PAUL SCHERRER INSTITUT

PSI

Wir schaffen Wissen – heute für morgen

# Experience and Competence in Severe Accident Research & Development and Application

S. Guentay

NRC Public Meeting

September 4th 2012, Rockville, MD

# Outline

- A short history
- Aerosol research in interest to FCVS
- Iodine Research in interest to FCVS
- Conclusions

# A brief history

1980s:

- Participation in LACE and DEMONA tests and development of on-line aerosol concentration measurement devices
- OECD-LOFT LP-FP-2: core melt and FP release: start of Fission product research
- Containment iodine behavior code development, start of experimental iodine research (basic iodine behavior under radiation), pool scrubbing of iodine in water pools, support SULZER in FCVS development
- ACE projects

1990's:

- Development of PSI suit of codes for source term analysis
- Start of aerosol research (aerosol generation by plasma, POSEIDON pool scrubbing, GE-SBWR PCCS and SIEMENS SWR100 PCCS behavior under SA, Sulzer FCVS qualification tests, …
- ACEX and MACE projects
- Phebus project

# A brief history (cont)

2000s:

- ARTIST project studying aerosol and droplet retention in steam generators
- Qualification tests for CCI-FCVS
- Severe Accident Safety studies for some Swiss plants in support of PSA
- Hydrogen behavior in containments (PANDA)
- Continued iodine research (effect of impurities and silver), code development
- Fundamental research on aerosol behaviour (deagglomeration, etc) and fluid turbulence interactions
- Development of a novel process to efficiently decontaminate iodine laden gas and effective retention of all iodine species in aqueous solutions
- MELCOR and SCDAP/RELAP5: intensive assent and model development efforts and plant application

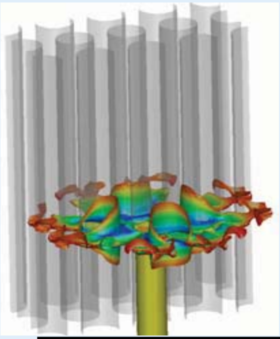
2010s:

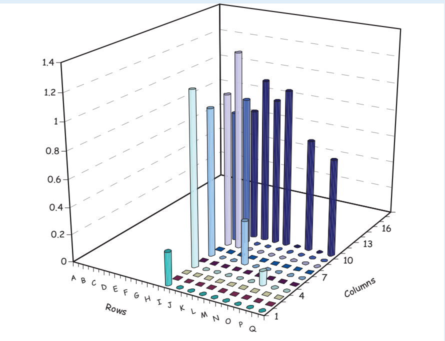
- Support IMI (CCI-AG) for demonstration of FCVS performance under utility specified conditions
- Steam Generator Safety Issues (reflux cooling, conditions leading to failure of RCS components) in process of launching
- Hydrogen mitigation: multi-experiment and analyses efforts (ERCOSAM, SAMARA)

# R& D related to Aerosols in Wet Scrubber based FCVSs

# ARTIST: Aerosol Retention in Steam Generator (2002-2011)

12 International organizations (safety authorities (including NRC), utilities, universities)

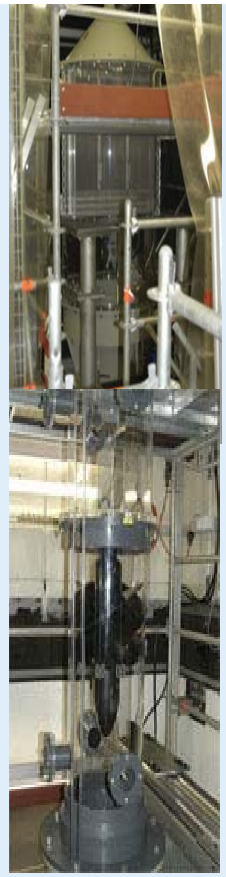





# ARTIST Facilities

Break stage

Larger scale-bundle

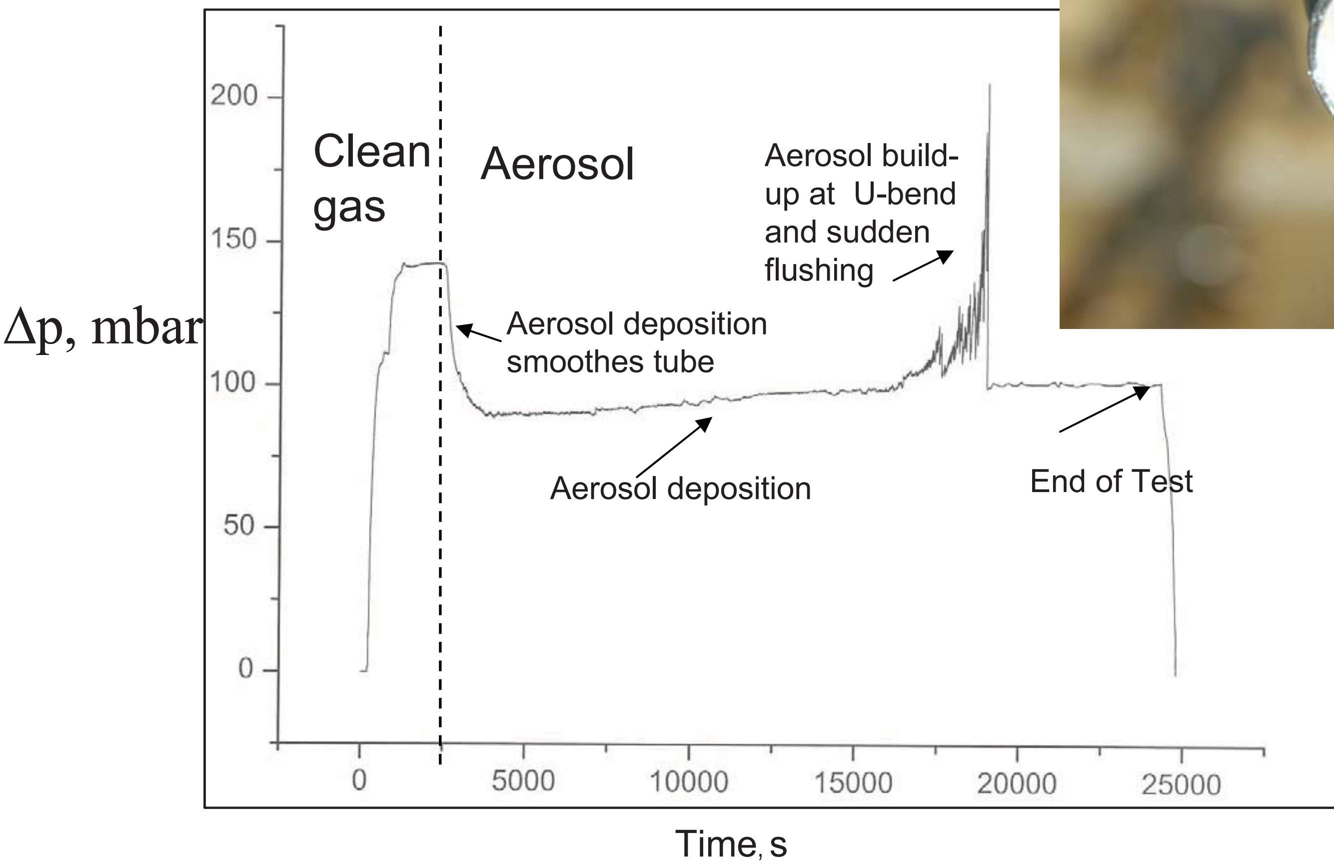


Droplet retention

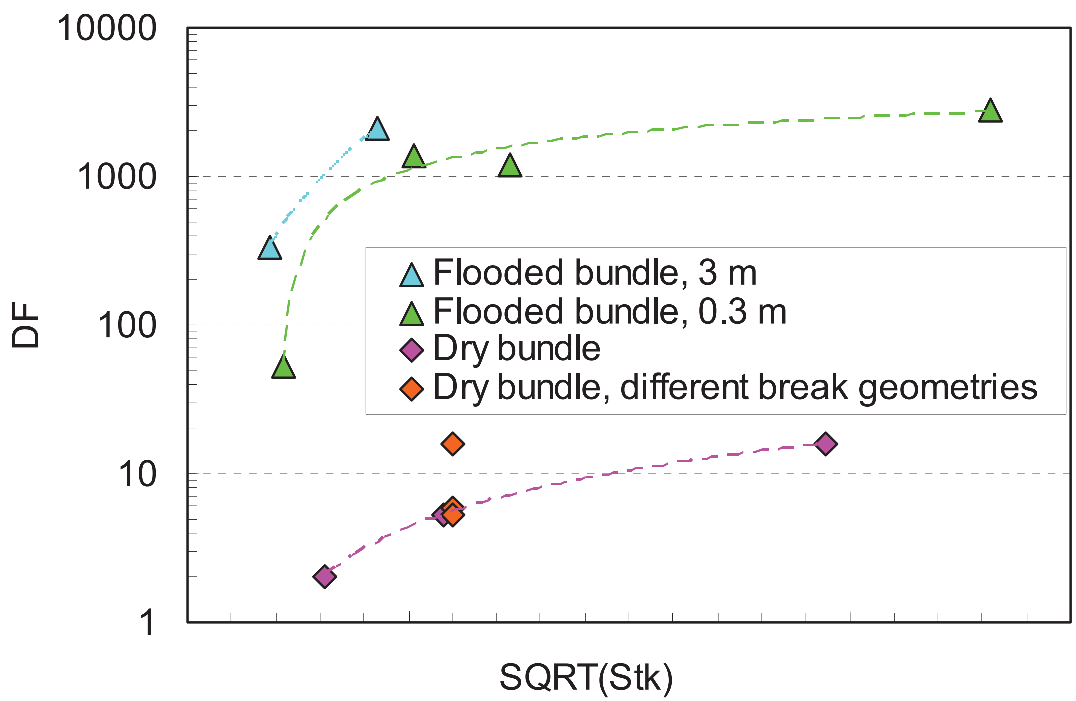
Integral mock-up facility

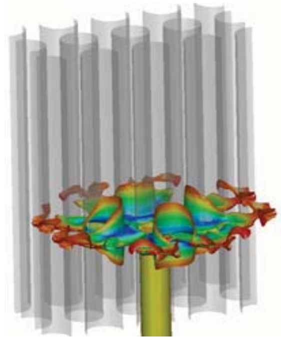
# ARTIST: Aerosol Retention in Steam Generator tubes

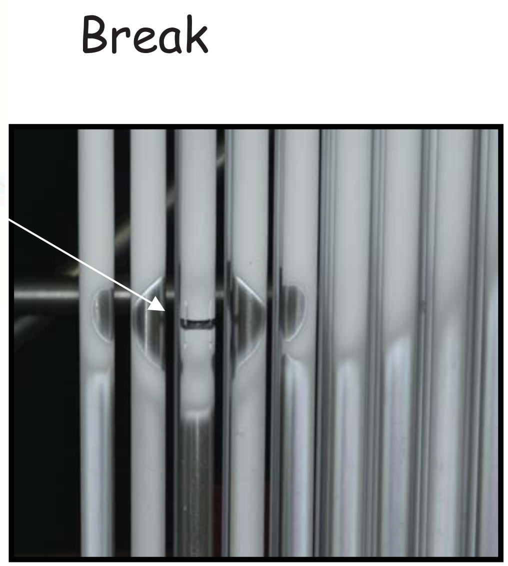
- Pressure drop across steam generator tube U-bend by aerosol deposition strongly dynamic




# Aerosol retention in flooded or dry SG bundle

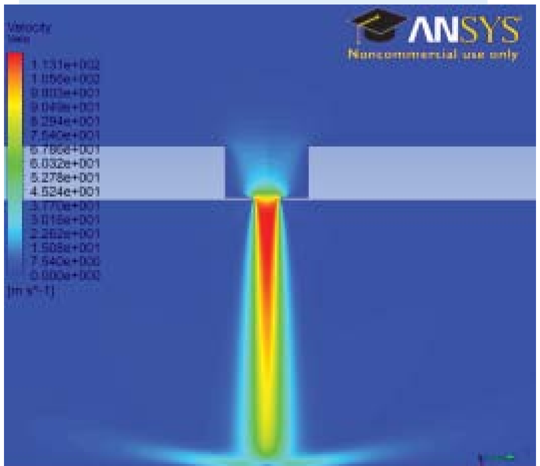


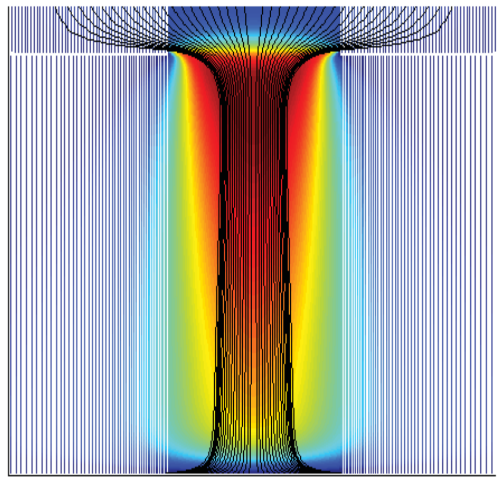





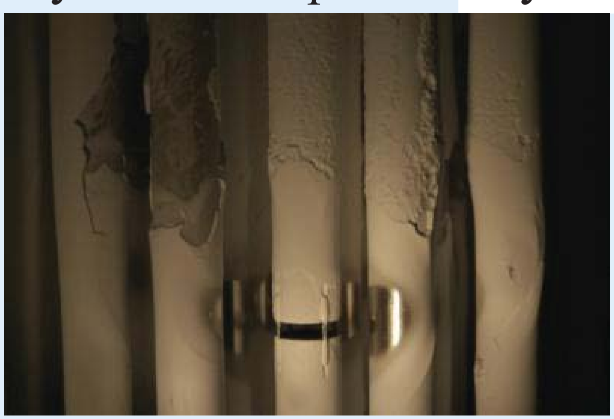
# Particle impaction and trajectories

**Flow simulation** → **Particle trajectories**

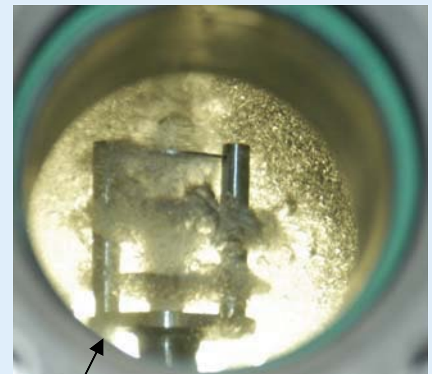


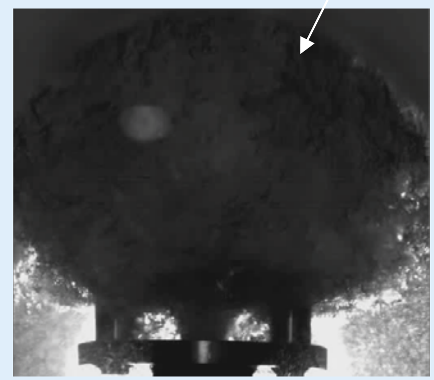
Dry aerosol deposition by impaction



Relevance to CCI-FCVS

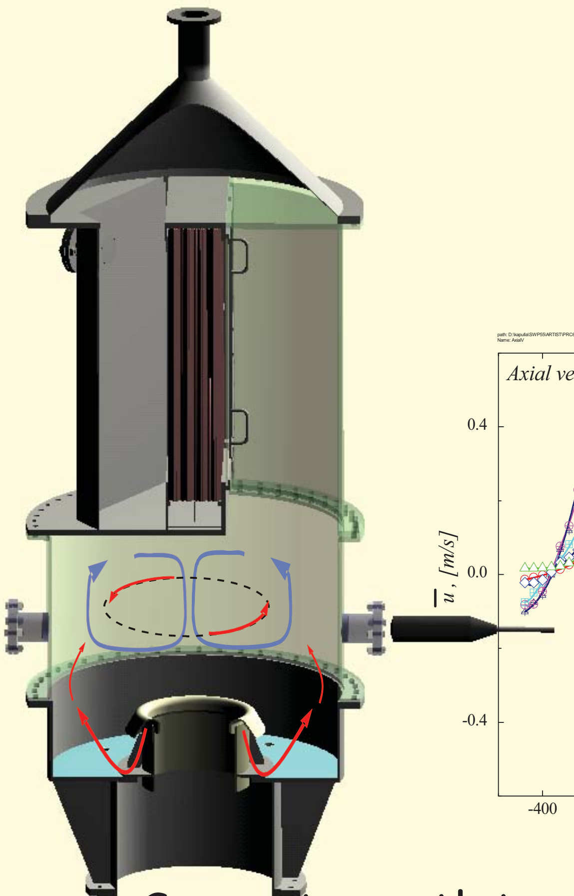


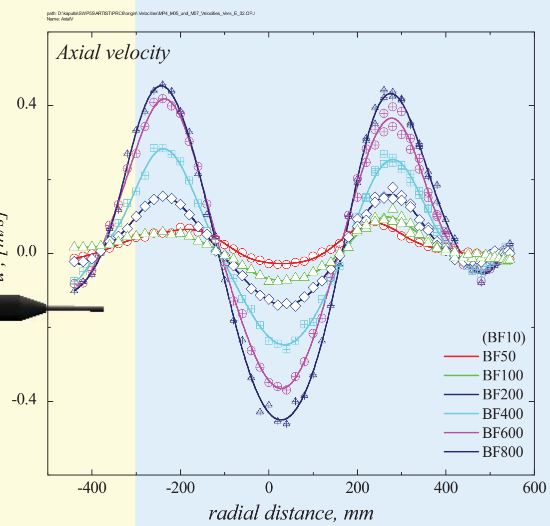
Small and high flow



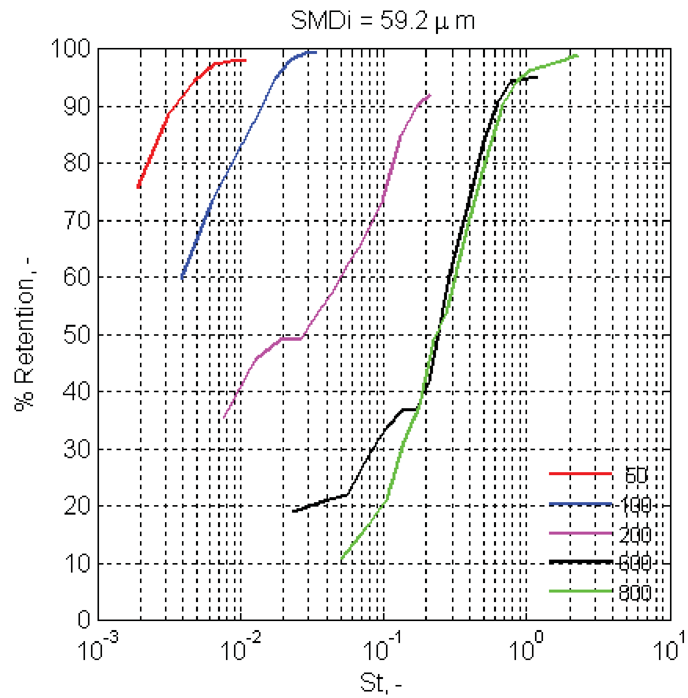
Sparger assembly acting like a multi stage impactor

# Physical understanding and modeling complex problems



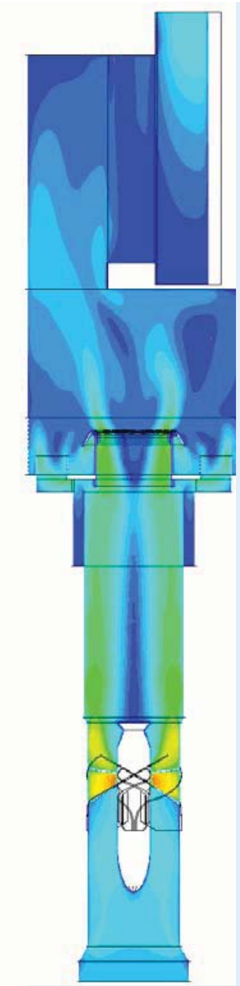


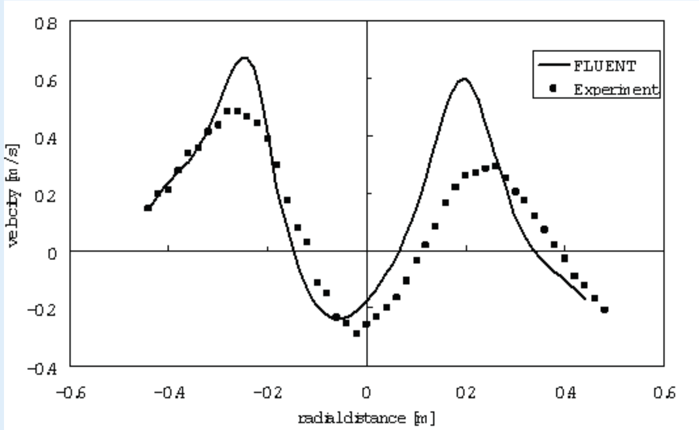

Separator outlet




Droplet retention efficiency

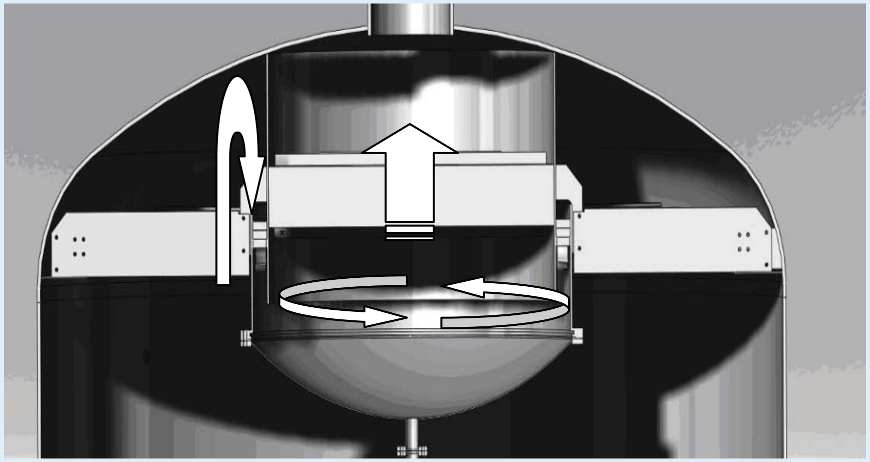
# Aerosol transport/retention in droplet separators






Measured and predicted velocity profiles in PWR Full Size Droplet Separator (ARTIST Project)

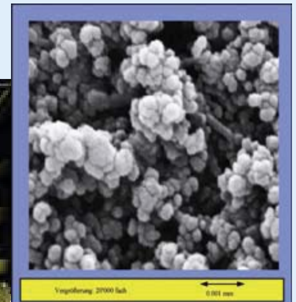
Experience important for evaluating performance



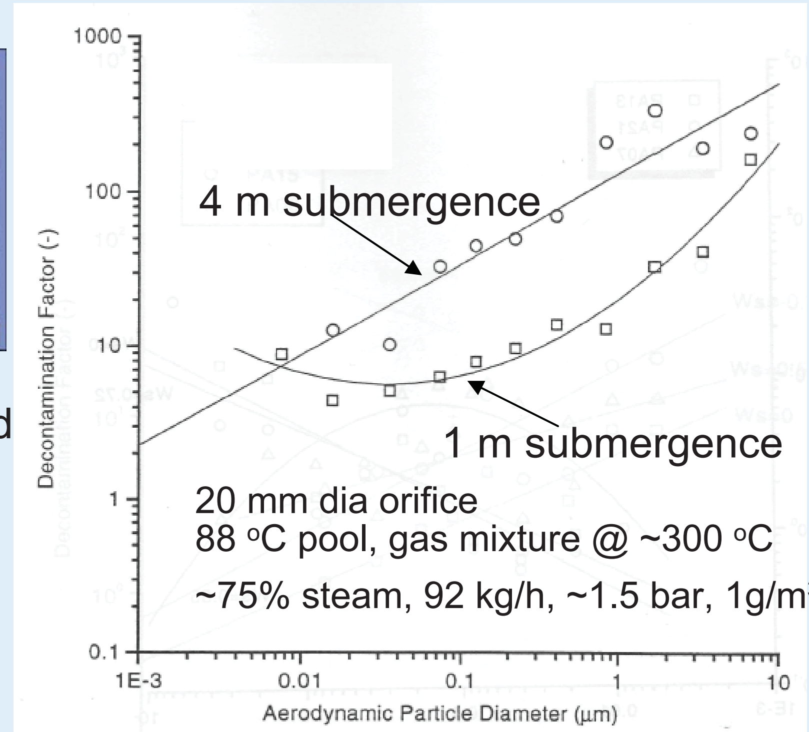
**CCI FCVS Droplet Separator**

# Aerosol removal by pool scrubbing in hot pool

POSEIDON II tests, 1994-1996

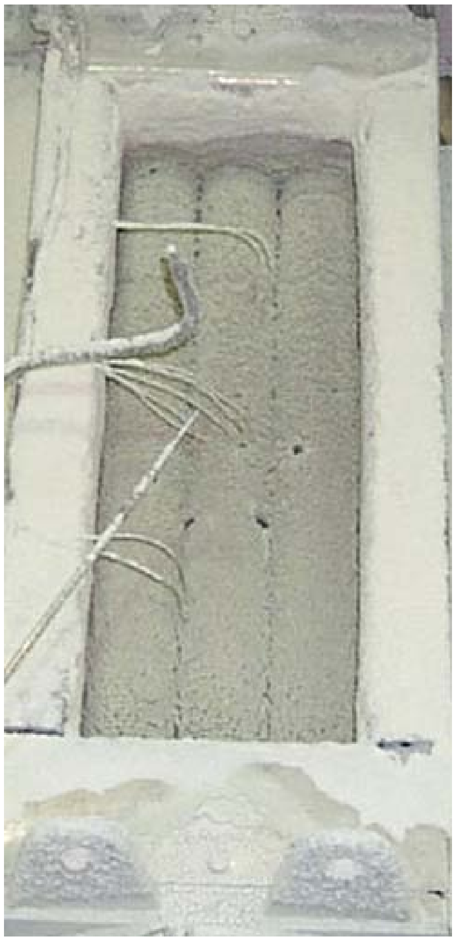


Plasma generated particles




Particle size dependent DF mostly of bubbles in Rise zone

# Aerosol deposition on steam condensing surfaces



SWR1000-PCCS model

- Aerosol particles: CsOH + SnO2
- Condenser
- Steam and non-condensable gas as the carrier gas
- ➢ Steam condensation not able to drain the aerosol particles depositing on the condenser surface

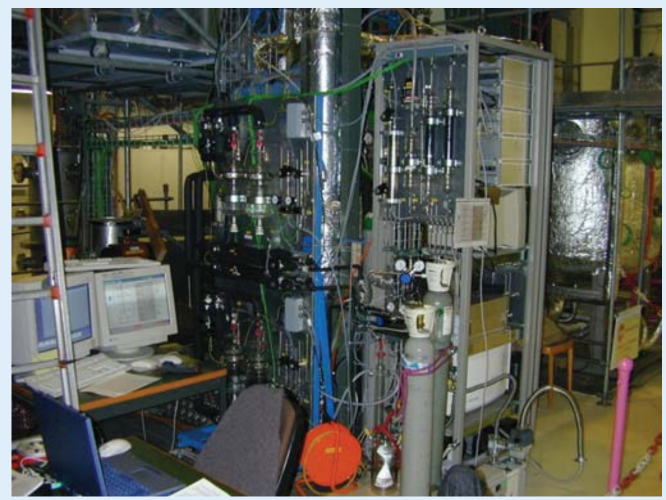
# Airlift reactor: demonstration facility with CCI-mixing element and sparger unit


Solution contains all chemicals

With I2 laden gas sparging

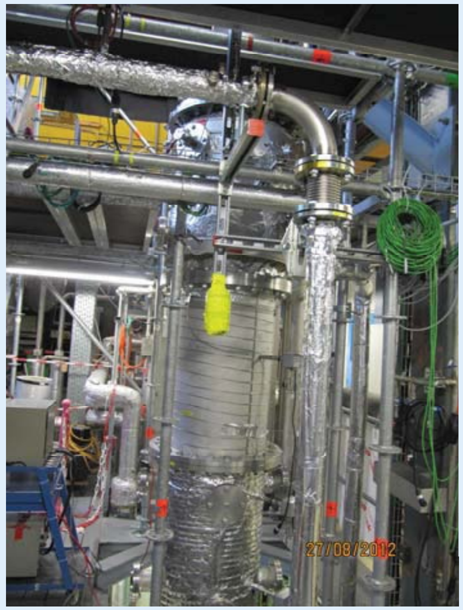
# CCI-FCVS mock-ups tested in 1990s and 2000s

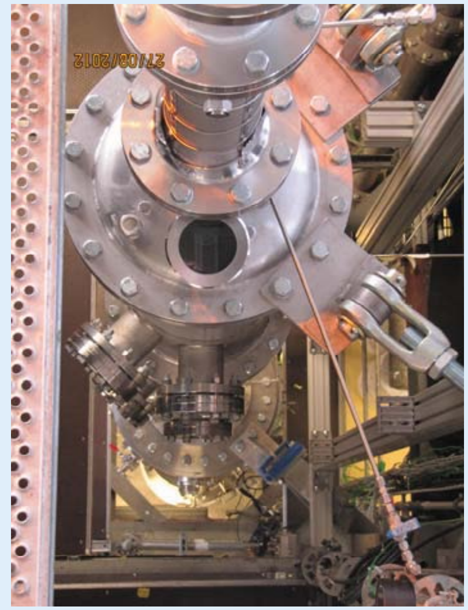
1993-1995

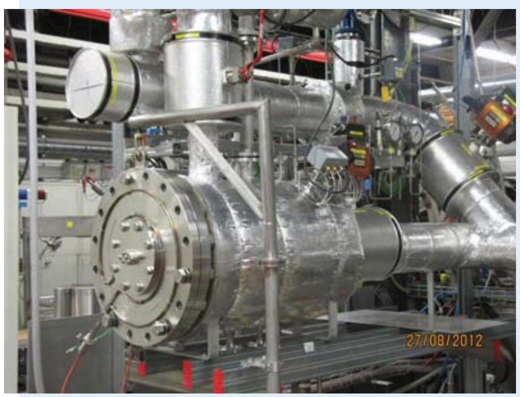



2000-2002

# CCI FCVS: Full-height Mock-up: today





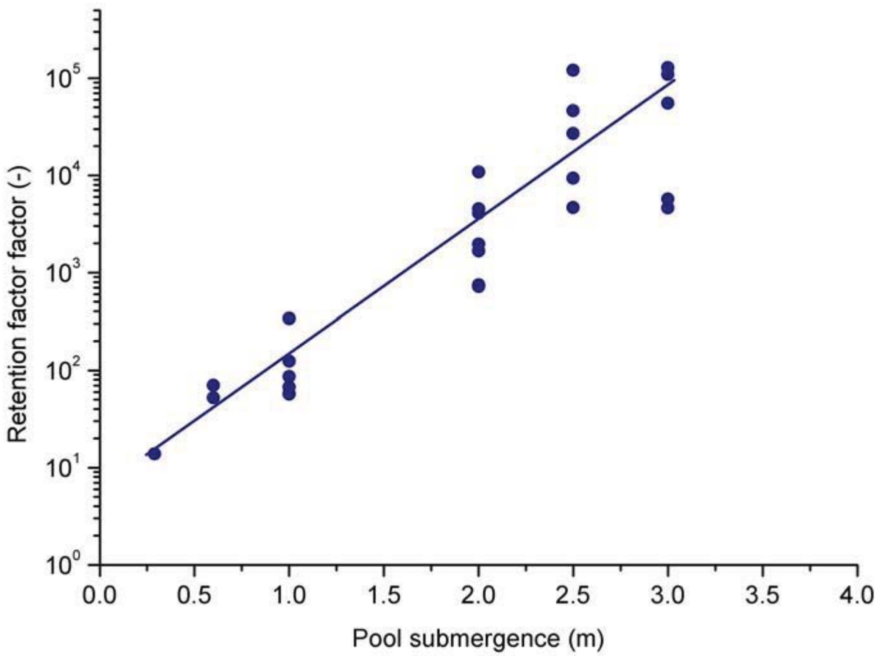
Steam/$N_2$, aerosol and iodine feed system

# R& D related to Iodine behavior in Wet Scrubber based FCVSs

# Iodine pool scrubbing: PSI POSEIDON I experiments (1989-1991)

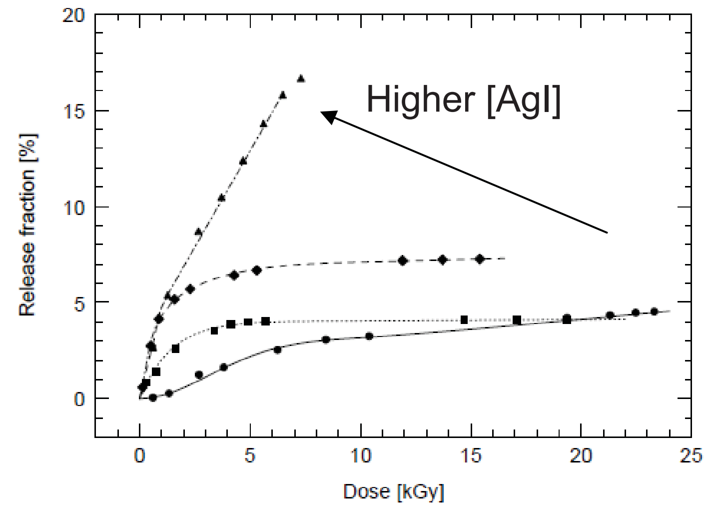
- Initial bubble size
- Water temperature

**$I_2$ scrubbing as a function of submergence**



# Radiation Iodine Chemistry R&D

- Radiolytic oxidation of Iodide ions
- Sudden release from sump close to dryness
- Radiation stability of colloidal silver iodide
- In-situ beta and external electron beam and gamma irradiation facilities
- Fast iodine species analysis Effect of sump impurities (NOx, Cl) on iodine volatility
- Effect of other sump impurities (Cd, Sn, U)
- Organic iodide hydrolysis and radiolysis
- Development of fast and efficient decomposition of organic iodide and fixation of iodine in aqueous solutions
- Containment iodine code IMPAIR3
- Iodine mechanistic code PSIODINE for aqueous iodine radiation chemistry




Release of iodine from decomposing AgI under in-situ $\beta$-irradiation at different initial AgI concentrations

# Development of Fast and Efficient Decomposition of Organic Iodide and Fixation of Iodine in Aqueous Solutions*

## Motivation for PSI research (2000-2008)

**L. F. Parsly, "Chemical and physical properties of methyl iodide and its occurrence under reactor accident conditions (A summary and annotated bibliography)", ORNL-NSIC-82, (1971).**

"For typical PWR conditions the half-time for methyl iodide removal by spray droplets doped with thiosulfate solution is estimated to be approximately 2 hr. Hence the dose reduction factor is insignificant. A reagent that will give substantially faster reduction is needed."

**C. Cau Dit Coumes, J. Chopin-Dumas, and F. Devisme, "Kinetics of the Reaction of Methyl Iodide with Hydroxylamine in an aqueous solution within the framework of nuclear spent fuel reprocessing", Ind. Eng. Chem. Res. 2001, 40, 3721-3731.**

**Iodide ions prone to revolatilize under Irradiation & prevailing conditions.**

***Patented (PSI/2006)**

# Steps for Efficient removal of gaseous iodine species from gas bubbles

a) Reduction of volatile elemental iodine and organic iodides to iodide by a reducing agent (thiosulfate):

$$I_2 + 2S_2O_3^{2-} \Rightarrow 2I^- + S_4O_6^{2-}$$

$$CH_3I + S_2O_3^{2-} \Rightarrow I^- + CH_3S_2O_3^-$$

<u>+ two concurrent reactions:</u>

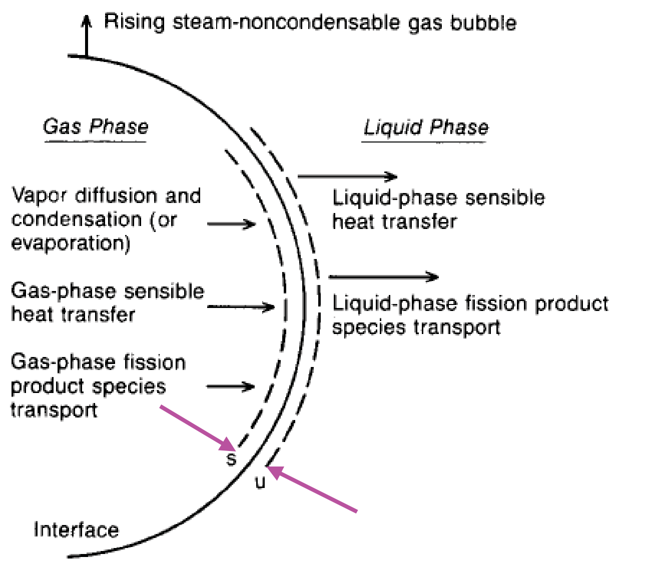
b) Catalysing the reduction reaction by use of a long-chain amin with a special structure (ALIQUAT 336®)

- Fast decomposition

c) Concurrent reduction of organic iodide:

$$(C_8H_{17})_3CH_3N{+}Cl^- + CH_3I \Rightarrow (C_8H_{17})_3CH_3N^+\, I^-$$

# Catalysing the reduction reaction by use of a special structure long-chain amin (Quaternary ammonium salt ALIQUAT336®)




Schematic diagram of heat and mass transfer phenomena in a rising steam-noncondensable gas bubble.

## Relative $CH_3I$ decomposition rates

| $CH_3I$ Decomposition process | 25 °C | 80 °C |
|---|---|---|
| Hydrolysis | 1 | 1’400 |
| Radiolysis + hydrolysis | 11’000 | 12’000 |
| Hydrolysis + additive alone | 15’000 | 290’000 |
| Hydrolysis + additive+ catalyst | >200’000 | >860’000 |
| Hydrolysis + radiolysis + additive+ catalyst | >210’000 | >870’000 |

Key transfer rate determining parameter: concentration gradient

# Sustained retention of iodine species in aqueous solution

Metallic iodides (CsI, RbI, etc)

- $I_2$

Organic iodides

Scrubbed in aqueous solution

$I^-$

Iodide ions
- Under irradiation
- Low pH
- Oxidation conditions

→ Volatile species ($I_2$)

Mass transfer from aqueous phase into gas phase (into gas bubbles/from water surface)

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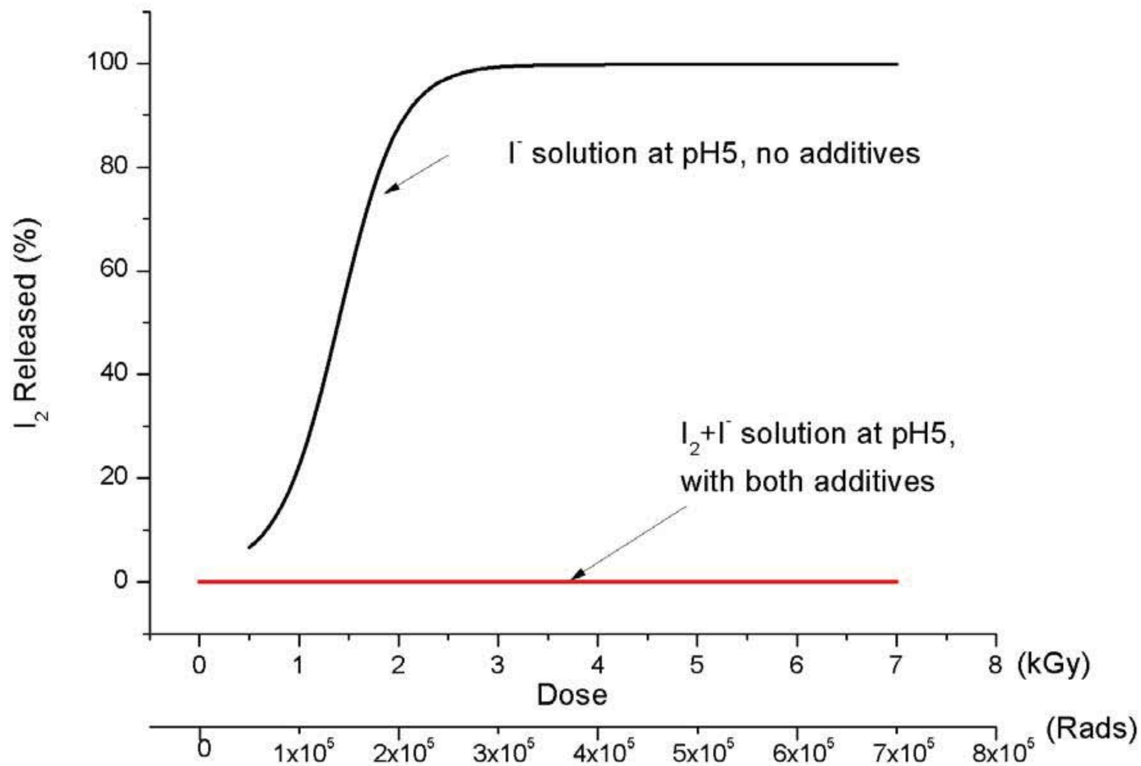
- ➢Iodine (gaseous/bound to particles) will be removed from gas flow: temporary retention
- ➢Aqueous solution will act a source of gaseous iodine afterwards if iodine not fixed

# Suppression of Iodine Release

- No evidence of Oxidation of iodide ions at pH5.
- Iodine remained bound to Aliquat336 under irradiation



# Conclusion

Large Experience and extensive data base and modeling of physics and chemistry

- Aerosol/droplet removal/transport
  - In complex geometries covering a large flow conditions
  - Pool scrubbing
  - Steam condensation
    - Prototypical accident conditions
- Thermal and radiation iodine chemistry
  - Data base and models
    - Prototypical accident conditions
- Laboratory infrastructure and experience in building new facilities

# Thank you for your attention