prescribed in EPA Method 5.[22] The sampling time at each point was 7.5 minutes. Preliminary counts of the air filters

---

[22]U.S. Environmental Protection Agency, 1981, Title 40 Part 60 Code of Federal Regulations, pp. 298-323.

from these burns yielded no detectable activity, so an alternative method was employed for the remaining burns.

For sets 3, 4, and 5, (9 individual burns) the sample probe was placed at the one point with the highest air flow rate. This method was used in an attempt to sample from the area where the highest concentration of effluents might exist.

At the end of each burn, the sample probe was removed from the stack and covered with aluminum foil. After cooling, the entire sample case was carried to the Office of Radiation Safety Laboratory for disassembly.

The particulate filter was removed and placed in a petri dish. The sample probe was then disassembled and cleaned with a brush and acetone wash. The washings from this cleaning procedure were collected directly on the particulate filter. In this manner, all of the particulates sampled were now on one filter. The filter was placed in a desiccator for 48 hours and then sealed in the petri dish.

The contents of all of the impingers were removed and measured. The water volume collected in the water-filled impingers was measured by pouring the impinger contents into a graduated cylinder. The desiccant was weighed on an electronic balance. The additional water in the bubblers and the added weight of the desiccant was used to determine the water vapor content of the effluent. The contents of the impingers were not analyzed for radioactivity for several reasons. As stated earlier, the study performed by Brekke, et. al. found all of the microsphere

radioactivity sampled on the particulate filter, and no detectable amounts in the impinger contents. With the lowest boiling point of all of the microsphere elements being greater than 2700 $^{o}C$, it was doubtful that any vapors would be formed in the incinerator environment. Any vapors that might form would probably react with the available $O_2$ to form oxide particles that would be trapped on the particulate filter. Condensation of any effluent vapors onto the relatively cooler particulate filter (approximately 38 to 66 $^{o}C$) also would play a role is preventing microsphere radioactivity from reaching the impinger section of the sampling train.

6. Ash Removal and Sample Preparation

After cooling, the ash from the incinerator was removed in two ways. The largest debris was manually collected onto a tray placed beneath the cleanout door using a hoe-like device. The remaining ash was collected using the HEPA filtered vacuum system which deposited the fine silt directly into a 55 gallon drum lined with a plastic bag. The manually removed material was then added to the vacuumed material in the drum. Any visible fine dust that was collected on the vacuum filter system was brushed off into the plastic bag. The plastic bag was then sealed and brought to the Radiation Safety Laboratory for weighing and spectral analysis.

In the lab, the ash was transferred to 1-liter Marinelli Beakers and weighed. Usually, all of the ash would fit into one beaker. If any excess ash remained, it was placed in plastic jugs for weighing. Mixing both the bulk ash and the vacuumed silt into one plastic bag produced a homogeneous sample matrix. It was

assumed for radioanalysis purposes that the radioactivity was homogeneously distributed in each of the samples counted.

Special precautionary procedures were followed while removing all incinerator ash. Because a danger of ingestion and personnel contamination existed, full face particulate respirators were worn, as well as complete sets of anti-contamination clothing. After ash cleanout was accomplished, contamination surveys around the incinerator area were performed and documented.

**7. Determination of Radioactivity in Ash Samples**

Once the ash samples were prepared for counting, the Marinelli beakers were counted on the MCA system. At the completion of the twelve hour counting period, each peak on the spectrum that was produced was marked and integrated as previously described in Part II, Section D. The MCA peak analysis program generated a report for each sample which contained the net counts and background counts in each region of interest. The net count rate and associated standard deviation for each energy region was then calculated and used to obtain the percent activity that was retained. Appendix B details the method used to calculate the retained fraction of radioactivity in ash.

**8. Refractory Swipes**

Filter paper swipes were taken on the interior refractory surface of the incinerator after each burn. The filter papers used were the same size and weight of the air filter papers used in air sampling. Each swipe covered approximately 100 $cm^2$. Although the exact location of each swipe varied, the general locations surveyed

were 1) the incinerator floor at the base of the ram door, 2) the center of the floor, 3) the floor area just inside the cleanout door where material was often accumulated, and 4) two feet up the wall of the incinerator near the ram door.

After each swipe was taken, it was placed in a petri dish and sealed for counting.

# III. RESULTS AND DISCUSSION

## A. Radioactivity Detected in Ash Samples

The overall objective of this research project was to document the retention and subsequent releases of radioactivity resulting from the incineration of microspheres. Ultimately, the data from the analysis of the radioactivity in the ash samples proved to be the most useful in meeting this goal.

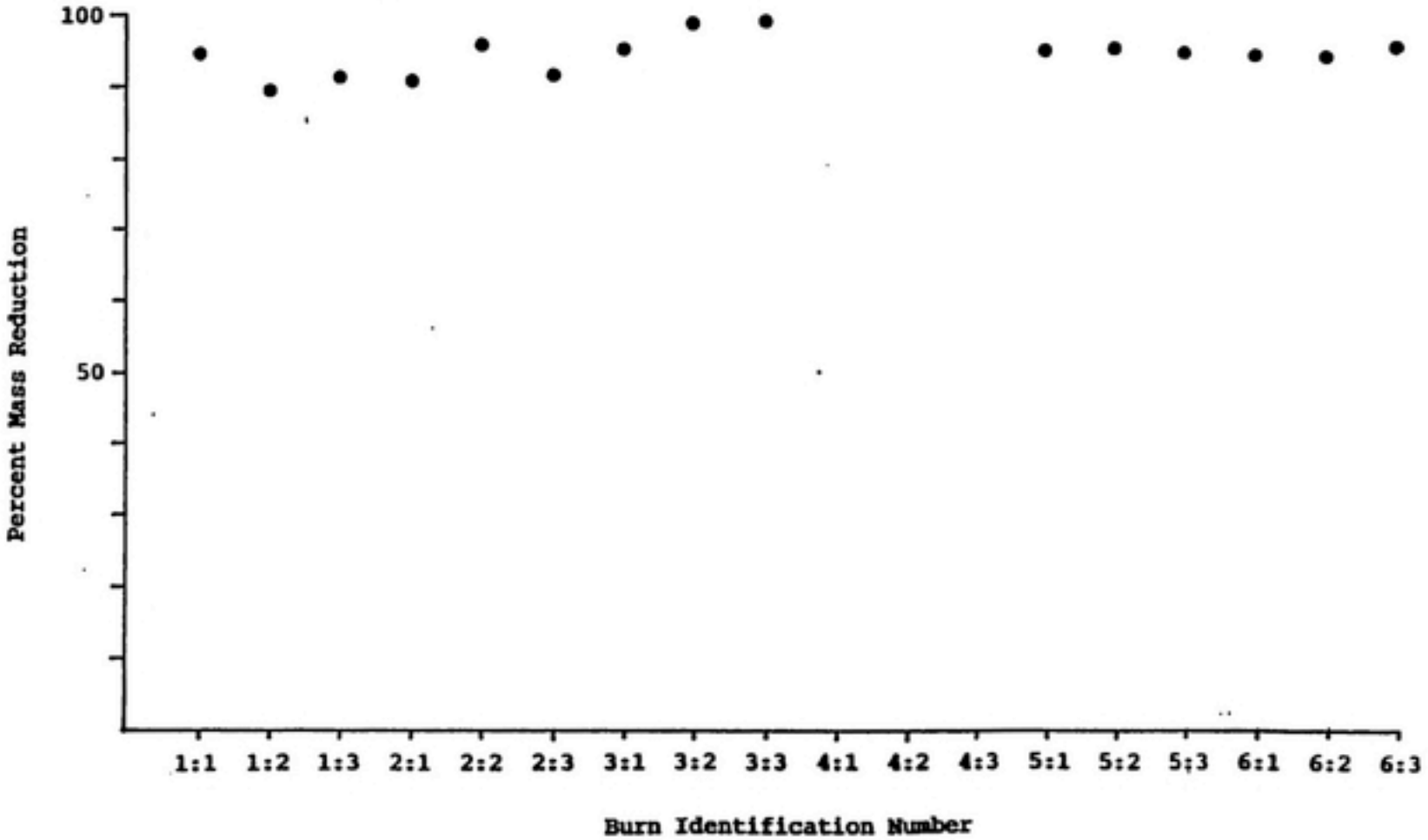
Each incineration series consisted of three burns. A coding system was established which would identify each sample as to its burn series and sequential number. For example, a sample with the identification number 2:1 indicates that the sample was from the first burn of the second series of burns. 2:2 indicates the second burn in the second series, and so on. This system proved to be most helpful in the identification of the large number of samples collected.

The incineration process routinely reduced the mass of the wastes by more than 90%. The average mass reduction for the 15 experimental burns performed was 94.1%. These results agree with the findings of several previous incineration studies, which reported reductions of 90-95%. Figure 3 shows the percent mass reduction for each of the fifteen experimental burns.

The fourth burn series, to be indicated by numbers 4:1, 4:2, and 4:3, was cancelled due to incinerator malfunction. During the initial burn of the series, which contained a non-radioactive carcass, the blowers for both the upper and lower chambers of the incinerator malfunctioned. This carcass was never completely

Figure 3

Percent Mass Reduction of Type IV Wastes
from One Hour Burn
(Initial Weight 11 to 31 Kg)



burned, and had to be removed for disposal. Repairs for the incinerator took a week to complete, so the remainder of the fourth series of burns was cancelled, and the fifth series was initiated. A sixth burn series was added to provide data which was lost due to this malfunction.

The ash from the five initial burns, 1:1, 2:1, 3:1, 5:1, and 6:1, all contained microsphere radioactivity, even though a non-radioactive carcass was burned. This contamination of non-radioactive ash apparently resulted from the release of radioactivity from the incinerator refractory from previous microsphere burns. For the five initial burns, the following ranges of gross activities were measured: Ce-141, 12.5 to 1326 Bq; Sn-113, 20 to 5515 Bq; Ru-103, 24 to 1729 Bq; Nb-95, 0 to 215 Bq; and Sc-46, 181 to 28,528 Bq. The higher activities were detected in the later series of burns, indicating a buildup of activity in the refractory was occurring as a result of the experimental burns.

The quantities of radioactivity found in the initial burns were not subtracted from the subsequent ash data as an indication of background levels of radioactivity. After examining all of the data from the first three series of burns, it became obvious that the amount of microsphere radioactivity released from the refractory into the ash decreased with successive burns. Since the rate of this release was not previously documented, it was assumed that the amount of activity contributed from previous burns would be negligible compared to the activity burned experimentally.

Cerium-141 was detected in all of the ash samples collected.

The average retention of Ce-141 in the ash from the burns of radioactive carcasses was 29.2±9.8%. The range of retention values was 17.5±1.2% to 43.1±3%. The average retention value was obtained from the ash results from burns 1:2, 2:2, 3:2, 5:2 and 6:2. The percent retention value and associated error for each burn was determined using the calculations shown in Appendix B. It should be noted that a range of ash retention values for each microsphere isotope was anticipated because of the inherent nature of industrial incinerators. Since these devices are explicitly designed and used for the processing of large amounts of institutional wastes, they should not be considered as precise laboratory equipment. Fluctuations in the various operating parameters of incinerator such as chamber temperature and induction of forced air, do indeed occur, and may effect the retention values of each specific burn. All of the previous studies performed on microsphere incineration make note of the retention variations that occurred from burn to burn. The variations in ash retention values warranted special consideration when calculating average retention values. The difference between samples produced errors more significant than the errors within each sample, so a mean and standard deviation for the mean retention values of each burn was calculated. This same calculation was also performed to summarize the results of each of the subsequent burns. The average retention value obtained using this technique adequately addressed the variation between individual samples.

The ash from the burns which immediately followed the

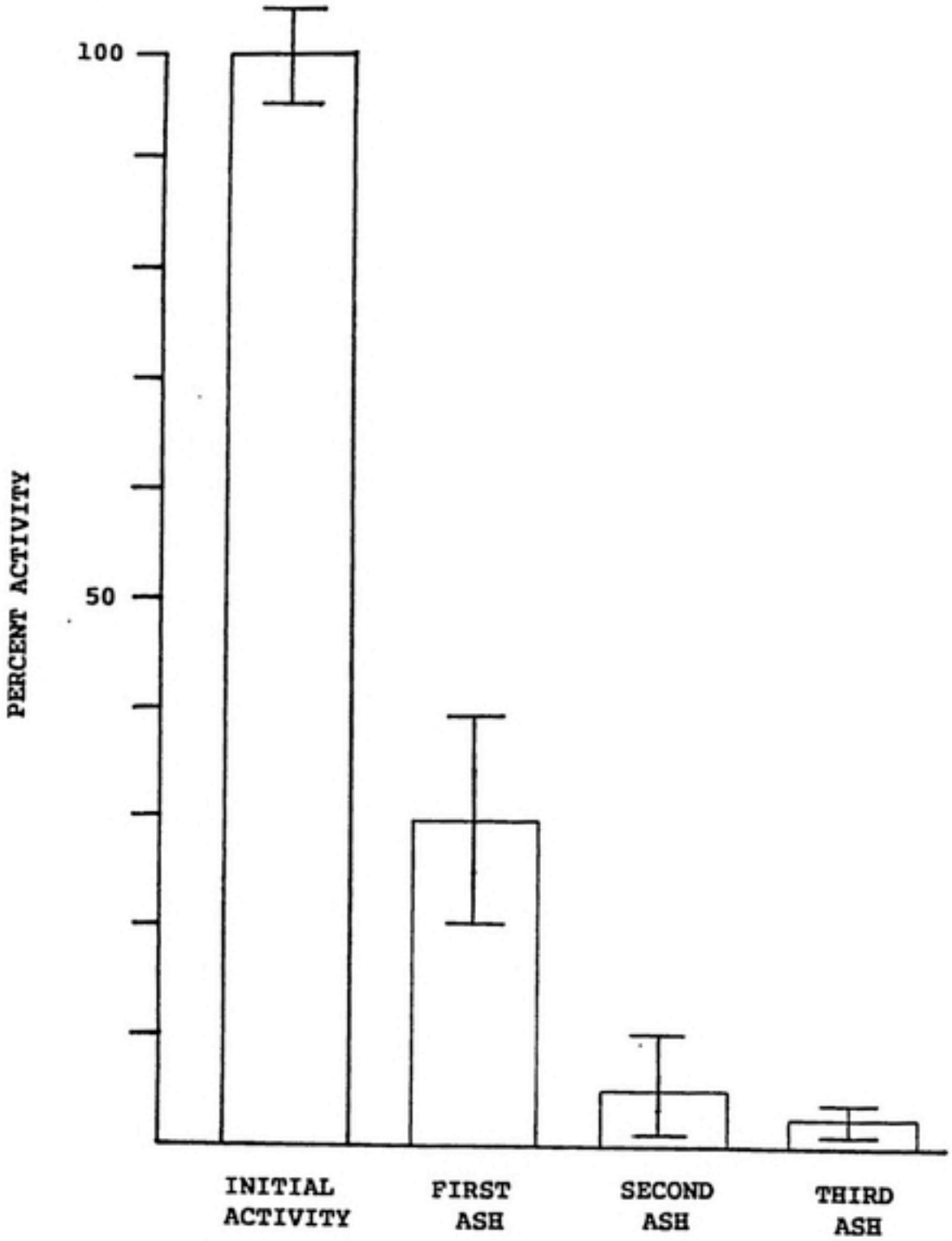
microsphere burn retained an average of 6.1±4.1% of the Ce-141 activity. The range of values was 1.4±0.1% to 12.4±0.9%. The radioactivity detected in these burns was undoubtedly due to the release of radioactivity from the incinerator refractory since the wastes contained no Ce-141.

Of the five series of burns performed, several were run in succession. The first, second, and third burn series were performed consecutively, without any interruption by the normal waste processing of the University. The fifth and sixth series of burns were also conducted consecutively. Since each burn series started with the incineration of a non-radioactive carcass, successive series of burns permitted the monitoring of ash from a second non-radioactive burn after the incineration of microspheres. As an example, the initial burn for series two, indicated by 2:1, also produced data as the second non-radioactive burn following the radioactive burn 1:2. For the three burns which could be considered as the secondary non-radioactive burns for Ce-141, an average of 3.0±1.9% of the original activity was recovered.

The average total Ce-141 activity recovered from all of the ash monitored was 38.3±10.8%. The only previous research with this microsphere isotope, performed by Finnegan, reported retention values of 83-100%. The previously reported values are higher than the results found in this study because the carcasses were burned while on collection trays, permitting almost complete ash retrieval, while preventing refractory contamination. Figure 4 displays the average retention values of each for Ce-141.

Figure 4

Retention in Ash From Incineration of Ce-141 Microspheres
Average Total Activity Retained: 38.3 ± 10.8%



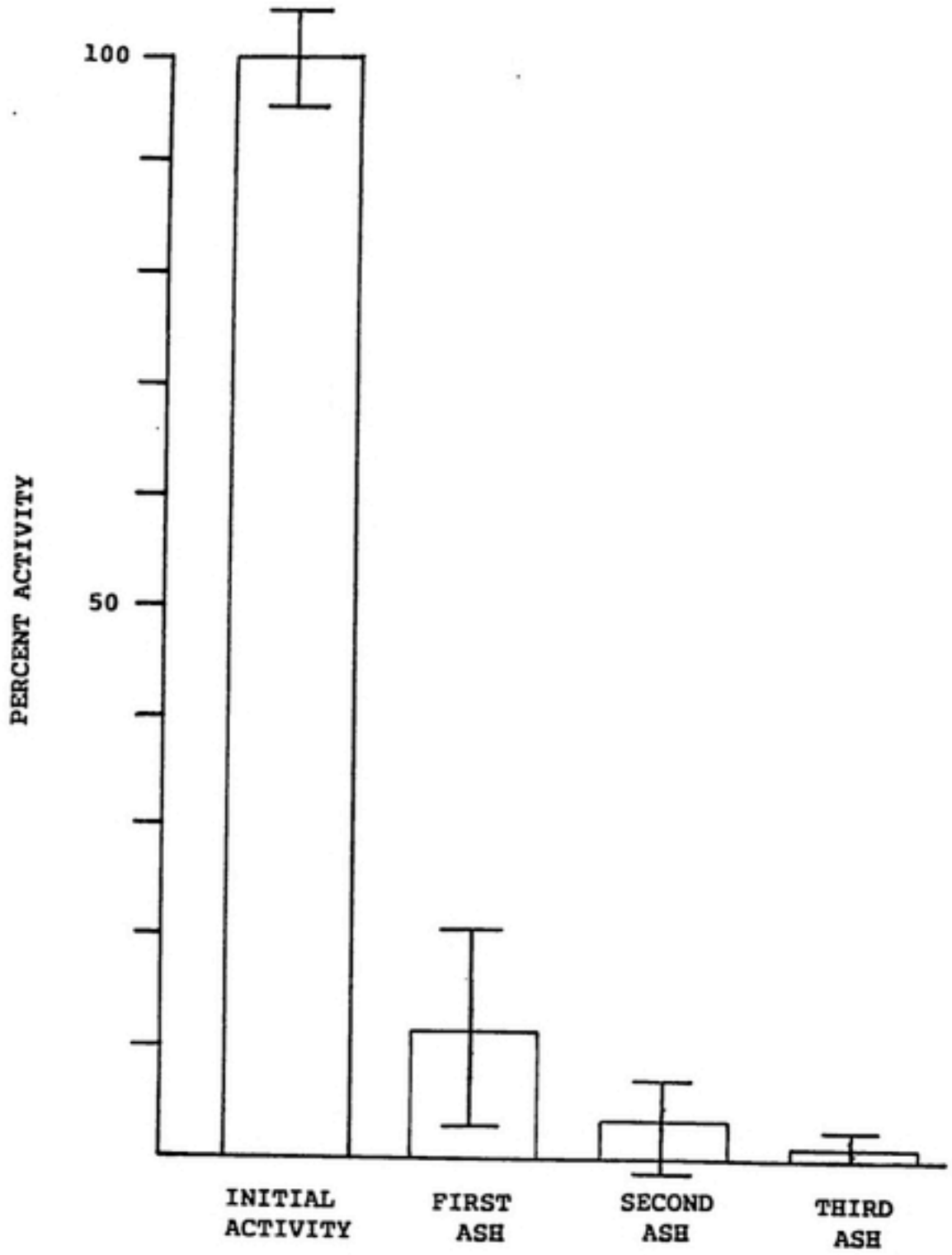
Tin-113 was also detected in all of the ash samples collected. The average retention value from the five microsphere burns was 11.6±9.1%. The range of values was 2.5±0.2% to 21.6±1.5%. The ash from the burns which immediately followed the radioactive burn yielded and average value of 2.8±3.8%, with a range of 0.9±0.06% to 9.5±0.2%. Of the three burns which permitted monitoring of a second non-radioactive burn, an average of 1.7±1.7% of the initial microsphere activity was detected, with a range of 0.4±0.03% to 3.6±0.3%.

The average total recovery of Sn-113 activity for the burn series was 16.1±10%. Two previous studies also examined Sn-113 microsphere incineration. Finnegan, et al. reported ash retention values of 80-100%, but these results are not comparable to this experiment because of reasons stated earlier. Krueger and McLaughlin reported an ash recovery value of 80% from the incineration of sheep carcasses. Although the carcasses were incinerated in the same manner as in this experiment, ash samples of only 10 grams were collected for analysis. Krueger and McLaughlin also reported the suspicion of large errors associated with the activity indicated to be in the carcass, estimating that activities could be incorrectly estimated "by as much as 100%". Figure 5 shows the average retention values of each burn for Sn-113

An average of 22.2±13.9% of Ru-103 was retained in the ash from the burns of radioactive carcasses. The range of values was 10.7±0.8% to 38.8±2.7%. Of the original activity, 4.8±4% was found

Figure 5

Retention in Ash From Incineration of Sn-113 Microspheres
Average Total Activity Retained: 16.1 ± 10%



in the first non-radioactive burn following the microsphere burn, with a range of 1.0±0.07% to 11.3±0.8%. The second non-radioactive burns contained, on average, 2.5±1.5% of the activity.

The total percent recovery for the burn series was 29.5±14.5%. Brekke reported ash retention values for Ru-103 of 58±8% and releases in air of 16.5±5%. It is felt that these two ash retention results could be directly compared because the experiments were similar in design and procedure. Figure 6 shows the average retention values of each burn for Ru-103.

Niobium-95 activity was not found in detectable quantities in several of the ash samples collected. The ash samples from the entire first and second series of burns showed no detectable activity due to Nb-95. Series 3, 5, and 6 revealed some activity, but in relatively small amounts.

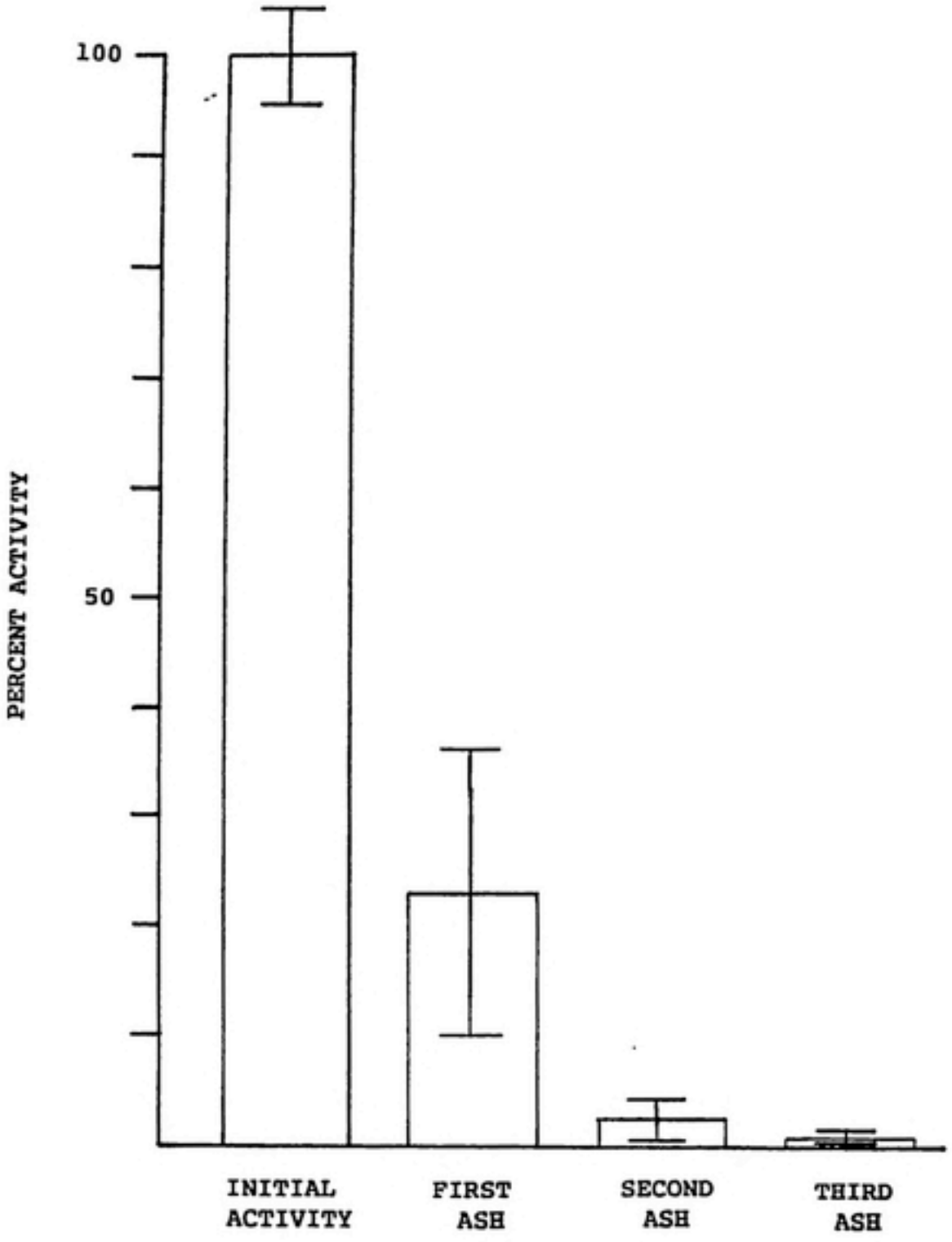
Using the data from the three burn series which contained significant Nb-95 activity, an average ash retention value for the initial radioactive burn of 6.6±3.8% was found, with a range of values from 3.5±0.2% to 10.8±0.8%.

The ash from the first successive non-radioactive burn contained, on average, 0.5±0.23%, with a range of 0.4±0.03% to 0.8±0.06%. No activity due to Nb-95 was detected above the LLD in any of the secondary non-radioactive burns (burns 2:1, 3:1, or 6:1).

The average total recovery of Nb-95 for the three series which produced activity was 7.1±3.8%. This is in stark contrast to the reported ash retention value of 91±24% by Brekke. It is important

Figure 6

Retention in Ash From Incineration of Ru-103 Microspheres
Average Total Activity Retained: 29.5 ± 14.5%



to remember that all of the results obtained for Nb-95 are based on spectral evaluations made with interference from one of the gamma emissions of Sc-46. Even though the method of detection used is admittedly poor for resolving two closely spaced peaks, Nb-95 could be recognized if present in appreciable amounts. No other explanation can be readily provided for this wide difference in results. Figure 7 displays the average retention values of each burn for Nb-95.

Scandium-46 was detected in sizable amounts in all of the ash samples collected. The average ash retention value for the radioactive burn was 60±20.8%, with a range of values from 41.3±3% to 93.5±6.6%. The average retention in ash for the first non-radioactive burn following the microspheres was 18±12.9%. The range of values was 4.6±0.3% to 38.2±2.7%. Ash collected from the secondary non-radioactive burns contained, on average, 10.0±6.1% of the initial activity, with a range of 3.1±0.2% to 14.4±1.0%.

The average total recovery of Sc-46 activity was found to be 88±25%. This figure is in agreement with all of the previous studies which included examination of this isotope. Landholt, et. al. reported ash retention of 97.6±8%, Finnegan 83-100%, Classic, et. al. 95.4±1%, Van Swearingen 79.7±13%, and Krueger and McLaughlin 92%. It is interesting to note that even though these studies incinerated carcasses in different ways, the Sc-46 retention results are still very similar. Figure 8 displays the average retention values of the burn series for Sc-46.

Figure 7

Retention in Ash From Incineration of Nb-95 Microspheres
Average Total Activity Retained: 7.1 ± 17.6%*

*based on arithmetic mean of values from three burns producing detectable activity in ash.

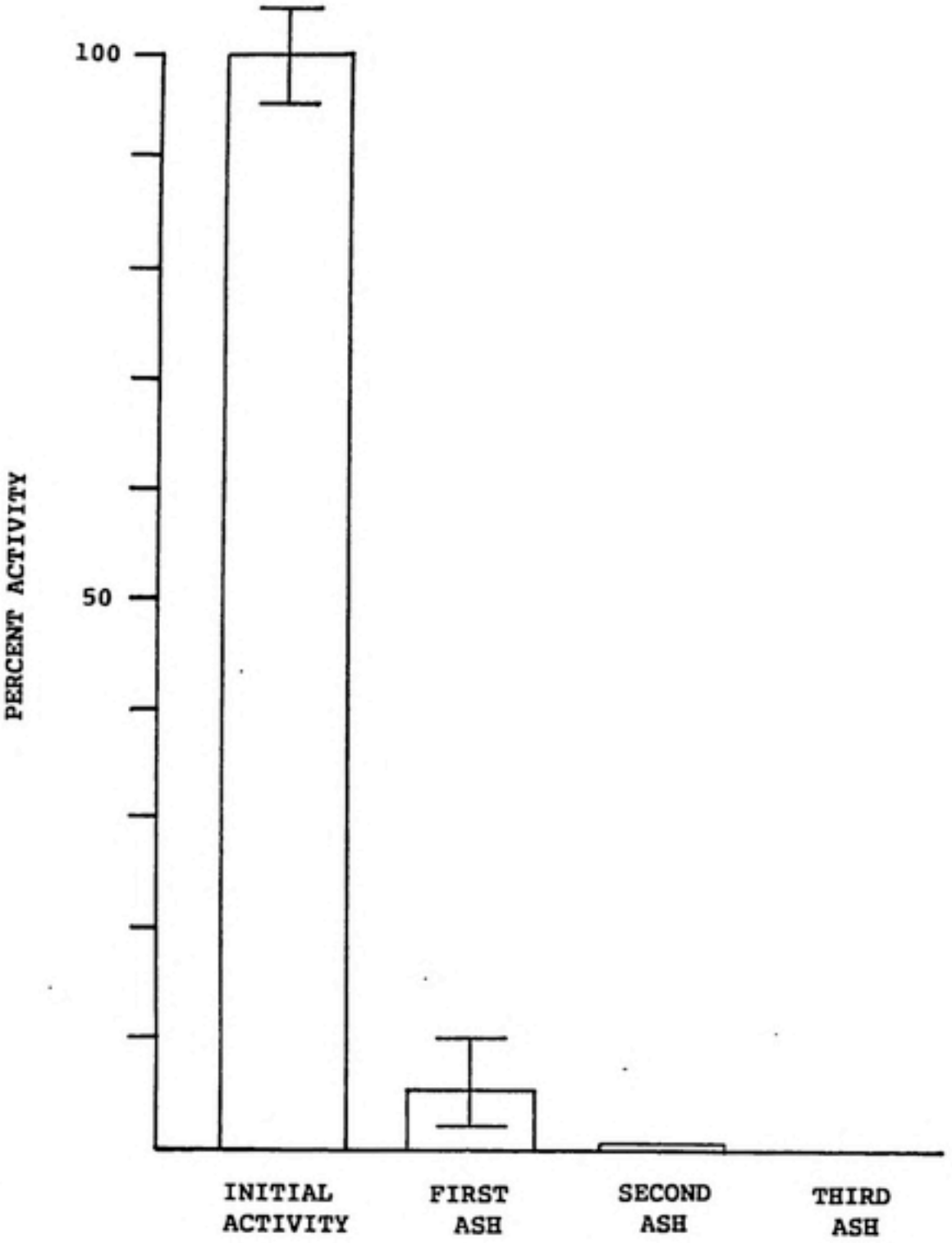


Figure 8

Retention in Ash From Incineration of Sc-46 Microspheres
Average Total Activity Retained: 88 ± 25%

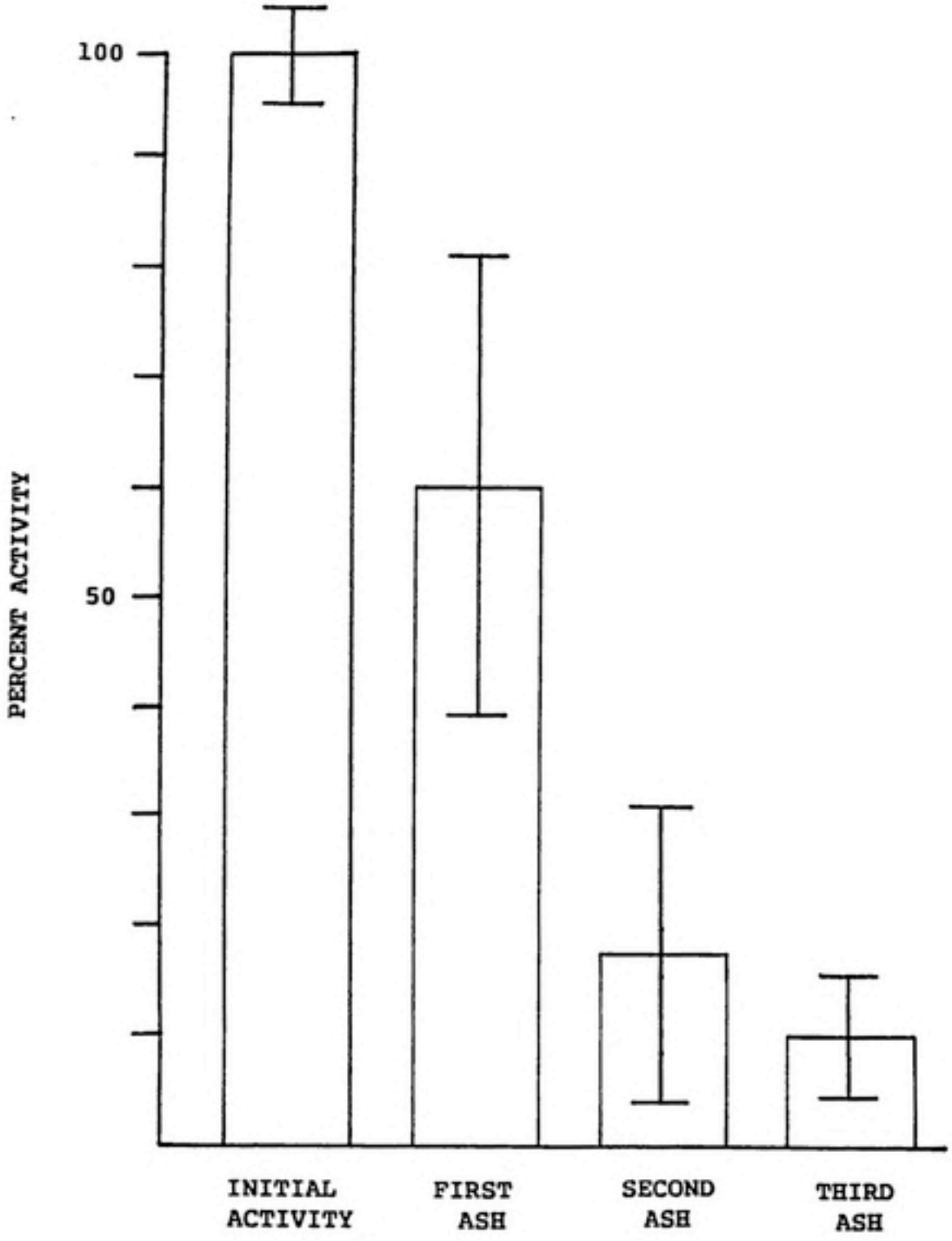


Table 9 contains the total percent retention of microsphere radioactivity for each radionuclide.

### B. Radioactivity Detected in Swipe Samples

Of the 60 swipe samples taken on the incinerator refractory, none contained any radioactivity above the lower detection limit of the multichannel analyzer system. These results are in agreement with the results of other studies where swipes were taken and no activity was found.

Two factors are thought to contribute to the reason no loose contamination was detected. First, the swipes were taken after the incinerator was completely cleaned out. Since the cleaning procedure included vacuuming, it is felt that any loose contamination would have been collected prior to the swipe survey. Secondly, the refractory is not only a very porous surface, but a very rough one as well. The metal tools used for routine manual cleanout of the incinerator have given the refractory a rough and convoluted surface with many cracks and crevasses. These areas could effectively trap particles, thus preventing their collection on a filter paper swipe.

### C. Radioactivity Detected in Stack Emissions

No radioactivity above the lower limit of detection was found on any of the fifteen particulate air samples collected. This does not demonstrate that radioactive emissions did not occur, but rather that emissions did not occur in detectable quantities.

The primary reason that no activity was detected was that only small amounts of radioactivity were incinerated in each burn. As

Table 9.

Total Percent Retention of Microsphere Radioactivity in Incinerator Ash

| | Burn Series | | | | |
|---|---|---|---|---|---|
| Isotope | 1 | 2 | 3 | 5 | 6 |
| Ce-141 | 49.2±3 | 48.2±2.4 | 18.9±1.2 | 41.9±2.4 | 27.3±1.6 |
| Sn-113 | 22.5±1.4 | 34.7±1.7 | 3.5±0.2 | 3.9±0.2 | 12.3±0.8 |
| Ru-103 | 44.6±2.7 | 51.1±2.6 | 11.7±0.8 | 22.3±1 | 13.0±0.8 |
| Nb-95 | <LLD | <LLD | 11.2±0.8 | 6.3±0.4 | 3.8±0.2 |
| Sc-46 | 108.0±6.7 | 96.3±4.2 | 45.9±3 | 93.7±4.5 | 75±4.4 |

explained in the section on Limiting Factors in this report, burn times were limited to one hour, which effectively limited the quantities of radioactivity which could be burned. Considering the flow rate of the effluent, and that the stack sampling probe only represents a small fraction of the total area of the incinerator stack, it is not surprising that no detectable amounts of radioactivity were collected. The stack sampling portion of this experiment also revealed that there is an uneven flow rate and temperature gradient across the sampling location of the stack, which lead to a turbulent stack flow atmosphere. This further reduced the chances of collecting a representative sample. Appendix C explains, in detail, the stack sampling air flow and temperature calculations and results.

# IV. CONCLUSIONS AND RECOMMENDATIONS

This investigation documents that retention and subsequent releases of radioactivity from the incinerator do occur when wastes are burned directly on the refractory floor. This conclusion is significant because subsequent releases of microsphere radioactivity have not been previously addressed. Other studies which evaluated microsphere retention concentrated on the ash produced from the initial burn of radioactive waste, and often did not simulate normal operating conditions by burning carcasses on trays or beds of ash.

It is apparent from the data collected that some of the radioactivity contained in the microsphere wastes becomes trapped in the refractory lining during the incineration process. The results of the swipe tests indicate that the activity is temporarily fixed in the refractory, and requires some sort of stimulation for removal. During the next consecutive burn after microsphere incineration, some of the activity is released from the refractory. It is theorized that the high temperatures and the turbulent atmosphere in the incineration chamber combine to loosen some radioactive particles and allow for their release. The results of this investigation cannot conclude if all of the activity released from each subsequent burn is retained in the ash produced. Some portion of the activity may well be released into the atmosphere. The effluent sampling equipment used in this study was unable to detect any radioactivity which may have been subsequently released.

A portion of the microsphere activity also remains trapped in the refractory. Five weeks after the last experimental burn series, the entire multichannel analyzer system was transported to the incinerator, and the detector was placed directly into the burning chamber about 1 cm over the general area where most of the carcasses were burned. A 40 minute count recorded activity apparently from Sn-113, Ru-103, Nb-95 and Sc-46 trapped in the refractory lining. Since no standard source for this counting configuration existed, no definitive identification or quantitation could be made. The familiar pattern of the spectrum obtained suggested the presence of microsphere radionuclides, but the possibility exists that some of the peaks could have resulted from the incineration of wastes which were considered non-radioactive, but actually contained various other radionuclides.

Supportive of the theory that some activity remains trapped in the refractory are the findings reported by Stan Wadsworth, Radiation Safety Officer, John Hopkins University. At the 1988 Southeastern Campus Radiation Safety Officer's Conference, he reported that microsphere activity was detected in samples of the old refractory material that was removed from their incinerator. No attempt was made to quantify the amounts of radioactivity detected.[23]

---

[23]Personal Communication, S. Wadsworth, March 8, 1988, Southeastern University Radiation Safety Officers Conference, Durham, NC.

Based on the findings of this report, several recommendations can be made concerning the procedures and precautions involved with the incineration of microspheres:

1) As demonstrated in this study, contamination of ash from burns following microsphere incineration does occur. Depending upon the amounts of activity initially burned, the specific activity in subsequent ash could be significant. This is an important finding with regard to the ultimate disposal of incinerator ash. Since there are no established maximum permissible concentration limits of radioactivity in ash, many institutions make the assumption that 1 gm of ash is equal to 1 ml of water, in order to apply Maximum Permissible Concentration values for water from Appendix B, 10 CFR 20 as a regulatory guide for disposal of incinerator ash.[24] If an institution incinerates microspheres, routine monitoring of all incinerator ash should be performed to verify that the specific activity of all of the ash generated is below regulatory and licensed limits prior to ultimate disposal.

2) The uncertainty associated with the actual amount of radioactivity in each microsphere is unknown. The retention values presented in this study are not effected by any error associated with the microsphere activity. However, an estimate

---

[24]U.S. Nuclear Regulatory Commission, Title 10 Part 20 Code of Federal Regulations. 1981.

of the absolute value of the activity retained in the ash would include the uncertainty in the activity in the microspheres. Considering this, prudent health physics practice mandates that conservative estimations be made concerning the original activity placed in to the incinerator for emission purposes.

3) Since institutional incinerators are not precise scientific instruments, it should not be assumed that ash retention factors will be constant over a period of time. Changes in operating characteristics of the incinerator are common, and may be the cause for fluctuations in microsphere activity retention. With this in mind, the only reliable way to determine ash retention factors for a particular burn is to actually analyze ash samples from each burn.

4) The air sampling portion of this study was unable to quantify initial or subsequent releases of microsphere activity in the effluent. However, it would be prudent to assume that some releases do occur. Therefore, only the activity actually documented as retained in the ash should be considered as not discharged into the atmosphere. The impetus for all of the studies about microsphere incineration was to demonstrate that activity is retained in the ash, thus effectively increasing the amount of microsphere wastes which could be burned. Because the ash retention values appear to vary with isotope,

those institutions operating under the blanket assumption that ash retention factors are constant should re-evaluate their microsphere incineration programs.

5) More stringent respiratory protection measures are needed for personnel who are involved with routine incinerator ash cleanout. Prior to this study, the concern about possible ingestion of airborne radioactivity centered around cleanout of the initial microsphere ash. It is now recognized that the potential for ingestion of radionuclides exists even from the ash of non-radioactive burns.

6) Comparison of the results from this study to previous works indicates that some measures may be available to reduce the amount of subsequent ash contamination which does occur. Although impractical for East Carolina University, the incineration of microsphere waste on trays apparently prevents refractory contamination, and aids in the collection of ash. Another abatement method would be the incineration of waste on a bed on non-radioactive ash. The ash layer may act as a barrier, preventing refractory contamination.

Many opportunities exist for further research in this field of microsphere incineration. Future studies are needed to examine several parameters, such as operating temperature and chamber turbulence, to determine if there is an effect on retention of

activity. Attention should also be directed towards different abatement procedures which could be used to maximize collection of the ashed portion of the radioactivity, and prevent subsequent contamination of ash. Methods of fixation of the radioactivity in the refractory may also be considered. As the technology of stack sampling advances, continuous monitoring of the incinerator effluent over burns of 6 to 8 hours could be possible. The data collected may indicate that subsequent releases into the atmosphere also occur.

# APPENDICES

# A. SUMMARY REPORTS OF ASH DATA

## ASH RESULTS

| ISOTOPE | Activity Incinerated | 1:1 First Burn (non-rad) | 1:2 Second Burn (rad) | 1:3 Third Burn (non-rad) | 2:1 Fourth Burn* (non-rad) | Total Ash Retention |
|---|---|---|---|---|---|---|
| Ce-141 | 100 ± 5 % $2.79 \times 10^4$ Bq | | 43.1 ± 3 % | 5.0 ± 0.4 % | 1.1 ± 0.07 % | 49.2 ± 3 % |
| Sn-113 | 100 ± 5 % $1.54 \times 10^5$ Bq | | 20 ± 1.4 % | 1.4 ± 0.1 % | 1.1 ± 0.07 % | 22.5 ± 1.4 % |
| Ru-103 | 100 ± 5 % $4.4 \times 10^4$ Bq | | 38.8 ± 2.7 % | 4.8 ± 0.3 % | 1.0 ± 0.07 % | 44.6 ± 2.7 % |
| Nb-95 | 100 ± 5 % $2.4 \times 10^4$ Bq | | < LLD | < LLD | < LLD | |
| Sc-46 | 100 ± 5 % $1.98 \times 10^5$ Bq | | 93.5 ± 6.6 % | 11.4 ± 0.8 % | 3.1 ± 0.2 % | 108 ± 6.7 % |
| Net Weight of Ash Samples: | | 739.3 g | 903.3 g | 803.6 g | 801.3 g | |

*If Data in This Column, Also Initial Burn for Next Burn Series

## ASH RESULTS

| ISOTOPE | Activity Incinerated | 2:1 First Burn (non-rad) | 2:2 Second Burn (rad) | 2:3 Third Burn (non-rad) | 3:1 Fourth Burn* (non-rad) | Total Ash Retention |
|---|---|---|---|---|---|---|
| Ce-141 | 100±5% $2.79 \times 10^4$ Bq | | 30.9±2.2% | 12.4±0.9% | 4.9 ± 0.3% | 48.2 ± 2.4% |
| Sn-113 | 100 ± 5% $1.54 \times 10^5$ Bq | | 21.6±1.5% | 9.5±0.7% | 3.6 ± 0.3% | 34.7±1.7% |
| Ru-103 | 100 ± 5% $4.4 \times 10^4$ Bq | | 359± 2.5% | 11.3 ±0.8% | 3.9±0.3% | 51.1 ± 2.6% |
| Nb-95 | 100± 5% $2.4 \times 10^4$ Bq | | <LLD | <LLD | <LLD | |
| Sc-46 | 100± 5% $1.98 \times 10^5$ Bq | | 43.7± 3.1% | 38.2 ± 2.7% | 14.4 ± 1.0% | 96.3±4.2% |
| Net Weight of Ash Samples: | | 801.2 g | 976.2 g | 827.8 g | 513.8 g | |

*If Data in This Column, Also Initial Burn for Next Burn Series

## ASH RESULTS

| ISOTOPE | Activity Incinerated | 3:1 First Burn (non-rad) | 3:2 Second Burn (rad) | 3:3 Third Burn (non-rad) | Fourth Burn* (non-rad) | Total Ash Retention |
|---|---|---|---|---|---|---|
| Ce-141 | 100 ± 5 % $5.93 \times 10^4$ Bq | | 17.5 ± 1.2 % | 1.4 ± 0.1 % | | 18.9 ± 1.2 % |
| Sn-113 | 100 ± 5 % $3.18 \times 10^5$ Bq | | 2.6 ± 0.2 % | 0.9 ± 0.06 % | | 3.5 ± 0.2 % |
| Ru-103 | 100 ± 5 % $3.55 \times 10^4$ Bq | | 10.7 ± 0.8 % | 1.0 ± 0.07 % | | 11.7 ± 0.8 % |
| Nb-95 | 100 ± 5 % $2.0 \times 10^4$ Bq | | 10.8 ± 0.8 % | 0.4 ± 0.03 % | | 11.2 ± 0.8 % |
| Sc-46 | 100 ± 5 % $1.83 \times 10^5$ Bq | | 41.3 ± 3.0 % | 4.6 ± 0.3 % | | 45.9 ± 3 % |
| Net Weight of Ash Samples: | | 513.8 g | 610.5 g | 479.1 g | | |

*If Data in This Column, Also Initial Burn for Next Burn Series

## ASH RESULTS

| ISOTOPE | Activity Incinerated | 5:1 First Burn (non-rad) | 5:2 Second Burn (rad) | 5:3 Third Burn (non-rad) | 6:1 Fourth Burn* (non-rad) | Total Ash Retention |
|---|---|---|---|---|---|---|
| Ce-141 | 100±5% $4.13 \times 10^4$ Bq | | 31.9±2.3% | 7.0±0.5% | 3.0±0.2% | 41.9±2.4% |
| Sn-113 | 100±5% $2.9 \times 10^5$ Bq | | 2.5±0.2% | 1.0±0.07% | 0.4±0.03% | 3.9±0.2% |
| Ru-103 | 100±5% $2.8 \times 10^4$ Bq | | 15.1±1.0% | 4.7±0.3% | 2.5±0.2% | 22.3±1.0% |
| Nb-95 | 100±5% $1.49 \times 10^4$ Bq | | 5.5±0.4% | 0.8±0.06% | <LLD | 6.3±0.4% |
| Sc-46 | 100±5% $1.62 \times 10^5$ Bq | | 59.2±4.2% | 21.9±1.5% | 12.6±0.9% | 93.7±4.5% |
| Net Weight of Ash Samples: | | 658 g | 500.6 g | 641.5 g | 780 g | |

*If Data in This Column, Also Initial Burn for Next Burn Series

## ASH RESULTS

| ISOTOPE | Activity Incinerated | 6:1 First Burn (non-rad) | 6:2 Second Burn (rad) | 6:3 Third Burn (non-rad) | Fourth Burn* (non-rad) | Total Ash Retention |
|---|---|---|---|---|---|---|
| Ce-141 | 100 ± 5% $4.13 \times 10^4$ Bq | | 22.8 ± 1.6% | 4.5 ± 0.3% | | 27.3 ± 1.6% |
| Sn-113 | 100 ± 5% $2.86 \times 10^5$ Bq | | 11.1 ± 0.8% | 1.2 ± 0.1% | | 12.3 ± 0.8% |
| Ru-103 | 100 ± 5% $2.72 \times 10^4$ Bq | | 10.7 ± 0.8% | 2.3 ± 0.2% | | 13 ± 0.8% |
| Nb-95 | 100 ± 5% $1.43 \times 10^4$ Bq | | 3.4 ± 0.2% | 0.4 ± 0.02% | | 3.8 ± 0.2% |
| Sc-46 | 100 ± 5% $1.51 \times 10^5$ Bq | | 60.7 ± 4.3% | 14.1 ± 1.0% | | 75 ± 4.4% |

Net Weight of Ash Samples: 780 g 708 g 847.9 g

*If Data in This Column, Also Initial Burn for Next Burn Series

## B. SOURCES OF ERROR AND DETERMINATION OF RETAINED FRACTION

In order to accurately assign an error to the percent retention values for each burn, the following sources of error were identified for consideration:

A) The error associated with the actual amount of radioactivity in each microsphere.

B) The error associated with the injection of the microspheres into the animal.

C) The error associated with the weighing of the ash.

D) The error associated with the counting of the sample.

E) The error associated with the counting efficiency of the detection system.

The following values were used for the sources of error identified above:

A) As described in the text, the associated error of the activity in each microsphere was unknown. Only the mean specific activity was supplied in the assay information for each microsphere batch.

B) An error of $\pm$0.05 ml for a 1 ml injection was determined by measuring 25 injections of 1 cc of microsphere suspension solution on filter papers in planchets. The variation in the weight of each planchet was used to derive the standard deviation.

C) The manufacturers specifications for the scale used to weigh the ash samples states an accuracy of $\pm$0.01 gm for a 1 gm measurement. This fractional error was considered insignificant in comparison to the error associated with the ash sample counts, and thus was ignored.

D) The net count rate ($C_n$) and associated error ($c_n$) of each ash sample was obtained using the following expressions:

$$C_n = \frac{(C_g - C_b)}{t}$$

$$c_n = \sqrt{\frac{C_g}{t} + \frac{C_b}{t}}$$

Where:

$C_n$: Net Count Rate

$c_n$: Standard Deviation of Net Count Rate

$C_g$: Gross Count Rate

$C_b$: Background Count Rate

t: Time

E) The counting efficiency (E) was expressed in the following manner in order to provide for the error introduced by the counting of the microsphere standard, and the error associated with the injection of the microsphere suspension solution:

$$E = \frac{(C_n' \pm c_n')}{(K)(F')(V' \pm v')}$$

Where:

E: detection efficiency

$C_n'$: net count rate of standard

$c_n'$: standard deviation of standard net count rate

K: constant to convert from volume to activity

F': factor to correct for decay of isotope in standard

V': volume of suspension liquid injected into standard

v': standard deviation of volume injected into standard

To calculate the percent activity retained in each ash sample, the activity in each sample ($A_2$) was divided by the initial activity incinerated ($A_1$). To accomplish this, a single expression was created from the formulas listed previously:

Using:

$$A_2 \pm a_2 = \frac{(C_n \pm c_n)}{(E)}$$

Where:

$A_2$: Activity in Ash Sample

$a_2$: Standard Deviation of Activity in Ash Sample

And:

$$E = \frac{(C_n' \pm c_n')}{(K)(F')(V' \pm v')}$$

The activity in each ash sample can be expressed as:

$$A_2 \pm a_2 = \frac{(C_n \pm c_n)}{(C_n' \pm c_n')} \quad (K)(F')(V' \pm v')$$

The initial activity in the waste ($A_1$) is represented by a volume of suspension liquid and a conversion constant, just as the activity in the standard:

$$A_1 \pm a_1 = (K)(F)(V \pm v)$$

Where:

$A_1$: initial activity in the waste

$a_1$: standard deviation of initial activity in waste

K: constant to convert from volume to activity

F: factor to correct for decay of isotope in waste

V: volume of microsphere injection solution

v: standard deviation of microsphere injection solution

Hence, the complete expression for the fraction of activity retained is:

$$\frac{A_2 \pm a_2}{A_1 \pm a_1} = \frac{(C_n \pm c_n)\ (K)(F')(V' \pm v')}{(C_n' \pm c_n')\ (K)(F)(V \pm v)}\ (f)$$

Where:

f: Correction factor for ash produced but not counted because of size limitation of Marinelli Beaker

As an example, the entire calculational procedure for data from burn 1:2 for Ce-141 is shown below:

Determination of LLD for Ce-141 in Ash Sample:

Typical Background Counts: 15120000 c

Counting time: 43200 sec

Standard Deviation of Background Count Rate: 0.090 c/s

LLD: 17.5 Bq

Determination of Detection Efficiency:

Gross Counts: 21475858 c

Background Counts: 14909994 c

Time: 1800 sec

Activity in Standard: $(K)(V' \pm v')(F') =$

$(4.75 \times 10^5\ Bq/ml)(1.0 \pm 0.05\ ml)(0.68) = 3.2 \times 10^5\ Bq$

Percent Gamma Emission: 48%

$$\frac{(3647.7\ c/s)}{(3.2 \times 10^5\ Bq)(0.48)} = 0.024$$

Detection Efficiency: 2.4%

Determination of Count Rate in Marinelli Beaker Standard:

Gross Counts: 21475858 c

Counting Time: 1800 sec

Gross Count Rate: 11931 c/s

Background Counts: 14909994 c

Background Count Rate: 8283 c/s

Net Count Rate: 3647.7 c/s

Standard Deviation of Count Rate:

$$= \sqrt{\frac{11931\ c/s}{1800\ sec} + \frac{8283\ c/s}{1800\ sec}}$$

Count Rate in Marinelli Beaker ($C_n' \pm c_n'$): $3647.7 \pm 3.4$ c/s

Decay Factor (F'): 0.68

Determination of Activity in Ash Sample:

Gross Counts: 18967826 c

Counting Time: 43200 sec

Gross Count Rate: 439 c/s

Background Counts: 13598688 c

Background Count Rate: 314.8 c/s

Net Count Rate: 124 c/s

Standard Deviation of Count Rate:

$$= \sqrt{\frac{439 \text{ c/s}}{43200 \text{ sec}} + \frac{314.8 \text{ c/s}}{43200 \text{ sec}}}$$

Net Count Rate in Sample ($C_n \pm c_n$): $124 \pm 0.13$ c/s

Decay Factor (F): 0.12

Correction Factor for Ash Not Counted (f): 2.28

Determination of Retained Fraction: $\frac{A_2 \pm a_2}{A_1 \pm a_1} =$

$$\frac{(124 \pm 0.13 \text{ c/s})(0.68)(K)(1.0 \pm 0.05 \text{ ml})}{(3647 \pm 3.4 \text{ c/s})(0.12)(K)(1.0 \pm 0.05 \text{ ml})} \quad (2.28)$$

$$A_2 \pm a_2 = (124)(0.68)(K)\left\{1 \pm \sqrt{\left\{\frac{0.13}{124}\right\}^2 + \left\{\frac{0.05}{1}\right\}^2}\right\}$$

$$= 84.3 \pm 4.2 \text{ c/s}$$

$$A_1 \pm a_1 = (3647)(0.12)(K)\left\{1 \pm \sqrt{\left\{\frac{3.3}{3647}\right\}^2 + \left\{\frac{0.05}{1}\right\}^2}\right\}$$

$$= 438 \pm 21.9 \text{ c/s}$$

$$= \left[\left\{\frac{84.3}{438}\right\}\left\{1 \pm \sqrt{\left\{\frac{4.2}{84.3}\right\}^2 + \left\{\frac{21.9}{438}\right\}^2}\right\}\right](2.28)$$

$$= (0.19 \pm 0.0134)(2.28)$$

$$= (0.433 \pm 0.0305)(100\%)$$

$$= 43.3 \pm 3\%$$

It is important to remember that this retention value does not require consideration of any error associated with the amount of radioactivity per microsphere.

To obtain the associated error for the total ash retention value for each isotope in each burn series, the variances associated with each ash sample in the series were summed, and the square root taken.[25]

To determine an average retention value of any particular isotope in any particular sequental burn (for instance, Ce-141, first non-radioactive burn) the mean and standard deviation of the retention values for that sequence was calculated. The standard deviation of the means was used rather than a sum of the associated errors because the errors between samples were more significant

[25]Knoll, G.F., Radiation Detection and Measurement, John Wiley& Sons, Inc., New York, N.Y., 1979, p. 132.

than the errors within each sample, with respect to indicating an average retention value. The only exception to this procedure was Nb-95 because of the large associated error in each ash sample.

## C. STACK SAMPLING CALCULATIONS AND RESULTS

The stack sampling portion of this study did not collect any detectable particulate activities. However, several interesting parameters concerning the flow dynamics of the stack were discovered.

The first two series of burns were sampled using the traverse method described in EPA Method 5[26]. The inside diameter of the stack warranted eight different sampling points on two traverses, as shown in Figure 9. To better describe the air flow profile of the stack, the data collected from each sampling point was averaged over the six burns that this method was used. The equations used to calculate the average stack gas velocity for each sample point are listed below:[27]

Eq. 1: Dry Gas Volume Measured Corrected to Standard Conditions = $V_{m\ std}$.

$$V_{m\ std} = \frac{(V_m)(T_{std})(P_{bar} + H/13.6)}{(T_m)(P_{std})}$$

---

[26]U.S. Environmental Protection Agency, 1982, Code of Federal Regulations, Title 40, Part 60.

[27]U.S. Environmental Protection Agency, 1979, Source Sampling for Particulate Pollutants: Student Manual for APTI Course 450, EPA 450/2-79-006.

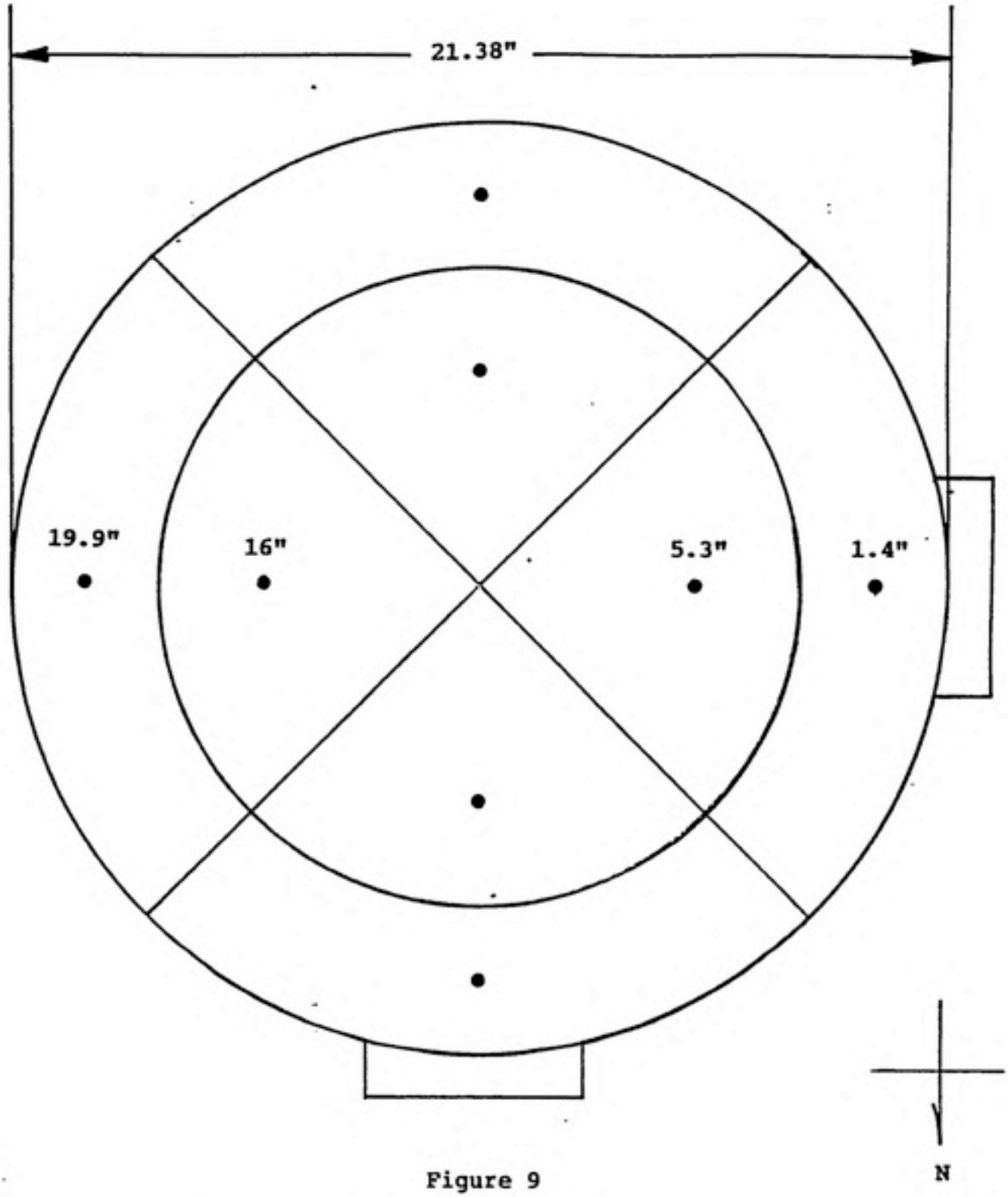



Figure 9

Eight Sampling Locations
in Incinerator Stack
as Perscribed in EPA Method 5

Where:

$V_m$ = dry gas volume measured by dry gas meter

$T_{std}$ = standard absolute temperature

$P_{bar}$ = barometric pressure at dry gas meter

$H$ = average pressure at dry gas meter

$T_m$ = absolute temperature at dry gas meter

$P_{std}$ = standard absolute pressure

Eq. 2: Proportion of Water Vapor in Stack Gas Stream by Volume = $B_{ws}$

$$B_{ws} = \frac{V_{wc(std)} + V_{wsg(std)}}{V_{wc(std)} + V_{wsg(std)} + V_{m(std)}}$$

Where:

$V_{wc(std)}$ = volume of water collected at standard conditions

$V_{wsg(std)}$ = volume of water desiccated at standard conditions

$V_{m(std)}$ = dry gas volume measured at standard conditions

Eq. 3: Wet Molecular Weight of Stack Gas = $M_s$ (lb/lb-mole)

$$M_s = M_d(1-B_{ws}) + 18(B_{ws})$$

Where:

$M_d$ = dry molecular weight of stack gas (assumed value of 29.0 lb/lb-mole because of lack of Orsat capability)

$B_{ws}$ = proportion of water vapor in stack gas

Eq. 4: Average Stack Gas Velocity = $V_s$ (ft/sec)

$$V_s = K_p C_p (T_s/P_s M_s)^{1/2} (\Delta p_{avg})^{1/2}$$

Where:

$K_p$ = pitot tube constant

$C_p$ = pitot tube coefficient

$T_s$ = absolute average temperature of stack gas

$P_s$ = absolute stack gas pressure

$M_s$ = wet molecular weight of stack gas (assumed value of 28.03 lb/lb-mole)

$\Delta p_{avg}$ = average velocity pressure of stack gas

The data from the first six burns indicated that a severe temperature and flow rate gradient existed across the stack. Figure 10 shows the average temperatures measured at each sample point, and Figure 11 shows the corresponding average flow rates.

Using the results of the first burns, the sampling point with the highest flow rate was identified. Since the traverse method was not collecting detectable amounts of activity, the point with the highest flow rate was then designated as the sole sampling site in hopes that this area would also contain the highest concentration of particulates. This method was used for the remaining nine burns. None of the samples collected using this method produced any detectable activity.

The limiting factors described in Part II E of this report prevented the collection of any useful effluent data concerning releases of radioactivity. Had the sampling probe not been so temperature sensitive, a time constraint would not have been

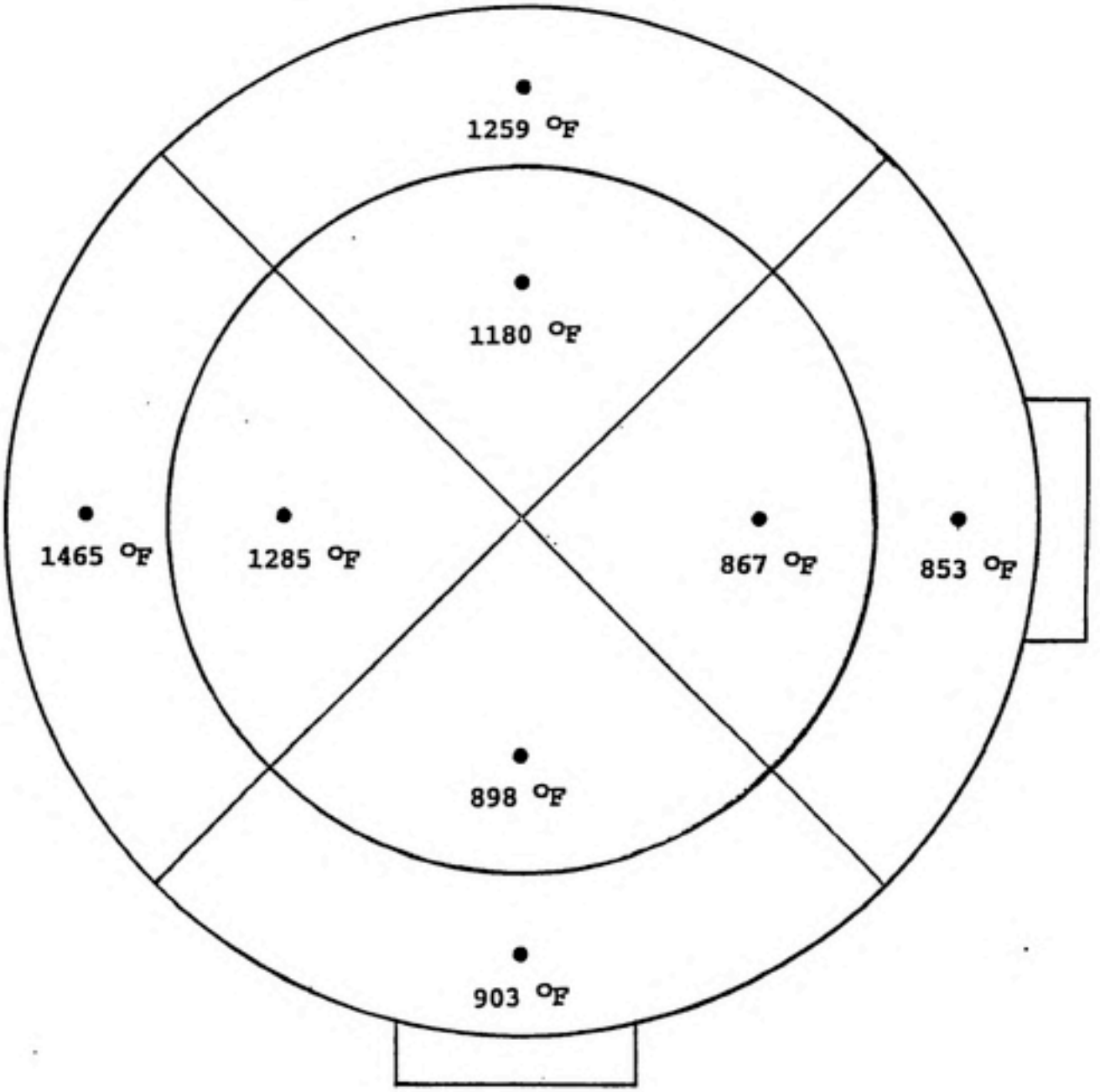



Figure 10

Average Stack Temperatures
From Six Experimental Burns

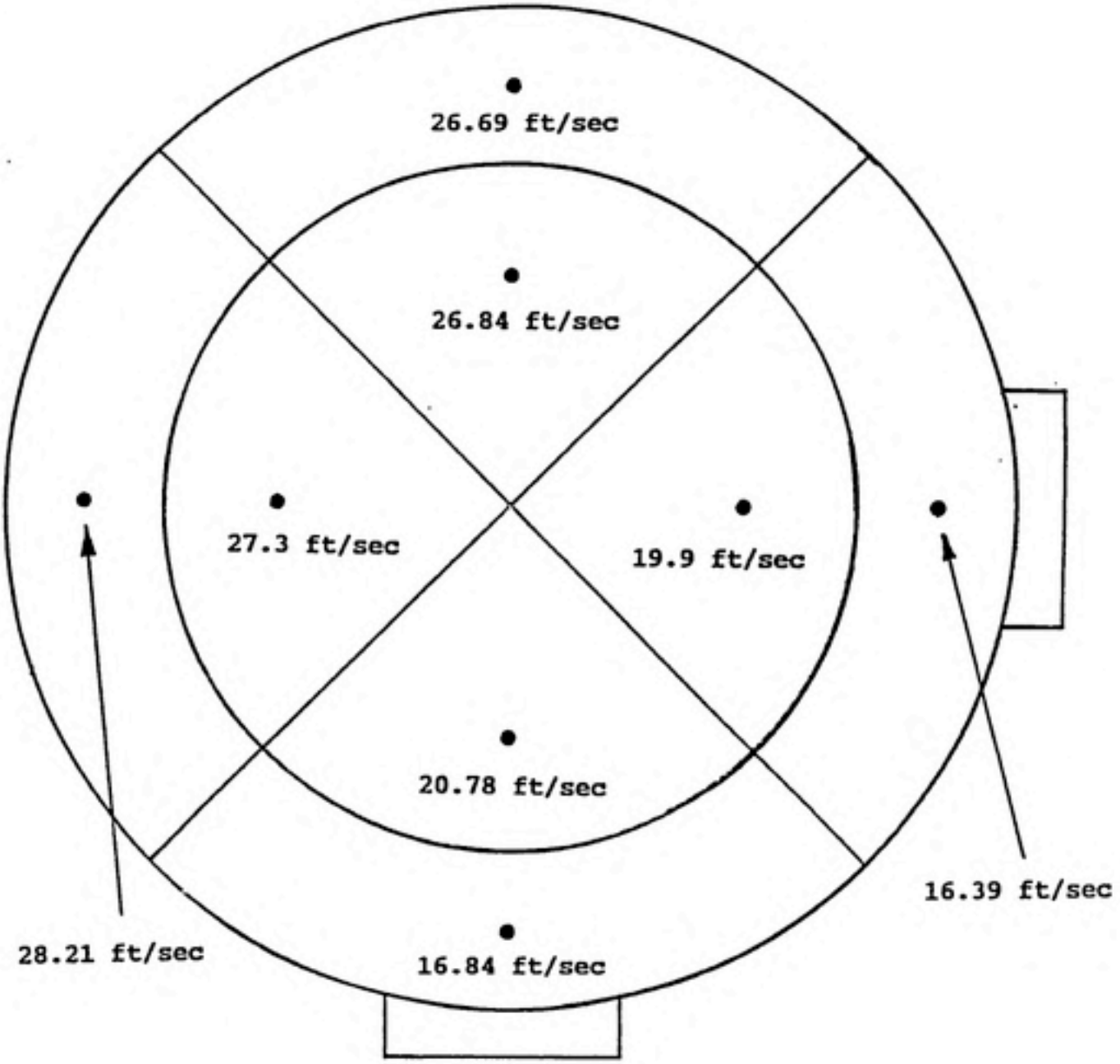



Figure 11

Average Stack Velocity
From Six Experimental Burns

imposed on the burns. Limiting the burns to one hour effectively reduced the amount of activity that could be incinerated during each burn. Larger amounts of activity incinerated would have permitted the air sampling portion of this study to be more effective. The sample calculation shown below, demonstrates how the amount of radioactivity in the animal carcass hampered the sampling capabilities of the stack sampling system:

Cross Sectional Area of Stack Sampling Probe: $7.2 \times 10^{-4}$ $ft^2$

Cross Sectional Area of Stack: 2.5 $ft^2$

Fraction of Stack Area Sampled: 0.00029

As an example, assume that the typical amount of one microsphere isotope is burned, which would be approximately $2.8 \times 10^4$ Bq (For this example, Ce-141 is used). Assuming 100% release of activity into the effluent, and assuming that the an even effluent flow rate existed across the sampling point, the amount of Ce-141 that would be collected would be 8.1 Bq. The LLD for Ce-141 was 1.4 Bq. These assumptions did not take into account any retention of activity in the ash or refractory, and that a severe flow rate gradient existed across the sampling point.