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SAMPLING AND CHARACTERIZATION OF AEROSOLS PRODUCED UNDER SIMULATED NUCLEAR REACTOR ACCIDENT CONDITIONS¹

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INTRODUCTION

A significant portion of the radioactive material available for release to the environment from a nuclear reactor in the event of a severe accident is postulated to be in aerosol form. Such an accident occurs when removal of heat from the reactor core is impaired or eliminated by failure of one or more components of the coolant system. The most common type of commercial reactors are water cooled. The uranium fuel and its cladding melt, allowing the release of fission products, the radioactive isotopes that are the by-products of the fission process. The volatile fission products will be transported into cooler regions of the reactor by steam, produced by the heating of coolant water present in the core at the time of the accident. Some of the most chemically reactive and biologically important volatile fission products, cesium, iodine, and to some degree tellurium, are believed to form compounds that will condense into aerosol particles in the cooler regions of the reactor. The zirconium-tin cladding will be oxidized by the steam, producing hydrogen and additional aerosol species.

Argonne National Laboratory is engaged in research to characterize the potential releases from water cooled reactors. The Laboratory has conducted a series of four experiments in which reactor fuel was heated to the point of severe cladding disruption in flowing steam environments under temperature and pressure conditions predicted for hypothetical accident scenarios. The experiments were conducted in Argonne's TREAT research reactor facility. The released fission products were sampled close to the fuel. Posttest analyses of the samples are being conducted to obtain information on their physical and chemical characteristics. This information, in conjunction with the thermal-hydraulic history of the tests, will be used in the development and evaluation of models that describe releases for different accident scenarios.

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Challenges

The aerosol sampling system was designed to yield information on the size, composition, morphology, and concentration of aerosol particles in the steam/hydrogen gas streams. The operational requirements of the system imposed by the test environments were as follows. It had to function under conditions of high temperature (approximately 650K), and high pressure (two of the tests were conducted at approximately 0.3 MPa and the other two were conducted at approximately 8 MPa). The system had to be resistant to the reactive carrier gas. Since the tests were conducted in a reactor radioactivity levels were very high. Space inside the reactor for the test vehicles was very limited. Gas velocities were relatively low, prohibiting the use of certain inertial samplers.

Experiments of this kind had not been performed before. Therefore, data was not available that could have been used for predicting the size range and concentrations of the particles. This resulted in the requirement that the system have adequate collection efficiencies for a wide size range of particles, that it not be susceptible to overloading by a high concentration aerosol, and that it collect sufficient numbers of particles for analysis if the concentrations were low. Expected particle compositions based on smaller scale research efforts were available, but the actual compositions for these large-scale integral experiments was uncertain. The expected chemical species were chemically reactive and radioactive.

Reliability was very important; these experiments required several years of preparation and were very costly. The experiments could not be repeated once the fuel was degraded. Due to the radioactive nature of the experiments, failure of the system resulting in the release of radioactivity was not acceptable. The same system had to be used for each test; schedule constraints did not allow for the feedback of information obtained by analysis of previous test's samples.

Canister Design

A review of commercially available aerosol sampling systems was made to evaluate the applicability of these systems based on the requirements listed above. It was concluded that no particular system was acceptable. Therefore, the following system was designed, incorporating a number of collection mechanisms applied independently in commercial systems.

The system consisted of two passive sampling canisters, with entrances from the main plenum of the test vehicle at the minimum and maximum distances from the fuel. Each canister contained three chambers; each chamber consisted of a series of stages that were configured in such a way that they created a long (3.2 m), winding channel in a space that was only 0.16 m high. The channel had a generally square cross-section. A portion of the steam/hydrogen mixture passed through one or more of the chambers in each canister, and particles were collected on collection devices located in the stages. The individual chambers were exposed to flow during different periods of the tests, allowing limited temporal separation of the collected material. Velocities in the main plenum of the test vehicle and at the entrances to the canisters were such that the sampling was isokinetic. The canisters were maintained at a constant temperature (650K), which was above the steam dew point but below the dew

point of the predominant expected species.

The primary collection devices were settling plates and fine-wires. The settling plates were positioned on the floors of the stages to collect larger particles by gravitational settling and smaller particles by diffusion. They were 0.9 sq. cm stainless steel plates. The fine-wires were suspended perpendicular to the gas stream. They collected larger particles by impaction and interception and smaller particles by diffusion. They were composed of a variety of materials, primarily platinum, and ranged in size from 2.5 to 250 μm . By collecting particles by a variety of mechanisms, the system had adequate collection efficiencies for particles across a relatively large size range. These collection devices also collected material by direct condensation and sorption, allowing the study of those species that did not travel in aerosol form.

In addition, coupons of various materials were attached to settling plates. Analyzing the reaction products of the coupon materials and the particulates and vapor species present in the gas provides information on the chemical nature of the releases. Similar information is also obtainable from examining the reaction products on the fine-wires that were composed of non-inert materials.

A schematic diagram of the canisters is shown in Figure 1. Figure 2 shows the details of a stage and Figure 3 illustrates the collection devices.

SAMPLE ANALYSIS

Analysis of the samples is being conducted posttest. The collection devices were designed to be readily removable from the stages, and were sized for examination by microanalytical techniques. The handling and examination of the samples are being conducted in radiation shielded environments with remote handling equipment due to their radioactive nature.

Qualitative Analysis

The primary examination technique is scanning electron microscopy (SEM). This technique yields information on the morphology of the deposited material in the form of micrograph images. The elemental composition of the deposits are obtained by analysis of the energy spectrum of the X-rays that are generated by the electron bombardment.

Additional microanalytical methods that are used to determine elemental composition as well as to identify chemical species include electron microprobe analysis, secondary ion mass spectrometry, X-ray diffraction and bulk analysis. The bulk analysis yields somewhat quantitative information on the masses of the different elements on individual samples. Small-scale laboratory experiments are being carried out to deduce some of the chemical species that existed during the test by generating products that are similar to those on the samples under controlled conditions.

Quantitative Aerosol Analysis

Quantitative analysis is being performed on that portion of the deposited material that appears to have traveled in aerosol form; i.e., the particulate matter. The procedure for the analysis consists of first, measuring the

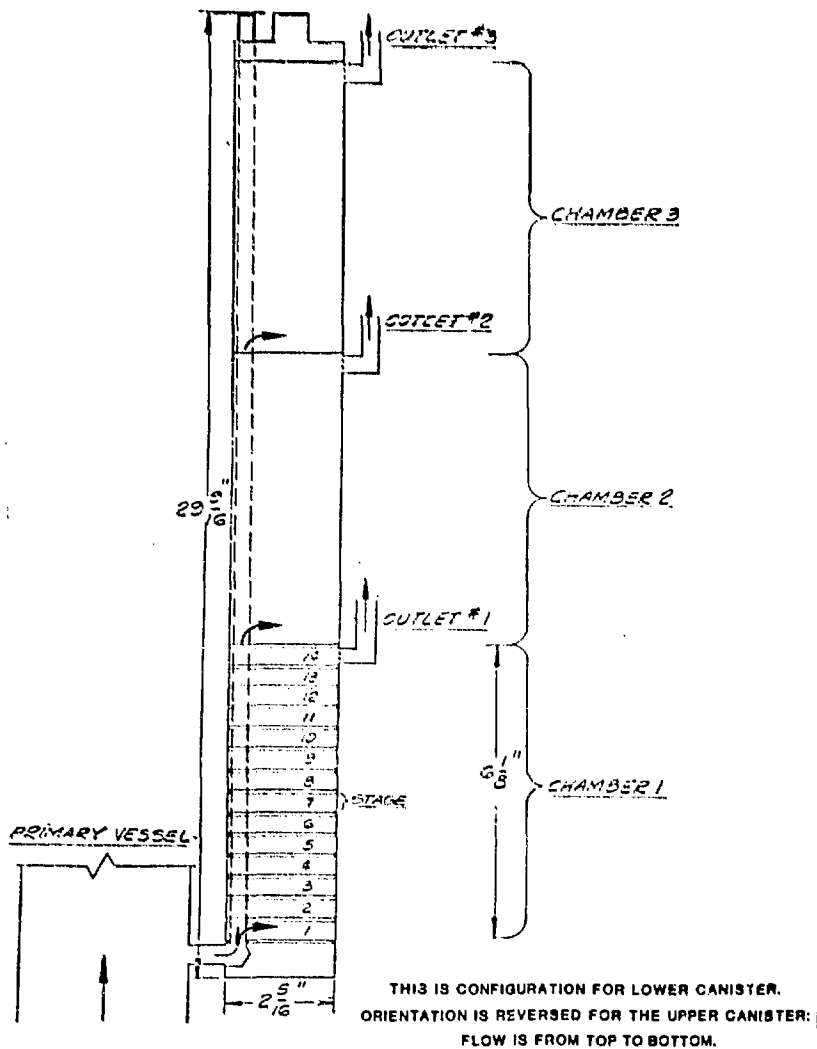


FIGURE 1. Aerosol sampling canister configuration

diameter of each particle in a selected area of a SEM micrograph using a computer-based grid. The results are sorted by size to determine the number of particles in selected size intervals. The particle counts are geometrically scaled to represent the number of particles in each size range on the surface of the settling plate or wire. Collection efficiencies of the wires and the plates for each average particle size are applied to the particle count data to determine the number of particles in each size range that passed by the collection device during the sampling period. The number of particles in each size range that entered the canister is calculated by applying collection efficiencies of attenuation along the flow path upstream of the collection device to the results of the previous computation. These values are then divided by the total gas flow to yield particle concentrations and distribution functions. Mass concentrations are calculated by applying a particle density estimated from the microanalysis. Since sampling was done isokinetically, these results also describe the aerosol in the main plenum of the test vehicle. The collection efficiencies used in these calculations were

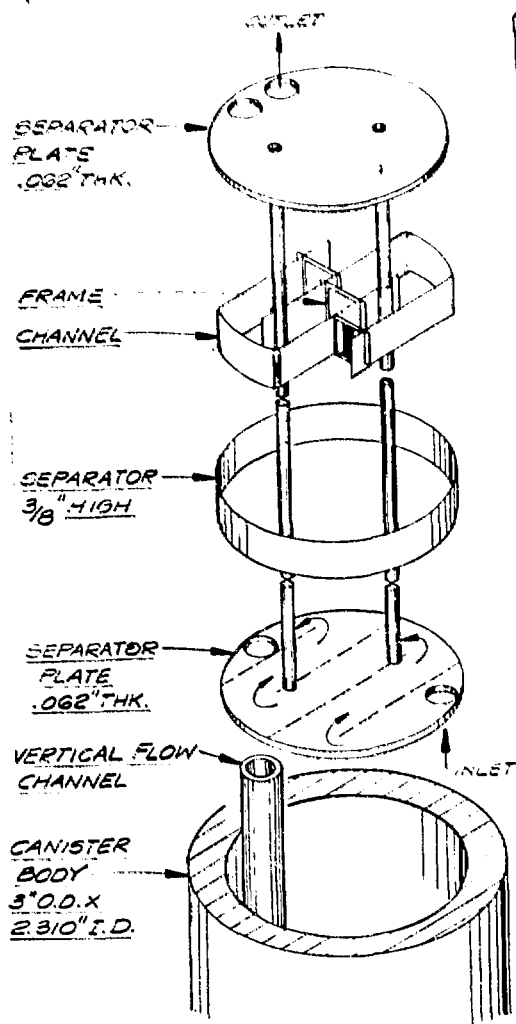


FIGURE 2. Canister stage, expanded

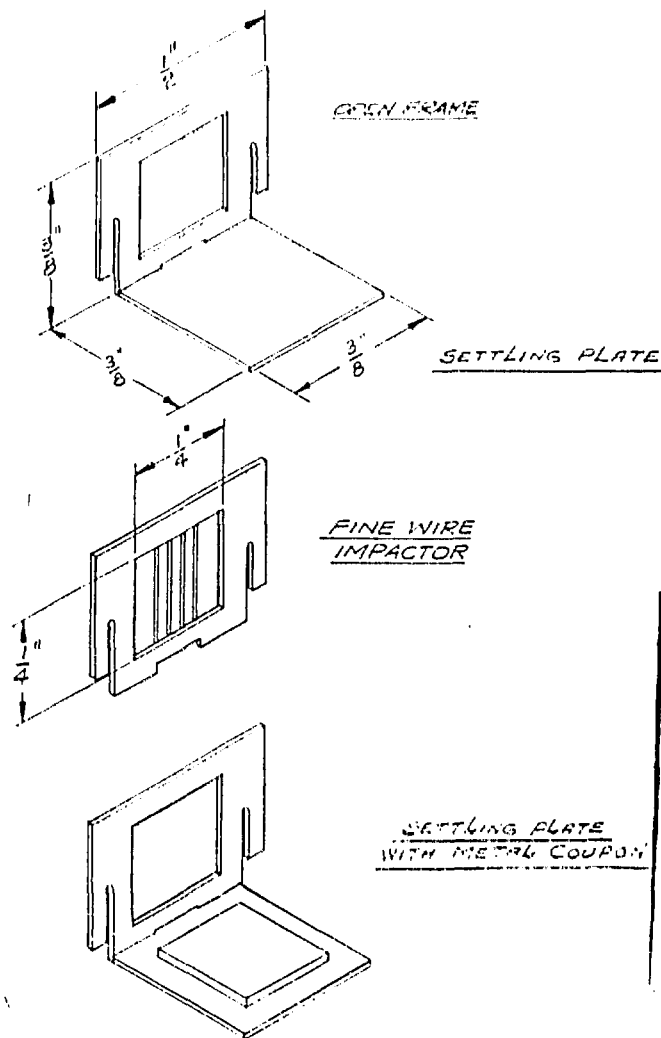


FIGURE 3. Collection devices

compiled from the literature. The selected equations were determined to be the most applicable for the range of flow conditions and system geometry.

Theoretically, the analysis of the particle data from each micrograph corresponding to collection devices in a particular chamber should yield the same distribution functions. In practice, however, there will be variations in the results due to nonuniformity of the samples, errors introduced by the particle counting, and the approximate nature of the efficiency equations. By looking at a number of micrographs from different collection devices in a chamber, sufficient data can be generated to determine reasonable average values for the distribution functions.

Examples of resultant particle and mass distribution functions that are generated by counting particles on micrographs from a chamber are shown in Figures 4 and 5.

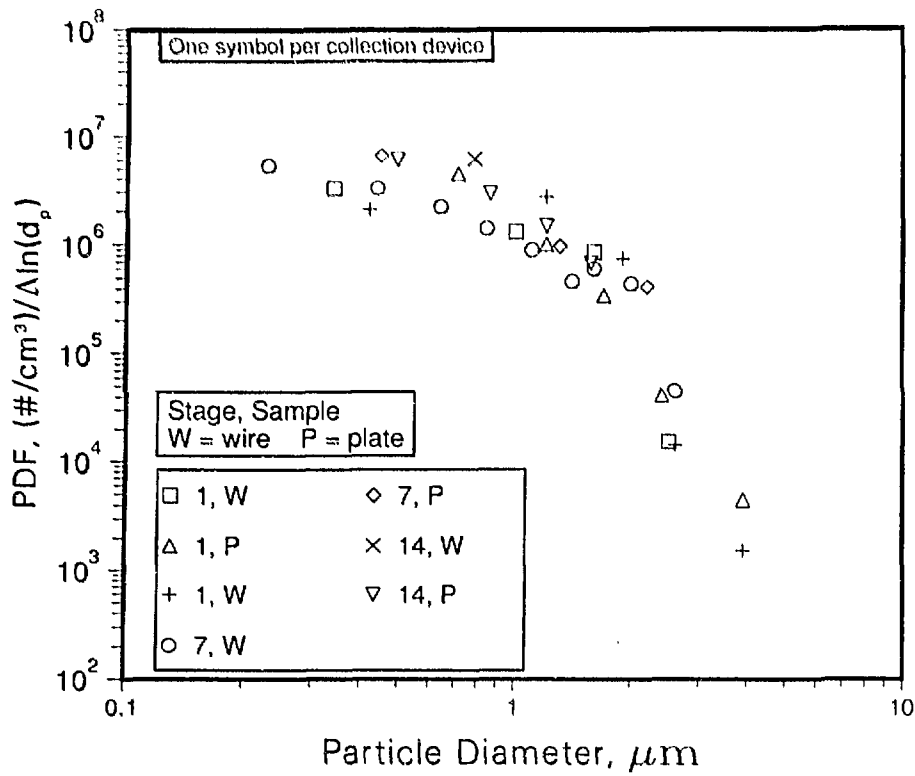


FIGURE 4. Particle distribution function, select chamber

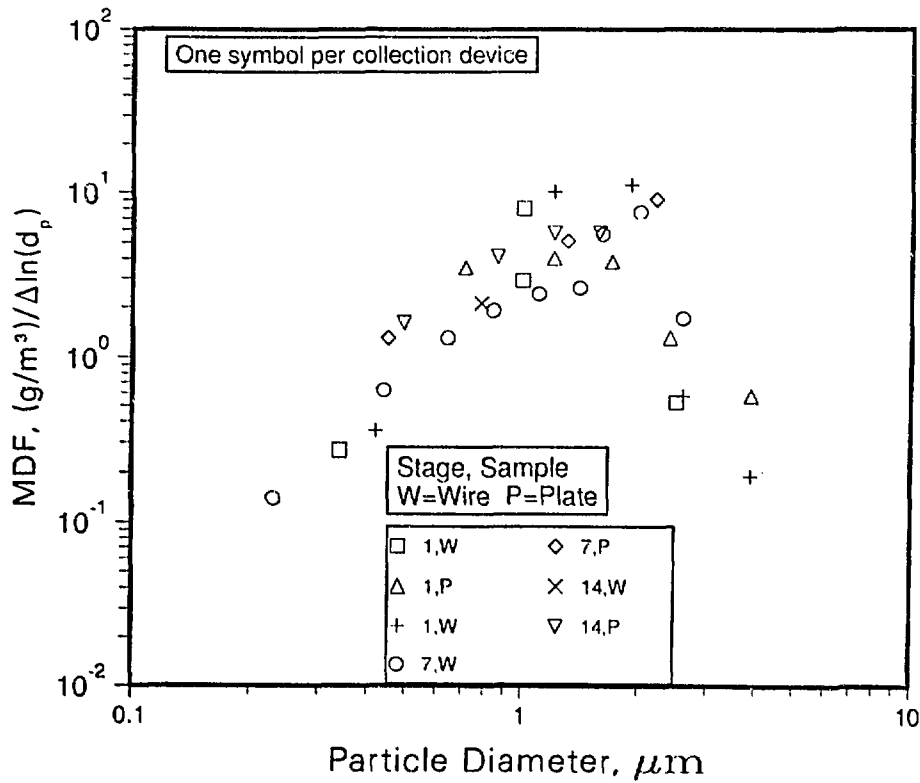


FIGURE 5. Mass distribution function, select chamber

SUMMARY

An aerosol sampling system was designed and used in a series of nuclear reactor safety experiments. The system was designed to sample radioactive and chemically reactive aerosols of unknown size distributions and concentrations in high temperature, high pressure steam/hydrogen environments. The aerosol samples are being analyzed posttest to determine their composition and morphology by microanalytical techniques. Main stream particle size distributions and loadings are being computed from particle data generated from SEM micrograph images and collection efficiencies calculated with measured thermal-hydraulic data. The system would be applicable to other types of experiments in which the sampling environment is severe and/or a priori knowledge of the general particle size range and loading are limited.

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