Iodine Behavior in Design Basis and Severe Accidents

D.A. Powers
Sandia National Laboratories
Albuquerque, NM
R.Y. Lee, M. Salay
US Nuclear Regulatory Commission
Washington, DC

Evolving View of Radioactive Iodine

- Important fission product
  - ~750 million Curies in typical reactor core
- Nearly all reactors licensed originally to the TID-14844 Source Term
  - 100 % noble gas release
  - 50 % of iodine in gaseous form (1/2 deposits)
  - 1 % of all other radionuclides as particulate
- Following TMI a belief all iodine released to containment as particulate
  - Focus was on CsI
- Nagging evidence of some gaseous iodine

Alternative Source Term

- Iodine released to containment over time:
  - Gap Release (~5% of inventory)
  - In-vessel Release (25 to 35% of inventory)
- Most iodine released as unspecified particulate
  - usually interpreted as CsI
- 5% released as gaseous iodine
  - I_2
  - HOI
  - HI
  - CH_3I

What Happens to Iodine in Containment ?

- Particulate Iodine
  - Agglomerates with other aerosol particulate
  - Gravitational settling
  - Diffusiophoresis to cool surfaces
  - Removal by engineered safety systems
- Gaseous Iodine
  - Voluminous research on iodine chemistry in chemically simple systems (UK, France, Canada, Poland, Germany, Switzerland)

Iodine Chemistry Complicated

- Most iodides very water soluble
  - Exceptions are AgI, TlI, CuI
- Aqueous iodine in water can assume any of 8 different oxidation states
  - Iodide, I^-, can be oxidized to molecular iodine, $I_2(aq)$, which can partition from water back into the gas phase
  - Sensitive to pH
  - Other reactions possible

Expectations

- Particulate and gaseous iodine released to containment would end up in sump waters
- Iodine would remain in sump if alkaline conditions maintained
  - Acidification from nitric acid formation and other radiolytic processes (cable insultation degradation, radiolysis of solvents from paint)
- If acidified, molecular iodine would partition back into the atmosphere
  - Might form volatile organic iodides, CH3I
  - Rate of release from sump to atmosphere would increase as the sump approached boiling

PHÉBUS-FP Tests

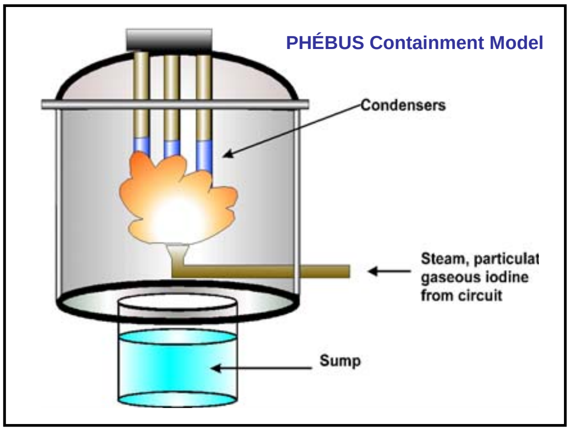
- Opportunity to test expectations
  - Realistic configurations and chemical complexities

Pertinent Tests

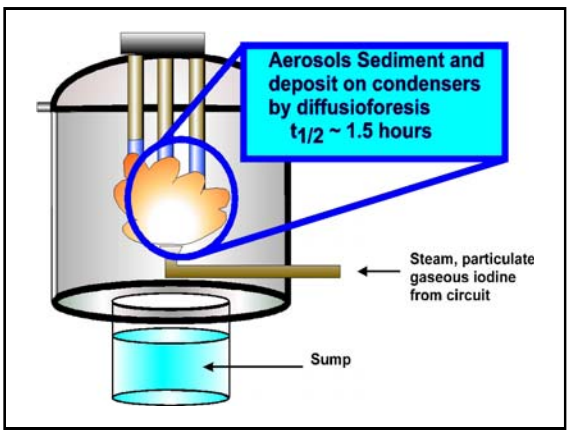
	FPT-0	FPT-1	FPT-2	FPT-3
Fuel	Irradiated 9 days	23 GWd/t	32 GWd/t	24 GWd/t
Flow	Steam rich	Steam poor	Steam poor + boric acid	Steam poor
Control Rod	Ag-In-Cd	Ag-In-Cd	Ag-In-Cd	B_4C
Sump pH	Acid (pH = 5)	Acid (pH = 5)	Alkaline (pH = 9)	Acid (pH=5)
Sump	Condensing	Condensing	Condensing initially; Evaporating in Chemistry Phase	Condensing initially; Evaporating in Chemistry Phase

Test Phases

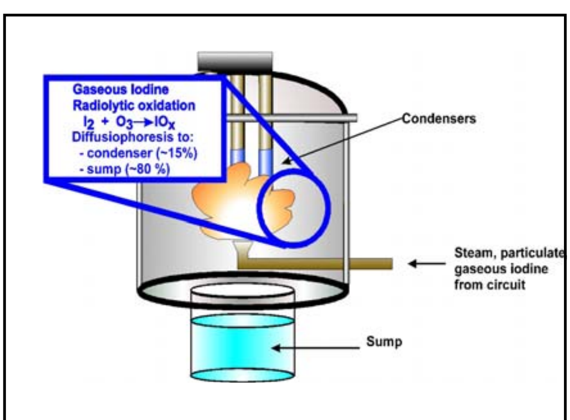
- Release Phase ~2.5 hours
- First aerosol phase with continued steam injection ~ 1 hour
- Second aerosol phase with no continued steam injection ~3.5 hours
- Washing phase ~20 minutes
- Chemistry phase ~10s of hours



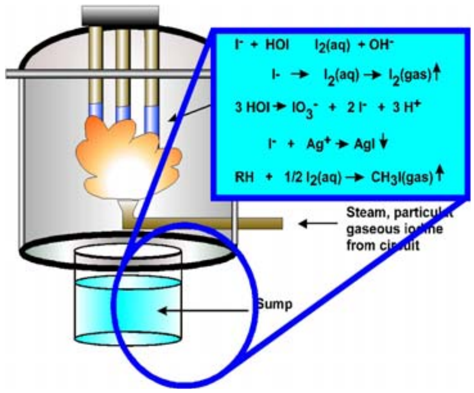
PHÉBUS Containment Model
Condensers
Steam, particulat
gaseous iodine
from circuit
Sump

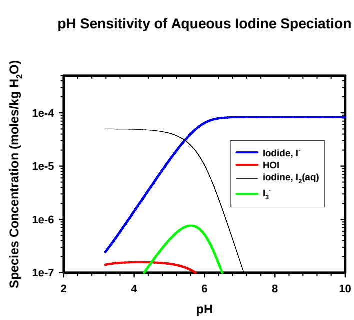


Aerosols Sediment and
deposit on condensers
by diffusioforesis
$t_{1/2}$ ~ 1.5 hours
Steam, particulate
gaseous iodine
from circuit
Sump



Gaseous Iodine
Radiolytic oxidation
$I_2 + O_3 \rightarrow IO_x$
Diffusiophoresis to:
- condenser (~15%)
- sump (~80 %)
Condensers
Steam, particulate
gaseous iodine
from circuit
Sump



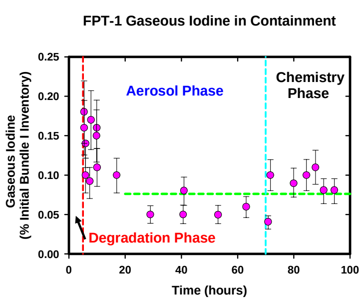


Pertinent Results

	FPT-0	FPT-1	FPT-2	FPT-3
Fraction of iodine released to containment as a gas	< 0.25	~0.02	~0.01	~0.85
Aerosol Sedimentation half life Phase 1 (hr) Phase 2 (hr)	1 1.5	1 1.5	1.5 1.8	4.5
Sump pH	Acid	Acid	Alkaline	Acid
Ag : I in sump	2000	45	10	< 0.5
Steady state iodine concentration in containment	Yes	Yes	Yes	Yes

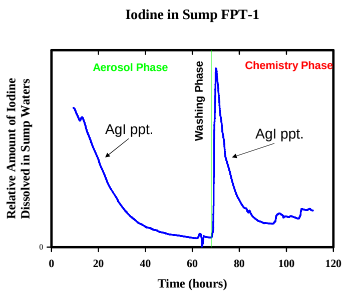
Conclusions

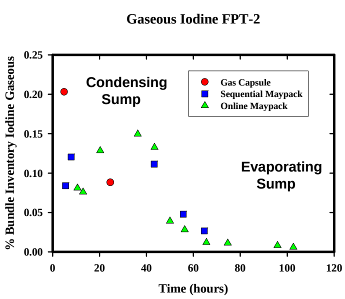
- Indeed some fraction of iodine released to containment model as a gaseous species
  - Consistent with AST
  - CsI not the only particulate form of iodine (CdI_2, NiI_2, InI_2, AgI, etc.)
- Overall Iodine release rate consistent with expectations based on current severe accident modeling
- Aerosol sedimentation consistent with expectations



Observations

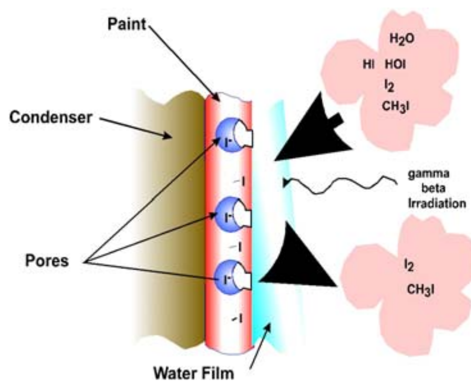
- A steady-state concentration of gaseous iodine developed in the containment model for all tests
  - Acid or alkaline sump did not affect qualitative observation
  - Ag precipitated iodine when abundant in sump (will iodine absorb on other materials in sumps following accidents?)
- Behavior contrary to expectations
  - Increases when sump condensing
  - Decreases when sump evaporating
- Complicated variations between molecular iodine (I_2) and volatile organic iodide (CH_3I)





Conclusion

- **Observed behavior of gaseous iodine in the PHEBUS tests is not driven by iodine partitioning from the sump!**
  - **Behavior likely involves interactions with painted condenser surfaces above the sump**
  - **The condensers simulate in some sense cooler structures with reactor containments during accidents**



Working Hypothesis

- Iodine deposited on condensers interacts with paint
  - Dissolves in pore water, reacts with polymer or with residual solvent
- Irradiation releases I_2(gas) or CH_3I(gas)
- Gaseous iodine radiolytically destroyed to form IO, IO_2, IO_3 (IO_x) or INO_y
- IO_x and INO_y nucleate to form very fine particles
  - Agglomerate with other particles and deposit
  - Driven by diffusiophoresis back into condenser

Scaling Issue

- How do observations of PHÉBUS tests translate to reactor accidents?
  - Original scaling of tests based on a different set of expectations
- Need a mechanistic understanding of the observed phenomenon of a steady-state concentration of gaseous iodine.

PHÉBUS-ST (EPICUR) and BIP Iodine Testing

- Examine the effects of prototypic sump debris on the partitioning of aqueous iodide back into the atmosphere as gaseous iodine
  - How well does debris inhibit gaseous iodine formation in a radiation field?
  - Use debris prescription from tests with paint chips and progressive development of corrosion products
- Validate the mechanism of gaseous iodine formation such that extrapolation to reactor accidents can be done confidently.
  - Formation of iodine oxide particulate
  - Nature of iodine interactions with paint
  - Characterization of reactive surface
  - Nature of radiolytic desorprtion of iodine from surfaces as a gaseous species

NRC Plan – Iodine Behavior

- Establish an analysis framework
  - Assemble mechanistic models to analyze iodine behavior in a containment
- Validate models
  - Obtain data to validate critical elements of models
- Understand the steady state gaseous iodine observed in the PHÉBUS tests
  - Analyze iodine behavior in PHÉBUS tests
- Scale to prototypic conditions
  - Analyze behavior to be expected in PWR containments
- Document
  - Publish
  - Peer Review
- Determine appropriate regulatory changes, if any (Regulatory Guidance, Rulemaking, etc.)