Pool Scrubbing Models for Iodine Components

Karsten Fischer[1]

Abstract

Pool scrubbing is an important mechanism to retain radioactive fission products from being carried into the containment atmosphere or into the secondary piping system. A number of models and computer codes has been developed to predict the retention of aerosols and fission product vapours that are released from the core and injected into water pools of BWR and PWR type reactors during severe accidents. Important codes in this field are BUSCA, SPARC and SUPRA. The present paper summarizes the models for pool scrubbing of gaseous Iodine components in these codes, discusses the experimental validation, and gives an assessment of the state of knowledge reached and the open questions which persist.

The retention of gaseous Iodine components is modelled by the various codes in a very heterogeneous manner. Differences show up in the chemical species considered, the treatment of mass transfer boundary layers on the gaseous and liquid sides, the gas-liquid interface geometry, calculation of equilibrium concentrations and numerical procedures.

Especially important is the determination of the pool water pH value. This value is affected by basic aerosols deposited in the water, e.g. Cesium and Rubidium compounds. A consistent model requires a mass balance of these compounds in the pool, thus effectively coupling the pool scrubbing phenomena of aerosols and gaseous Iodine species. Since the water pool conditions are also affected by drainage flow of condensate water from different regions in the containment, and desorption of dissolved gases on the pool surface is determined by the gas concentrations above the pool, some basic limitations of specialized pool scrubbing codes are given. The paper draws conclusions about the necessity of coupling between containment thermal-hydraulics and pool scrubbing models, and proposes ways of further simulation model development in order to improve source term predictions.

---

[1]) Battelle Ingenieurtechnik GmbH, D-65760 Eschborn, Germany

1. INTRODUCTION

In water reactor severe accident analysis, several scenarios are under consideration where a steam/gas flow is injected into a water pool, and radioactive fission products are washed out partly from the gas mixture by interacting with the water. The efficiency of this pool scrubbing process determines the contamination of the space above the pool surface, which could be part of the primary system loop, the containment building, or even the environment (for certain containment bypass scenarios); the classical case is pool scrubbing in a BWR pressure suppression chamber. Computer codes have been developed to estimate the pool decontamination factors, and they have been validated on the basis of experiments. Important codes in this field are BUSCA [RAMSDALE 1995], SPARC [OWCZARSKI 1991] and SUPRA [WASSEL 1991]. These codes put their main emphasis on models for aerosol interactions; pool scrubbing of gaseous Iodine appears to be treated only as a by-product.

2. GASEOUS IODINE SCRUBBING MODELS

2.1 Pool scrubbing hydrodynamics

Three different hydrodynamic zones can be distinguished when a steam/gas mixture is injected into a water pool:

- The injection zone.
This zone is characterized by the formation of large primary bubbles at the injection orifices; alternatively, under high injection velocities a submerged jet is formed. During bubble formation or jet expansion, heat and mass transfer between the injected gas and the water occurs. After some time the primary bubble detaches from the orifice and loses its velocity of motion relative to the water.

- The bubble rise zone.
After dissipation of the injection momentum, the gas rises to the pool surface in a bubble column or swarm. Large unstable primary bubbles decay into smaller ones. Heat and mass transfer lead to a thermodynamic equilibrium between gas and water. Bubbles are subject to a slight expansion due to the hydrostatic pressure decrease while the bubbles are rising. A recirculation flow of the pool water is introduced, which enhances the effective bubble rising velocity.

- The pool surface zone.
At the pool surface, the bubbles break up and release their gas to the space above the pool. Due to the breakup process, water droplets are generated and carried into the atmosphere. This entrainment of contaminated water droplets may reduce the effective retention capacity of the pool.

The interactions of the gaseous Iodine species (which are part of the injected gas/steam mixture) with the pool water are embedded in these hydrodynamic processes. Furthermore, interactions of these species with the aerosols exist. In general, Iodine will be present as a tracer in a gas which is predominantly insoluble, like Hydrogen or Nitrogen. The steam concentration is governed by the pool saturation conditions.

2.2 Iodine models in BUSCA

In BUSCA, the Iodine gas species I_2 and HI are considered; however, pool scrubbing is only modelled for I_2, and HI is treated as a non-soluble gas.

No special models are used for the injection zone.

In the bubble rise zone, removal of I_2 is calculated for a bubble of spherical cap shape, where diffusion coefficients D_G and D_L are determined for the gas and liquid sides of the upper curved surface, while for the flat base only the liquid side mass transfer is taken into account. A semi-analytical solution for the diffusion equations gives the decontamination factor for a bubble during the time interval δt as follows [CLOUGH 1984]:

$$DF_{I_2} = \frac{2\pi^{1/2}}{\lambda} (\delta t \ / \ \tau_D)^{3/2} \tag{1}$$

with

$$\tau_D = \frac{\pi^{1/2} \ \beta \ r_c^{5/4} \left(He + (D_L \ / \ D_G)^{1/2}\right)}{6 \ D_L^{1/2} \ g^{1/4} \ f(\varphi)} \tag{2}$$

$$\beta = 1 - \frac{3}{2} \cos \varphi + \frac{1}{2} \cos^3 \varphi \tag{3}$$

$$f(\varphi) = \left(\frac{2}{3} + \frac{1}{3} \cos \varphi\right)^{1/2} (1 - \cos \varphi) \tag{4}$$

$$\lambda = \frac{3}{2} \ \frac{\sin^2\varphi \ D_L^{1/2} \ \tau_D^{1/2}}{\beta \ r_c \ (He + (D_L \ / \ D_G)^{1/2})} \tag{5}$$

where r_c is the curvature radius of the upper bubble surface, and φ the bubble semi-angle. The Henry number He is proportional to the inverse of the partition coefficient; it is correlated as a function of the water temperature. For spherical bubbles, the calculation reduces to

$$DF_{I_2} = \exp \ (\delta t / \tau_D) \tag{6}$$

The following simplifications are involved in this treatment:

1. Diffusion boundary layer changes due to steam condensation or evaporation are neglected.
2. The diffusion coefficients are not correlated with the gas component concentrations.
3. The pool bulk concentration of dissolved I_2 is neglected.
4. The partition coefficient is a function of temperature only.
5. Surface renewal by the water circulation flow is neglected.

This means that the pool is treated as an I_2 sink with unlimited capacity, and neither chemical effects (pH) nor resuspension are taken into account.

The pool surface zone is not modelled.

2.3 Iodine models in SPARC

In SPARC, the Iodine gas species I_2 and CH_3I are taken into account.

The code has a model to estimate the deposition of I_2 on hygroscopic aerosols in the primary system before the injection into the pool. The primary system is modelled like a plug-flow reactor. Given the total volume V_p of the primary system ($V_p = 198\ m^3$ for the Peach Bottom reactor), and the volumetric flow rate $\dot{V}_{in}$ of the released steam/gas mixture, the residence time of gases and particles in the primary system is

$$t_{res} = V_p/\dot{V}_{in}. \tag{7}$$

Steam deposition on the hygroscopic aerosols is estimated by calculating the equilibrium droplet diameter from the atmospheric relative humidity, assuming that Cs and Rb components in the solid aerosol mass are hygroscopic like Na. After having determined the size of the wet aerosol droplets, diffusion of molecular Iodine to the droplets is calculated by the absorption coefficient

$$\alpha_i = n_i\ \pi\ D\ r_i\ t_{res} \tag{8}$$

In this relation, D is the diffusivity of I_2 in a steam-hydrogen atmosphere, n_i the number concentration of particle size class i, and r_i the particle radius. The decontamination factor DF_{I2} for I_2 in the primary system is

$$DF_{I2} = 1 / \sum e^{-\alpha i} \tag{9}$$

The weakness of this model is the fact that the droplets act as an infinite sink for I_2. Furthermore, even for particles without hygroscopic mass fraction or adsorbed water the same correlations are used to calculate I_2 removal.

In the injection zone, it is assumed that instantaneous thermal equilibrium is reached by steam condensation. The removal of I_2 due to this condensation is assumed proportional to the volume reduction of the injected flow. In contrast to this assumption, the aerosol decontamination factors are taken a factor 3 higher, which should account roughly for additional removal by diffusiophoresis. Removal of CH_3I is calculated assuming concentration equilibrium between gas and liquid phases, using a temperature dependent Henry number.

In the bubble rise zone, removal processes are calculated for I_2 and CH_3I by using gas and liquid side diffusion coefficients. Diffusive mass transport velocities u_D are calculated as

$$u_D = \sqrt{D / \pi\ t_x} \tag{10}$$

where D is the diffusivity of I_2 or CH_3I in the gas or liquid phase, and t_x is the time of surface exposition. The interfacial concentration c_i is calculated from the continuity of the diffusive mass flow as

$$c_i = (c_L u_{D,L} + c_G u_{D,G}) / (He\, u_{D,L} + u_{D,G}) \tag{11}$$

where $u_{D,L}$ and $u_{D,G}$ are the diffusive transport velocities at the liquid and gas side, respectively; c_L and c_G are the associated bulk molar concentrations, and He is the Henry number. The diffusion boundary layer changes due to steam condensation or evaporation are taken into account by corrections to the transport velocities. Also, the bubble surface renewal by water recirculation is taken into account by numerical integration over the bubble surface which has an oblate ellipsoidic shape. A mass balance of dissolved gases in the pool is calculated.

In order to estimate the partition coefficient or Henry number for I_2 at the bubble interface, the following fast chemical reactions in the water are considered:

$$I_{2,gas} \leftrightarrow I_{2,aq} \tag{12}$$

$$I_{2,aq} + I^- \leftrightarrow I_3^- \tag{13}$$

$$I_{2,aq} + H_2O \leftrightarrow H^+ + I^- + HIO \tag{14}$$

$$I_{2,aq} + H_2O \leftrightarrow H_2OI^+ + I^- \tag{15}$$

$$H_2O \leftrightarrow H^+ + OH^- \tag{16}$$

For each of these reactions, temperature dependent equilibrium constants are evaluated from the literature [EGGLETON 1967]. The presence of additional I^- and OH^- ions from dissolved aerosol material (CsI, CsOH, RbOH) is taken into account, using the pool mass balances of deposited aerosol components. Thus, the effects of temperature and pool pH are considered via the Henry number for I_2. In the case of CH_3I, a simplified treatment using a temperature dependent partition coefficient correlation is done.

In the <u>pool surface zone</u>, equilibrium conditions between the dissolved I_2 and the I_2 concentration of the gas leaving the pool are assumed, taking into account the Henry number He to determine the concentration ratio at the pool/gas interface:

$$c_G = c_L / He \tag{17}$$

where c_G and c_L are the molar I_2 concentrations in the gas and liquid phase, respectively. This formulation allows to simulate I_2 resuspension from the pool if the I_2 concentration in the injected gas is lower than the equilibrium concentration in the water.

2.4 Iodine models in SUPRA

Iodine gas species in SUPRA are I_2, CsI, and CH_3I.

The code does not simulate deposition of gaseous Iodine components in the injection zone.

In the bubble rise zone, models similar to BUSCA and SPARC are applied. Diffusive mass transfer coefficients are calculated for the gas and liquid sides of the interface, taking into account the boundary layer changes due to steam condensation or evaporation. Surface renewal by water circulation is not modelled. The bubble shape is approximated as an oblate ellipsoid. The Henry numbers describing the concentration jump at the bubble surface are correlated as follows:

For I_2, He is determined from a tabular interpolation with respect to the concentration of dissolved I_2 at the interface. Then, the influences of temperature and pool pH are accounted for by appropriate correlation factors. The pH used has to be specified in the code input.

For CH_3I, He is determined from a temperature-dependent correlation.

For CsI, He is set to a small constant value, which leads to a gas side controlled deposition rate.

Heat and mass transfer at the pool surface are influenced by the thermal hydraulic conditions of the pool and the gas space above the pool. In SUPRA, evaporation of steam as well as desorption of tracer gases are calculated using Nusselt number correlations for natural convection at horizontal surfaces [EDWARDS 1979]:

$$\mathrm{Nu}_M = 0.12\ (\mathrm{Gr\ Sc})^{1/3} \tag{18}$$

where Gr is the Grashof number

$$Gr = \frac{\Delta\rho}{\rho_G}\ g\ L^3/\nu_G \tag{19}$$

and L is a characteristic length of the pool surface; $\Delta\rho$ is the density difference between the bulk gas and the gas at the pool surface, driving the natural convection flow in the gas space; g is the gravitational acceleration, ρ_G the gas density, and ν_G the gas viscosity. On the liquid side, mass transfer coefficients are approximated by [DAVIES 1979]

$$K_L = D_L^{1/2}\ \rho_L \tag{20}$$

where D_L is the diffusivity of the dissolved tracer gas, given in m^2/s; ρ_L in kg/m^3, and K_L in kg/m^2s. The mass transfer coefficients are corrected for steam condensation, and Henry number correlations are supplied for the species concentration ratio at the pool surface. The species concentrations in the gas space above the pool are specified in the code input data.

In SUPRA, mass balances for the substances dissolved in the pool are calculated. However, this information is not used to determine the pool pH value.

3. MODEL VALIDATIONS

The BUSCA models for Iodine scrubbing were validated by post-calculations of two I_2 tests by Diffey et al. [DIFFEY 1965]; this work was reported in [LÓPEZ-JIMÉNEZ 1995].

SPARC validation on the Diffey tests for I_2 and CH_3I is mentioned in the SPARC-90 manual [OWCZARSKI 1991]. The same data set was post-calculated in [LÓPEZ-JIMÉNEZ 1995].

Also SUPRA was validated in comparison with the Diffey data on CH_3I.

More recent experimental data, partly taken in larger scaled facilities, are existing [LÓPEZ-JIMÉNEZ 1995], but no published code validations based on such data were found. Obviously, the present validation covers only a small fraction of the relevant parameter ranges.

4. DISCUSSION

Table 1 gives an overview of the species and zones considered in the codes.

Table 1. Species and Zones Considered in the Codes

Species	BUSCA	SPARC	SUPRA
I_2	Bubble rise zone	Primary system Injection zone Bubble rise zone Pool surface zone	Bubble rise zone Pool surface zone
CH_3I		Injection zone Bubble rise zone	Bubble rise zone Pool surface zone
CsI_{gas}			Bubble rise zone Pool surface zone
HI	Non-soluble		

It is evident that the only common feature for all codes is the treatment of I_2 in the bubble rise zone. Other species or zones are considered less important and treated differently. The models for the bubble rise zone are similar in all codes: diffusive transport on the liquid and gas sides, coupled by a Henry number at the interface. It is worth mentioning that the models for the pool surface zone differ between SPARC and SUPRA. While SPARC simulates concentration equilibrium between released gas and water, SUPRA simulates equilibrium between the atmosphere above the pool and the water, where the atmospheric concentration has to be specified. While SPARC has the possibility to model resuspension of dissolved I_2, SUPRA can calculate gas absorption by the pool surface.

Differences show up in the following submodels:
- model for diffusive mass transfer in the gas or liquid side boundary layer,
- calculation of species diffusivities,
- consideration of steam condensation-induced corrections to species diffusion,
- shape of bubbles,
- consideration of bubble surface water renewal,
- calculation of Henry numbers.

The Henry number is of central importance for the I_2 deposition. Table 2 shows the different formulations.

Table 2. Henry Number Correlations Used in the Codes

BUSCA	SPARC	SUPRA
Function of temperature	Temperature-dependent equilibria of: $I_{2,gas} \leftrightarrow I_{2,aq}$ $I_{2,aq} + I^- \leftrightarrow I_3^-$ $I_{2,aq} + H_2O \leftrightarrow H^+ + I^- + HIO$ $I_{2,aq} + H_2O \leftrightarrow H_2OI^+ + I^-$ $H_2O \leftrightarrow H^+ + OH^-$ OH^- calculated from dissolved basic components	F(c) · F(t) · F(pH) F(c) Function of I_2 concentration F(t) Function of temperature F(pH) Function of pH pH specified in input

In comparison to more elaborate models for Iodine behaviour in a LWR containment, like IMPAIR [GÜNTAY 1992], the simulation of Iodine component interactions appears to be strongly simplified even in SPARC. While IMPAIR simulates dynamic rate reactions, the pool scrubbing codes assume chemical equilibrium. IMPAIR considers the following reactions in the pool:

$$I_{2,gas} \leftrightarrow I_{2,aq} \qquad (12)$$

$$I_{2,aq} + H_2O \leftrightarrow H^+ + I^- + HIO \qquad (14)$$

$$3\ HOI \leftrightarrow IO_3^- + 2\ I^- + 3\ H^+ \qquad (21)$$

$$H_2O \leftrightarrow H^+ + OH^- \qquad (16)$$

$$2\ I^- + \tfrac{1}{2}\ O_2 + 2\ H^+ \rightarrow I_{2,aq} + H_2O \qquad (22)$$

$$I_{2,aq} + 2\ Ag \leftrightarrow 2\ AgI \qquad (23)$$

Equations 12, 14 and 16 are the same in SPARC and IMPAIR. The reactions involving I_3^- and H_2OI^+ are used in SPARC only, while the reactions with IO_3^-, O_2 and Ag are used in IMPAIR only. IMPAIR takes into account many other rections, like gas space interactions, radiolysis, and deposition on walls, which should be important for the pool scrubbing processes also. Unlike SPARC, IMPAIR does not simulate the dependency of pH on the dissolved Cs and Rb mass, instead the pH is a specified input value.

It is generally accepted that the pool pH is of great importance for the Iodine distribution. This entity is influenced by the concentrations of boric acid and basic aerosol components (Cs, Rb) as well as water radiolysis. The concentrations are related to the thermal-hydraulics and transport processes in the containment or primary system, which govern the distribution of steam, water, condensate flows, suspended and deposited aerosols. The radiolysis is determined by the radiation levels of the fission product distribution. A tight feedback between I_2 partitioning, I_2-generated radiation and pH is expected.

Obviously the operation of specialized, limited-purpose codes like BUSCA, SPARC, SUPRA or IMPAIR is not satisfactory because important parameters have to be specified as user input. A consistent model should take care of all important interactions. Such a model would preferably be based on a thermal-hydraulic system code, which can simulate all transport processes and associated mass balances. This code would then form a suitable framework for special models like pool scrubbing, giving all necessary boundary conditions and parameters that would otherwise require user input when running a self-standing specialized code. A first step in this direction was done with the development of FIPLOC 3.0 [WEBER 1996], where a thermal-hydraulic containment code was coupled with an aerosol module, a pool scrubbing code (SPARC) and an Iodine behaviour code (IMPAIR). This code combines and extends the capabilities of its constituting code elements, and it opens up the possibility of more advanced, mechanistic and consistent predictions for severe accident transients. Some of the submodels, like the primary system I_2 interaction in SPARC, or the pool surface-atmosphere interaction in SUPRA, are more consistently handled in such a system code framework. However, there is still a number of questions to be resolved. As shown above, the validation of the Iodine pool scrubbing models is based on a single series of experiments (Diffey), and the validated parameter range is small. Further work appears necessary in the following fields:

- Extension of Iodine scrubbing models to cover all relevant chemical species and reactions,
- Extension to include scrubbing in the injection zone,
- Extension to include resuspension in the pool surface zone,
- Consideration of chemical kinetics where necessary,
- Integration with IMPAIR-like pool models for deposition on submerged surfaces,
- Addition of decay heat release model,
- Extension of the code validation using more recent available data.

5. REFERENCES

[CLOUGH 1984] Clough P.N; Brighton P.W.M. and Ramsdale S.A; Decontamination factors for gas mixtures in spherical cap bubbles. Report SRD R 311, 1984

[DAVIES 1979] Davies J.T. and Lozano F.J; Turbulent Characteristics and Mass Transfer at Air-Water Surfaces. *A.I.Ch.E. J.* 1979, **26**, 405-415

[DIFFEY 1965] Diffey H.R; Rumary C.H; Smith M.J.S. and Stinchcombe R.A; Iodine Clean-Up in a Steam Suppression System. Atomic Energy Research Establishment Report AERE-R-4882, 1965

[EDWARDS 1979] Edwards D.K; Denny V.E. and Mills A.F; Transfer Processes. McGraw-Hill 2nd ed., 159-161, 1979

[EGGLETON 1967] Eggleton A.E.J; A Theoretical Examination of Iodine-Water Partition Coefficients. Report AERE-R 4887, Harwell 1967

[GÜNTAY 1992] Güntay S. and Cripps R; IMPAIR/3: A Computer Program to Analyse the Iodine Behaviour in Multi-Compartments of a LWR Containment. PSI-Bericht 128, September 1992

[LÓPEZ-JIMÉNEZ 1995] López-Jiménez J; Herranz L.E; Escudero M.J; Espigares M.M; Peyrés V; Polo J; Kortz Ch; Koch M.K; Brockmeier U; Unger H; Dutton L.M.C; Smedley Ch; Trow W; Jones A.V; Bonnani E; Calvo M. and Alonso A; Pool Scrubbing. CEC Report ST(95)-P195, CIEMAT Report ITN/TS-12/SP-95, Madrid, June 1995

[OWCZARSKI 1991] Owczarski P.C. and Burk K.W; SPARC-90: A Code for Calculating Fission Product Capture in Suppression Pools. Report NUREG/CR-5765, October 1991

[RAMSDALE 1995] Ramsdale S.A; Güntay S. and Friederichs H.-G; BUSCA-JUN91 Reference Manual for the Calculation of Radionuclide Scrubbing in Water Pools. Report GRS-116, ISBN 3-923875-66-5, Köln 1995

[WASSEL 1991] Wassel A.T; Hoseyni M.S; Farr Jr. J.L.; Bugby D.C. and Mills A.F; SUPRA: Suppression Pool Retention Analysis Computer Code. Report EPRI NP-7518-CCML, September 1991

[WEBER 1996] Weber G; Schwarz S; Ewig F and Fischer K; Benutzerhandbuch für FIPLOC 3.0. GRS Report, to be published, March 1996

6. ACKNOWLEDGEMENTS

This work was sponsored by the German Federal Minister of Research and Technology, project 1500 920A.