

## FISSION PRODUCT TRANSPORT ANALYSIS

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Battelle-Columbus

### Summary

In hypothetical meltdown accidents of light water reactors (LWR's), it is expected that radionuclides released from the melt will undergo chemical and physical changes and will deposit on various surfaces as they are transported through the primary system to the containment. It is of considerable interest to know what fraction and what form of these released nuclides actually reaches the containment and is available for leakage to the environment.

The escape of radionuclides to the environment under meltdown conditions requires release from successive barriers with transport along flow and leak paths. A radionuclide species can transport in the vapor phase, the condensed (particulate) phases or combinations of these, and transformations among the phases are possible. The purpose of this study has been to provide a methodology for analyzing transport and deposition along flow and leak paths for fission products released from fuel materials under a range of accident conditions in LWR's with emphasis on meltdown situations.

The major effort in developing an analytical methodology has been the formulation of a computer code, TRAP-MELT. This code considers an arbitrary number of control volumes connected by fluid flow in an arbitrary manner. Within each control volume a radionuclide species can be associated with at least two carriers or locations either in particle (liquid or solid) or vapor (molecular) form. Combining the phase of the fission product species with the concept of carrier, one can describe four states in which the species may reside: steam-molecular, steam-particle, walls-molecular, walls-particle. This list of states is not necessarily exhaustive (for instance, for two-phase flow, the carrier water must be considered) and the logic of the code has been chosen to readily accept an arbitrary number of states.

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Radionuclide transport can occur among the four states of an individual control volume or between certain states of different control volumes if these are connected by fluid flow. The former types of transport are generally limited by molecular effects and are modeled and correlated in the code itself. Transport of fission products between control volumes is assumed to occur in phase with fluid transport. This transport is imposed on the code by time-dependent thermal-hydraulic data read into the code.

It is assumed that the flow system under consideration can be subdivided into a sufficient number of control volumes such that the radionuclide population in each is expected to be homogeneously distributed (well mixed). Further, the transport rates among states are assumed to be proportional to the amount of radionuclide in the state from which the transport occurs. Rate coefficients for transport among states are determined from correlations for vapor and particulate deposition velocities. Phase transitions of a given species are modeled mechanistically using typical mass transfer correlations. Particle growth by agglomeration is also included.

General calculations with the TRAP-MELT code and evaluations of available input data suggest that an uncertainty analysis currently underway will be of considerable importance in critically assessing radionuclide retention in primary systems. Further, uncertainties in the code are expected to be reduced considerably after results from experimental vapor and particulate deposition studies currently being performed are incorporated into the code.

Initial baseline calculations of retention within primary systems for TMLB', AB, and TC meltdown accident sequences indicate little retention of iodine, with retention of CsOH and particulate species varying considerably with accident conditions. A significant result noted in these calculations is that condensation of materials of intermediate vapor pressure onto existing aerosol particles may lead to considerable growth thereby altering the character of the transported materials during passage through the primary system.

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FISSION PRODUCT TRANSPORT ANALYSIS

NOTES FOR PRESENTATION

SEVENTH WATER REACTOR SAFETY  
RESEARCH INFORMATION MEETING  
U.S. NUCLEAR REGULATORY COMMISSION  
WASHINGTON, D.C.  
NOVEMBER 5 - 9, 1979

 **Battelle**  
Columbus Laboratories

FISSION PRODUCT TRANSPORT ANALYSIS

PURPOSE: TO PROVIDE METHODOLOGY FOR ANALYZING TRANSPORT AND DEPOSITION ALONG FLOW AND LEAK PATHS FOR FISSION PRODUCTS RELEASED FROM FUEL MATERIALS UNDER A RANGE OF ACCIDENT CONDITIONS IN LWR'S WITH EMPHASIS ON MELTDOWN SITUATIONS.

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SIGNIFICANT RESULTS IN FY1979

- 1) COMPLETED REVISIONS TO TRAP CODE THAT ALLOW IT TO BE USED FOR ANALYSIS OF MELTDOWN ACCIDENTS.
- 2) PREPARED PROCEDURES AND INITIATED UNCERTAINTY ANALYSIS FOR TRAP-MELT CODE.
- 3) COMPLETED CHECKOUT OF AEROSOL DEPOSITION EQUIPMENT AND COMPLETED EXPERIMENTS.
- 4) COMPLETED PLANNING AND EQUIPMENT DESIGN FOR VAPOR DEPOSITION EXPERIMENTS.

TRAP-MELT CODE DEVELOPMENT

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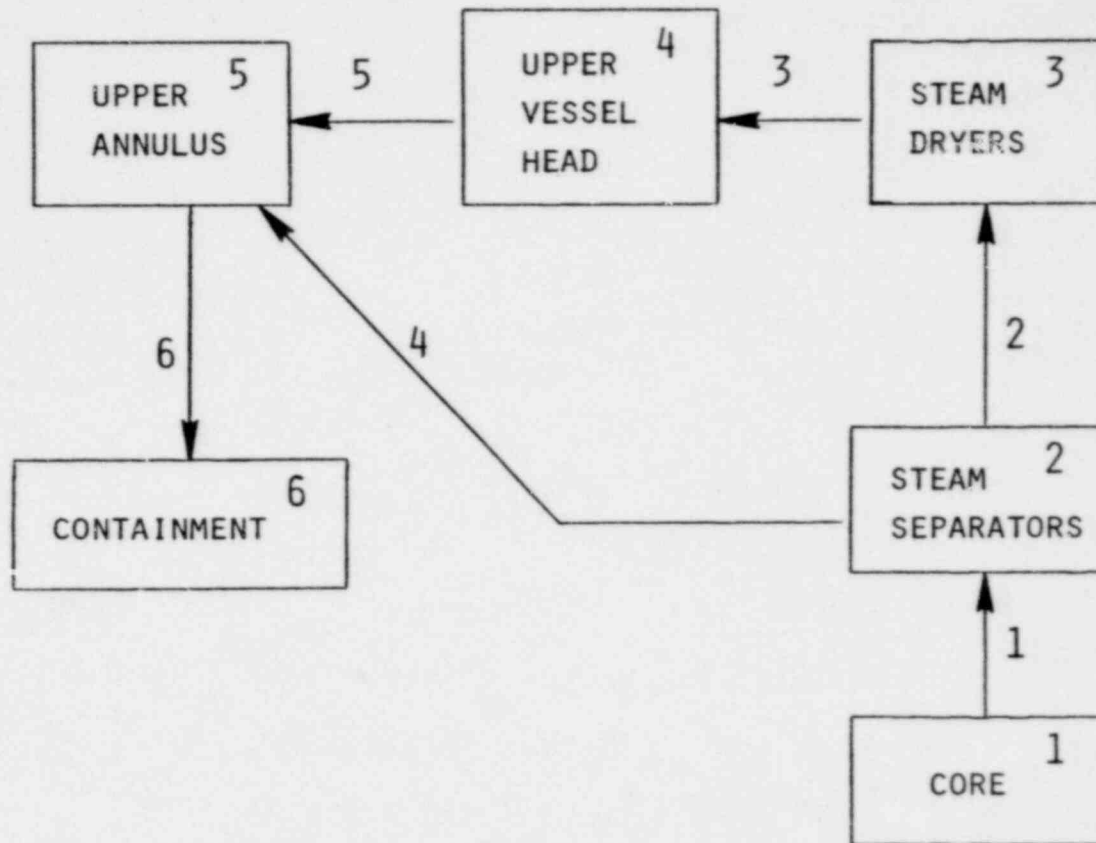
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OBJECTIVE

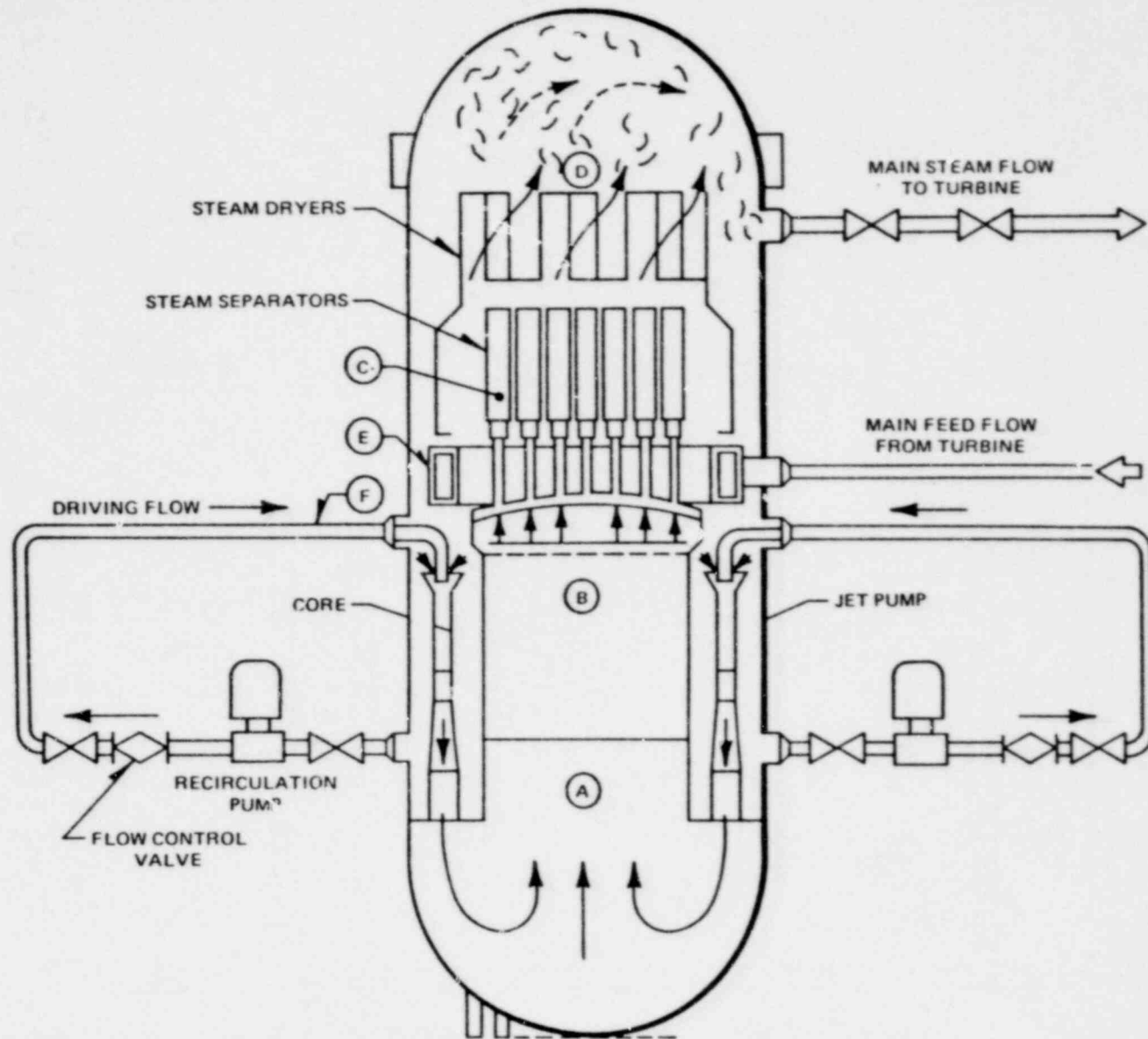
- CREATE A VEHICLE FOR ANALYZING  
RADIONUCLIDE TRANSPORT AND  
DEPOSITION IN LWR PRIMARY SYSTEMS
- IDENTIFY AREAS OF DATA AND MODEL  
UNCERTAINTY IN NEED OF FURTHER  
RESEARCH



CONTROL VOLUMES FOR TC ACCIDENT (BWR)

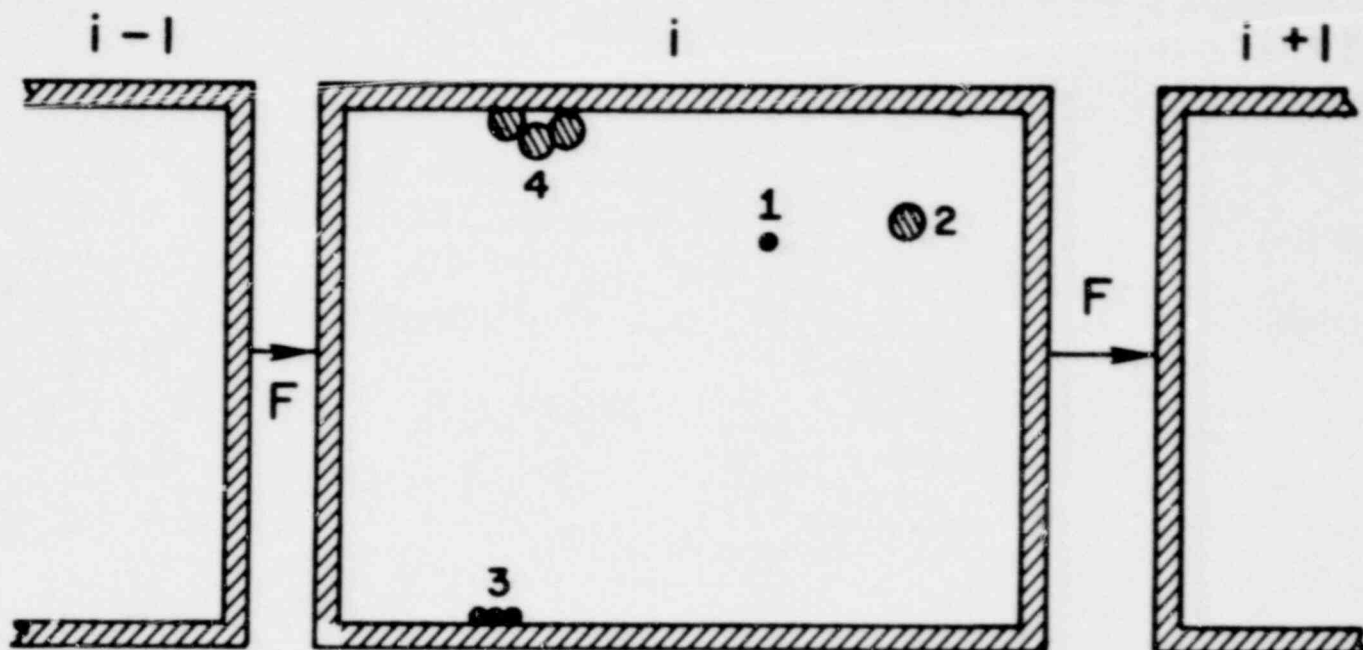


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SCHEMATIC OF A TYPICAL BWR PRIMARY SYSTEM

# FISSION PRODUCT STATES AND TRANSPORT



$m, n =$  Nuclide States

$i, j =$  Control Volumes

$k =$  Species

${}^n \beta_{im}^k =$  Deposition Rate Coefficients

${}^j F_{im} =$  Flow Coefficients

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## MASS TRANSPORT EQUATIONS

$$\begin{aligned} \frac{dM_{im}^k}{dt} = & S_{im}^k + \sum_{n \neq m} n_{\beta im}^k M_{in}^k \\ & - \sum_{n \neq m} m_{\beta im}^k M_{im}^k \\ & + \sum_{j \neq i} j_{F jm}^k M_{jm}^k \\ & - \sum_{j \neq i} i_{F im}^k M_{im}^k . \end{aligned}$$

where

$M_{im}^k$  = mass of radionuclide species  $k$  in volume  $i$   
and state  $m$

$S_{im}^k$  = source rate of species  $k$  in volume  $i$  and state  $m$

$n_{\beta im}^k$  = transfer coefficient for transport of species  $k$  in  
volume  $i$  from state  $m$  to state  $n$

$j_{F im}^k$  = transfer coefficient for transport of fission product  
in state  $m$  from volume  $i$  to volume  $j$ .

## II. MODELS IN TRAP-MELT

- DEPOSITION VELOCITIES ( $\beta$ 'S OR A MATRIX)
- PHASE TRANSITIONS OF NUCLIDES
- PARTICLE AGGLOMERATION

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## DEPOSITION VELOCITIES

- I<sub>2</sub> AND HI VAPOR SORPTION ON S.S. WALLS FROM STEAM FLOW
- PARTICLE (> 1 μM) DEPOSITION FROM TURBULENT FLOW
- PARTICLE (< 1 μM) DEPOSITION FROM TURBULENT FLOW
- PARTICLE DEPOSITION FROM LAMINAR FLOW
- THERMOPHORETIC DEPOSITION

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## PHASE TRANSITIONS OF NUCLIDES

$$\frac{dC_s}{dt} = -\frac{A_w k_w}{V} (C_s - C_w^s) - \frac{A_p k_p}{V} (C_s - C_p^s)$$

$$\frac{dM_w}{dt} = A_w k_w (C_s - C_w^s)$$

$$\frac{dM_p}{dt} = A_p k_p (C_s - C_p^s)$$

$$M_w = M_{w0} + A_w k_w \left( \frac{\beta}{\alpha} - C_w^s \right) \Delta t - A_w k_w \left( \frac{\beta}{\alpha} - C_{s0} \right) \cdot \frac{1}{\alpha} (1 - e^{-\alpha \Delta t})$$

$$M_p = M_{p0} + A_p k_p \left( \frac{\beta}{\alpha} - C_p^s \right) \Delta t - A_p k_p \left( \frac{\beta}{\alpha} - C_{s0} \right) \cdot \frac{1}{\alpha} (1 - e^{-\alpha \Delta t})$$

$$C_s = \frac{\beta}{\alpha} - \left( \frac{\beta}{\alpha} - C_{s0} \right) e^{-\alpha \Delta t}$$

where

$$\alpha = \frac{1}{V} (A_w k_w + A_p k_p)$$

$$\frac{\beta}{\alpha} = \frac{A_w k_w C_w^s + A_p k_p C_p^s}{A_w k_w + A_p k_p}$$

III. OUTPUT

FOR ANY DESIRED TIME:

FLUID DATA AND DEPOSITION VELOCITIES (EACH VOLUME)

- STEAM VELOCITY
- STEAM REYNOLDS NUMBER
- TURBULENT DEPOSITION VELOCITIES
- LAMINAR DEPOSITION VELOCITY
- THERMAL DEPOSITION VELOCITY
- DEPOSITION VELOCITY FOR  $I_2$

OUTPUT (CONT'D)

CUMULATIVE MASS (EACH SPECIES, EACH STATE, EACH VOLUME)

- FROM THIS, IN PARTICULAR, THE TOTAL MASS  
OF RADIONUCLIDE EMITTED FROM, AND RETAINED  
IN, THE PRIMARY SYSTEM

PARTICLE NUMBER CONCENTRATION (EACH VOLUME)

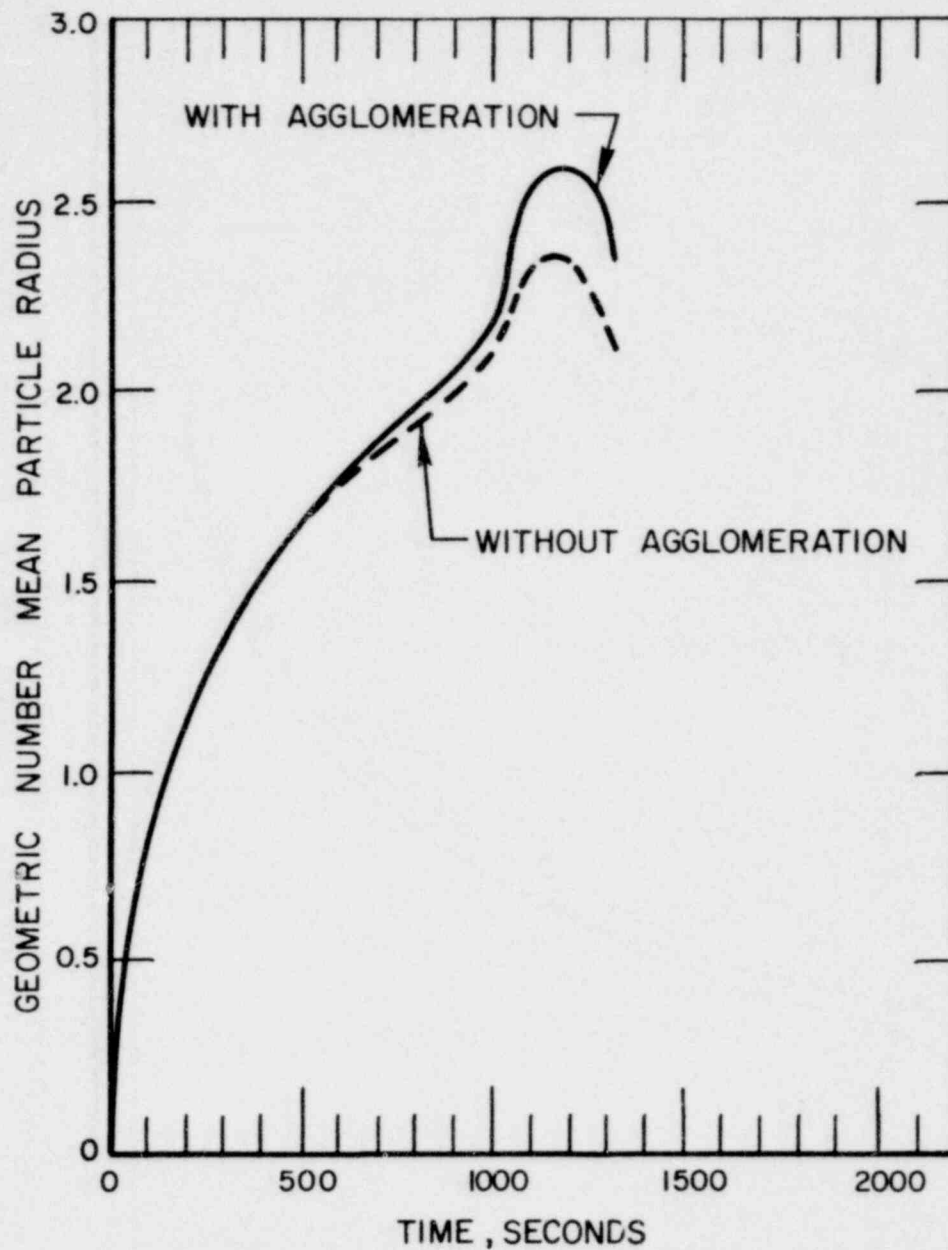
PARTICLE GEOMETRIC MEAN RADIUS (EACH VOLUME)

PARTICLE LOGARITHMIC STANDARD DEVIATION (EACH VOLUME)

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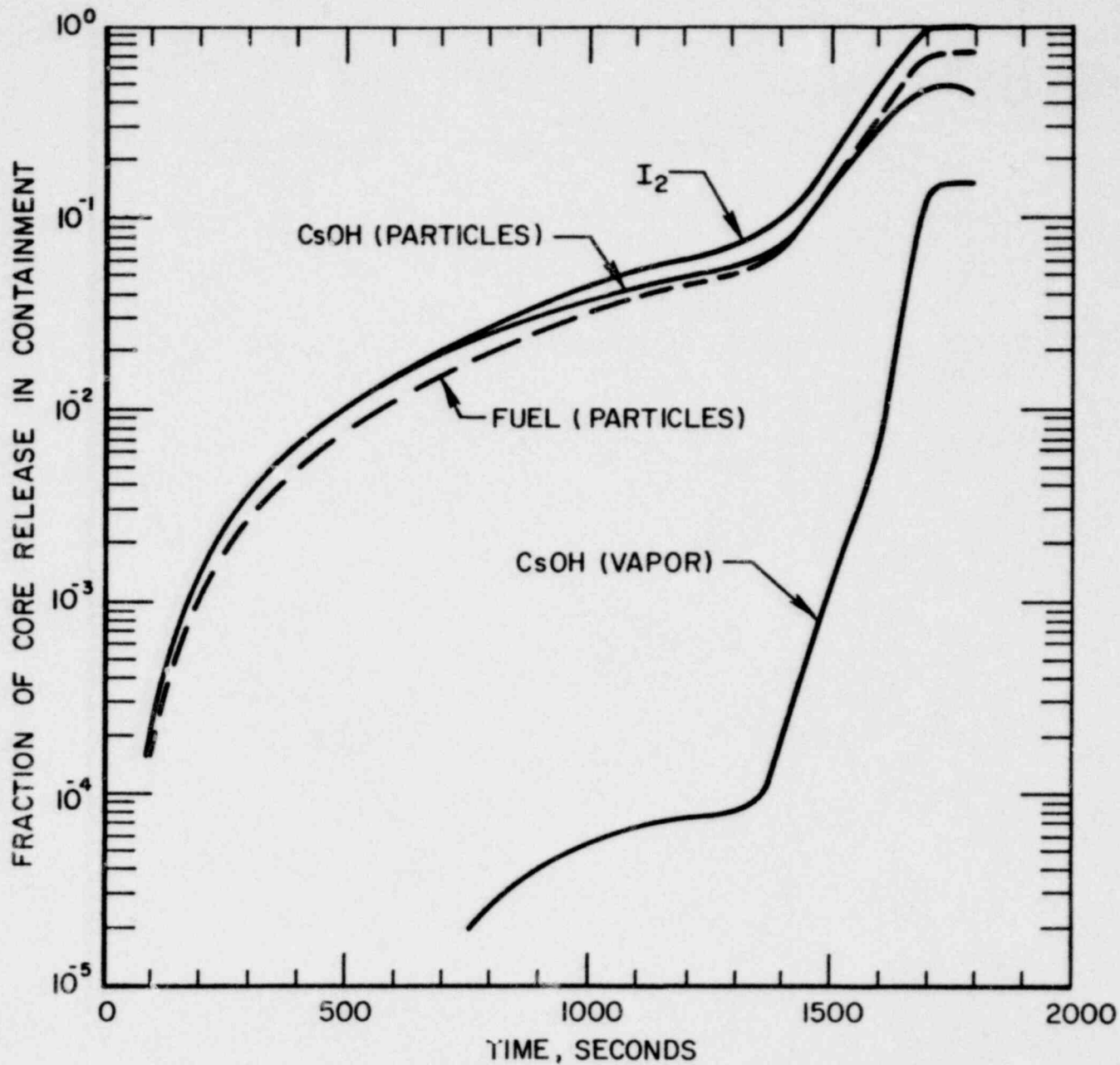
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PARTICLE SIZE LEAVING  
THE PRIMARY SYSTEM,  
TMLB' BASE CASE

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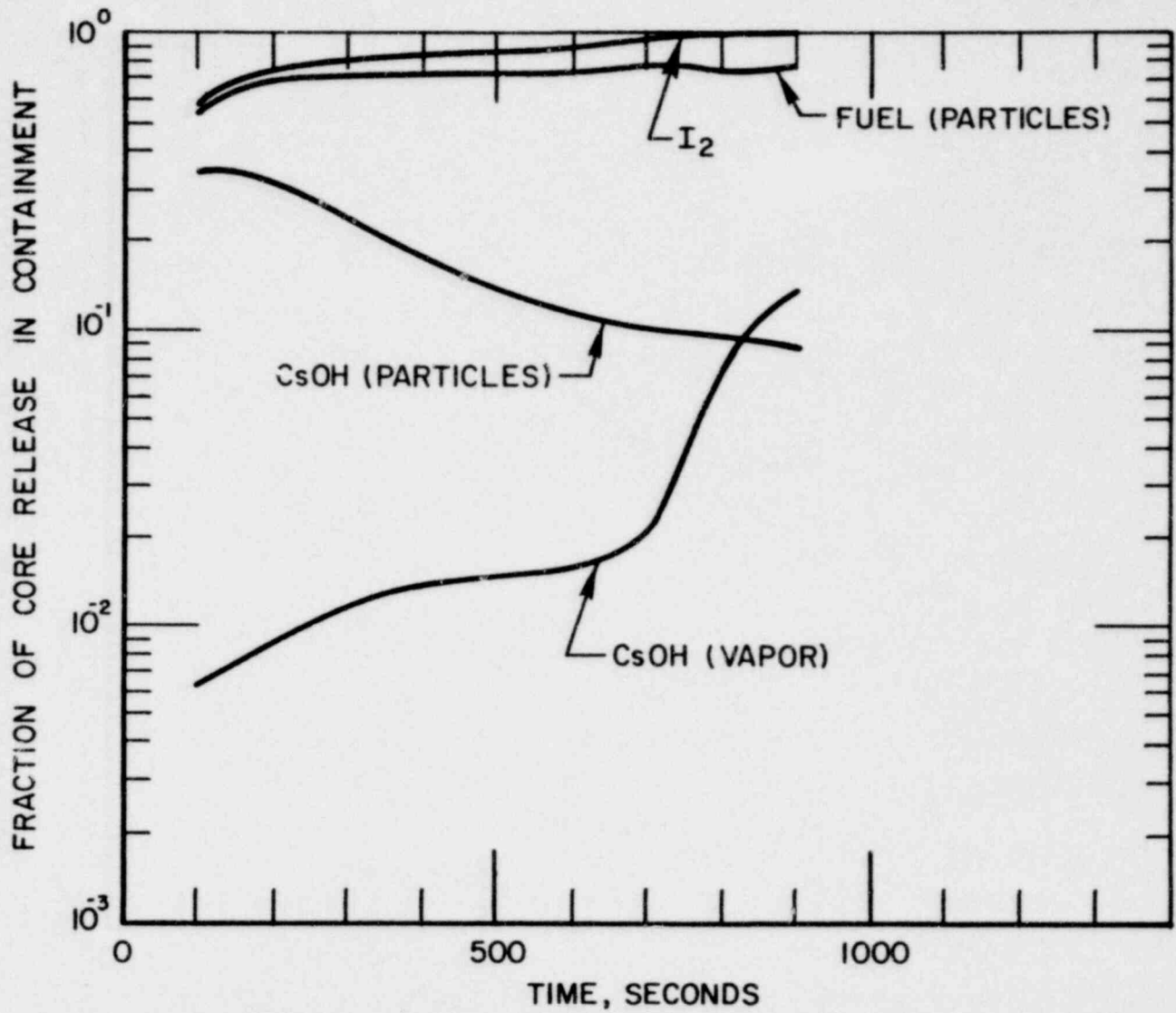
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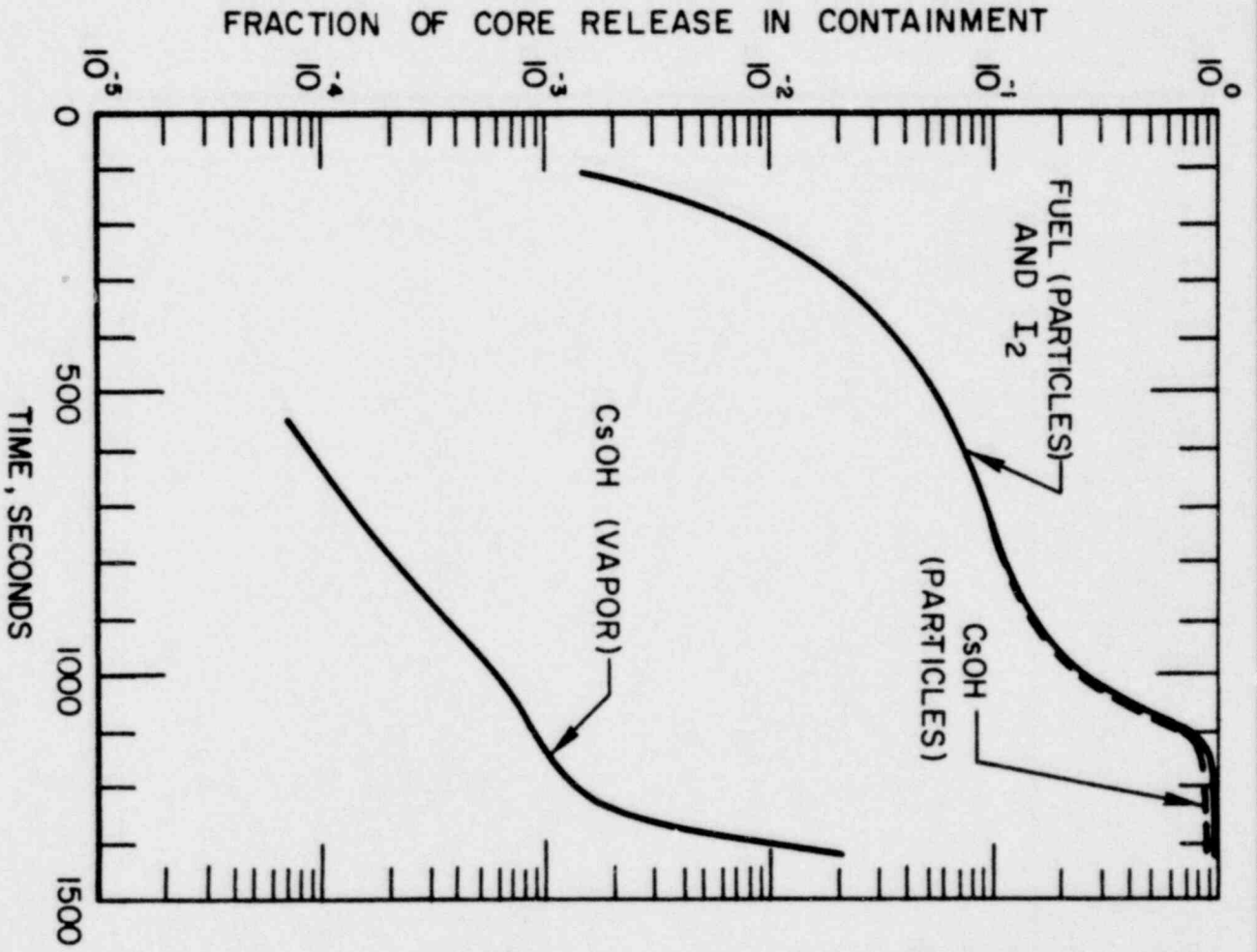
RELEASE FROM THE PRIMARY SYSTEM FOR TC BASE CASE

342 2081

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RELEASE FROM THE PRIMARY SYSTEM FOR AB BASE CASE



RELEASE FROM THE  
PRIMARY SYSTEM,  
TMLB' BASE CASE

842 2081

CONCLUSIONS ON PRELIMINARY TRAP-MELT CALCULATIONS

- A BETTER IMPRESSION OF RETENTION OF NUCLIDES IN THE PRIMARY SYSTEM MUST AWAIT THE UNCERTAINTY ANALYSIS IN VIEW OF THE LARGE UNCERTAINTIES OF SOME MODELS IN TRAP-MELT.

- THE CHARACTER OF THE MATERIAL EMITTED BY THE CORE IS SIGNIFICANTLY CHANGED BY PASSAGE THROUGH THE PRIMARY SYSTEM:

CsOH EMITTED AS VAPOR, TRANSPORTS ON PARTICLES

$r_g$ : 0.1  $\mu\text{m}$  +  $\sim 20$   $\mu\text{m}$

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EXPERIMENTAL STUDY OF SUBMICRON AEROSOL DEPOSITION

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OBJECTIVES

- DETERMINE CORRECT MECHANISMS OF AEROSOL DEPOSITION
- CORRELATE THE DATA WITH THEORETICAL MODELS
- INCORPORATE INTO TRAP CODE

AEROSOL DEPOSITION MECHANISMS

- BROWNIAN DIFFUSION
- INERTIAL IMPACTION
- GRAVITATIONAL SETTLING
- MIGRATION DUE TO THERMOPHORETIC FORCES

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IN GENERAL, DEPOSITION RATE DEPENDS ON

- PARTICLE SIZE AND DENSITY
- FLOW VELOCITY AND CHARACTERISTICS
- TUBE DIAMETER AND LENGTH.

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## EXPERIMENTAL TECHNIQUES

- AEROSOL PREPARATION

DI-OCTYLPHTHALATE (DOP) AEROSOLS

BY CONDENSATION TECHNIQUE

( $\sigma_g$ : 1.2 - 2.0)

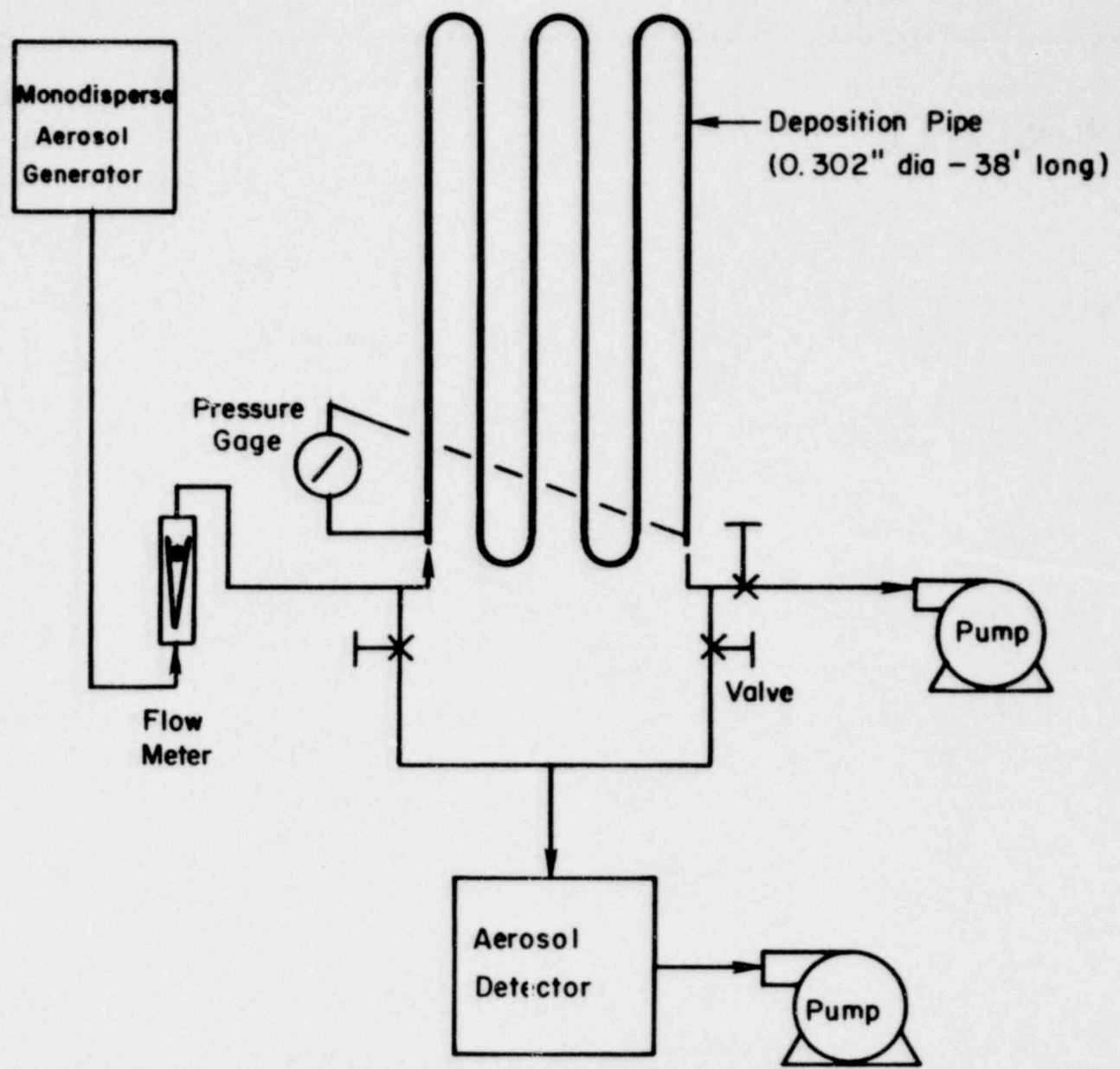
- AEROSOL DETECTION

- ELECTRICAL DETECTION

- WASH-OUT OF TUBE USING TAGGED  
PARTICLES

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AEROSOL DEPOSITION MEASUREMENT SYSTEM

EXPERIMENTAL CONDITIONS

● DEPOSITION SECTION

DIAMETER - 0.302 IN. ID

LENGTH - 38.5 FT.

● PARTICLE SIZE RANGE

0.035 - 1.3  $\mu\text{M}$

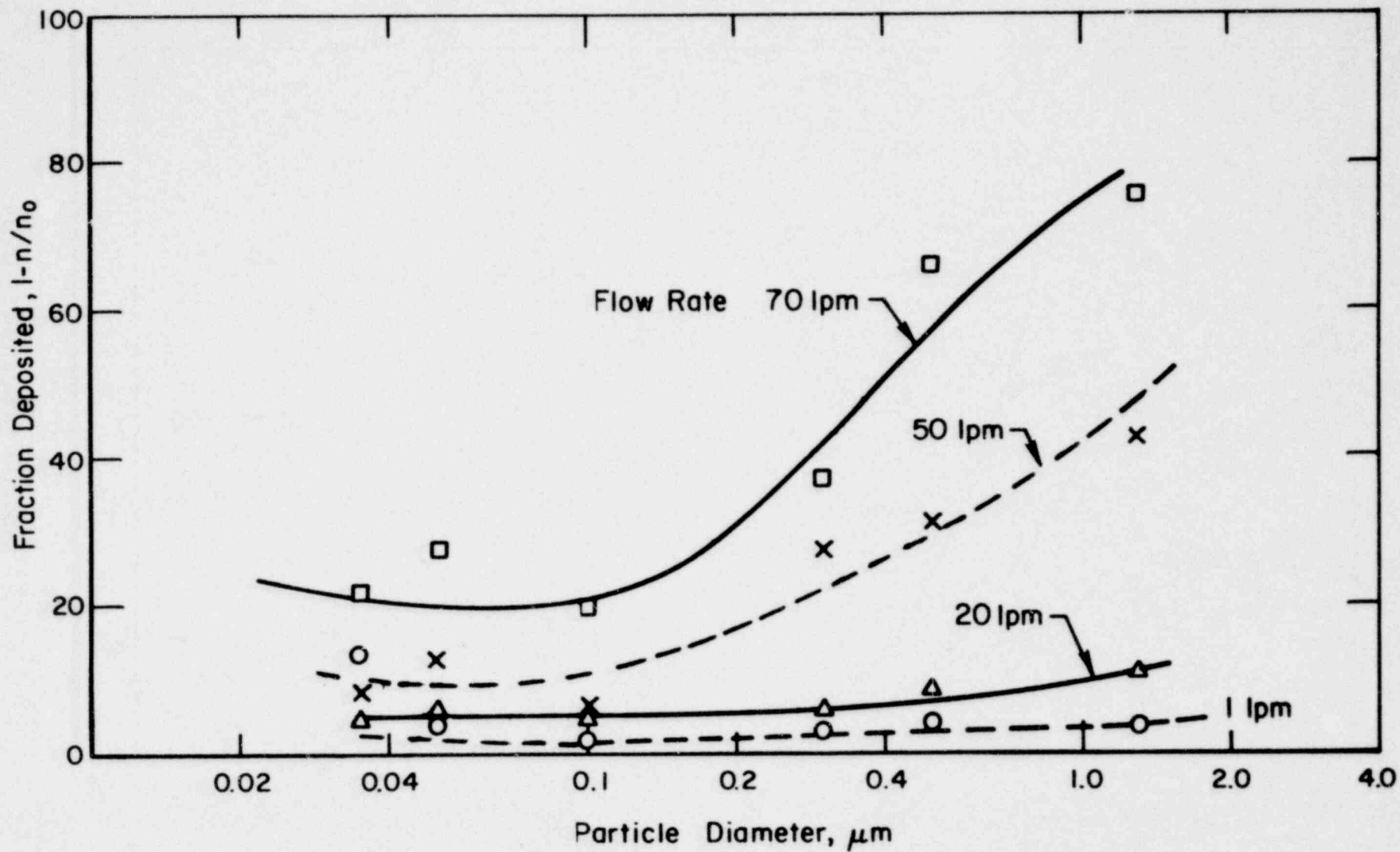
● FLOW RANGE

VOLUMETRIC: 1 - 70 LPM

VELOCITY: 0.36 - 25 M/SEC

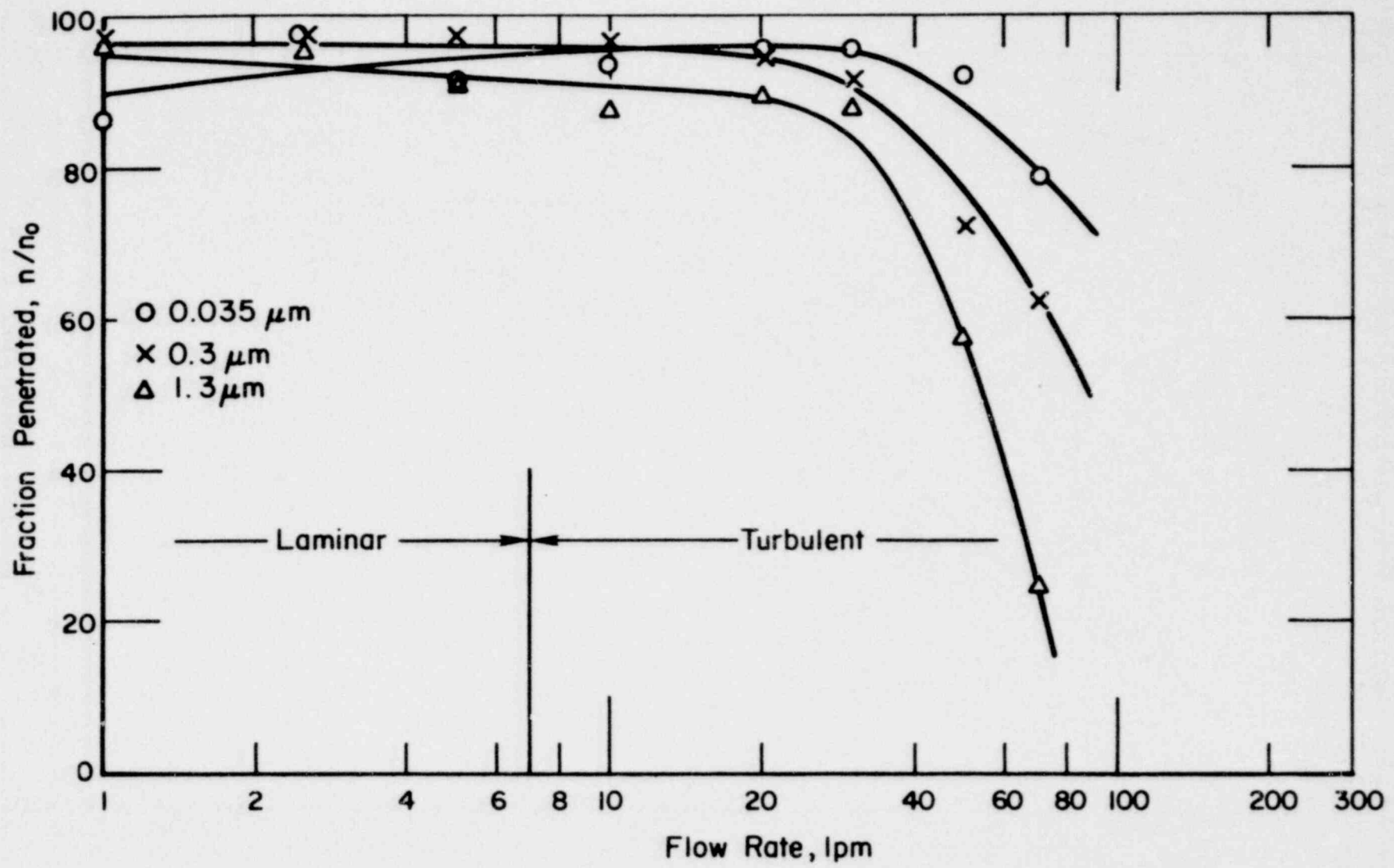
REYNOLDS NO.: 180 -  $10^4$

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VAPOR DEPOSITION EXPERIMENTS



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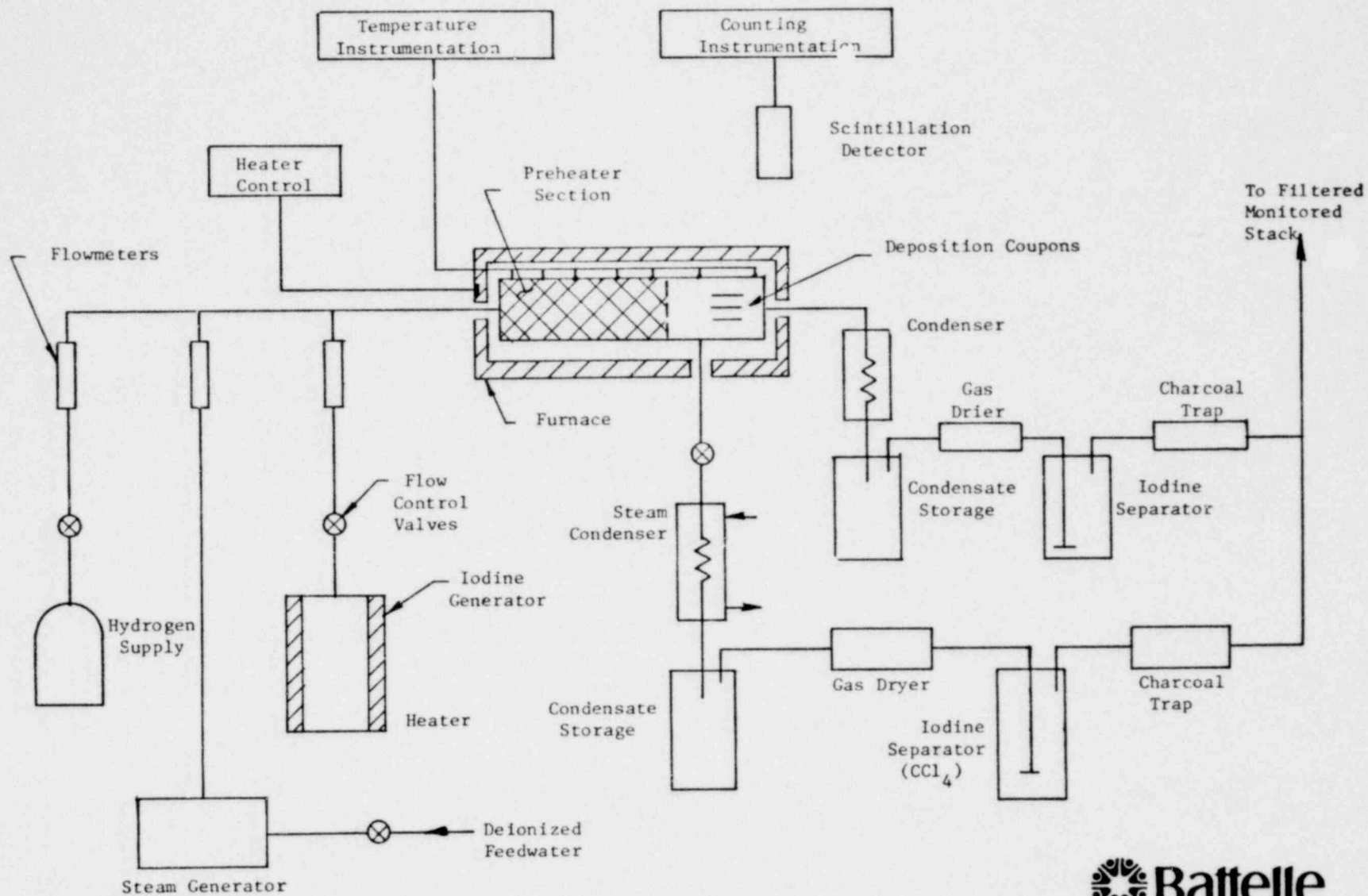
OBJECTIVES

- TO MEASURE DEPOSITION RATES FOR RADIONUCLIDE VAPORS ON LWR PRIMARY SYSTEM SURFACES
- TO PROVIDE INPUT CORRELATIONS FOR THE TRAP CODE
- TO INVESTIGATE CHEMICAL REACTIONS OF RADIONUCLIDES WITH SURFACES



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# SCHEMATIC OF I<sub>2</sub> EXPERIMENTAL SETUP



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PLANNED CONDITIONS FOR EXPERIMENTAL RUNS

SURFACE TEMPERATURES (C): 300, 600, 900, 1200

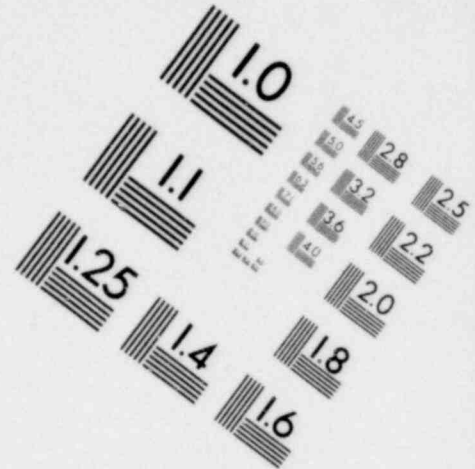
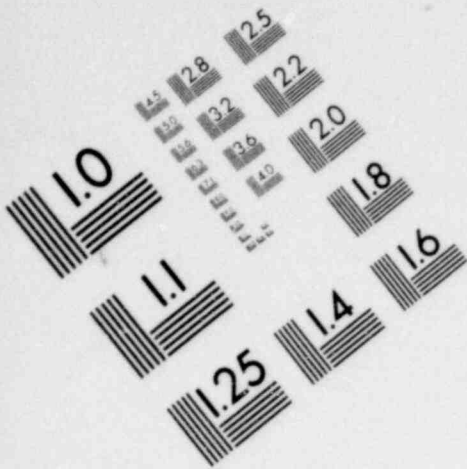
FLUID COMPOSITION (v/o H<sub>2</sub>): 0, 10

RADIONUCLIDE COMPOUNDS: I<sub>2</sub>, HI, Te

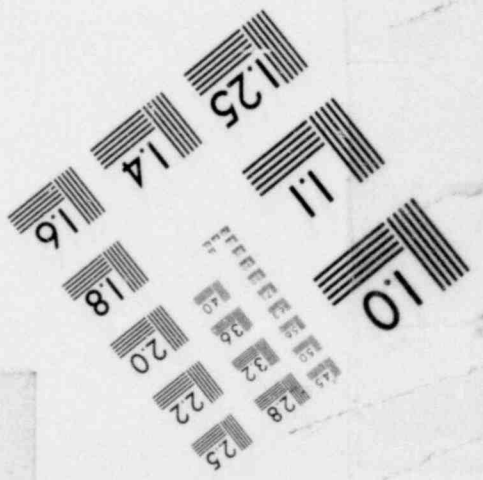
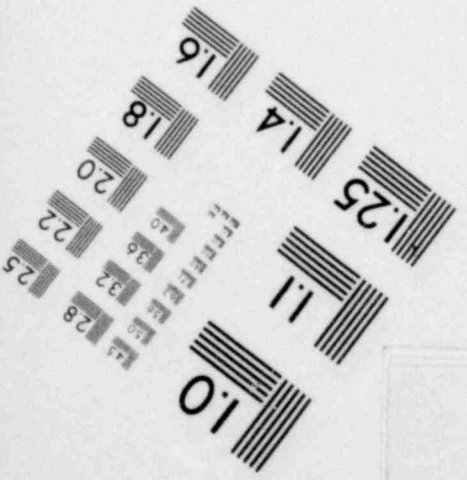
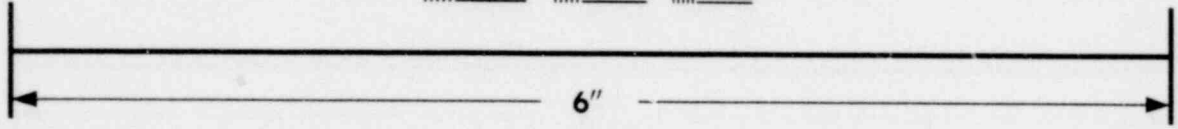
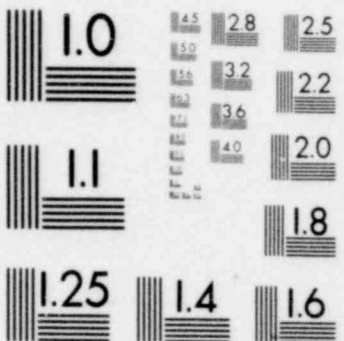
SURFACES: SS, INCONEL

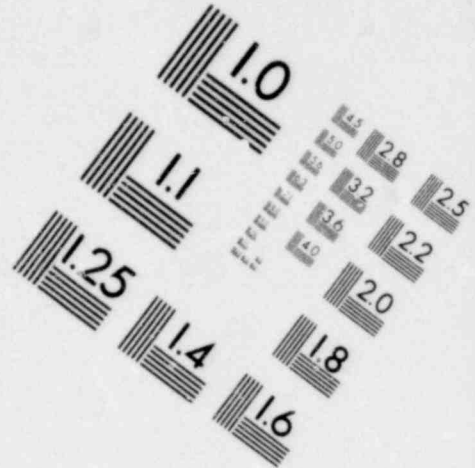
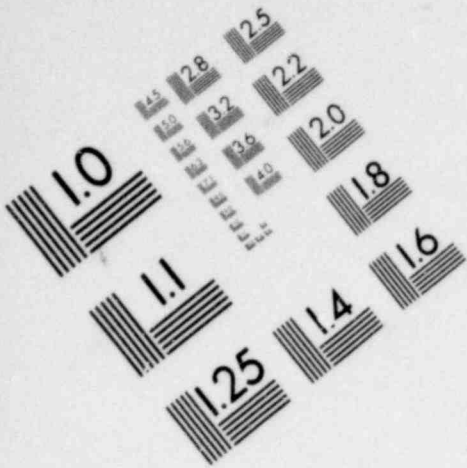
RADIONUCLIDE CONCENTRATIONS: BEST ESTIMATE

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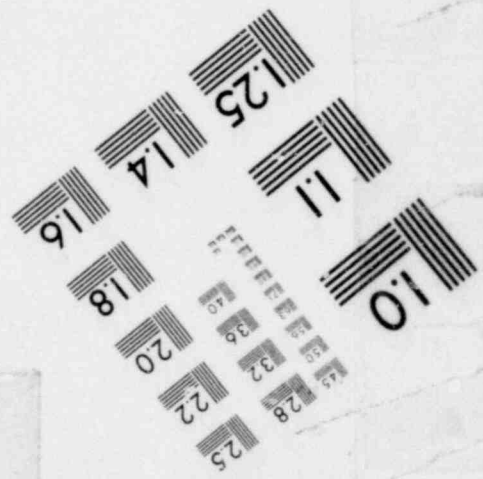
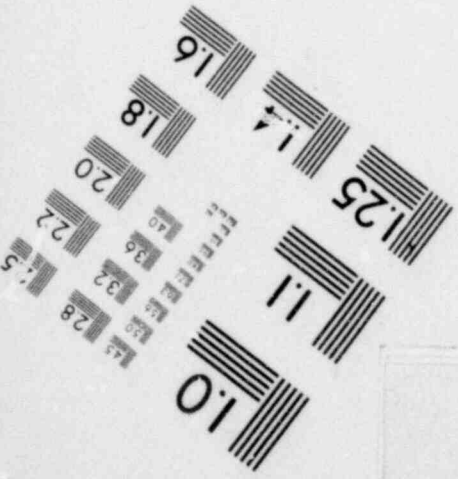
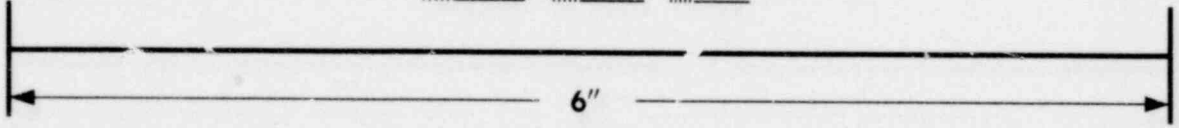
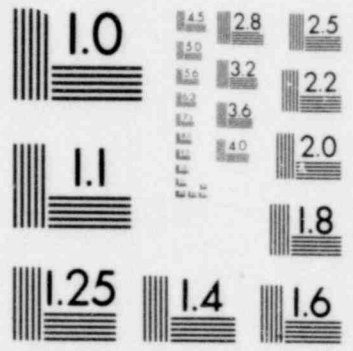


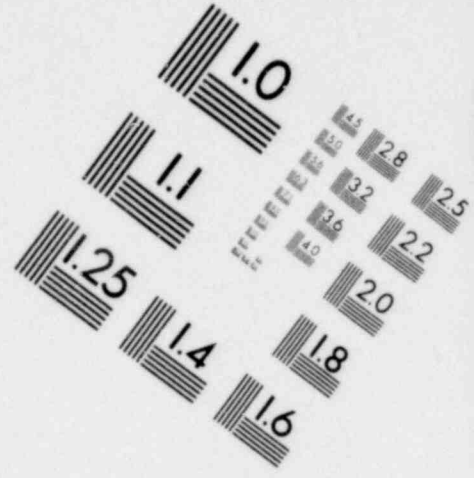
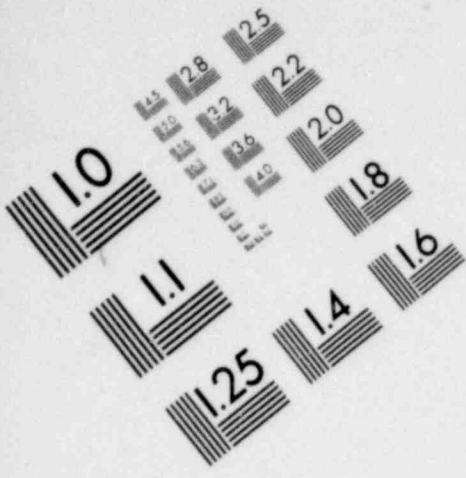
**IMAGE EVALUATION  
TEST TARGET (MT-3)**



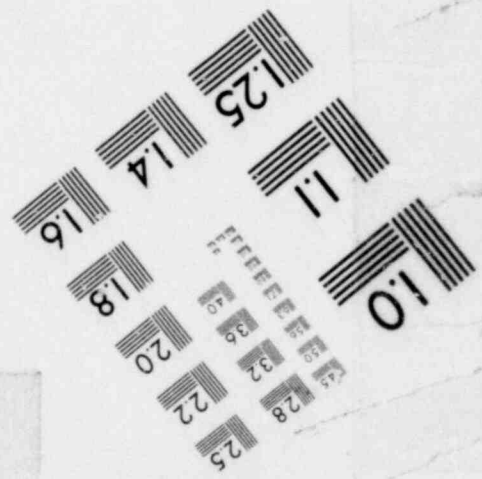
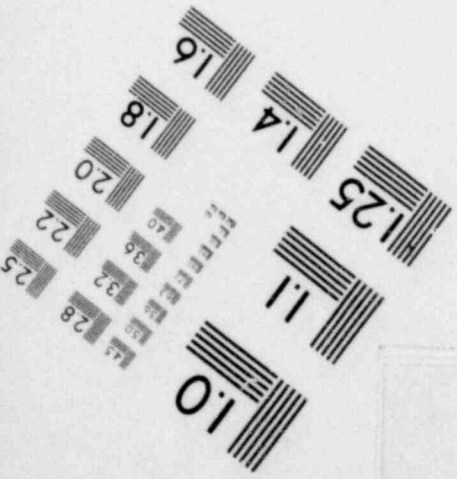
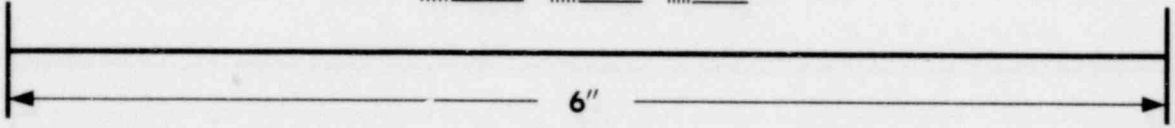
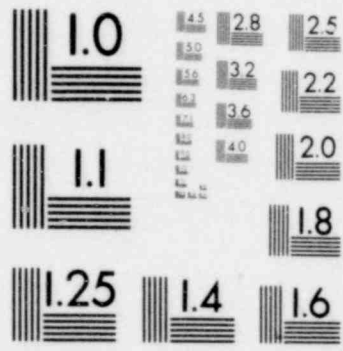


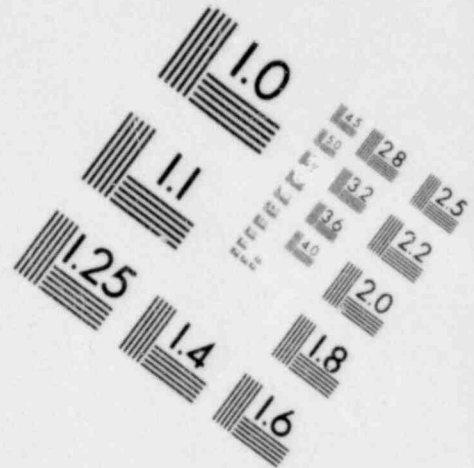
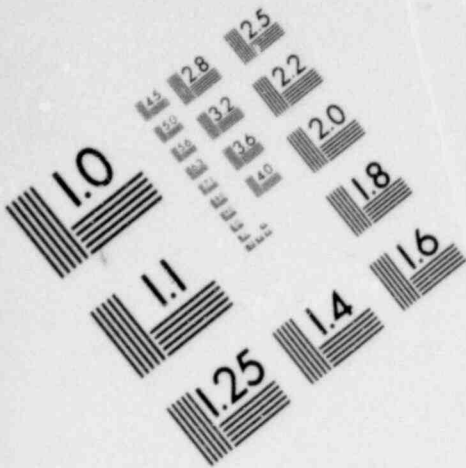
**IMAGE EVALUATION  
TEST TARGET (MT-3)**



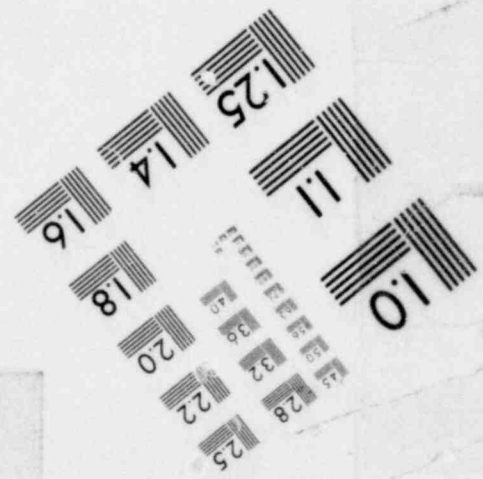
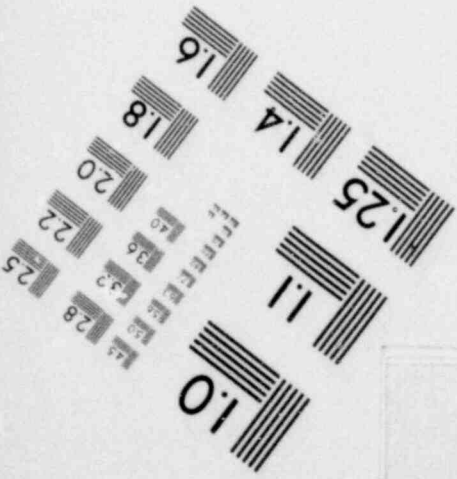
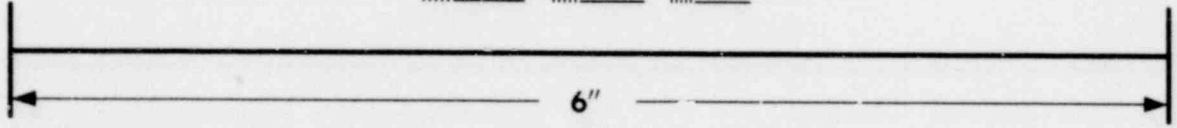
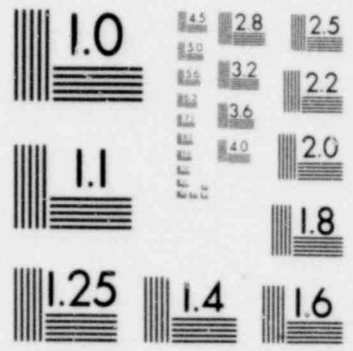


**IMAGE EVALUATION  
TEST TARGET (MT-3)**





**IMAGE EVALUATION  
TEST TARGET (MT-3)**



TRAP CODE UNCERTAINTY ANALYSIS

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## OBJECTIVES

- TO IDENTIFY AREAS OF UNCERTAINTY IN TRAP INPUT DATA AND MODELS
- TO DETERMINE THE CONTRIBUTIONS OF INPUT DATA AND MODEL UNCERTAINTIES TO THE OVERALL UNCERTAINTY IN RADIONUCLIDE DEPOSITION
- TO RANK INPUT VARIABLES AND MODELS ACCORDING TO THEIR RELATIVE IMPORTANCE
- TO RECOMMEND AREAS FOR FURTHER MODELING EFFORT OR DATA COLLECTION WHICH WILL REDUCE OVERALL UNCERTAINTY



UNCERTAINTY ANALYSIS PROCEDURE

- 1) IDENTIFY CRITICAL VARIABLES AND MODELS
- 2) ESTABLISH VARIABLE RANGES AT A GIVEN PROBABILITY LEVEL
- 3) SELECT A REPRESENTATIVE SET OF ACCIDENT SEQUENCES
- 4) IDENTIFY SPECIFIC VARIABLES AND MODELS TO BE CONSIDERED FOR EACH ACCIDENT SEQUENCE
- 5) DEVELOP AN APPROPRIATE STATISTICAL DESIGN FOR EACH ACCIDENT SEQUENCE
- 6) GENERATE A RESPONSE SURFACE FOR EACH STATISTICAL DESIGN
- 7) RANK THE VARIABLES AND MODELS

ACCIDENT SEQUENCES TO BE ANALYZED

	<u>TC</u>	<u>TMLB'</u>	<u>AB (COLD-LEG)</u>
SURFACE TEMPERATURE, C	300 - 345	300 - 595	300 - 895
FLUID TEMPERATURE, C	300 - 1590	370 - 595	300 - 1830
FLUID FLOW VELOCITY, M/SEC	.0021 - .0081	.0012 - .022	0.13 - 6
MELT PERIOD, MIN	55	20	15

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TC: BWR TRANSIENT WITH FAILURE OF RPS

TMLB': PWR TRANSIENT WITH LOSS OF OFFSITE AC POWER AND FAILURE OF FEEDWATER DELIVERY SYSTEM

AB (COLD-LEG): PWR LARGE LOCA WITH ELECTRIC POWER FAILURE TO ALL ESF'S

TRAP CODE INPUT VARIABLES TO BE INCLUDED IN UNCERTAINTY

ANALYSIS AND VARIABLE RANGES

	<u>Low</u>	<u>BE</u>	<u>High</u>
● VAPOR PRESSURES*			
● MASS FLOW RATES**			
● GAS TEMPERATURES**			
● SURFACE TEMPERATURES**			
● SOURCE TERM (MASS RELEASED)		RSS/WASH1400	
● MEAN PARTICLE SIZE ( $\mu\text{M}$ )	0.01		0.1
● VAPOR DEPOSITION VELOCITIES $I_2$ (MULTIPLIER)	0.2	1	5
● VAPOR MASS TRANSFER COEFFICIENTS (MULTIPLIER)	0.1	1	10
● THERMOPHORETIC PARTICLE DEPOSITION (MULTIPLIER)	1	1	4
● TURBULENT PARTICLE DEPOSITION (MULTIPLIER)	1/3	1	3

\* BOUNDING ARRAYS HAVE BEEN DEVELOPED FOR EACH RADIONUCLIDE SPECIES OF INTEREST.

\*\* BOUNDING ARRAYS HAVE BEEN DEVELOPED FOR EACH ACCIDENT SEQUENCE OF INTEREST.

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