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FISSION PRODUCT SOURCE TERM RESEARCH AT OAK RIDGE NATIONAL LABORATORY*

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EXECUTIVE CONFERENCE

RAMIFICATIONS OF THE SOURCE TERM

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I. INTRODUCTION

Estimates of the quantities of fission products which might escape into the biosphere during a nuclear reactor accident usually proceed from considerations of the physico-chemical behavior of the fission products along the escape pathway. The escape pathway, in turn, is customarily viewed as a series of geometric compartments through which the fission products are transported in the course of their release into the environment.

In a generic sense, these compartments are: (1) the fuel in which the fission products are born; (2) the void region, or "fuel-clad gap," that is bounded by the Zircaloy cladding; (3) the core of the reactor; (4) the primary system; and (5) the containment. Although the boundaries of each of these compartments are well-defined, at least prior to the accident, their internal geometrics can be extremely complex, and moreover can change with time. Furthermore, as an accident scenario develops, even the boundaries between

compartments, and hence the fission product escape pathway, may be altered. In spite of these complexities, the approach nonetheless provides a convenient basis for bookkeeping purposes.

Of prime concern are the physico-chemical processes involving the fission products which occur in each of these compartments or "control volumes," for it is precisely these processes which determine the extent, ultimately, to which a particular fission product species can enter the biosphere as a consequence of a nuclear reactor accident. And these processes can be relatively simple, such as the migration of the fission products through the UO_2 fuel lattice due to thermal motions, or quite complex, such as are considerations of chemical interactions of some of the fission products in the primary system and within the containment structures. Thus, in each compartment, the bookkeeping needs to be extended to include the various processes involving fission product behavior which occur in that compartment.

In addition, a thorough description of the transport, thermodynamic, and chemical processes that occur in a particular control volume require a detailed temporal and spatial characterization of the thermalhydraulics. Thus, for the type of approach outlined above, the bookkeeping becomes so complex that one must resort to the use of electronic computational techniques.

The development of a program of research which is directed toward establishing the necessary technical detail is truly a formidable task; nonetheless this has been a major thrust of the research sponsored by the U.S. Nuclear Regulatory Commission (NRC). Oak Ridge National Laboratory (ORNL), along with its sister institutions, has been a participant in this research endeavor.

The purpose of this work is to describe some of the research being performed at ORNL in support of the effort to describe, as realistically as possible, fission product source terms for nuclear reactor accidents. In order to make this presentation manageable, only those studies directly concerned with fission product behavior, as opposed to thermalhydraulics, accident sequence progression, etc., will be discussed.

II. ORNL STUDIES OF FISSION PRODUCT BEHAVIOR

For purposes of this presentation it is convenient to order the research activities using the concept of control volumes described earlier. In addition, the discussion is restricted only to considerations of severe degraded-core accident conditions. Under such conditions, processes that occur within the UO_2 matrix and the fuel-clad gap become unimportant, so that the first control volume of interest is the core region itself.

A. Fission Product Release from the Core Region

Studies of fission product releases from the core region can be conveniently subdivided into two types: releases from fuel rods with failed claddings but with no liquified material present, and releases from molten pools. For most purposes, the first classification is equivalent to investigations of the release of volatile fission products from fuel rods, whereas the second is tantamount to studies of aerosol production. Both types are currently under investigation at ORNL.

1. Fission product releases from fuel rods

One of the earliest source term-related research programs sponsored by the NRC (as opposed to its predecessor agency, the Atomic Energy Commission) has been the investigations at ORNL of the releases of fission products from commercial, irradiated, fuel rod segments. These studies,[1] which were originally intended to establish the levels of conservatism in the Reactor Safety Study (WASH-1400) approach to estimates of fission product release from Design Basis Accidents, utilized an experimental apparatus similar to that which is diagrammed in Fig. 1. As currently employed, the apparatus consists of a zirconia or thoria furnace that is inductively heated through the use of a tungsten susceptor. The irradiated fuel specimen, still in its cladding, is contained within the furnace and is exposed to a flowing steam-helium stream when the test temperature is attained. Fission products which are released from the fuel rod are transported down a gold- or stainless steel-lined quartz tube along which is imposed a nearly linear temperature gradient, where selected deposition occurs. Material still in the gas phase is then passed through a series of filters and sorption beds which are designed to collect particulates and to classify highly volatile forms of iodine that may be present (I_2, CH_3I). Lastly, after moisture has been removed from the gas stream, the ^{85}Kr that is released during the experiment is collected on charcoal traps and the amount collected is determined.

Even during the initial design phase it was recognized that cesium, as well as iodine, could exist in several gas-borne chemical forms. The thermal gradient tube was therefore incorporated into the design in an effort to

achieve a separation among the different cesium forms. In that regard, the effort was unsuccessful. The device nonetheless did play a significant role in elucidating the chemical form of iodine.[2] It was also instrumental in establishing that antimony is released in elemental form,[3] and that cesium hydroxide is the dominant chemical form of gas-borne cesium in the presence of steam.[4]

Some of the early data obtained with the apparatus described in Fig. 1 were used to construct the release rate curves that are presented in Fig. 2.[5] More recent data are in essential agreement with these results.[6]

Note that the graphical representation shown in Fig. 2 indicates identical releases of krypton, cesium, and iodine, as fractions of inventory present, at temperatures in excess of about 1300°C. This observation has been confirmed by the more recent measurements[6] and is supported by determinations of cesium and krypton releases during the accident at the Three Mile Island Unit 2 Reactor.[7] It also forms the basis for the contention that only half of the iodine released from the fuel during the accident has been accounted for. It is currently believed that the discrepancy is due either to errors in chemical analysis or that the remainder precipitated in the core region as silver iodide. If the latter had in fact occurred, then radiolytic reactions played a greater role than previously thought.

Concern has been raised recently regarding the "prototypicality" of the hot cell experiments, particularly with regard to axial and radial temperature gradients. It should be noted at the outset that the hot cell experiments were intended to be performed under strictly isothermal conditions, and the

data so obtained were to be input into computational programs, in which, presumably, effects of gradients in temperature would be taken into account.

In a severe core damage accident, temperature distributions will undoubtedly vary markedly from one region of the core to another, and from one period of time to another. Moreover, the same is true for both flow and gas composition. Hence, the question of "prototypicality" becomes moot; for some accident sequence in some region of the core at some period in time, the experimental conditions unquestionably duplicate the actual thermalhydraulic conditions exactly.

2. Fission Product Releases from Molten Pools

Unlike the studies of fission product release from irradiated fuel rod segments, in which only the fission products in large measure are transported beyond the furnace region, the investigations of releases form molten pools focus strongly on the behavior of non-radioactive materials. These materials, however, because they are the dominant substances of the aerosols which form in the core region in severe core damage accidents, are nonetheless of primary importance, for it is these aerosols which act as carriers for both volatile and less volatile radiotoxic species.

It is nonetheless interesting to compare axial and radial temperature gradients along a single rod as calculated for a severe core damage accident with those estimated for the ORNL hot cell tests and in-pile tests currently being conducted in the Power Burst Facility. The comparisons are presented in Table I. Although no inferences should be drawn from this comparison because,

as already noted, the question of prototypicality has no significance, it is nonetheless both interesting and surprising to note that the hot cell tests actually appear to be more representative of expected accident in-core thermal conditions than the in-pile tests.

Partly for this reason, the ORNL studies of releases from molten pools are performed with the radioactive fission products replaced by stable isotopes. The experiments performed to data[8] have been conducted with a smaller version of the apparatus that is depicted in Fig. 3.

The experiments involve heating an assembly like that shown in Fig. 4 inductively, by coupling the RF field directly to the urania. The test assembly is mounted on an elevator within the furnace, so that melt progression is simulated by passing the assembly from top to bottom through the radiofrequency coil. Also note that the tests are conducted in the presence of a steam-hydrogen atmosphere.

Aerosols that are produced in the tests are transported to various characterization devices which are designed to determine the quantities and compositions of the materials released from the molten pools.

In these experiments as well, prototypicality with regard to thermalhydraulic conditions is a moot issue; however, the rate of heating the bundle is generally selected to be representative of conditions calculated for specific accident sequences.

Differences in release characteristics of several fission products (Te, Ba, Sn) and some of the core components (Ag, Fe) have been noted between the

experiments conducted at ORNL[9] and those performed using a somewhat different experimental approach at Karlsruhe, West Germany.[10] These differences are believed to result primarily from differences in the chemical environment, especially as it relates to the oxidation of Zircaloy. This serves to underscore the significance of chemical factors, heretofore ignored, on the magnitude of accident source terms.

For example, the core melt release experiments indicate that, if the accident conditions are such that significant quantities of molten, unoxidized Zircaloy is present, strontium and barium species will be chemically reduced by the zirconium.[11] This promotes the release of these nuclides. Tellurium, on the other hand, would be almost quantitatively retained by the molten Zircaloy.[12]

B. Transport Through the Primary Circuit

Once the fission products escape the core region, either as a vapor or as an aerosol component, they can encounter a veritable jungle of metallic surfaces. The extent to which this occurs, of course, is dependent upon the type of reactor, the location of the breach in the primary system, and the thermalhydraulic conditions. Moreover, in the course of its transport through the primary circuit, a given fission product species can be involved in a number of possible interactions involving the gas phase, the aerosol particles, and the system surfaces. Of the many possibilities, perhaps the most important are fission product-surface interactions and aerosol behavior in the primary circuit. Studies concerning the first of these are being conducted at Sandia National Laboratory,[13] whereas the latter is being investigated at ORNL.

A key objective of the ORNL program is to establish the adequacy of the TRAP-MELT computer program in describing aerosol processes which occur in the primary circuit. The code itself, however, was developed at Battelle, Columbus Laboratories.[14]

A recently-modified version of the apparatus that is being used in the ORNL program is depicted in Fig. 5. In brief, the apparatus consists of an aerosol generator, presently a plasma torch, an instrumented test section, and a large, expandable volume to completely contain all of the aerosol produced in an experiment.

The test section contains instrumentation designed to continuously monitor the thermal hydraulic conditions during an experiment, and aerosol characterization devices which permit a classification of the aerosol with regard to size and with regard to the mode of deposition.

Results of two experiments that were performed using iron oxide aerosols are presented graphically in Fig. 6.[15] The first test was conducted under flow rate condition such that the average residence time of an aerosol particle in the test section was 32 secs. In the second test, the flow rate was reduced to yield a residence time of 64 secs. As one might expect intuitively, residence time has a profound influence on deposition by settling.

In the experiments conducted to date, comparison between experiment and computation has been only marginally satisfactory when the data are compared in a detailed manner.[16] This is due in part to difficulties in specifying the experimental thermalhydraulic conditions sufficiently precisely, in part to

deficiencies in aerosol characterization input parameters, and in part because of the strong coupling among the deposition processes through the aerosol concentration. Attempts are currently underway to eliminate the problems arising from the first two conditions.

The metal-foil deposition samples which are identified in Fig. 5 will also be employed in studies of aerosol resuspension. The apparatus which will be used for this purpose is currently under design.

C. Behavior within Containment Structures

In most accident sequences, the last control volume through which the fission products must be transported prior to their escape into the biosphere is the labryrinthine structure known loosely as the containment. In boiling water reactor plants, the containment usually consists of the dry-well, the wet-well, and the reactor building; in pressurized water reactor plants, this final control volume is generally the containment building itself, although in some accidents sequences, such as the TMI sequence, auxilliary and fuel handling buildings also become part of the containment control volume.

Even in so-called "dry accidents" it is first necessary to expel virtually the entire coolant inventory from the primary system in order to experience severe core damage. Moreover, in most situations the breach in the primary system through which the coolant is lost also serves to define the escape pathway along which the fission products subsequently become transported (prior to reactor vessel melt through). Hence, the presence of water in the containment control volume is generally an important determinant in source term considerations.

Furthermore, except for catastrophic failure of the containment structure, which is now generally regarded as an incredible event,[17] only material that is already gas-borne (as a vapor or aerosol component) is susceptible to escape into the environment. Thus two issues must be addressed in establishing fission product concentrations in the containment: (1) the extent to which gas-water interactions determine the quantities and forms of vapor phase fission product components, and (2) the persistence of accident-generated aerosols under high humidity conditions. Both facets are being addressed at ORNL; a related problem, which addresses the extent to which a hydrogen burn in the containment can perturb the physical and chemical status of the air-borne material, is being investigated at Sandia National Laboratory.[18]

1. Iodine aqueous chemistry

Elemental iodine reacts with water via a two-step process that is generally written in the form[19]

$$I_2 + H_2O = H^+ + I^- + HOI \ , \qquad (1)$$

$$3HOI = 3H^+ + 2I^- + IO_3^- \ . \qquad (2)$$

The net reaction is

$$3I_2 + 3H_2O = 6H^+ + 5I^- + IO_3^- \ . \qquad (3)$$

The first reaction is observed to proceed toward equilibrium extremely rapidly under conditions of concern to reactor accident analyses;[20] the second reaction, as one might expect from the stoichiometry, proceeds considerably

more slowly.[21] Thus, the initial focus of the ORNL studies concerned the existence of HOI and its volatility. Although the ORNL work was unsuccessful in establishing the existence of HOI unambiquously, the studies did demonstrate that the compound designated as "HOI" was indeed a neutral species in solution, and, more importantly, that its volatility is insignificant.[22] (Subsequent studies[23] at the Atomic Energy of Canada, Limited Laboratory at Whiteshell have established the chemical form of the species as HOI.) Thus, the only volatile iodine component of concern arising from considerations of iodine-water interactions is elemental iodine itself.

Within the primary system under severe core damage accident conditions, the environment is chemically reducing to iodine.[24] After the core has been recovered, or within the wet-well of a BWR, radiolysis reactions can result in conditions which are chemically oxidizing to iodine. Similarly, oxygen content in the containment building of a PWR will render conditions which are oxidizing to iodine (but the extreme slowness of the process requires the use of catalysts). For these reasons studies are being conducted of the volatility of iodine from aqueous solutions.

One of the experimental assemblies for this purpose is depicted in Fig. 7.[25] The apparatus consists simply of an inverted U-shaped quartz chamber, one leg of which contains the solution under investigation. The experiment involves adding radio-iodine in an appropriate chemical form to the solution, and by alternately positioning the vapor- and liquid-containing legs of the U-tube over a scintillation detector, monitoring the manner in which the radioactive species is distributed. Effects of radiolysis are studied

either by mounting the apparatus in a ^{60}Co radiation field prior to gamma radiation counting, or by performing the distribution measurements using a modified apparatus while the sample is still within the ^{60}Co irradiator.

The results are generally displayed in terms of the "partition coefficient," which is defined as the ratio of iodine concentration in the aqueous phase to that in the gas phase. Hence, a partition coefficient greater than unity signified for example that, for equal volumes of water and air, more iodine would be dissolved in the water than would be present in the air. Typical values of the partition coefficient for iodine under conditions of interest range between about 100 and 10,000.[26]

Some results of the ORNL studies are presented graphically in Fig. 8.[27] These experiments were performed beginning with solutions adjusted to pH 6.0 with boric acid and sodium hydroxide, and containing the concentrations of iodine and iodide ion indicated. The change in partition coefficient with time is due to the second stage of the two-step reaction described earlier.

The effects of the radiolysis of water on the oxidation of iodide ion in solution are clearly evident in Fig. 8. Also evident is the extent to which silver, if present in the system, negates radiolysis effects. This may in fact be the reason for the iodine inventory that is currently unaccounted for at TMI; it simply precipitated as silver iodide when the core was recovered.

The partition coefficient, unfortunately, is dependent upon a number of factors in addition to radiolytic effects. These include temperature, iodine concentration, pH, and redox potential. The current studies are attempting to

elucidate these dependences. The work has also been extended to include considerations of organic iodide formation in reactor accidents.

2. Aerosol behavior in high humidity environments

It is now recognized that, for most severe accident sequences, aerosol particulates are the primary form of radiotoxic species which have the potential for release into the biosphere.[28] Hence, the persistence of these air-borne entities from the time of release into the containment control volume to the time of failure of the containment is an important determinant in accident consequence analyses.

Studies of this nature are being performed at ORNL in the Nuclear Safety Pilot Plant. The facility consists of a M^3 vessel which is equipped, as shown schematically in Fig. 9, with an aerosol generator, various aerosol sampling devices, and instrumentation designed to monitor the physical characteristics of the vessel.[29]

The effects of moisture on U_3O_8 aerosols are shown pictorially in Fig. 10; under conditions of high humidity, the web-like agglomarates are spheroidized. One thus expects, intuitively, that such aerosols become less persistent in moist atmospheres, and this has in fact been observed.[30]

The shapes of aerosols formed by the vaporization of concrete powder are similarly modified by the presence of water vapor, as seen by examination of the photographs presented in Fig. 11. Surprisingly, however, this has little discernable effect on the persistence of these aerosols; this is evident from the data displayed graphically in Fig. 12, in which the fraction of aerosol

generated in the NSPP vessel is plotted as a function of time for three different experiments. This unexpected behavior, which continues to be investigated, can markedly affect the theoretical treatments thus far developed to describe the behavior of mixed aerosols.

D. Integral Studies — Severe Accident Sequence Analyses (SASA)

Results derived from the (primarily experimental) investigations presented above are immediately incorporated, where applicable, into the ORNL SASA Program. The purpose of this program is to perform realistic accident sequence analyses for a series of accidents beyond the design basis for selected Boiling Water Reactor Plants as they are actually configured.

Unlike probabalistic risk assessment approaches, which consider all possible accident sequences and which employ fault-tree and event-tree methodologies to rank sequences in terms of risk dominance, the SASA effort involves in-depth analyses, usually of accident sequences previously identified by PRAs as risk dominant, as they might occur at specific plants.

The studies performed to date relate solely to the Browns Ferry Unit One plant, which is a BWR-MKI plant that is owned and operated by the Tennessee Valley Authority. Thus for studies of station blackout,[31] scram discharge volume break,[32] loss of decay heat removal,[33] and loss of injection sequences[34] have been completed, and investigations of an anticipated transient without scram (ATWS) sequence is in progress.

For this type of study to be conducted effectively, it is imperative that there be unqualified cooperation with the utility that operates the facility.

We have been most fortunate to enjoy this type of interaction with the TVA staff, and this has in large measure been responsible both for the excellent progress and for the high quality of the work performed on ORNL's SASA program. Our results, in turn, have been made immediately available to TVA and have, in fact, been incorporated in their own PRA analyses.

III. CONCLUDING REMARKS

The Reactor Safety Study[35] is a landmark in nuclear reactor accident consequence analysis. It was the first serious attempt to address the question of accident risk in a systematic ways and with attention to realism. Even the question of fission product chemical form was addressed, although the results were unfortunately largely ignored, particularly for iodine, for reasons of conservatism.[36] Nonetheless, the participant in the Reactor Safety Study did much to advance the state of the technology; it is indeed to their credit that the results of WASH-1400 are still used as the basis for comparison.

Since the Study had been completed, considerable research has been performed in this country and elsewhere to further develop some of the methodologies which originated with the Reactor Safety Study, and to improve upon an admittedly deficient data base. Not surprisingly, the newer studies identified new phenomena, previously undetected, and developed new, more efficient approaches toward risk analyses.

Currently, there is a prevailing opinion that, while much new information has been obtained concerning the consequences, and indeed the risks, associated with nuclear reactor accidents, little change in the regulatory

process has in fact been effected. This opinion, which is shared by many who expect wholesale changes in the regulatory process because of a general reduction in fission product source term values is, like this expectation, ill-founded.

Results of current research have in fact already influenced regulatory decisions. For example, the Nuclear Regulatory Commission has failed to endorse the distribution of potassium iodide tablets to residents in the immediate vicinity of nuclear power plant,[37] and I am unaware of any probabilistic risk assessment (PRA) which employs Regulatory Guides 1.3[38] or 1.4[39] for the iodine source term.

Certainly more can be done. I cite as examples the chemical makeup of containment spray systems, the design and operation of off-gas cleanup systems, containment leak rate requirements and, of course, emergency preparedness criteria, all of which have been developed in some measure from outmoded considerations of fission product transport and behavior.

But the regulatory process, like the technology upon the decisions are based, is an evolutionary one. Changes will occur, changes are in fact occurring now, but they will be deliberate.

ACKNOWLEDGMENT

This work is a report of the activities of a number of dedicated, extremely capable staff at Oak Ridge National Laboratory who remain at the forefront of the source term issue. Their assistance in the preparation of this document is gratefully acknowledged.

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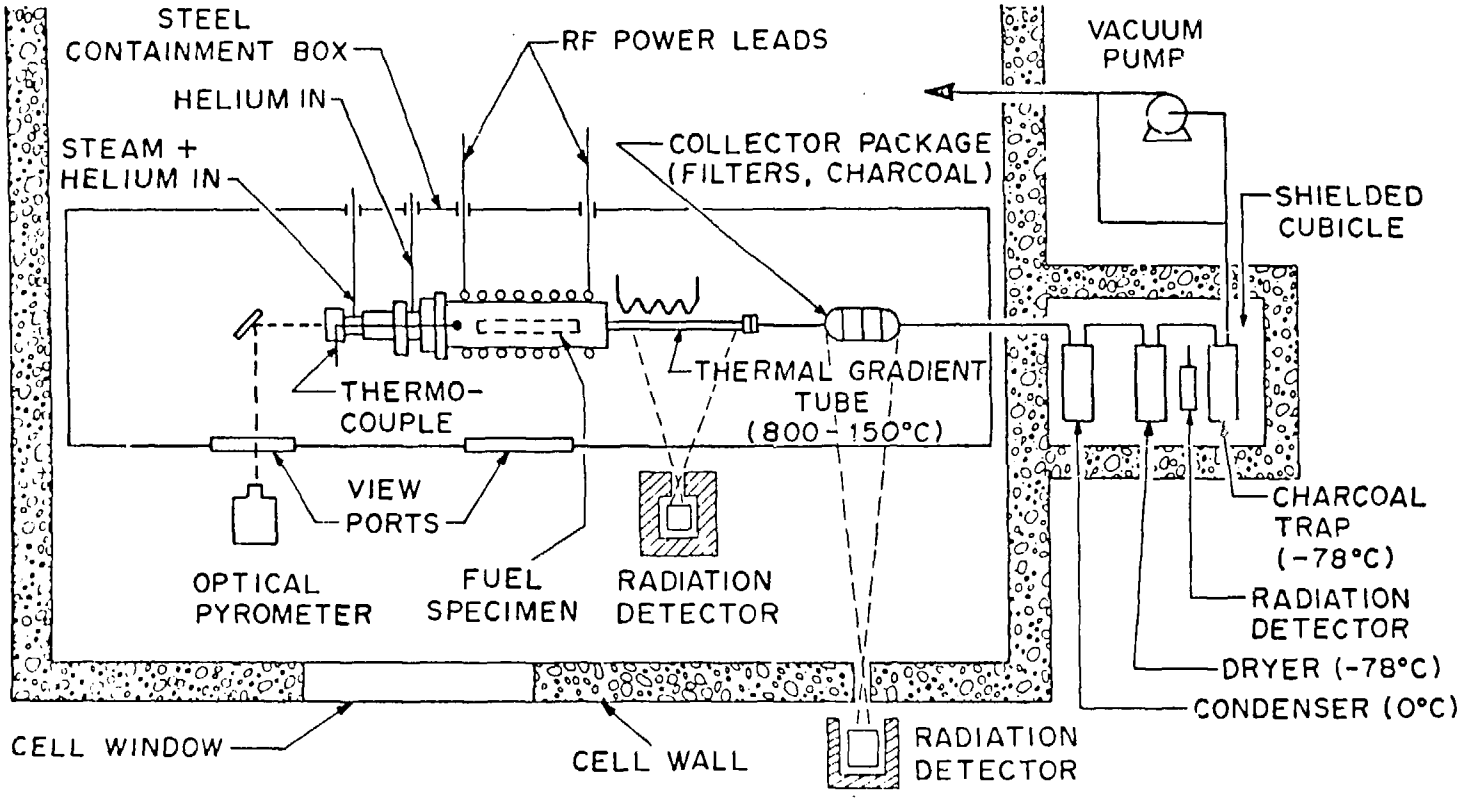
39. Reg. Guide 1.4

TABLE I. Comparison of Conditions for Fission Product Release Tests

	Reactor Accident	Hot Cell (ORNL)	In-Pile (PBF)
Radial ΔT (K)	7	0	70-200
Axial Gradient (K/cm)	0-10	4	25

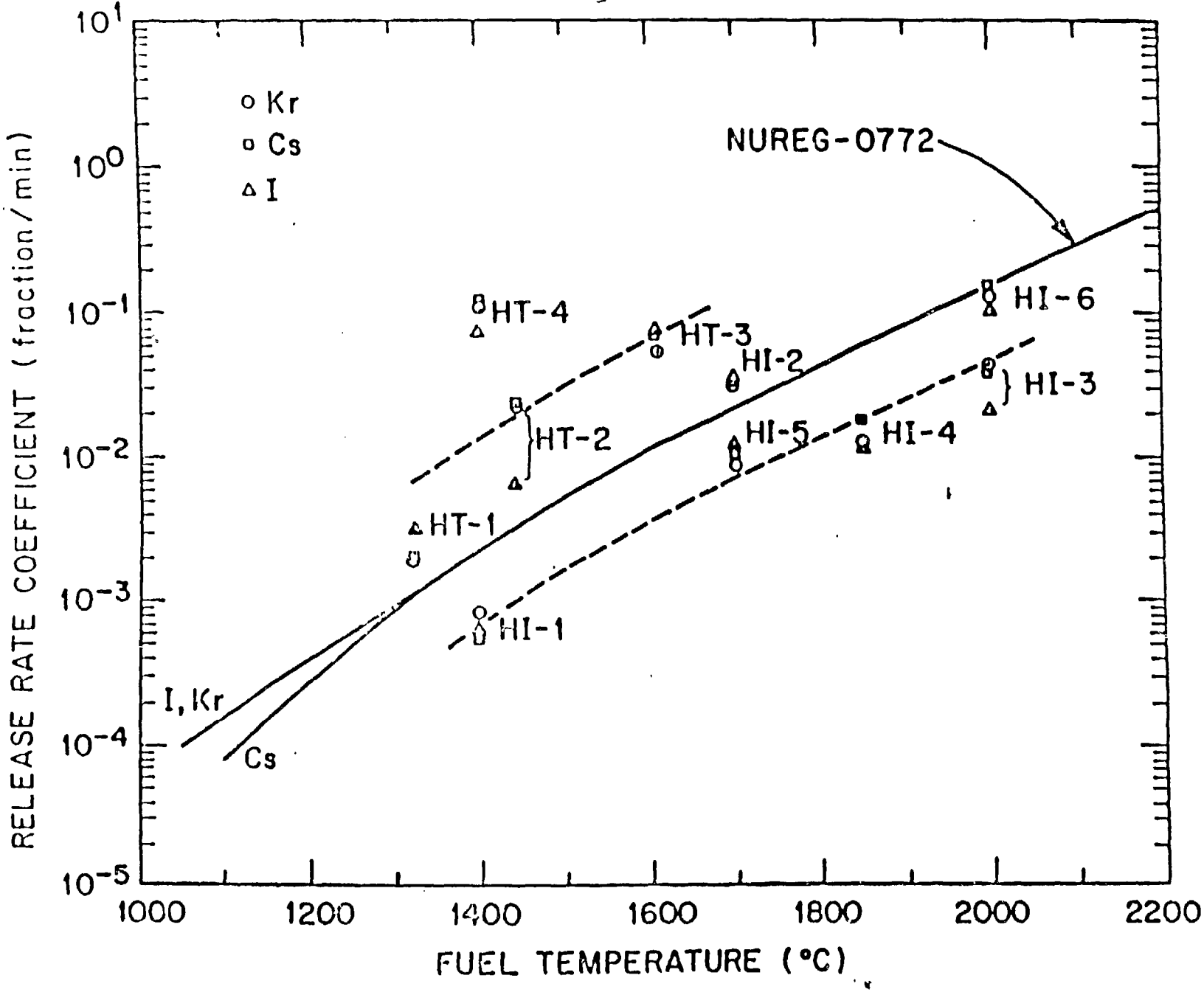
ORNL DWG 82-279R



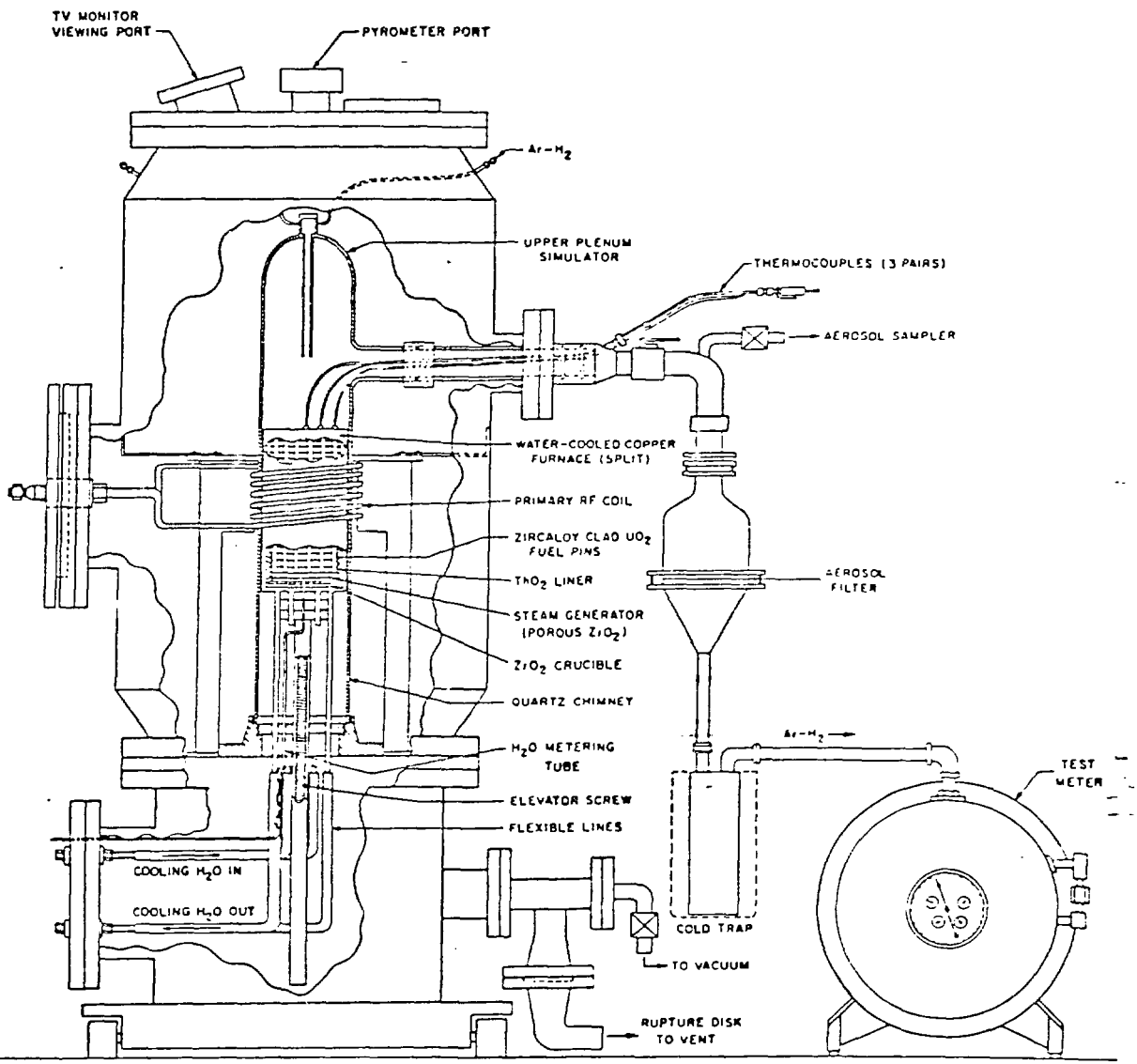
FISSION PRODUCT RELEASE AND COLLECTION SYSTEM

Fig. 1 Fig. 1

FIG. 2 (Only Ve NUREG curve)



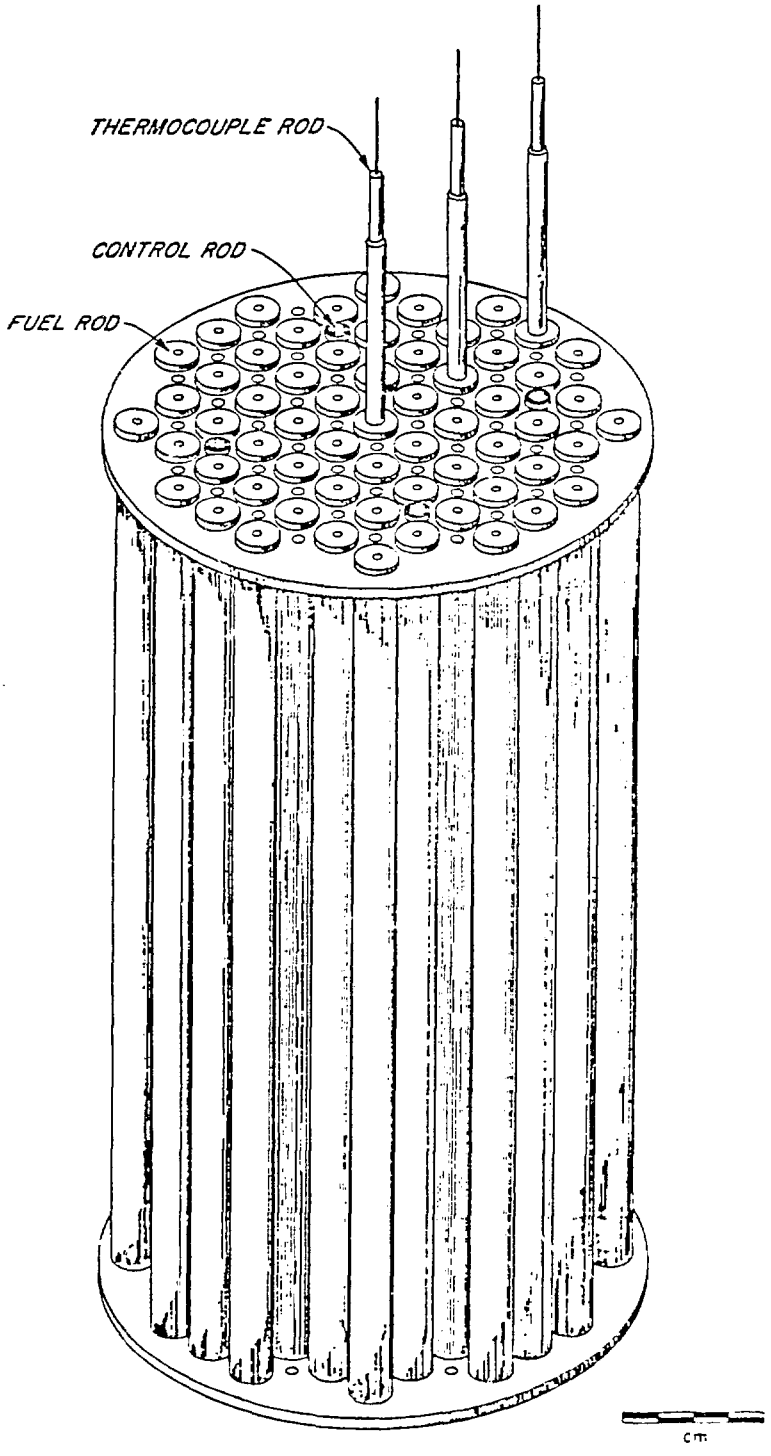
ORNL DWG 64-600R



TEN kg CORE MELT FURNACE ASSEMBLY

~~Fig. 1.~~

ORNL DWG 84-709

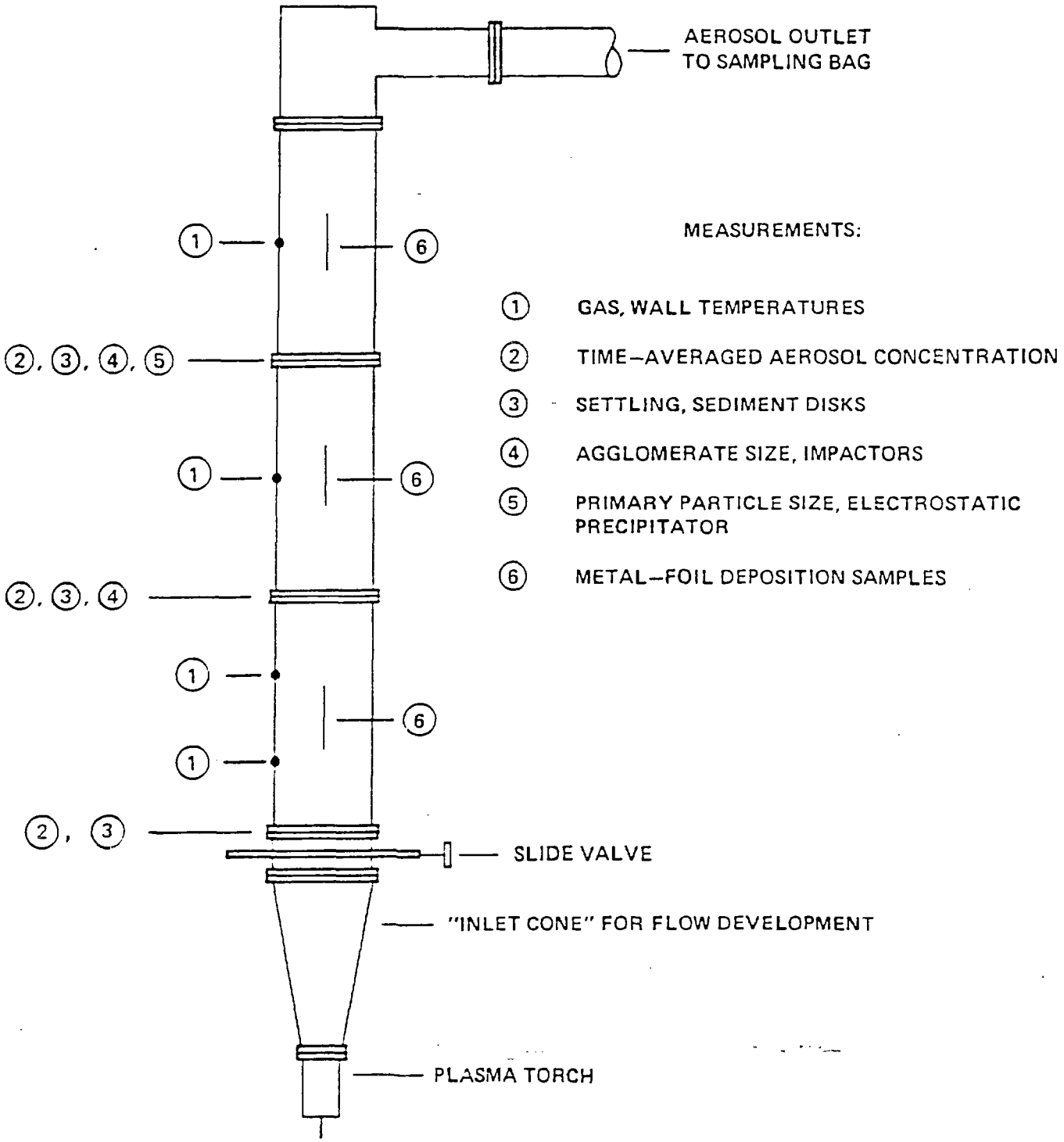


TEN kg CORE MELT FUEL BUNDLE

~~Fig. 3.~~

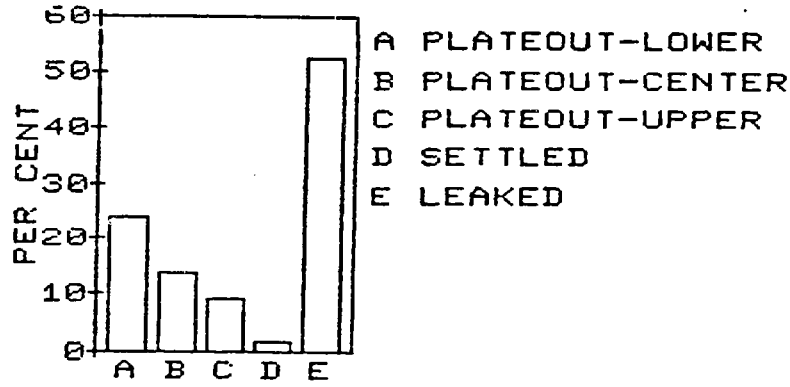
MODIFIED AEROSOL TRANSPORT TEST SECTION

ORNL DWG 84–113



VARIED RESIDENCE TIMES PRODUCED DIFFERENT RESULTS IN TESTS A103, A104

- A103: IRON-OXIDE, 32-SEC RESIDENCE TIME, 93 GRAMS OF AEROSOL



- A104: IRON-OXIDE, 64-SEC RESIDENCE TIME, 189 GRAMS OF AEROSOL

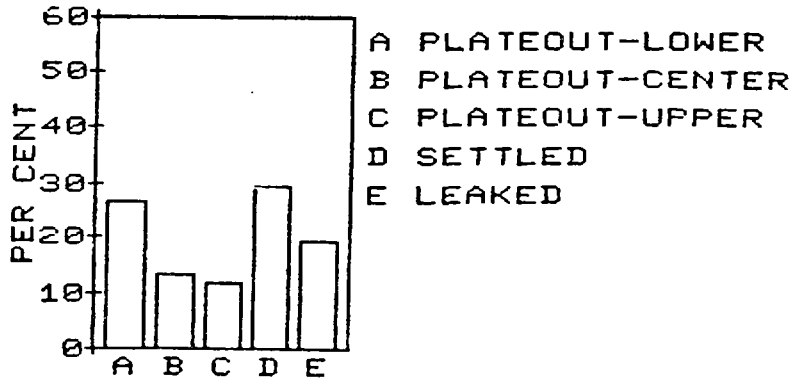
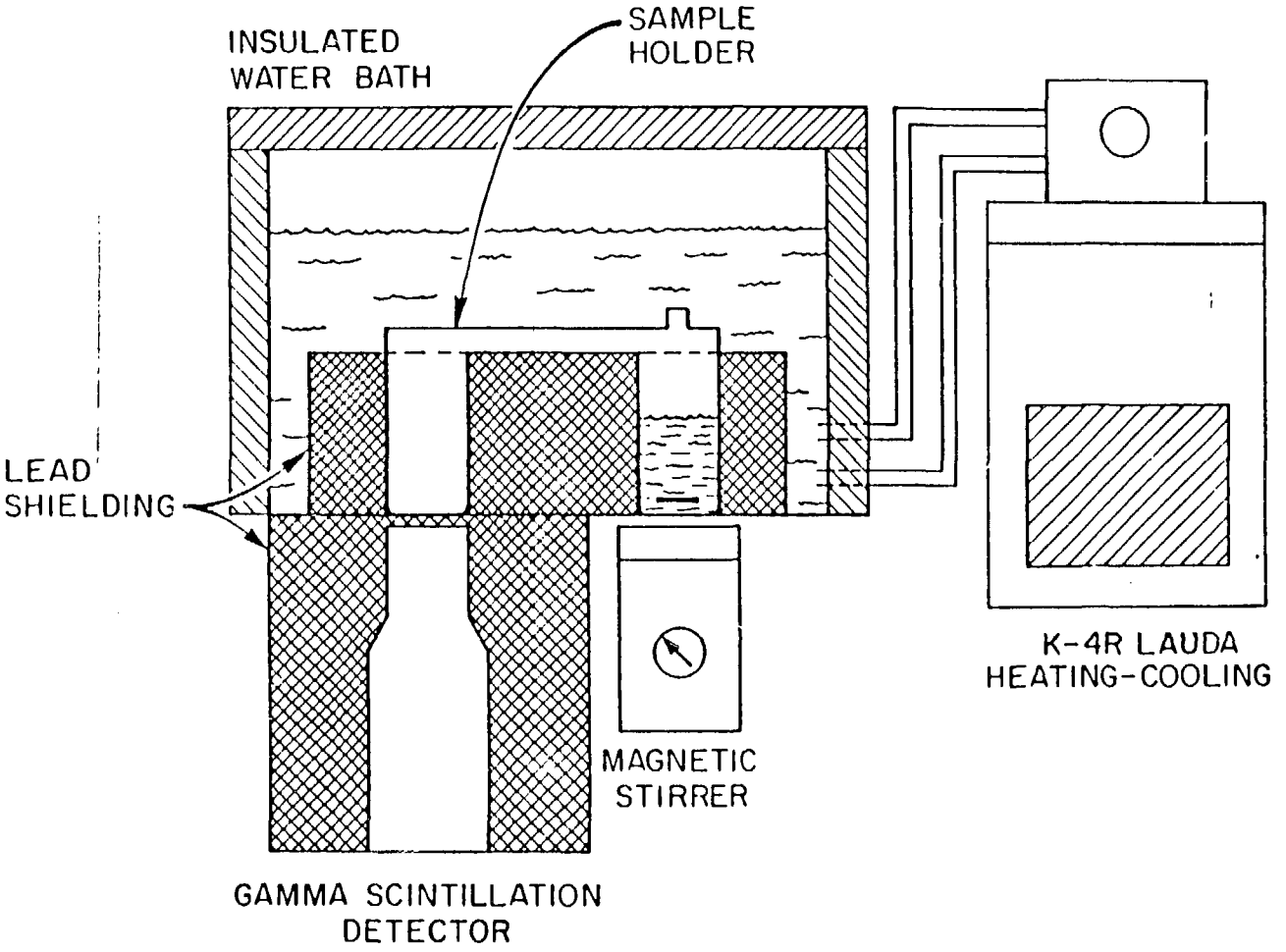
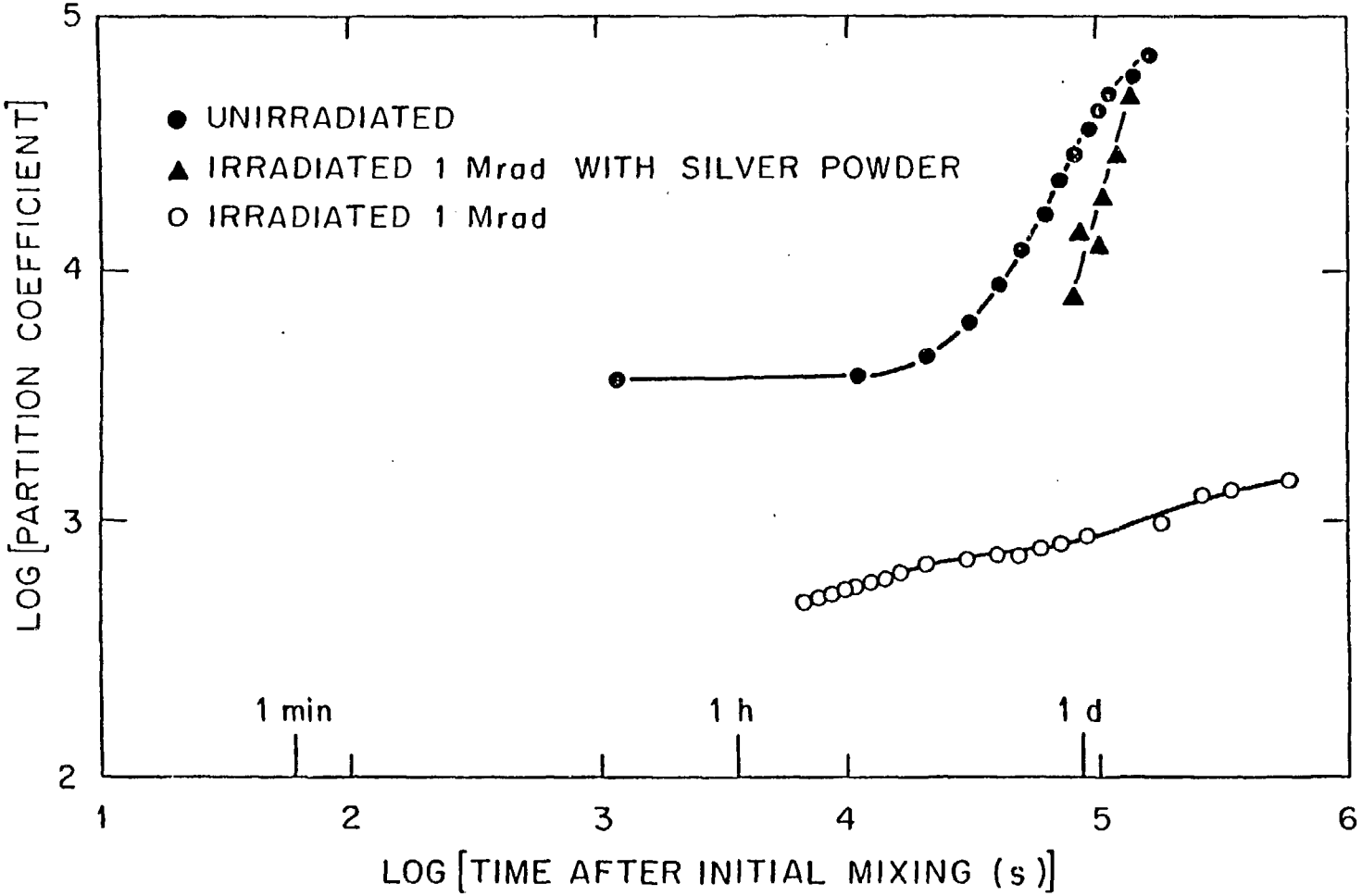


FIG. 6


INSULATED
WATER BATH
SAMPLE
HOLDER
LEAD
SHIELDING
K-4R LAUDA
HEATING-COOLING
MAGNETIC
STIRRER
GAMMA SCINTILLATION
DETECTOR



IODINE PARTITION COEFFICIENTS VS TIME
1×10^{-4} M I^-, 1×10^{-6} M I_2, pH 6.0, 323 K

Fig. 8

ORNL-DWG 84-5901 ETD

NSPP FACILITY SCHEMATIC

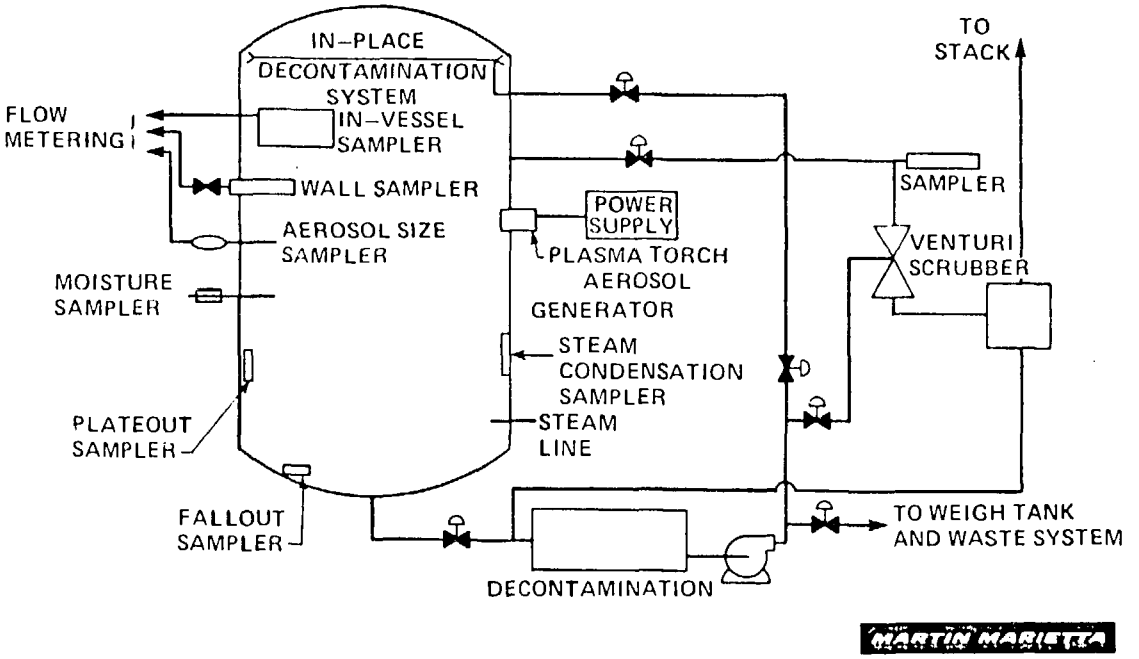
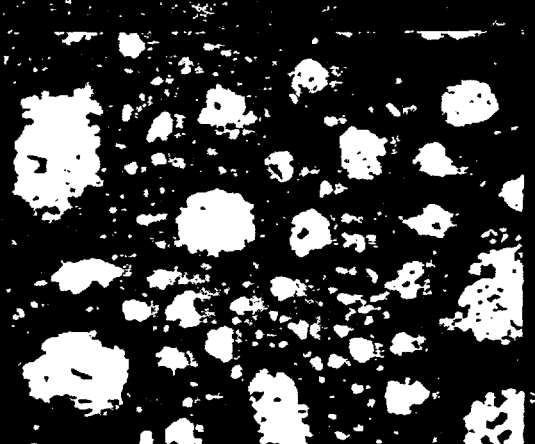


FIG. 9

MOISTURE/STEAM INFLUENCES PHYSICAL SHAPE OF U_3O_8 AEROSOL AGGLOMERATES

TEST 207 – DRY ATMOSPHERE
(RH ~ 20%)

TEST 208 – MOIST ATMOSPHERE
(RH ~ 95%)

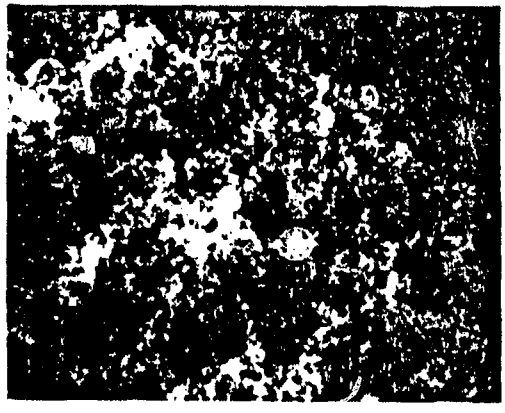


TEST 404 – STEAM–AIR ATMOSPHERE
(RH ~ 100%)

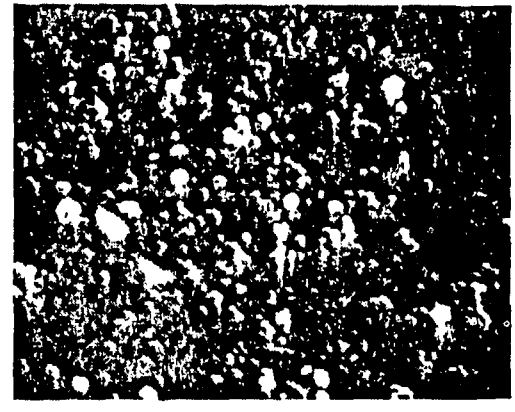
MARTIN MARIETTA

Fig. 10

STEAM INFLUENCES PHYSICAL SHAPE OF CONCRETE AEROSOL AGGLOMERATES



TEST 531 – DRY ATMOSPHERE
(RH ~ 20%)
6300X MAG.



TEST 052 – STEAM–AIR ATMOSPHERE
(RH ~ 100%)
9000X MAG.

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FIG. 11

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INFLUENCE OF STEAM ON BEHAVIOR OF CONCRETE AEROSOLS IN NSPP VESSEL

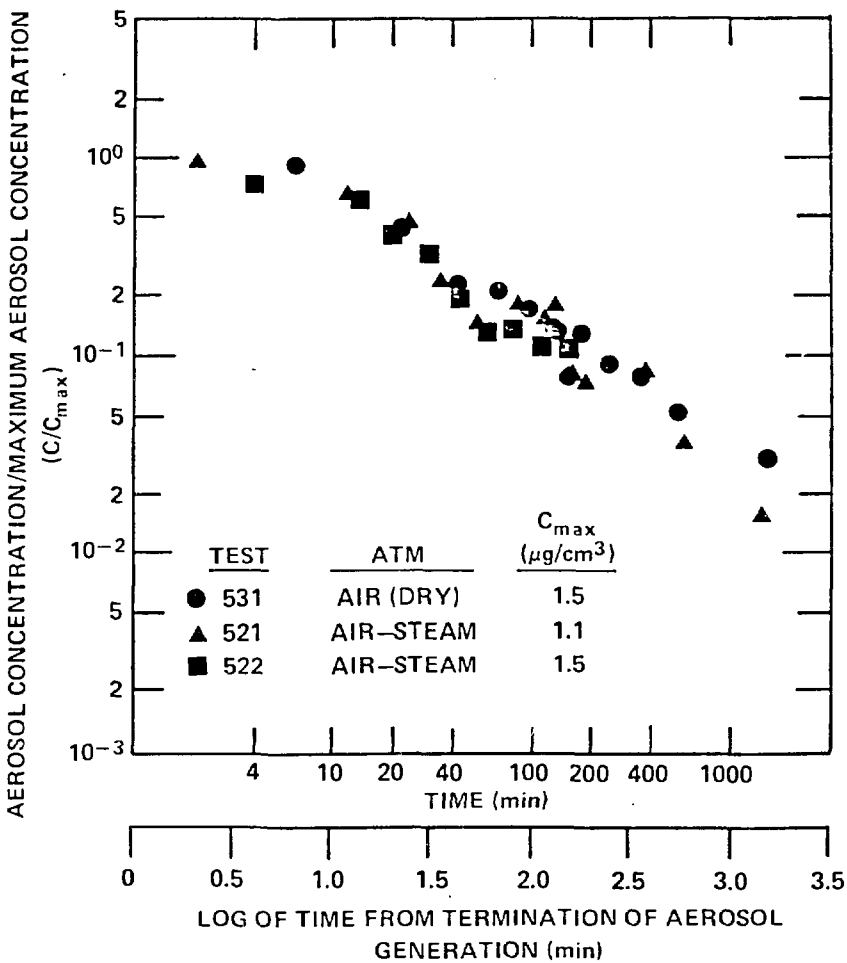

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FIG. 12