

CHARACTERIZATION OF AN AEROSOL SAMPLE FROM THREE MILE ISLAND REACTOR AUXILIARY BUILDING

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EXECUTIVE SUMMARY

The accident which occurred at the Three Mile Island Unit Two Nuclear Generating Plant on March 28, 1979 resulted in contamination of the containment and auxiliary buildings. Analyses were performed at this laboratory for: (1) radioisotopic composition and (2) dissolution characteristics of material collected on a filter during a week of continuous air sampling in the auxiliary building. This work was done to help characterize the environment in which cleanup operations are being performed. Gamma and alpha spectroscopy along with scanning electron microscopy were employed to determine the elemental and isotopic composition of the aerosol. The major radioisotopes found on the filter were ^{89}Sr , ^{90}Sr , ^{134}Cs and ^{137}Cs . Dissolution behavior of the aerosol in a synthetic serum ultrafiltrate containing DTPA was determined using both static and flow-through systems. In both systems, rapid dissolution of greater than 90% of both $^{89-90}\text{Sr}$ and $^{134-137}\text{Cs}$ was found. Only slight differences in the dissolution rates of Sr and Cs were observed. Scanning electron microscopic analysis showed the presence of respirable size particles as well as larger particles ranging up to 10 μm . The major matrix components were Fe, Ca, S, Mg, Al and Si. Except for Al and Si, all matrix materials dissolved in the *in vitro* dissolution systems. Although the radionuclides were present in a heterogeneous matrix, their observed rapid dissolution behavior justified their classification as Class D compounds.¹ Knowledge of such a classification enables better evaluation of bioassay data and predictions of dose distribution after inhalation exposure to this aerosol. The techniques used in this study may be applicable to other aerosols of unknown composition.

INTRODUCTION

The Three Mile Island nuclear reactor accident in March 1979, resulted in the contamination of the containment and auxiliary buildings. During the cleanup operations of the auxiliary building, airborne radioactive fission products were encountered. The potential inhalation of these aerosols by workers and others involved in the cleanup prompted the characterization of the aerosol. In addition, knowledge of the characteristics of this aerosol could provide some information on the nature of the release. Thus, the quantities, distributions and the physicochemical forms of the radioactive isotopes were the primary data needed. Data on elemental composition, size characteristics and other relevant physicochemical properties of the aerosols were also desirable.

The dissolution behavior of an inhaled aerosol is one of the major factors that determine its retention, translocation and resulting dose distribution.¹ Since the nature of the aerosol from the auxiliary building of the Three Mile Island (TMI) was not known, there was no basis for predicting its dissolution in the lung after inhalation deposition. An important variable affecting the dissolution rates is the solvent. A solvent which has been used extensively for *in vitro* dissolution studies of several radionuclides in a variety of chemical forms is a synthetic serum²⁻⁴ ultrafiltrate. The *in vitro* dissolution of many aerosols in this solvent were qualitatively comparable to their *in vivo* dissolution in the lungs of animals.^{3,4} Therefore, *in vitro* dissolution studies on this aerosol were conducted using this solvent. Other auxiliary studies for characterizing the aerosol included scanning electron microscopic (SEM) analysis, energy dispersive x-ray analysis (EDXA) and alpha and gamma spectroscopy.

Aerosol Sample

The aerosol sample, which TMI personnel supplied, was obtained by filtering about 10^9 cc of air from the auxiliary building on a 2-inch diameter, glass fiber filter during a period of about 8 days. According to the information supplied by personnel from the GPU Service Corporation, the isotopic concentrations were approximately 20% ^{137}Cs , 10% ^{134}Cs , 60% ^{89}Sr , 2-4% ^{90}Sr - ^{90}Y , 2-4% ^{140}Ba - ^{140}La and 1% of ^{58}Co and ^{60}Co . The sample appeared to be a thick, black deposit. Presumably, this material contained normal room aerosol, combustion aerosol and other unknown aerosols.

The total β and γ activity on the filter was about 3.5 μCi . The filter was divided into four segments (Table 1) with approximately 40% activity on each of segments 1 and 2 and 10% activity on each of segments 3 and 4. Segments 1 and 2 were used in the dissolution studies. Segment 3 was used for electron microscopic analysis and the fourth segment for determining isotopic composition by radiochemical analysis.

Table 1

Radionuclide Activity (nCi) in the TMI Aerosol Filter Samples as of 79305 (11/1/79)

Sample	^{137}Cs	^{134}Cs	^{90}Sr - ^{90}Y	^{89}Sr
Segment 1	657	157	124	440
Segment 2	634	164	120	424
Segment 3	157	37	23	105
Segment 4	<u>218</u>	<u>54</u>	<u>41</u>	<u>146</u>
Total	1666	412	308	1112

Dissolution

Two different dissolution systems, a flow-through system and a static system, were used in this study.² In the flow-through system, the solvent was directly in contact with the particles. This dynamic dissolution system provides estimates of early dissolution rates. The static dissolution system was a simpler system better adapted for long-term dissolution studies.

In both dissolution systems, the samples were sandwiched between two Nuclepore[®] filters (100 nm pore diameter) and secured in a filter holder. The flow-through filter holder was a 47 mm polypropylene filter holder (Millipore, Inc.). A Delrin[®] filter holder⁵ with both filter faces open to the solvent, was used in the static system.

The solvent used in this study was a synthetic ultrafiltrate containing DTPA (SUF + DTPA). The pH, ionic concentration, precipitating and chelating capacity of this solvent were designed to be similar to that of serum ultrafiltrate.^{2,3} The solvent pH was maintained between 7.3 and 7.4 by exposing it to 5% CO_2 in air. Both dissolution studies were conducted at 37°C by maintaining the dissolution systems in a water bath.

A schematic of the flow-through system is shown in Figure 1. The solvent in a lucite reservoir was pressurized with 5% CO_2 in air. The maintenance of a 0.5 ml/min flow rate required a pressure of 1.5 to 2.5 psig. A Gelman[®] filter cartridge with pore diameter of 0.2 μm was used as a prefilter for the solvent. The eluant fractions were collected at 20-minute intervals during the first day and then at 30-minute intervals by using a fraction collector.

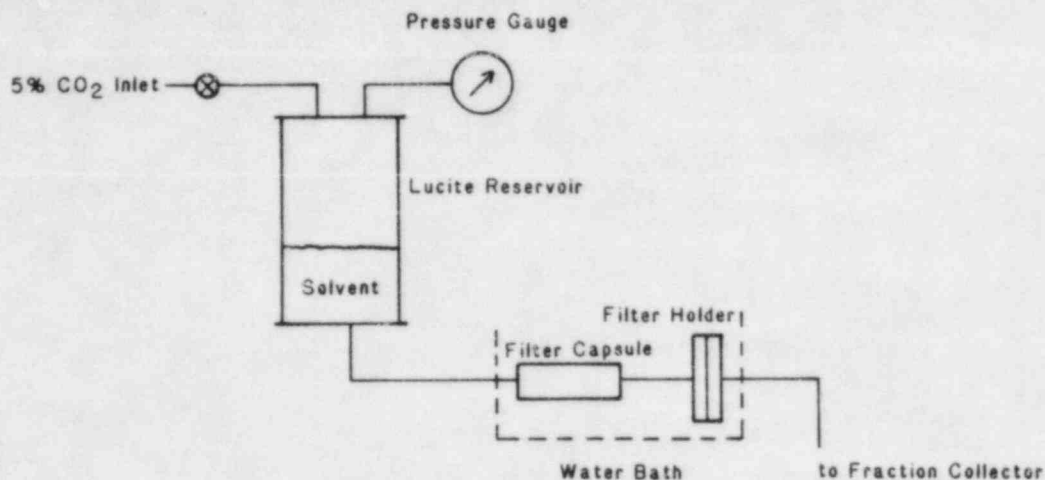


Figure 1. A schematic design of the flow-through dissolution system used to study the *in vitro* dissolution of radionuclides present on an air filter sample collected in the auxiliary building at the TMI facility.

In the static system, the filter assembly was placed in 100 ml of solvent in a 500 ml container, sealed with a lid and then placed in the water bath. Solvent changes were made at 2, 4, and 18 hours and then every 24 hours. The solvent was changed by removing the filter assembly from the solvent and placing it in fresh solvent.

Analysis of Dissolved Activity

Gamma activity dissolved in the flow-through system was determined by counting the fractions directly in a Beckman Autogamma Counter (Model 8000) with a NaI crystal detector. Ten milliliter aliquots of the static system samples were counted in the same detector. Total dissolved beta activities were determined by counting 100 μ l aliquots prepared on planchets in a Beckman Wide Beta II gas proportional counter.

Undissolved activity remaining on the filter or total sample on segment four of the filter sample were analyzed by dissolving the activity in 7 M HCl at 80°C. After dilution, aliquots were counted in the Wide Beta II counter and the Autogamma counter. The filter retained < 1% activity, although some black residue was still present.

The ^{90}Sr - ^{90}Y activities in the original aerosol sample and in the dissolution fractions were determined by precipitating ^{90}Y phosphate at pH 5⁹ and counting the associated beta emissions. Total Sr was precipitated from the remaining solution at pH 10.

Determination of Gamma Activities

Gamma activities in the aerosol samples were determined by counting for 17 hours in a coaxial Ge-Li detector with an active surface area of 16 cm² and a resolution of 1.4 keV at 622 keV. After the dissolution studies, the filter samples were also counted in this Ge-Li detector system to determine the remaining gamma activity.

Scanning Electron Microscopy Energy Dispersive X-ray Analysis

Samples were transferred onto polished carbon stubs by pressing the carbon stubs against the sample on the filter. Replicate carbon stub samples were obtained from the same aerosol filter and were observed in a JEOL Model JSM-35 scanning electron microscope (SEM). Energy dispersive x-ray analysis (EDXA) of selected areas was performed by using a Li-drifted Si detector (Kevex

Model 78) and Kevex Model 5100 multichannel analyzer. Micrographs were also obtained by using the secondary electron imaging mode of the SEM. All micrographs were taken using 25 kV accelerating voltage and about 1 na beam current.

Alpha Spectroscopy

An aliquot of the original aerosol sample in 7 M HCl solution was evaporated to dryness, redissolved in 3 M HCl and the iron was removed by ethyl ether extraction. The actinides were then electroplated on stainless steel discs from a $\text{NaHSO}_4 \cdot \text{Na}_2\text{SO}_4$ buffer at pH 2.¹⁰ The alpha activities were determined in a 128-channel pulse height analyzer with a resolution of 14.8 keV per channel. The detector was a 300 mm² surface barrier silicon detector (Princeton Gamma Tech). The duration of alpha counting and analysis was 19 hours.

RESULTS

A gamma spectrum of the aerosol sample is shown in Figure 2. All four segments had identical gamma peaks with the same ratios of ^{134}Cs to ^{137}Cs . The amounts of the major radionuclides present in the different segments and the total aerosol filter are shown in Table 1. The activities of ^{134}Cs and ^{137}Cs were determined directly from gamma spectroscopy and the activities of ^{90}Sr - ^{90}Y and ^{89}Sr on Segment 4 by radiochemical methods. The ^{89}Sr activity was calculated by subtracting ^{90}Sr - ^{90}Y activity from the total observed ^{89}Sr and ^{90}Sr - ^{90}Y activity. The activities of ^{90}Sr - ^{90}Y and ^{89}Sr in the other three segments were calculated from the ^{137}Cs activity in each segment and the ratios of ^{90}Sr - $^{90}\text{Y}/^{137}\text{Cs}$ and $^{89}\text{Sr}/^{137}\text{Cs}$ observed in Segment 4.

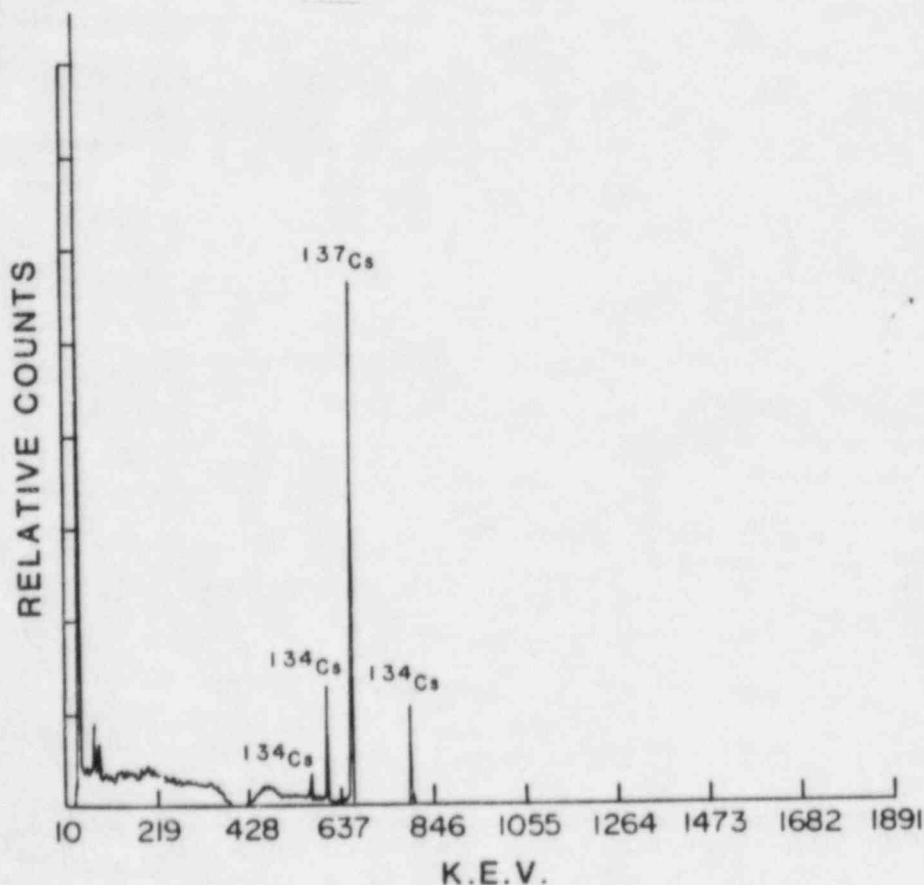


Figure 2. Gamma spectrum of Segment 1 of the TMI aerosol as determined with a coaxial Ge-Li detector.

The dissolution data obtained in the flow-through and static systems are summarized in Figures 3 and 4, respectively, as fractions of undissolved activity remaining on the filter versus elapsed time. Total gamma activity was directly determined on the filters before the samples were subjected to dissolution. The initial total beta activities on these filters were calculated from the ratio of total beta counts in the Wide Beta II counter to ^{137}Cs in segment 4 and the ^{137}Cs count on segment 1 (flow-through system) and segment 2 (static system). The undissolved fractions were calculated on the basis of beta counts. The ^{90}Sr - ^{90}Y activity, both initial and those in the solutions were determined radiochemically. Summation of all dissolved radioactivities and those remaining on the filter provided estimates of initial amounts of radioactivities within a few percent of those activities shown in Table 1.

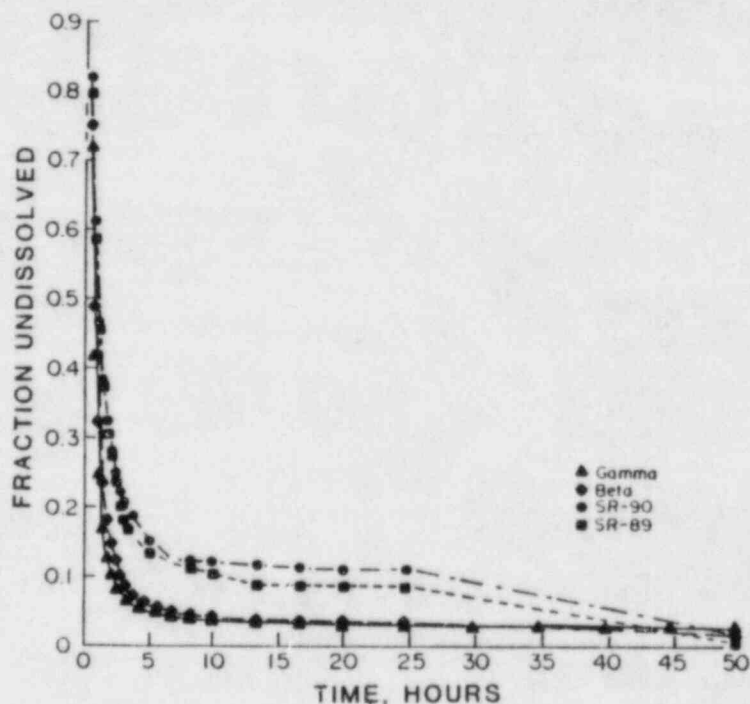


Figure 3. Dissolution of radionuclides in a flow-through system expressed as fraction dissolved versus elapsed time.

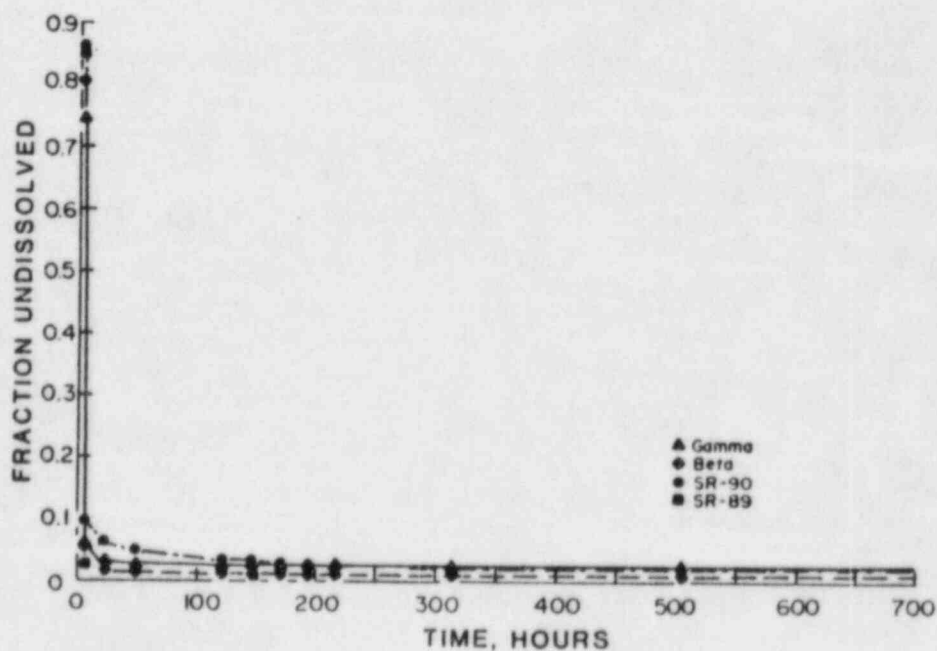


Figure 4. Dissolution of radionuclides in a static system expressed as fraction dissolved versus elapsed time.

Scanning electron micrographs of particles from Segment 3, which represents the original aerosol sample, and those from Segment 1 after dissolution in the flow-through system are shown in Figures 5 and 6, respectively. Average elemental composition of a large number of particles from Segment 3 and 1 are shown in Table 2.

An alpha spectrum of the original aerosol is shown in Figure 7. Based on ^{137}Cs counts, this sample represented about 4% of the total sample. The total activity of ^{239}Pu , ^{238}Pu or ^{241}Am , and ^{236}Pu in this sample is 0.50 dpm. On this basis, an estimate of total actinide alpha activity on the original aerosol was calculated to be 13 dpm.



Figure 5. Scanning electron micrographs of original aerosol sample from Segment 3 of the TMI filter. Magnification 3000 X.

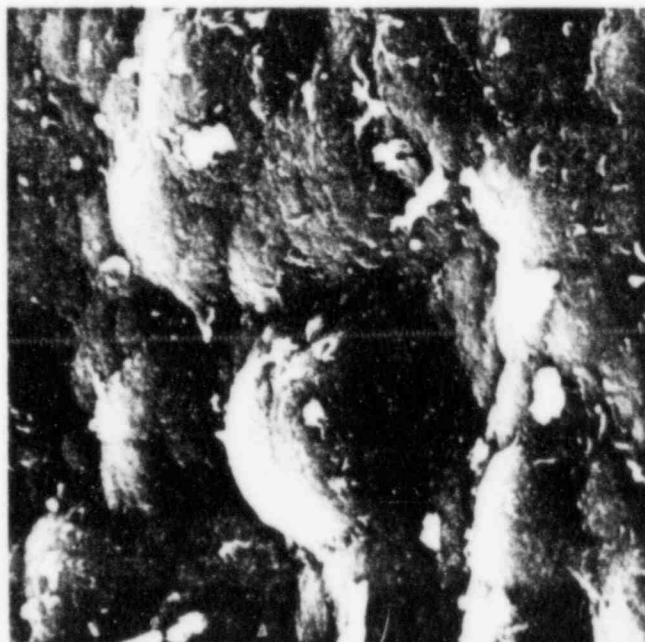


Figure 6. Scanning electron micrograph of aerosol particles from Segment 1 of the TMI filter after dissolution experiment in the flow-through system. Magnification 3000 X.

Table 2

Elemental Composition of the TMI Aerosol Filter Sample
as Determined by Energy Dispersive X-Ray Analysis

<u>Element</u>	<u>Weight (%)</u>	<u>Atomic (%)</u>	<u>Standard Deviation</u>
<u>Segment 3 Before Dissolution</u>			
Mg	7.2	9.6	2.9
Al	16.2	19.3	1.6
Si	27.1	31.1	1.5
S	17.3	17.4	1.1
Ca	18.0	14.4	0.93
Fe	14.3	8.2	1.0
<u>Segment 1 After Dissolution</u>			
Al	63.8	64.8	7.9
Si	36.2	35.3	3.2

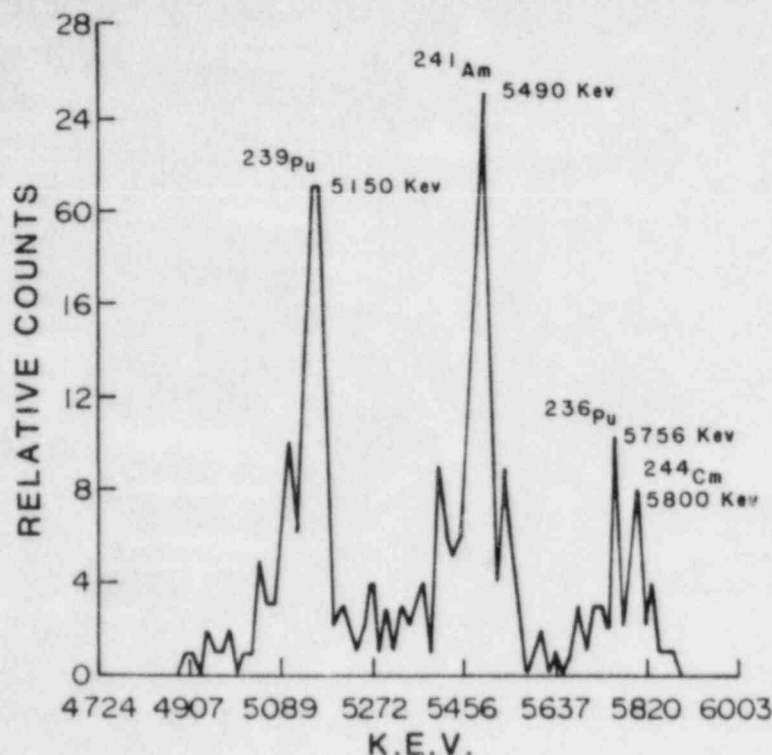


Figure 7. An alpha spectrum of the TMI aerosol sample determined on an electroplated sample by using a surface barrier detector and pulse height analyzer.

DISCUSSION

Analysis of the gamma spectrum (Fig. 2) indicated that ^{137}Cs and ^{134}Cs were the primary gamma-emitting radionuclides on the filter. The data supplied by GPU suggested the presence of small quantities of ^{140}Ba , ^{140}La and ^{60}Co . Since the gamma spectrum (Fig. 2) was obtained about a month after the aerosol was collected, it was expected that no ^{140}Ba or ^{140}La would be observed. The energy peaks at 1168 and 1365 keV, from ^{134}Cs , could have been interpreted by TMI personnel as that of ^{58}Co and ^{60}Co . It is noteworthy that there was no indication of any lanthanide isotopes, such as ^{144}Ce , present in the sample.

The quantities and proportions of ^{134}Cs , ^{90}Sr - ^{90}Y and ^{89}Sr (Table 1) were similar to the estimates provided by GPU. All data indicated that the major radionuclides were ^{134}Cs , ^{137}Cs , ^{89}Sr and ^{90}Sr - ^{90}Y . If there were other beta-emitting lanthanides, they would have been noticeable during the repeated counting of the ^{90}Y fractions obtained during the radiochemical analysis of ^{90}Sr - ^{90}Y . The half-time of decay of the ^{90}Y fraction indicated no major, long-lived lanthanide isotopes. The observed actinide radionuclides (Fig. 7) were unexpected. Although the alpha activity was only 13 dpm in the total aerosol sample, it is a significant observation that there were actinide nuclides present in the aerosol. Since the physicochemical behavior of lanthanide and actinides are similar, there may have been minute but undetectable amounts of gamma- and beta-emitting lanthanide radionuclides also present in the original aerosol.

The presence of major isotopes of alkali and alkaline earth elements, which are generally water soluble, but not other fission products such as ^{144}Ce and ^{91}Y in detectable quantities suggest that the major release mechanism was probably one of dissolution in water and then dispersion in the auxiliary building. In addition, the presence of the small amount of insoluble actinide suggests other possible mechanisms of particle release as a minor contributing factor. The elemental analysis (Table 2) suggests that the aerosol consisted of nonradioactive materials. Comparison of the composition before and after dissolution suggests that only Al and Si were insoluble and the other elements were present as soluble materials. The radionuclides were

present as minor components in a heterogeneous matrix consisting of Fe, Ca, S, Mg, Al, Si and carbonaceous material. The aluminum and silicon represented 43% of the total inorganic mass. It should be noted that the EDXA was incapable of determining C, O₂, H₂, etc. The filter sample was black both before and after dissolution which suggested the presence of soot or other carbonaceous materials.

The sizes of the aerosol particles were estimated from the electron micrographs (Figs. 5 and 6). Respirable-sized particles as well as larger particles ranging up to 10 μ m were observed. Although the relative number density of the particles in the electron micrograph after dissolution (Fig. 6) was less than that before dissolution (Fig. 5), there was no apparent size-dependent, preferential dissolution of particles from the filter. Therefore, it is not possible from our study to deduce the size distribution of the soluble fraction which contained the radionuclides. However, it is reasonable to conclude that at least a fraction of the radionuclides were associated with the respirable-size particles.

The dissolution data (Figs. 3 and 4) show rapid dissolution of > 90% of all isotopes. The Cs isotopes appear to dissolve almost entirely in both dissolution systems. Less than 2% of the Cs isotopes could be detected on the filters after the dissolution experiments. The data show that within 5 hours, more than 95% of the Cs isotopes dissolved in both systems. The total beta dissolution rates were somewhat slower than the dissolution rates of the Cs isotopes. It should be remembered that the total beta activity was the sum of ⁸⁹Sr, ⁹⁰Sr-⁹⁰Y, ¹³⁴Cs and ¹³⁷Cs beta activities. The ⁹⁰Sr-⁹⁰Y dissolution rates suggest that a slightly higher fraction, 5 to 10% of the activity, was relatively insoluble compared with Cs. This undissolved fraction of the Sr was present in both dissolution systems.

The International Commission on Radiological Protection (ICRP) has grouped specific chemical forms of radionuclides into three classes (D, W, and Y) to describe their expected patterns of retention in different portions of the respiratory tract.⁸ The three classes, days (D), weeks (W), and years (Y) describe both the expected clearance by mechanical and dissolution/absorption pathways. These classifications have been derived for single radionuclides in known chemical forms based on available *in vivo* and *in vitro* data. However, the ICRP solubility classification scheme does not include predictions for heterogeneous mixtures such as the one present on the sample analyzed here.

Results from the *in vitro* dissolution tests performed on this sample demonstrate that most of the radionuclides present dissolved very quickly (within 5 hr) in the SUF plus DTPA solvent. Although an exact correspondence between *in vitro* and *in vivo* dissolution characteristics has not been established for these radionuclides in this form and mixture, it seems reasonable to assume that a similar rapid dissolution would occur *in vivo*. Thus, based on this assumption and the observed *in vitro* dissolution characteristics of the present sample, it is appropriate to evaluate observed air concentrations with respect to derived air concentrations (DAC) by classifying the radionuclides as class D materials.

This study has demonstrated the applicability of the above techniques for characterizing airborne radionuclides of unknown physical and chemical composition. The data may be useful in determining the body burden of these radionuclides from bioassay data and helping to estimate the dose distributions resulting from inhalation of the aerosol.

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AFFIDAVIT

Randall C. Thompson

I, Randall C. Thompson, worked at Three Mile Island Unit II during the month of April, 1979. Before that, I had been a health physics and chemistry technician for PAD Services of Pittsburgh, PA and had held the title of Senior Plant Chemist at the Peach Bottom nuclear plant for the last two years of my nuclear career. For the first ten days to two weeks of my stay at TMI I was the only hp qualified as surveillance technician to change the vent stack monitor filters. I also did containment sampling and provided hp coverage for the task of changing the banks of auxiliary building charcoal filters.

While working at TMI, I changed the vent stack monitor filters twice a day. In every single case, the dose rate from these filters were very high, often constituting a "high radiation area" (more than 100 mr/hr. By way of comparison, consider that at Peach Bottom a four cubic ft./min filter would yield a dose rate of less than 10 mr. Based on this, I came to the conclusion that the plant was emitting large quantities of radioactive gas through the vent stack.

On two occasions, once in the ~~HP~~ST counting trailer and once at the HP checkpoint I was able to look at isotopic identification printout sheets from those filters. On both occasions iodine 131 was particularly high, showing concentrations of one to five x10/-5 mc/ml.

Often, because of the high dose rates of these filters, they were set aside so that no one could disturb it before it was decayed to some degree. I was asked specifically on two occasions to hide the filters. On both of these instances, when I went to retrieve the hidden filter it had already been taken.

In all cases, the daily reports of radiation released from Three Mile Island were much lower than I personally observed.

Randall C. Thompson

I, ^{all S/}~~Randolph~~ Thompson,

From 1976 through 1978, I was the senior health physics and chemical technician, senior plant chemist and surveillance technician (air release specialist) at the Peach Bottom Nuclear Generating facility in Peach Bottom, Pennsylvania.

As a health physics technician, it would be unthinkable not to check for alpha radiation on a daily basis, under normal operating conditions where the possibility of alpha presence is practically nil. However, at TMI, where the conditions present indicated a stronger possibility for alpha contamination than normal, we were not able to check for alpha on a routine or spot basis. This was because there were no alpha counters available.

I continued to suspect alpha presence at TMI and inquired of many people, including at least four NRC inspectors with whom I worked. I asked every utility person that I saw for an explanation as to why there were not alpha counters available. The explanation was that there was no alpha, and, thus, no reason to check. This was unacceptable as an explanation since checking is routine. Upon further questioning, it was explained by MetEd health physics employees that, indeed, during the first week after the accident, there was an "alpha scare", but after "independent analysis", it was determined that alpha was not alpha after all, but was "piggy-back beta".

Following the accident, planned releases were scheduled and monitored. There was one situation where the monitoring system failed before the release. I tried to set up an alternative sampling system. MetEd wanted to go ahead with the release prior to my setting up the alternative system which would not have taken more than 30 minutes. I pleaded my case with the head NRC person, arguing that legally or morally they could not go ahead. The NRC person stated that they had to go ahead, quipping, "Its the only game in town." The release occurred, unmonitored. I lodged a complaint. To further identify this occurrence, it was in connection with the sampling of reactor water through the Auxiliary Building.

Jaudall C. Thompson

AFFIDAVIT

Marian E. (Joy) Thompson

I, Joy Thompson, was employed by RAD Services, Inc. of Pittsburgh, Pennsylvania as a health physics technician assigned to the Dosimetry department at Three Mile Island Unit II for the last three weeks of April, 1979. My husband, Randall C. Thompson also worked at TMI as a ranking health physics technician and surveillance technician.

Before the accident, my husband and I were incorporated as a publishing company, and published a monthly magazine for children about skateboarding. Previous to that, my husband had spent seven years in the nuclear industry as a health physicist.

When Three Mile Island had its accident, we decided to investigate its causes and effects first-hand, as the discrepancies in what we were hearing on the news and what we were hearing from our friends who were already working at TMI were tremendous.

During my employment at TMI, our friend and co-worker, David Bloombaum, was accidentally contaminated with radioactive iodine on his face and forehead. Nasal smears were taken before and after decontamination efforts by both David and my husband, and the geli identified the contamination as iodine. The body scanner, however, found none on the day of the contamination. The next day, after David had developed beta burns on the inside of his nose and the upper part of his throat, the NRC body scanner also found no iodine. It was my conclusion in this instance that the body scanners were not programmed to detect iodine.

In my job as hp in the dosimetry department, I processed and recorded the daily TLD measurements for all personnel and visitors to TMI. The two TLD machines I used in that processing were the property of Metropolitan Edison and the NRC. Each concern was in charge of calibrating their own machine. I randomly placed groups of badges into one or the other machine to be read, depending on which was busy and which available. Often there would be discrepancies in one or the other machine's average dose measurement, when the average dosages should have been within a few MR of each other.

When I would inform my superiors of the discrepancies, the machines would be re-calibrated during the day shift, but the differences in average dose measurement would grow each day thereafter until I would again have to complain to my superiors. It was my belief that both machines were deliberately miscalibrated to underestimate actual dose.

Once in mid-April, I was brought the worker's badges who were assigned by my husband to the task of changing the banks of

charcoal filters in the auxiliary building. The badges were usually brought over to the trailer from the island as each group of workers emerged from the aux building in order to immediately determine how much dosage they had received so as not to accidentally overexpose them. Most of the time a health physics technician would bring the badges, I would read and record the dosage immediately, then he would take that information back to the island.

One time, a GPU executive engineer (management-level person) brought the badges for me to read. I was alone in the trailer and it was about two a.m. Despite the fact that the badges were in blue cases (indicating whole-body dose as opposed to yellow badges which were used as extremity measurements), I was given a direct order to intercept the computer documentation and enter the dosages as extremity. The doses ranged from 150 millirem to three hundred and fifty millirem...significant doses. Because those doses were entered as extremity doses, they did not become part of those worker's permanent dose record.

The fact that the computer was programmed to disregard any dose reading under 10 millirem might not be significant in the operation of a normal nuclear power plant, but at TMI the doses were high and the badges were read daily instead of once or twice a month. Thus a person receiving 10 mr or less each day would have no record of any radiation exposure. In some instances, this practice could result in significant underestimation of dose and negation of legal rights if the worker were later to develop diseases caused by exposure to radiation.

The underestimation of dosage and the deliberate cover-up of high doses at Three Mile Island is unethical behavior. I believe that evidence of unethical behavior by the utility should be a consideration in any decision to approve the start-up of TMI Unit I.

Marian E. (Jay) Thompson