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# AQ1 Experimental Investigation on the AQ2<sub>1</sub> Retention of Soluble Particles by Pool Scrubbing

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In the late stage of a severe loss-of-coolant accident, the pressure in the containment building of a nuclear power plant could rise 28 beyond the design limits and thus endanger its structural integrity. Therefore, to avoid pressure failure, it may be necessary to per-30 form controlled venting of the containment. During the event of 31 an accident, a large amount of fission and activation products are 32 released into the containment as airborne particles (aerosols). 33 These particles are filtered during the venting process, usually 34 with the help of wet filters, in order to keep risks to the surround-35 ing environment to a minimum. Consequently, the knowledge of 36 the retention processes in a water reservoir (pool scrubbing) is of 37 central importance for such filtered containment venting systems 38 and for reactor concepts in which water reservoirs are used for 39 pressure reduction (e.g., condensation chamber of a boiling water reactor (BWR)). Investigations on pool scrubbing are carried out 41 in the severe accident aerosol behavior (SAAB) test facility at the Juelich Research Center in the framework of a national project 43 ("severe accident aerosol behavior-II"). SAAB is a unique largescale facility with the ability to perform a great variation of 45 experiments using various measurement tools. The influence of 46 numerous parameters, such as the height of the water pool, solu-47 bility of aerosols and concentration on the retention capacity, is 48 investigated by means of separate effect studies on both insoluble 49 and soluble particles. This technical brief gives a detailed over-50 view over the facility and includes example results of the first test series with soluble particles including cesium iodine (CsI). [DOI: 10.1115/1.4051250]

Keywords: severe accident, source term, aerosols, experimental research, pool scrubbing

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### 1 Introduction

In case of an at least temporarily uncontrolled loss-of-coolant accident in a water-cooled nuclear reactor, the reactor core can be partially exposed and destroyed. This leads to the release of radioactive fission products (aerosols and noble gases) first into the reactor cooling circuit and then into the containment atmosphere. In addition to the fission products and hydrogen, a large amount of steam also enters the containment, which leads to a pressure increase. In such an accident, there is a possibility that the pressure will reach the design limit of the containment and thus endanger its structural integrity. A failure of the containment should be avoided in any case because this would lead to an uncontrolled release of radioactive material into the environment. To prevent this, the containment should be depressurized before critical pressure limits are reached. Filtered containment venting systems are installed in numerous European nuclear power plants to reduce the pressure inside the containment with minimum release of radioactive fission products. One filter stage of this system usually consists of a wet filter, where airborne particles flow through a water seal. Therefore, a better understanding of the phenomenology of pool scrubbing is necessary in order to be able to make reliable assertions about the radioactive release into the environment. The condensation chamber of a boiling water reactor (BWR) not only serves to reduce pressure but also, in the sense of pool scrubbing, to retain fission products. In addition, water accumulations with small depths that have arisen during the course of an accident can also contribute to particle retention. [1,2]

Experimental studies in the past almost exclusively used insoluble particles [3]. Most of the pool scrubbing investigation focused on experiments with the insoluble substance SnO2 which is revealed in the passive and active systems on severe accident source term mitigation project [4]. Although it has been disclosed in the state-of-the-art review of fission product aerosol that the bulk of the aerosol released is likely to be soluble particles such as CsI and CsOH [5]. One of the most common integral test facilities in Europe "THAI" focused with their pool scrubbing investigations also on SnO<sub>2</sub> [6]. Only the Poseidon Project at the Paul-Scherrer-Institute in Switzerland focused besides SnO2 also on iodine [7]. The first results of the investigation on the retention of soluble particles presented briefly in the following, generating data for model development or validation, is a part of future work to allow for robust simulations of pool scrubbing related phenomena.

### 2 Pool Scrubbing Phenomenon

Aerosol retention through pool scrubbing is expressed in terms of a decontamination factor (**DF**), which is defined as the ratio of aerosol mass flow rate at the inlet to that at the outlet.

$$\mathbf{DF} = \frac{\dot{m}_{\rm in}}{\dot{m}_{\rm out}}$$

The path of the aerosols through the pool height may be split 100 into three regions: injection, bubble rise, and pool surface and 102 subsequently the overall DF is a multiplication of individual DFs and defined as in [1] 104

$$DF = DF_{inj} \times DF_{rise} \times DF_{sur}$$

2.1 Injection Zone. When the carrier gases along with the aerosol enter the pool at the orifice, the aerosols due to their inertia, impact on the liquid surface and are trapped, i.e., aerosol 108 retention due to jet impaction. Gas injection velocity and the orifice diameter determine the flow regime. For this purpose, a non-dimensional Weber number is defined as [1]

$$\mathbf{We} = \frac{\rho_l \times D_{\text{inj}} \times u_{\text{inj}}^2}{\sigma}$$

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Stage:

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113 If We < 105, the flow is in the globule regime, and if 114 We > 105, the flow is in the jet regime. For the experiments pre-115 sented in this technical brief (We = 9.104), we restrict the discussion to only the globule regime. As the carrier gas enters the water 117 pool through the orifice, an initial globule is formed. This globule, 118 too large to be stable, breaks into smaller bubbles, and they rise 119 like a swarm through the water pool. For the globular flow regime, 120 aerosol size and density are the main contributors to the decon-121 tamination factor in the injection zone.

122 **2.2 Bubble Rise Zone.** In this region, the dynamics of aero-123 sols trapped in the small rising bubbles is the primary contributor 124 to the decontamination factor

$$\mathbf{DF_{rise}} = \exp\left(\left(\frac{1}{V} \times \sum_{n} \int_{A} v_{n}(r) \times dA\right) \times \frac{H}{u_{rise}}\right)$$

The key factors are the residence time of the bubbles  $(H/u_{rise})$ and the relative motion of aerosols within the bubbles due to the following mechanisms: Diffusiophoresis, Thermophoresis, Sedimentation, Centrifugal impaction, and Brownian Diffusion. Each of the phenomena results in a relative velocity of aerosols with the bubble, causing the aerosols to drift toward the bubble interface (gas-liquid interface), where they are captured. Condensation/ evaporation at the interface causes gradients in steam concentration and temperature, promoting aerosol transport due to diffusiophoresis and thermophoresis, respectively. Sedimentation accounts for the relative motion of aerosols due to gravitational acceleration, i.e., the heavier the particle, the better the retention. Centrifugal force on the aerosol is a result of the bubble's rotating motion. Bubble's rotating motions causes centrifugal forces on the aerosols inside, promoting migration toward the interface. Brownian diffusion results in aerosol transport from regions of high concentration to low concentration and is important for small particles ( $d < 0.1 \,\mu\text{m}$ ) [1,8].

Also, the particle size distribution within the rising bubbles impacts the gravitational settling, centrifugal impaction, and Brownian diffusion. Particle size distribution varies because of agglomeration and steam condensation on hygroscopic aerosols

2.3 Pool Surface Zone. The bubbles rise to the surface of the water pool and rupture, producing microdroplets. The gas flow entrains some of these droplets, and others fall back due to gravity. Entrained droplets, in turn, transport fine aerosol particles [1].

To summarize, the factors affecting aerosol retention in a water pool are the inlet gas velocity, gas composition (steam concentration), gas temperature, particle Stokes number (aerosol density and size), hygroscopicity, or solubility of the aerosol material and pool height. In this work, the gas velocity, temperature, and composition (no steam) are kept constant. Only the impact of particle density and size distribution, solubility, and pool height on the pool scrubbing phenomenon is studied.

### 3 Facility Description 161

The severe accident aerosol behavior (SAAB) test facility (Fig. 1) consists of three parts—aerosol generation, aerosol conditioning, and tank with water reservoir. First, the aerosols are produced in the aerosol generating unit and then fed into the mixing chamber. In the mixing chamber, different aerosol streams are combined and further mixed with the carrier gas. The well-mixed aerosol is then led into the water reservoir, where aerosol particles are filtered. The properties of the aerosol are measured at the inlet and outlet of the water seal by sampling with measuring instruments, such as an electric low-pressure impactor (ELPI), an aerodynamic particle sizer or the scanning mobility particle sizer [3].

The tank in Fig. 1 is the heart of the test facility and contains the water seal. The tank itself is of modular design and consists of

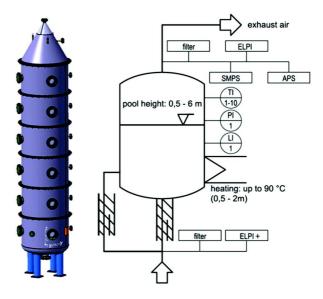


Fig. 1 SAAB test facility [3]

up to five separate segments, each segment measuring 1 m in 175 height and 1.5 m in diameter. The aerosol flows into the lowest 176 segment, through the tank, and out at the uppermost segment. It 177 offers the possibility of varying the height of the water seal from a 178 minimum of 0.5 m to a maximum of 5.5 m above the feed under 179 identical sampling conditions. The maximum possible volume is 180 10 m<sup>3</sup>. The top part of the test facility is conical and has provisions for the extraction of the filtered aerosol into an air filter system and a sampling line for measurements at the outlet. The right 183 part of Fig. 1 shows the instrumentation of the vessel. The aerosol 184 is directed into the SAAB container through an opening (1 in. diameter for the experiments presented here) located in the middle 186 of the bottom segment. Additionally, it is possible to feed the aerosol downward or sideways to the container. Each segment, 188 except the top one, has four flanged openings for instrumentation or optical access. The container and water reservoir can be heated 190 to a temperature of 90 °C by feeding steam. It is also equipped 191 with a trace heating system for both the lower and the upper segments. As also shown in Fig. 1, a water-level sensor (see Fig. 1 longitude indicator (LI)), a pressure sensor ("PI"), and a humidity 194 sensor ("MI") with integrated temperature sensor are installed in 195 the upper, conical segment. In addition, a sample is taken at a height of about 0.65 m above the water surface.

### 4 Test Description and Execution

Cesium iodide (CsI) and cesium hydroxide (CsOH) are among 199 aerosols present in the containment atmosphere during a late stage 200 of a severe accident [1]. CsI (natural isotop) with a solubility of 669.7 g/l can only be used in the experiments after a number of precautions have been taken. CsOH is highly alkaline (pH = 14 at 203 500 g/l) and it is therefore extremely difficult to be utilize in 204 experiments. Due to these limitations, substitutes were selected for the first series of experiments. Instead of CsI, sodium chloride 206 (NaCl) with a water solubility of 358 g/l was chosen. Instead of 207 CsOH, potassium acetate ( $C_2H_3KO_2$ ) a water solubility of 2560 g/  $^{208}$ l was chosen [9]. After conducting the tests with NaCl, the facility 209 was equipped with additional safety precautions to allow for the 210 safe testing of CsI.

For the generation of soluble aerosols, a two fluid spraying sys- 212 tem was used. The water-salt solution of the aerosol is mixed with 213 N2 gas in the nozzle. The gas at the inlet to the nozzle system is 214 under a higher pressure (5–10 bar). When it expands through the 215 nozzle to a lower pressure (~1 bar), the gas accelerates. The 216 impulse transfer between gas and liquid breaks the liquid into fine 217

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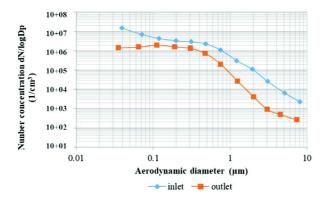


Fig. 2 Particle size class number concentration of NaCl aerosol measured at the inlet and outlet for a flow rate of 250 g/h and a pool height of 1.5 m

droplets. During the subsequent evaporation of the liquid droplets by means of a heating system, the particles crystallize and are then mixed with the carrier gas stream (N2) to produce the desired

In addition to the spraying system for soluble particles, the SAAB facility also uses a particle disperser with brush, which produces insoluble particles, such as SnO2. A detailed description can be found in the SAAB final project report [3].

The parameters of the experimental test matrix are:

- solubility (NaCl, C<sub>2</sub>H<sub>3</sub>KO<sub>2</sub>, CsI)
- aerosol concentration (NaCl, C2H3KO2, CsI)
- pool height (NaCl, C<sub>2</sub>H<sub>3</sub>KO<sub>2</sub>, CsI)
- injection speed (CsI, NaCl)

The height of the water above the aerosol inlet is referred to here as the water level or pool height. The water heights considered in the experiments are 1.5 m and 5.5 m for NaCl and 1.5 m to 5.5 m for CsI. The aerosol concentration in the carrier gas is varied by changing the mass flow of the liquid solution into the two fluid spraying system (150-300 g/h). The N2 carrier gas volume flow is kept constant in all experiments (20 m<sup>3</sup>/h). The gas temperature before the inlet is 30 °C, the water temperature averages 22 °C, and the sampling temperature is 60 °C to keep the relative humidity at a similar level as at the inlet. All tests are repeated at least two times to ensure the reproducibility of results.

Table 1 Uncertainties of the measured retention efficiencies

di (µm)	0.1	0.2	0.3	0.5	0.8	1.2	2.0	3.1	5.1
$\begin{array}{l} -\bar{\eta} \\ +\bar{\eta} \end{array}$							0.2 % 0.2 %		

### Results

About 100 experiments were performed at the SAAB facility to 239 study the aerosol retention capability of pool scrubbing for multi- 240 ple combinations of test parameters—aerosol material (solubility), aerosol concentration (injection rate), and pool height. Presenting 242 the results of all the experiments is outside the scope of this tech- 243 nical brief; therefore, we restrict the discussion on retention efficiency of aerosols only for the following cases:

NaCl aerosol for four concentrations (mass flow rate of the liquid solution: 150 g/h, 200 g/h, 250 g/h, and 300 g/h), at two pool heights (1.5 m and 5.5 m).

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- CsI at 250 g/h for five pool heights—1.5 m, 2.5 m, 3.5 m, 4.5 m, and 5.5 m.
- Soluble aerosols (NaCl, C<sub>2</sub>H<sub>3</sub>KO<sub>2</sub>, and CsI) and Insoluble aerosol (SnO<sub>2</sub>) at 250 g/h and for a pool height of 1.5 m.

Figure 2 shows an example measurement output of the ELPIs. 250 It shows the number concentrations for a basin height of 1.5 m 251 before (blue) entry into and after (orange) exit from the water reservoir. Each measuring point is assigned to a certain size class. The number of concentration describes the quantity of particles of 254 a certain size class per cm<sup>3</sup> counted during the experiment. With 255 the help of this data, conclusions can ultimately be drawn about the retention efficiency described in more detail below.

If  $\mathbf{DF} = 1$ , no retention has taken place. In the case of especially 258 large DF values where retention is mainly the result of large particles, the figures are not very clear. More illustrative is the so- 260 called retention efficiency  $(\eta)$ , which is linked to **DF** as

$$\eta = 1 - \frac{1}{\mathbf{DF}}$$

Therefore,  $\eta$  is preferred in this document for the representation 263 of the retention. The equation of the retention efficiency can thus 264 also be set as a function of the mass flow or the measured mass 265 concentration cm. This leads to the following equation: 266

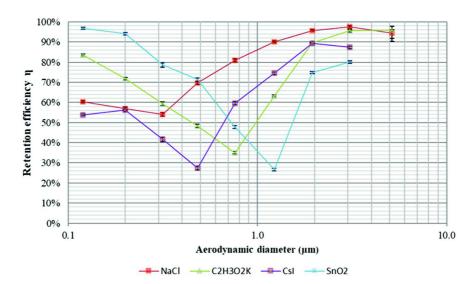


Fig. 3 Particle size class retention efficiency for soluble (NaCl, CH3CO2K, Csl) and insoluble aerosols (SnO2) for an injection rate of 250 g/h and pool height of 1.5 m

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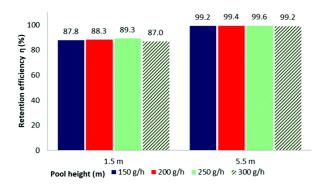


Fig. 4 Impact of aerosol concentration (injection rate) on retention efficiency for NaCl aerosol at pool heights of 1.5 m and 5.5 m

$$\eta = 1 - \frac{c_{m \text{ out}}}{c_{m \text{ in}}}$$

268 The used measurement device ELPI measures and calculates the mass concentration for each diameter as follows [10]:

$$c_{m,\text{elpi},i} = \frac{d_i^3 \times \pi \times \gamma \times \rho}{X_i \times 6}$$

270 After 100 and more experiments have been carried out on the 272 SAAB system, it is finally possible to calculate and predict the 273 measurement uncertainty of the SAAB facility. For this purpose, 274 the model of the Gaussian error propagation law [11] was used and applied in the following equation:

$$s_{n,i}^2 = \frac{\partial c_{\mathrm{elpi},i}}{\partial d_{\mathrm{stk},i}} \times s_{\mathrm{dstk},i}^2 + \frac{\partial c_{\mathrm{elpi},i}}{\partial \gamma} \times s_{\gamma}^2$$

278 These preparations finally lead to the following equation to 278 determine the uncertainty as:

$$\eta = 1 - \frac{c_{m \text{ out},i} \pm s_{n \text{ out},i}}{c_{m \text{ in},i} \pm s_{n \text{ in},i}}$$

289 If the largest and smallest quotients of the equation are used, 281 the raw values of the uncertainty are obtained. Because particles

with a larger diameter have more mass, their uncertainty is 282 greater. However, only measured values up to max. 5 µm are 283 included in the following diagrams. The calculated uncertainties 284 of the measured values are shown as an example in Fig. 3. Table 1 285 displays the uncertainties in positive and negative directions for 286 diameters, which could be measured with the ELPI.

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Figure 4 shows the integral retention efficiencies over the feed 288 concentration for sodium chloride. The concentration variations 289 are shown both for a water column height of 1.5 m and for a 290 height of 5.5 m. The different concentrations differ in color. Based 291 on these results, a concentration variation due to a change in the 292 feed mass flows of 150 g/h up to a concentration of 300 g/h leads 293 to almost identical results. For a height of 5.5 m, the retention efficiency for all concentrations is approx. 99.4% ±0.2%. For a height 295 of 1.5 m, the results of the retention efficiency are around 296  $88\% \pm 1\%$ .

Figure 5 shows the preliminary results of the first experiments 298 with CsI. The retention efficiency for each size class is shown for 299 a water-level height of 1.5 m-5.5 m. The influence of height on 300 the retention efficiency is clearly recognizable, especially a substantial increase in filtration efficiency between pool heights of 302 1.5 m and 2.5 m for all particle sizes; for the smaller particles 303  $(0.1-1.0 \,\mu\text{m})$ , a significant rise in retention efficiency between 304 pool heights of 4.5 m and 5.5 m (Fig. 5).

A comparison of different substances tested in the SAAB facil- 306 ity is shown in Fig. 3. The retention efficiency  $\eta$  for a pool height 307 of 1.5 m is graphed over the particle diameter. The height of 1.5 m 308 was chosen because the greatest difference in retention can be recognized at this level. Similar size distribution of aerosol feed is 310 maintained to ensure the comparability of retention efficiency for 311 different soluble materials. The blue curve shows the course of the 312 size classes of the retention efficiency for NaCl, the red curve for 313 C<sub>2</sub>H<sub>3</sub>KO<sub>2</sub>, the green curve for CsI, and the purple curve for the 314 insoluble SnO2 for comparison. All four substances show a 315 substance-specific minimum in the filter efficiency (so-called filter 316 gap). For sodium chloride, the minimum is 54% at a particle diameter of about 0.3  $\mu$ m, for potassium acetate 35% at about 0.8  $\mu$ m, for 318 cesium iodine 27% at 0.5  $\mu$ m, and finally for tin dioxide 26% at a 319 particle diameter of about 1.2  $\mu$ m. Contrary to our expectations, the 320 substance with the highest solubility was not retained the best.

Another unexpected result is that the insoluble SnO2 exhibits 322 the best retention for particles sizes lower than  $0.5 \,\mu\text{m}$ . With 323decreasing the density, the retention efficiency increases for all 324 substances, except for potassium acetate. When considering 325

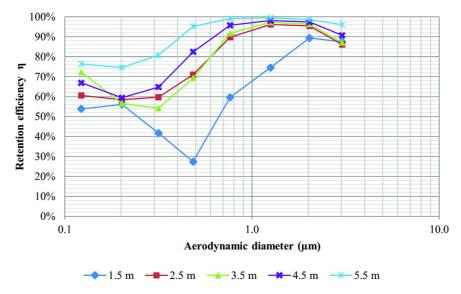


Fig. 5 Particle size class retention efficiency for CsI aerosol at 250 g/h for different pool heights—1.5 m, 2.5 m, 3.5 m, 4.5 m, and 5.5 m

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	diameters above $0.7 \mu\text{m}$ , NaCl $(2.16 \text{g/cm}^3  [9])$ is retained the
	best, followed by CsI (4.51 g/cm <sup>3</sup> [9]). The material with the high-
328	est density, SnO <sub>2</sub> (6.95 g/cm <sup>3</sup> [9]), shows the worst performance
329	of the three from this size class up.

### **Summary and Conclusion** 330 6

The investigations were carried out at the SAAB test facility in Research Center Juelich, to understand the impact of aerosol material solubility on pool scrubbing retention efficiency. For this purpose, three soluble materials—NaCl, C<sub>2</sub>H<sub>3</sub>KO<sub>2</sub>, and CsI—and one insoluble substance—SnO<sub>2</sub>—were used. In addition to the material solubility, the parameters investigated were the pool height (1.5 m-5.5 m) and aerosol mass concentration at the inlet orifice (150 g/h-300 g/h). Integral as well as particle sizewise  $(dp \sim 0.04 \,\mu\text{m} - 8 \,\mu\text{m})$  retention efficiencies are presented. The estimation of measurement uncertainties is still a work in progress

For NaCl aerosol, the retention efficiency remained independent of the inlet mass concentration, contrary to the expected behavior of enhanced retention due to a higher number of particle-bubble surface interactions.

For the same mass concentration and pool height, NaCl, having a comparatively lower density than CsI (a higher number of particles), was retained better in the water pool. Between NaCl and CsI, Brownian diffusion due to particle number concentration overwhelms gravitational settling and Centrifugal impaction proportional to particle density.

For insoluble and soluble aerosols, the highest retention occurs within the first 1.5 m of pool height, confirming that the jet impaction at the orifice is the dominant phenomenon. Despite our expectations, solubility was not a key factor in pool scrubbing. Rather, density and, therefore, number concentration seem to be of more importance. For NaCl and CsI aerosols, with increasing pool heights, the residence time of the aerosol in water increases, and consequently, the retention efficiency increases.

The results from this work indicate that an integral DF (based on mass) does not reveal an accurate picture of aerosol retention; the larger, heavier particles are best filtered, thereby masking the poor retention of small, respirable aerosols.

Future work will focus on studying the retention efficiency of pool scrubbing methods for mixed component aerosols and different gas compositions and temperatures. Furthermore, investigations on bubble behavior in SAAB are planned to gain deeper insights into the pool scrubbing phenomenon.

### AO7 **Funding Data**

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### Nomenclature

373 374  $c = \text{concentration, kg/m}^3$ 375 D/d = diameter, m376 H = height, m $\dot{m} = \text{mass flow, kg/s}$ 377 r = radius, m378 s =standard deviation/uncertainty 379 u = velocity, m/s380 v = depletion mechanism velocity, m/s

X = measurement accuracy

V = volume, m

### **Greek Symbols**

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$\gamma = \text{dilution factor}$	384
$\eta = \text{retention efficiency}; \left(1 - \frac{1}{\text{DF}} = 1 - \frac{c_{m \text{ out}}}{c_{m \text{ in}}}\right)$	385
$\rho = \text{density, kg/m}^3$ DF $c_{m \text{ in}}$	386
$\sigma = \text{surface tension N/m}$	387

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### Nondimensional Numbers

$$\begin{aligned} \mathbf{DF} &= \text{decontamination factor;} \\ &\left(\frac{\dot{m}_{\text{in}}}{\dot{m}_{\text{out}}} = \mathrm{DF}_{\text{inj}} \times \mathrm{DF}_{\text{rise}} \times \mathrm{DF}_{\text{sur}} \right) \end{aligned}$$

$$\mathbf{We} = \text{Weber number; } \left( \frac{\rho_l \times D_{\text{inj}} \times v_{\text{inj}}^2}{\sigma} \right)$$

### Subscripts or Superscripts

elpi = electric low-pressure impactor	393
in = in/inlet	394
inj = injection	395
l = liquid	396
m = mass(-based)	397
out = out/outlet	398
rise = (bubble) rising zone	399
stk = Stokes	400
sur = surface	401

Acronyms	402
BWR = boiling water reactor	403
$C_2H_3KO_2 = potassium acetate$	404
CsI = cesium iodine	405
CsOH = cesium hydroxide	406
ELPI = electric low-pressure impactor	407
MI = hygrometer	408
$N_2 = nitrogen$	409
NaCl = sodium chloride	410
PI = pressure indicator	411
SAAB = severe accident aerosol behavior (facility)	412
$SnO_2 = tin dioxide$	413

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