



## Experimental studies on retention of iodine in a wet scrubber

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### ABSTRACT

In order to develop an iodine scrubbing model for nuclear severe accidents, experimental data on filtration efficiency of a wet scrubber for gaseous molecular iodine are collected to improve understanding of the basic mechanisms involved in iodine chemical scrubbing. A bubble column reactor 1.5 m high and 0.2 m in diameter equipped with an injection nozzle and a bubble breaker is used to investigate retention under relevant flow regimes. Scrubber is loaded with a scrubbing solution containing sodium hydroxide and sodium thiosulphate, no irradiation is provided. Retention efficiency is found to be strongly dependent on flow regime and on residence time as models predict. It was found that jet-like injection has a strong effect on filtration efficiency introducing a significant dependence on iodine initial concentration in gas phase believed to be due to nozzle hydrodynamics.

### 1. Introduction

Severe Accidents in nuclear power plants might lead to fuel element melting and fission product (FP) release into the containment and subsequently beyond the plant boundaries. Nuclear power plants are designed with engineered safety systems and associated operational procedures that provide an in-depth defence against these releases. The strategy proposed to prevent the release of radioactive species as a consequence of containment failure due to overpressure is a controlled venting for releasing the pressure. To allow the containment atmosphere to be vented into the environment, it is advisable that the vent gas be decontaminated to retain the largest possible fraction of source term inside the plant boundaries. Thus, Filtered Containment Venting Systems (FCVS), such as wet scrubbers or sand bed filters, have been installed on venting lines of nuclear power plants in many countries to provide the necessary flow purification. The release of iodine compounds is a major concern due to their volatility, radiotoxicity and body accumulation making its isotopes the most significant ones in determining the dose delivered to population in the early phase of the accident (Soffer et al., 1995). Hence, it is of paramount importance to provide effective removal of iodine by FCVS.

Wet scrubbers are commonly used in FCVS due to their high collection efficiency for aerosol particles, and due to the possibility to simultaneously retain gas phase species, such as elemental iodine (I<sub>2</sub>). To quantify the effectiveness of a scrubber, a generally accepted parameter is the Decontamination Factor (DF) measuring the ratio of the contaminant mass entering M<sub>in</sub> the scrubber to the mass leaving it M<sub>out</sub>:

$$DF = \frac{M_{in}}{M_{out}}$$

This definition is adopted by most countries to define the regulatory requirements that NPPs have to meet when installing FCVS. Regulators specify minimum DF to be guaranteed for aerosols and elemental iodine. European countries commonly require for elemental iodine DFs from 10 to 1000 depending on the regulation (Hillrichs et al., 2012) resulting in a collection efficiency from 90 to 99.9%, respectively.

Due to its radiological impact, iodine chemistry, transport and retention in the water have been studied since the late 60's mainly in regard to spray systems installed in the containment (Row et al., 1969). Following the Three Mile Island accident it was realized that fission products might be released beyond nuclear power plant boundaries. Further, the Fukushima Dai-ichi accident demonstrated the need of long term venting capabilities of containment as well as the importance of an appropriate mitigation of iodine releases (Lebel et al., 2016). Extensive experimental work on iodine chemistry and speciation under irradiation was conducted in the last decades and provided wide knowledge of significant reactions occurring in the reactor containment in severe accidents (Clément et al., 2007), however only limited work was conducted on the specific topic of iodine pool scrubbing.

Experiments aiming at quantifying the scrubbing efficiency have recently been conducted mainly on specific commercial designs involving the commonly used Venturi nozzle (Gulhane et al., 2015; Ali et al., 2013) and previously on a more generic pool geometry as summarized in (Polo et al., 1996). Nonetheless, the lack of a systematic assessment of iodine scrubbing and a comprehensible data base for an appropriate

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model development still persists. Codes developed to calculate iodine scrubbing in severe accident scenarios, i.e. SPARC, SUPRA and BUSCA, are acknowledged to rely on tentative modelling. As pointed out by Fisher (1996) the validation of these codes is based on a single series of experiments (Diffey et al., 1965), and the validated parameter range is limited.

The aim of this work is to further understand the process of iodine scrubbing in a chemical reactor through experiments in a reduced scale wet scrubber. Only gaseous elemental iodine scrubbing is investigated in a prototypical scrubber without irradiation. The scope is to assess the influence of two main parameters affecting the scrubbing efficiency, namely, iodine concentration in the gas stream and the residence time in the scrubber. Experiments are conducted with pure, dry nitrogen as carrier gas and at pool temperature of 21 °C. No steam fraction is employed. Chemicals involved are representative of the actual chemical conditions in commercial FCVS units. It is noted that the experimental conditions in these tests differ from those typical of commercial FCVS in which the steam mass fraction and the water temperature are high. It is foreseen that further tests will be conducted with a mixture of steam and a non-condensable gas, and at a higher water temperature, to determine the effect of those parameters on iodine retention.

## 2. Experimental work

The work reported here was conducted in a bubble column reactor Mini-VEFITA which simulates the vessel of a FCVS. Mini-VEFITA is a simplified down-scaled model of the industrial scale PSI FCVS facility VEFITA (Suckow et al., 2015). This section presents a description of the facility, as well as the measurement systems and techniques.

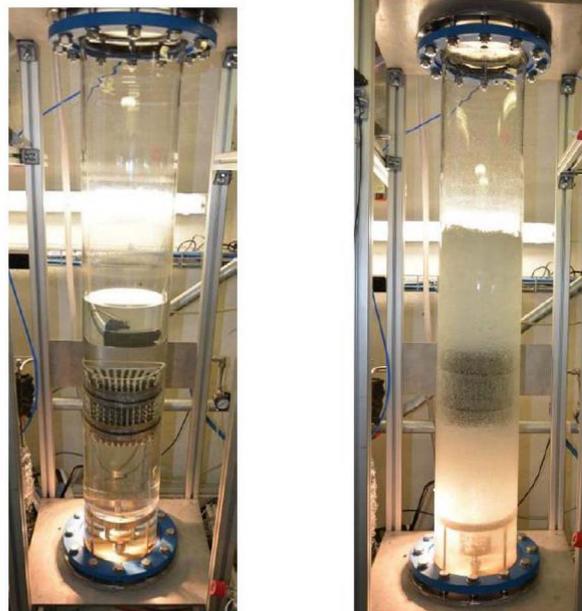
### 2.1. Mini-VEFITA facility

The Mini-VEFITA facility, Fig. 1, consists of a 1.5 m borosilicate glass tube of 0.2 m inner diameter representing the filter vessel equipped with the following internals: a) a IMI-CCI nozzle (Jacquemain et al., 2014) of 10 mm in diameter with 3 perforated impaction plates designed for jet swarm break-up; b) an acrylic glass riser of 0.15 m inner diameter, 0.5 m height and 5 mm wall thickness supported by three thin 0.15 m long legs which holds the mixing element and provides a 20 mm gap between glass vessel and internals for promotion of recirculation; and c) a 0.2 m thick Sulzer Chemtech mixing element, i.e. a stainless steel mesh designed for optimized bubble breaking. Internals are according to commercially employed FCVS installed in Swiss nuclear power plants (Jacquemain et al., 2014). The vessel is connected to a nitrogen source (d) and to an iodine generator (e). Gaseous iodine is generated by evaporating molecular iodine crystals in a small heated glass vessel. The iodine generator is connected to the main nitrogen feed line via bypass valves enabling an accurate control of the feeding. The inlet pipe is made of stainless steel upstream of the iodine generator. To avoid iodine deposition on the feed and sampling lines downstream of the iodine injection point, polytetrafluoroethylene (PTFE) is used with the sole exception of valves and connection parts which are made of stainless steel. The short outlet pipe connected to the outlet sampling is stainless steel. All pipes are insulated and heated to prevent iodine condensation and deposition.

### 2.2. Sampling set-up

Gas phase samples were collected at the inlet (f) and the outlet (g) by impingers. Each sampling line was connected to the main line via a sampling valve followed by a critical orifice (h) which both reduced the pressure, especially necessary at the inlet location, and controlled the sample mass flow rate enabling easy and accurate sampling. Orifice and valves were made of stainless steel while the rest of the pipes were polytetrafluoroethylene (PTFE) or glass. A 500 ml bottle impinger loaded with 300 ml water solution containing ascorbic acid ( $H_2A$ ), at a

a)



b)

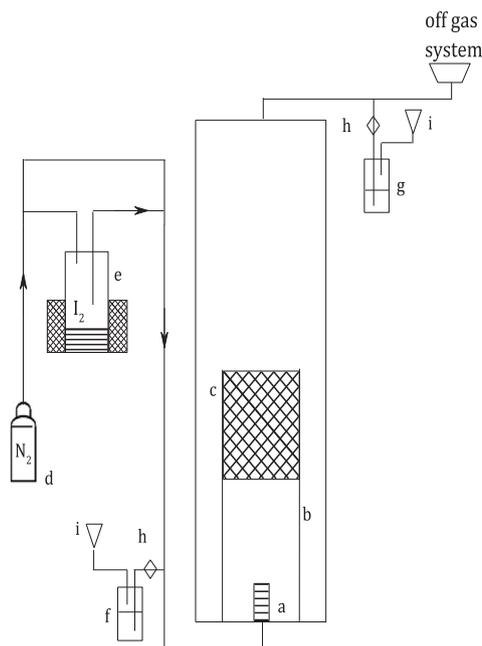


Fig. 1. Mini-VEFITA a) Set-up: Bubble breaker and riser holder are visible at stagnant condition (left) in the lower part of the scrubber while riser support and nozzle can be seen with gas flow (right), and b) Simplified schematic diagram of the facility showing iodine feed system and sampling lines.

concentration of 50 mmol/L and 1 mmol/L for inlet and outlet, respectively, and 6 ml of Ion Strength Adjuster (ISA) was placed downstream of the critical orifice.

For the iodide detection, ascorbic acid was oxidised to dehydroascorbic acid (D) via the redox reaction (Eq. (1)) used to reduce iodine to soluble and measurable iodide:

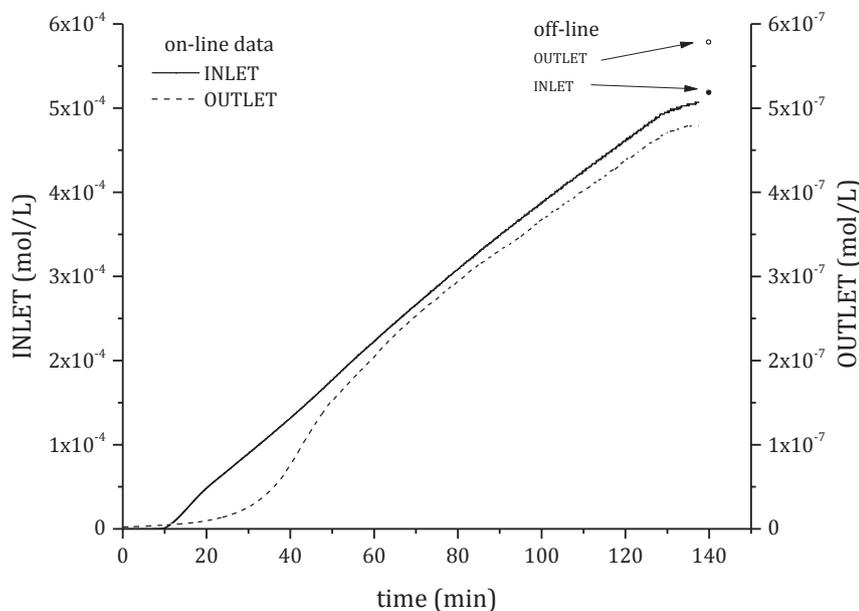


Fig. 2. ISE signals converted to concentration data for impingers located at inlet and outlet showing a linear increase of concentration over test time. Note different scale for inlet and outlet. Off line data are added as a point at the corresponding time, i.e. the end of the test.



The sample impingers scrubbed the iodine out of the gas phase collecting more than 99.9% of the iodine in the solution, as confirmed by placing a second sampling impinger downstream of the first one. Downstream of the impinger bottle, a connection to vacuum system (i) secured a critical pressure ratio at the orifice location for an accurate mass flow rate control.

### 2.3. Test procedure

To perform each test, Mini-VEFITA vessel was drained and washed twice with de-ionized (DI) water before being filled with the desired amount of DI-water and chemicals. For the present set of tests, the chemical conditioning of the scrubber was always the same, namely: 0.01 mol/L NaOH and 0.06 mol/L  $Na_2S_2O_3$ , a scrubber solution composition used in commercial units (Jacquemain et al., 2014).

Once the vessel was filled, the gas flow and the heating of the sampling lines were started. When the sampling lines reached the desired temperature, i.e. 60 °C for inlet and 80 °C for outlet, the impinger bottles for the sampling of the iodine were connected to the sampling lines. Then sampling started and on-line signals of iodine concentration were acquired using Ion Selective Electrode (ISE; Mettler Toledo perfectION™ comb I Lemo Combination Electrode nr. 51344818) for about 10 min to verify that no residual iodine was present in the pipes. Once the ISE signal stabilized and showed no residual contamination, the iodine feed was started. Tests were run for 2–3 h in order to collect sufficient amount of iodine at the outlet impinger to secure a reliable reading. The scrubber temperature was kept constant in the range between 19 and 21 °C for every test. The carrier gas was always dry nitrogen at 60 °C, no steam was used. Once the iodine collection was completed, the iodine feed was stopped while the sampling valves were left open. During this phase lasting 10 min, the ISE signals stabilized to the final asymptotic value corresponding to the total accumulated iodine quantity scrubbed by the impingers.

### 2.4. Iodine quantification

The iodide concentration in the impinger solution was measured with an Ion Selective Electrode (ISE; Mettler Toledo perfectION™ comb I Lemo Combination Electrode nr. 51344818) both on-line (h) and off-

line after the test. Calibration was performed both before and after the test with a dilution series of Orion™ ISE Calibration Standard nr. 945306 in the range  $10^{-7}$ – $10^{-3}$  mol/L. The ISE response to the iodide concentration was linear down to  $10^{-7}$ – $5 \times 10^{-7}$  mol/L of iodide ( $I^-$ ). The calibration was performed at a controlled temperature and at constant stirring with the solution containing a 1:50 vol ratio of Ionic Strength Adjuster (ISA). ISA was required to adjust the ionic strength of the solution as ISEs do not measure directly the concentration but rather the activity of the ions. Hence reproducible results need to be independent of the matrix solution. The ISA guaranteed a constant background ion activity which provided reproducible measurements. Due to the very sensitive response of ISEs, for actual iodine quantification only the off-line measurements were used. Off-line measurements were taken at the end of the test and were conducted in a laboratory environment right after post-test calibration at the exactly same conditions, i.e. the same stirring velocity, sample volume and temperature within 1 K variation. On-line data acquired at not strictly controlled temperature, with bubbly flow affecting the sensor membrane and without sufficient response time for low concentrations were treated rather as a qualitative monitoring of the test progress.

### 2.5. Iodine concentration data

The data consist of on-line inlet and outlet impinger concentrations and the final concentration of iodide in the impingers measured at well-defined laboratory conditions. The data shown in Figs. 2 and 3 are representative of all the data collected during Mini-VEFITA tests. Similar trends were observed in every test hence observations are generally true for all the results obtained in this work.

Fig. 2 shows the increase of iodide concentration in sampling impingers at the test section inlet and outlet. The increase at the inlet (solid line) started after 10 min, i.e. when iodine feed began, and the concentration increased linearly for the entire test proving constant iodine content in the inlet flow. The outlet concentration (dashed line) had a non-linear behavior in the first phase followed by a constant increase starting at approximately 50 min. When comparing the concentration range in the non-linear zone of the outlet measurements with the calibration curve, Fig. 3, it appears that the deviation from the linear growth occurred in a concentration region below the linear response range of the ISE. Hence in this range the primary signal could

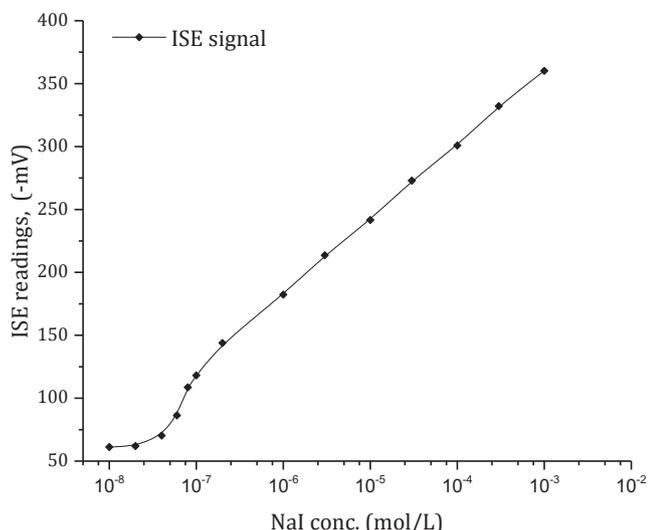


Fig. 3. A typical ISE calibration curve.

not be properly converted into concentration value and needed to be discarded. The linear growth of both impinger concentrations implies constant iodine content in both sample gas streams. Using known sample flow rates and scrubber solution volumes in the impingers, the slope of the curves in Fig. 2 could therefore be converted into constant iodine concentration in the gas flow. The effect of bubble disturbance and lack of proper mixing inside the impingers during the ongoing test was seen as a difference between the on-line readings and the laboratory off-line measurements. The difference was always larger at the outlet as the ISEs are more sensitive at low concentrations.

From the on-line data, an instantaneous DF can be deduced, while off-line data provides an integral DF, which is cumulative for the whole test span, both being shown in Fig. 4. DF could not be calculated for the

initial 20 min of feeding because the iodide concentration was below the linear response range of the ISEs at the outlet. The integral DF calculated on the basis of the off-line measurements was slightly lower than the online value because the online measurement tended to underestimate the concentration at the outlet. However, the assumption that a constant DF which is equal to the integral value is representative of the entire test period seems justified by the history of the instantaneous DF which is nearly constant throughout the test.

Significant efforts were taken to prevent losses due to iodine condensation and deposition on inner walls of the facility. The sampling lines were made of non-reactive PTFE, accompanying heating and thermal insulation was installed for each pipe and sampling flows were as high as possible to reduce the residence time in sampling lines. Furthermore, to assess the accuracy of the set-up, preliminary tests were carried out to test the reliability of the measurements.

For evaluation of the accuracy of the inlet iodine concentration measurement, the inlet concentration was compared with the evaporated iodine mass in the generator. This assessment was feasible only with an evaporated mass over 10 g, otherwise the losses in the generator pipe introduced excessive errors. When comparing the evaporated mass calculated via the ISE reading and the weight loss of the generator, the former was 5–10% less than the latter. The difference was more pronounced for the tests with low flow rates through the generator line and it was almost negligible for higher flow rates. Thus, discrepancies are believed to be mostly due to losses in the generator line in which steel was used to withstand the high temperature of the evaporator. Consequently losses occurred mainly upstream of the injection point and had only an insignificant effect on the inlet concentration.

For the outlet measurements, the absence of contamination of the line was controlled for each test in the initial test stage where no iodine feed was provided. The absence of contamination was indicated by the absence of an iodine signal in this phase of the experiment. To assess losses in the outlet sampling line, a test was run with pure DI water as a scrubber solution, i.e., without the nominal additives NaOH and Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub>. The outlet concentration showed an initial plateau due to the

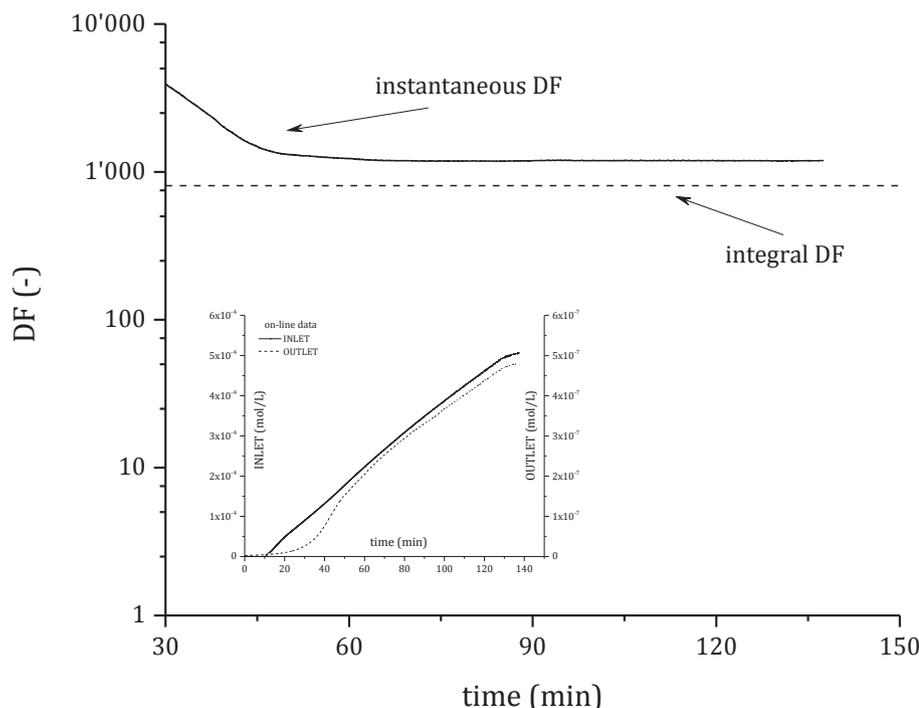


Fig. 4. Instantaneous DF compared to integral value. Outlet concentration in the impinger bottle was too low for the first 20 min of feeding hence instantaneous DF is given only after 30 min.

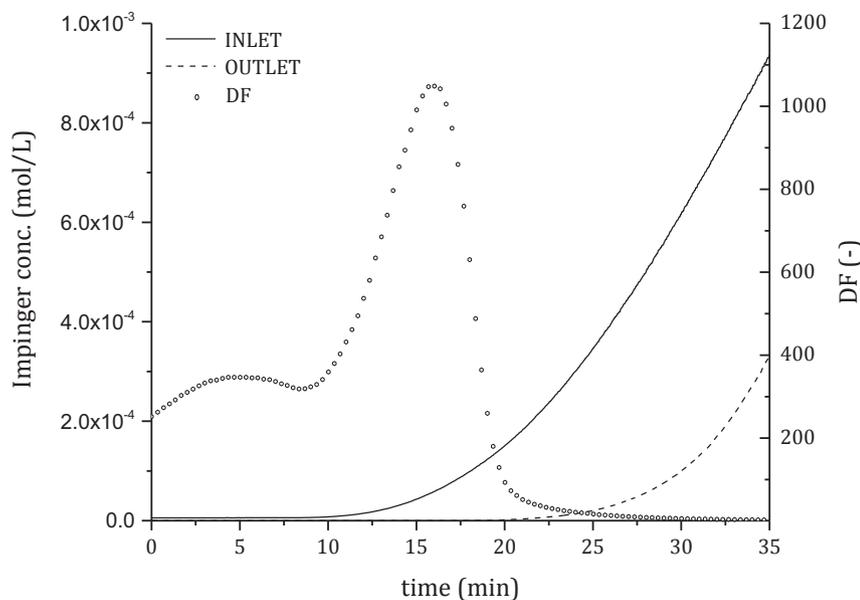


Fig. 5. Impinger concentration trends for a test using a DI water scrubber.

solubility of iodine in water, Fig. 5, which provided some scrubbing capacity. Once the solutions became saturated, the DF dropped to  $DF = 1$  at around 30 min after the start of the feeding, which means that all iodine passed through the scrubber and appeared at the outlet. The inlet (solid line) and outlet (dashed line) have the same slope, i.e., the iodine concentrations in both streams are equal during the last period of the test. This proves that outlet pipe was not retaining any significant amount of iodine.

### 2.6. Swell level

The transparent walls of the Mini-VEFITA glass vessel allow visual observation of the flow pattern. Additionally, the swell level was measured visually. A rough estimate of the bubble size was obtained by comparing the bubbles close to the glass wall to a meter scale. It is acknowledged that this is a very rough estimation of the bubble size, and should be considered only as an estimate for the change of bubble size with the flow regime. Indeed, the bubble size was observed to increase when moving from the bubbly regime to the churn-turbulent, as expected.

The recorded swell level was used to calculate an average residence time ( $\tau$ ) of the gas phase:

$$\tau = \frac{V_{swell}}{\dot{V}_{flow}} \quad (2)$$

where  $V_{swell}$  is the volume of the two-phase column, determined by multiplying the swell level with the inner cross-section of the Mini-VEFITA cylinder, and is the volumetric flow rate.

### 2.7. Uncertainty estimation

Main error sources were discussed in the facility description; here the estimation of these sources is used to assess the overall uncertainty of the DF of elemental iodine ( $I_2$ ). It is defined as the iodine mass entering the test section  $m_{I_2,in}$  divided by the iodine mass released from the test section  $m_{I_2,out}$ :

$$DF = \frac{m_{I_2,in}}{m_{I_2,out}} \quad (3)$$

The mass of iodine into the facility  $m_{I_2,in}$  can be determined based on the iodine concentration in the gas at the inlet  $C_{I_2,in}$  and the total gas

volume  $V_{gas,in}$  entering the facility during the experiment. The mass of iodine released from the facility is correspondingly the iodine concentration at the facility outlet  $C_{I_2,out}$  in the total gas volume  $V_{gas,out}$ :

$$m_{I_2,in} = C_{I_2,in} \times V_{gas,in} = C_{I_2,in} \times Q_{gas,in} \times t \quad (4)$$

$$m_{I_2,out} = C_{I_2,out} \times V_{gas,out} = C_{I_2,out} \times Q_{gas,out} \times t \quad (5)$$

where  $Q$  is the volumetric flow rate of the gas ( $L_N/min$ ) and  $t$  is test time.

Hence, the decontamination factor, as described above, can be written:

$$DF = \frac{m_{I_2,in}}{m_{I_2,out}} = \frac{C_{I_2,in} \times Q_{gas,in} \times t}{C_{I_2,out} \times Q_{gas,out} \times t} \quad (6)$$

It is assumed that the time can be determined with a very high accuracy.

The uncertainty of the flow  $\Delta Q$  depends on the calibration of the mass flow controllers for the main carrier gas and the feed flow. The nameplate uncertainty of the mass flow controllers was about  $\Delta Q = \pm 1.0\%$  within the flow rate range used in this study. As can be seen later, this is insignificant compared to the uncertainties in the iodine sampling and measurement.

The uncertainty of concentration measurement  $\Delta C$  depends on ISE measurements, impinger gas flow rate, impinger efficiency and pipe losses.

ISE measurements are extremely accurate at high concentrations while the accuracy decreases at lower concentrations. For this reason, a sample is always measured with two different electrodes which have different calibration curves. To further test the reproducibility of the data, for some tests the iodine measurement was performed a second time after repeating the calibration after the first measurement. The comparison of the results of both measurements, in total four values for both inlet and outlet, proved a negligible difference for inlet values and differences of up to 10% for outlet.

Impinger gas flow rate was controlled by critical orifices. As the critical orifices are very steady for flow control, and they were calibrated under the test conditions, the uncertainty caused by flow control was estimated to be less than 2%. This is low compared to other uncertainties in iodine concentration measurement.

The high efficiency of the iodine collection by the impingers was tested by passing an iodine contaminated flow through two impingers

in series at a flow rate similar to the one used during the tests. The first bottle showed an iodide concentration around 10 000 times higher than then the second one, hence only a negligible share of 0.01% of iodine was not collected in the first impinger. The high retention is a result of the generation of tiny bubbles by the micro-sized glass frit of the impinger and of the fast redox reaction occurring between iodine and ascorbic acid.

Sampling error was estimated to be not more than 10% due to losses in sampling lines as described in Section 2.5.

The total uncertainty  $\Delta C$  can then be calculated as the sum of different partly systematic uncertainties:

$$\Delta C_{in} = 10\%$$

$$\Delta C_{out} = 20\%$$

### 3. Results and discussion

The experimental work was focused on investigating the effect of three parameters found to significantly affect the retention efficiency of the wet scrubber, namely: a) the flow regime, b) the residence time of bubbles in the pool, and c) the iodine concentration in the feed gas. Dedicated experimental campaigns were carried out to study the effect of these variables on the overall decontamination factor.

#### 3.1. Flow regime

A preliminary hydrodynamic characterization of the flow in Mini-VEFITA was conducted to assess the flow rates of interest. The collapsed water level was kept constant at 0.96 m. The scrubber was loaded with DI water and chemicals as used in the tests, i.e., 0.01 mol/L NaOH and 0.06 mol/L  $\text{Na}_2\text{S}_2\text{O}_3$ , in order to capture the effect of these additives have on the surface tension. The parameters for the hydrodynamic characterization are summarized in Table 1. Fig. 6 shows the normalized swell level as function of the flow rate. A flow rate of 100  $\text{L}_N/\text{min}$  (normal liters per minute) in Mini-VEFITA corresponds to a superficial gas velocity of 5.2 cm/s.

Up to about 200  $\text{L}_N/\text{min}$ , i.e. gas phase velocity ( $U_G$ ) of 0.1 m/s, swell increased almost linearly with increasing flow rate and gas phase consisted of rather homogenous bubbles about 5–10 mm in diameter with similar terminal velocities. According to Shah et al. (1982), transition from bubbly to churn-turbulent flow in a column of 0.20 m diameter occurs for  $U_G$  between 0.05 and 0.08 m/s in water and in dilute aqueous solution. In Mini-VEFITA transition occurred at a slightly higher velocity, likely due to the homogenising effect of the bubble breaker. At flow rates higher than 200  $\text{L}_N/\text{min}$ , larger bubbles of 20–30 mm with significantly higher rise velocity appeared along with a turbulent churn-like flow pattern. For flows higher than 200  $\text{L}_N/\text{min}$ , the circulation of water in the riser-downcomer loop became significant and several bubbles were trapped in the downcomer.

To test the effect of the flow regime on the scrubbing process, three flow rates, Table 2, were chosen. The lowest flow (150  $\text{L}_N/\text{min}$ ) was in the heterogeneous bubbly flow regime while the other two (300 and 450  $\text{L}_N/\text{min}$ ) were in the churn-turbulent regime. The tests were carried out at a constant inlet iodine content, which was set at 10 ppm,

Table 1

Test matrix for the hydrodynamic characterization.

Item	value	unit
Pool height	0.96	m
Pool volume	30	L
Pool temperature	20 ± 1	°C
Gas temperature	60	°C
I <sub>2</sub> evaporator temperature	50	°C
Na <sub>2</sub> S <sub>2</sub> O <sub>3</sub> * 5H <sub>2</sub> O mass (molarity)	447 (0.06)	g (mol/L)
NaOH mass (molarity)	12 (0.01)	g (mol/L)

corresponding to  $1.42 \times 10^{-6}$  mol/L for all three cases, to exclude an effect of the inlet concentration on the DF.

The integral decontamination factor as a function of the average residence time, Fig. 7, shows a clear distinction between the bubbly and churn-turbulent flow regimes. A semi-logarithmic scale of the DF plotted against the residence time is more representative than a plot against flow rates. It allows a better interpretation of the DF trends based on the assumption that the mass transfer is driven by concentration gradients (Lewis and Whitman, 1924). This assumption leads to exponential decay of the contaminant concentration during the scrubbing process, which should be reflected as a linear dependence in semi-logarithmic coordinates. There is a significant change in the behaviour at the transition from bubbly to churn-turbulent flow. The significant reduction of the DF in the churn-turbulent flow cannot be explained only by the reduction of residence time. There is obviously a strong influence of the interfacial area concentration, which significantly decreases at transition from the bubbly to the churn-turbulent flow regime.

#### 3.2. Residence time

In the subsequent test series, we tried to keep the flow regime constant and change the residence time by varying the liquid level. A set of 16 tests was conducted according to Tables 3 and 4. For this series, the flow rate was kept constant at 150  $\text{L}_N/\text{min}$  to avoid influence of flow regime. Each test was repeated twice at identical conditions to confirm reproducibility of the results. Four residence times were combined with two different iodine inlet concentrations of 30 ppm and 100 ppm i.e.,  $4.25 \times 10^{-6}$  and  $1.42 \times 10^{-5}$  mol/L, respectively.

The results are shown in Fig. 8 and Table 5. The DF followed an exponential increase with increasing residence time. Exponential functions were successfully fitted to the experimental points, whereby the test results obtained at constant inlet concentration and varied flow rate are well approximated by unique model coefficients. In particular, the same residence time constant of  $\tau = 1.5$  s was found to well describe both series with different inlet concentrations, i.e., matching both cases of 30 and 100 ppm, while the scaling factor differed significantly, i.e. A = 24 and B = 117.

Assuming the process to be mass transfer controlled, as suggested by the two-film model (Lewis and Whitman, 1924),  $\tau$  is related to mass transfer coefficients at the gas and liquid side of the interface. The total mass transfer rate is controlled by iodine diffusion through these media and by the interfacial area which depends on the bubble size distribution. Hence, tests run at similar conditions, i.e., at the same flow rate and with the same water height and identical chemical conditioning of the scrubber, are expected to have the same characteristic time constant. A dependence on initial concentration is not expected. However, the measured data show a very strong concentration effect: the scrubbing efficiency is five times higher for a three times higher initial iodine concentration at otherwise identical experimental conditions.

#### 3.3. Iodine concentration

In order to confirm the observed concentration effect, a set of tests covering a wide range of iodine concentrations in the inlet gas was carried out at different flow conditions. The water level was kept constant for every test thus tests with similar flow rates had also the same residence time. The test conditions were the ones presented in Tables 1 and 2.

Measured DFs show a strong correlation between inlet iodine concentration and scrubbing efficiency with higher collection efficiency for high iodine concentration in the inlet gas, Fig. 9. Noticeably, exactly same flow conditions, e.g., a flow rate of 300  $\text{L}_N/\text{min}$ , resulted in a DF of about 1000 when the concentration of iodine in the feed gas was high (300 ppm), whereas it was only DF = 10 with an iodine content of 10 ppm. Residence time and flow regime played a role which can be

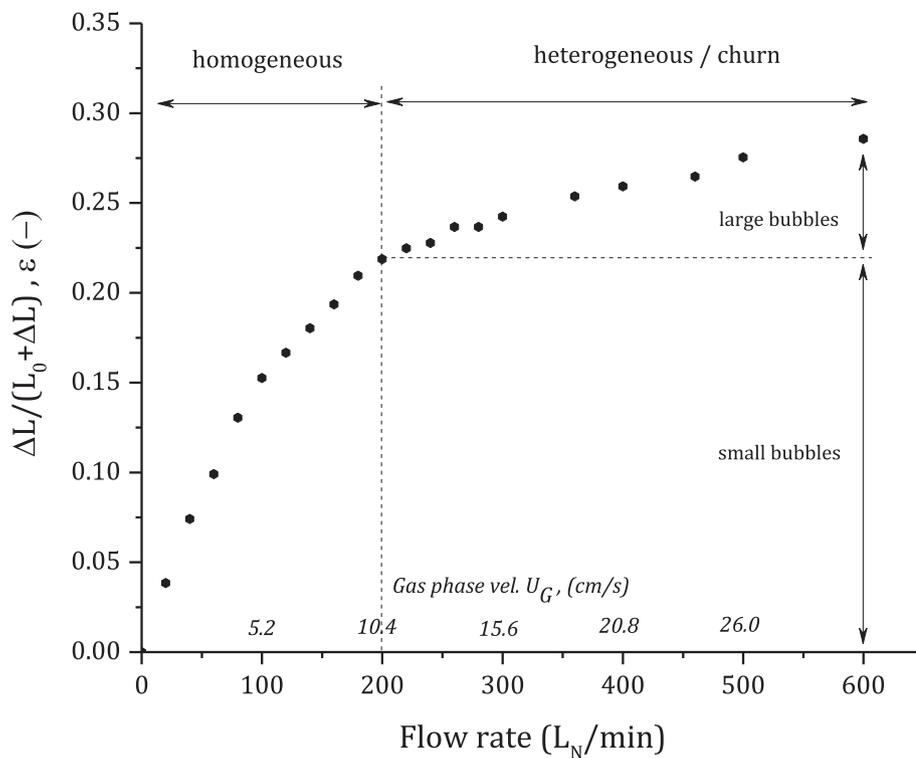


Fig. 6. Normalized swell level vs. mass flow rate for Mini-VEFITA facility loaded with the chemical scrubber solution used during the test and an initial pool height of 0.96 m.

Table 2

Experimental matrix for the tests to study the effect of flow regime on iodine retention.

Series ID	Swell [m]	Residence time [s]	Volume flux [cm/s]
150 L <sub>N</sub> /min	0.21	2.65	8.0
300 L <sub>N</sub> /min	0.30	1.90	15.9
450 L <sub>N</sub> /min	0.34	1.41	23.9

seen especially when comparing the data collected at 150 L<sub>N</sub>/min, with those at 300 L<sub>N</sub>/min, the latter being systematically lower than the former. With increasing flow rate the effect was less pronounced with DFs at 450 L<sub>N</sub>/min rather comparable to those obtained at 300 L<sub>N</sub>/min. This is consistent with the previous results where the DF was proven to be significantly dependent on flow regime. In this series, the inlet concentration increased by two orders of magnitude from 10 ppm to 1000 ppm which was reflected as a similar 100-fold increase in the DF.

A general slightly increasing trend for outlet concentration with increasing inlet concentration implies a weak correlation with the initial iodine content, Fig. 10. This contradicts to the linearity expected from an assumption of a purely mass transfer controlled reaction. The outlet concentration appears to be determined mainly by the residence time and the flow regime, the latter being dictated by the flow rate, as can be seen by the well separated points belonging to the 150 L<sub>N</sub>/min and 300 L<sub>N</sub>/min series.

### 3.4. Injection zone

To identify the reason for the dependence of the iodine retention on the inlet iodine concentration, the injection zone including the injection orifice was separated from the bubble-rise zone. For this, we assumed that a measurement at the lowest water level of 0.34 m represented the scrubbing efficiency of the injection zone, and the water pool above this level represented the bubble-rise zone. Following this concept, the DF measured for the lowest water level, i.e., the shortest residence time, was used to calculate the iodine concentration in the gaseous phase at the beginning of the bubble-rise zone above the injection zone. Assuming this as the starting point of pool scrubbing process in the

bubble-rise zone, DFs and residence times are recalculated for the bubble-rise zone, Table 6.

The resulting decontamination factors for the bubble-rise zone are plotted in Fig. 11 against the recalculated residence time and interpolated by an exponential function. It was found that the iodine retention in the bubble-rise zone is described by an almost identical exponential function for both inlet iodine concentrations of 30 and 100 ppm. This indicates that the DF in the bubble-rise zone was not affected by the inlet iodine concentration, at least with the tested iodine concentrations 30 and 100 ppm. There, the assumption of a mass transfer controlled reaction is obviously a good approximation.

The observed concentration effect for iodine retention in a wet scrubber was pointed out by Ali et al. (2013) who measured decontamination factor in a facility equipped with a Venturi nozzle i.e., where injection hydrodynamics was strongly affected by the nozzle performance. However, no further analysis was provided to explain the reason for the increasing DF with increasing iodine concentration. Our results suggest that the inlet iodine concentration has an influence on the iodine retention in the injection zone, but not in the bubble rise zone. The reason for this localized effect might be found in the jet regime characterized by liquid film disintegration and droplet formation. Under these conditions, a large interfacial area is available accompanied by fast film renewal rate, hence the mass transfer from the gas phase to the liquid phase might not be the limiting step in the scrubbing process, and thereby chemical kinetics, even though fast, might become the limiting factor for iodine retention. Studies on iodine reduction via thiosulphate (Dodd and Griffith, 1949) have shown that the reaction rate is increased by an excess of I<sup>-</sup> in the water. The resulting non-linear reaction kinetics is a possible explanation for the observed concentration effect which has the character of an autocatalytic phenomenon. Further work is planned to investigate this effect.

In bubble-rise zone, on the other hand, the interfacial area concentration and the renewal rate are much lower, and consequently, the mass transfer through bubble surface is presumably the limiting step in the scrubbing process. Therefore, mass transfer scales with concentration gradient and total concentration have no influence on the time constants. Inlet concentration has no effect on the scrubbing in the

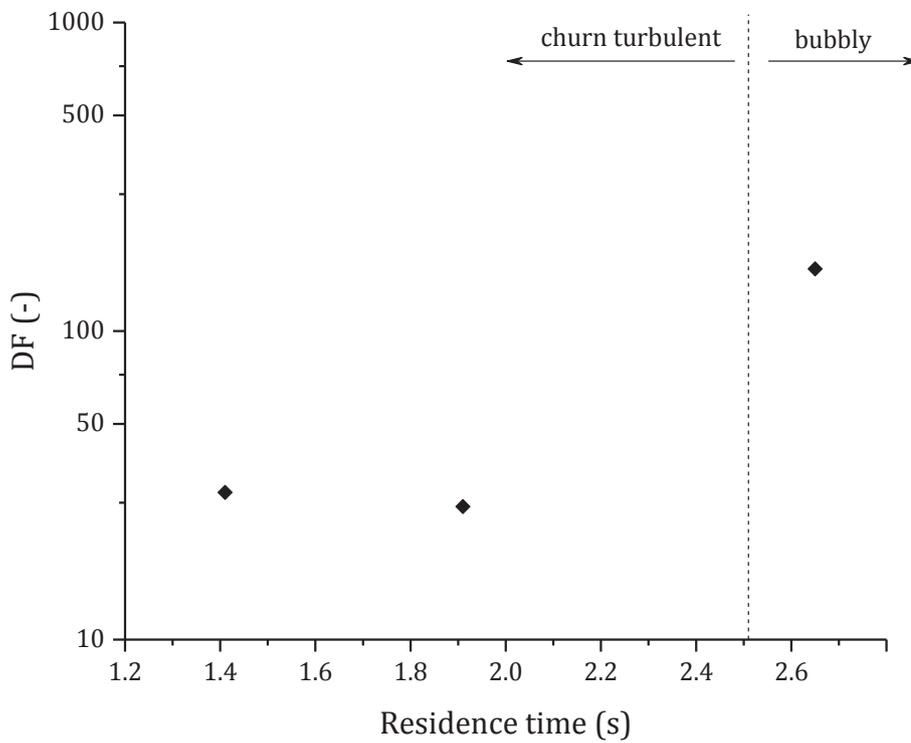


Fig. 7. DF as a function of residence time showing significant change in the flow regime transition.

**Table 3**  
Experimental parameters for the residence time tests.

Item	value	unit
Gas flow rate	150	L <sub>N</sub> /min
Inlet gas temp.	60°	°C
Pool temperature	20°	°C
Gas species	Nitrogen	-
Iodine content	30 and 100	ppm

**Table 4**  
Experimental matrix for residence time tests at 30 and 100 ppm of inlet iodine concentration. Residence time is calculated according to pool swell height and diameter.

Water level [m]	Swell [m]	τ [s]
0.34	0.11	0.78
0.57	0.12	1.51
0.77	0.24	3.02
0.95	0.32	4.02

bubbly flow regime, as the data in this investigation confirm.

**4. Conclusions**

Elemental iodine retention was investigated in a small-scale wet scrubber which represents a model of a commercial filtered containment venting system with slightly simplified internals. The effect of the flow regime and the residence time of the gas in the wet scrubber as well as of the iodine concentration in feed gas on the iodine retention was determined.

Based on the test results, the following main conclusions can be drawn:

- The efficiency of a wet scrubber for retention of elemental iodine depends strongly on the flow regime, as expected. Bubbly flow was

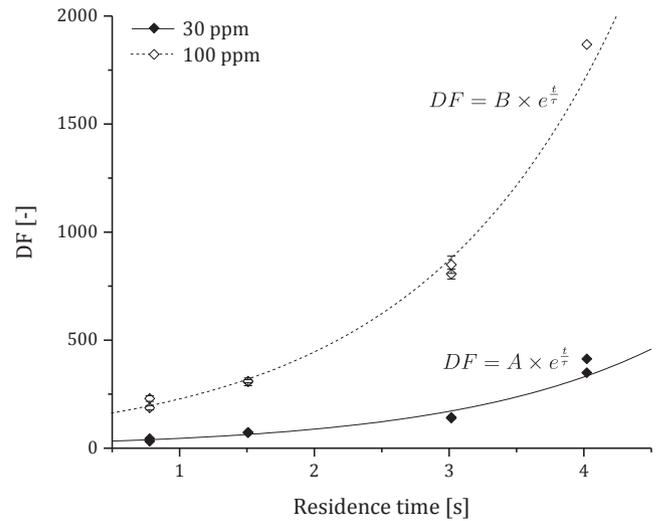


Fig. 8. DF showing exponential increase with residence time for different concentrations.

**Table 5**  
DF values for the residence time experiments. Each test is identified by its residence time given in the first column, while DF depends on the initial iodine concentration in gas phase.

τ [s]	DF	
	30 ppm	100 ppm
0.78	38	208
1.51	73	308
3.02	140	830
4.02	381	1850

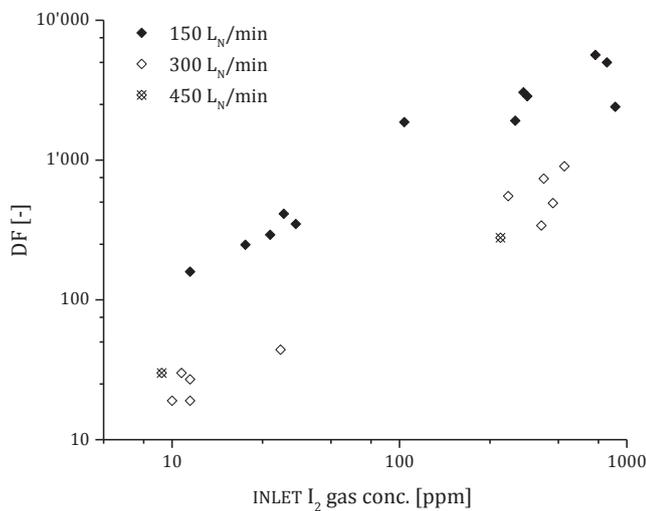


Fig. 9. DF dependence on iodine content in the inlet gas. Tests performed at identical chemical conditions with initial water level of 0.96 m, with different flow rates and correspondingly, different flow regimes.

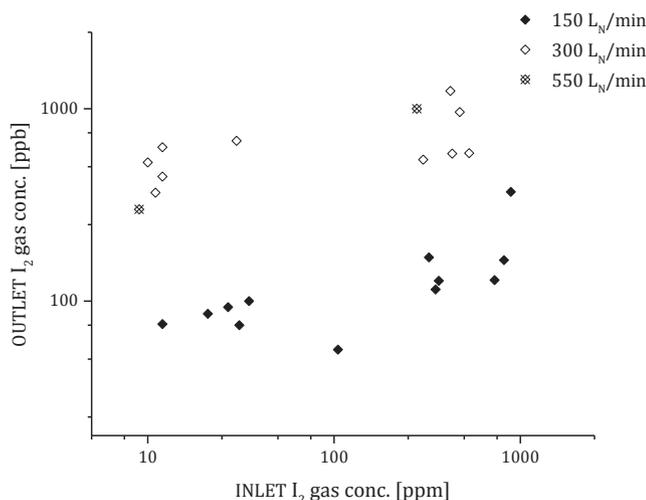


Fig. 10. Outlet iodine concentration for different flow rates and inlet concentrations.

Table 6

Recalculated DF values for the residence time experiments using as initial concentration the iodine gas concentration downstream the nozzle. Residence times are recalculated accordingly.

$\tau$ [s]	DF	
	30 ppm	100 ppm
0.73	1.9	1.5
2.24	3.7	4.0
3.24	10.0	8.9

proven to provide better scrubbing when compared to a churn-turbulent flow even if accounting for residence time variation caused by different flow rates. As the flow regime in commercial FCVS wet scrubbers is almost invariably in the churn-turbulent region, this is an important result regarding the retention of gas phase iodine in commercial wet scrubbers.

- The injection nozzle involving jet formation and droplet generation causes high mass transfer rates, at which chemical reaction kinetics

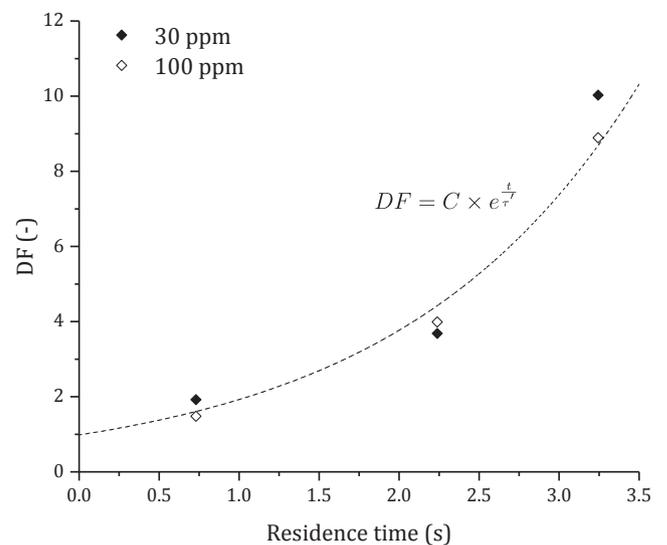


Fig. 11. DF for the bubble-rise zone, i.e., pool without the injection zone.

seem to become limiting. The result is an increase of the decontamination factor with increasing iodine concentration in the feed gas. The higher iodine retention at higher inlet iodine concentration is particularly relevant for testing FCVS as experiments are often carried out at high concentration for measurement accuracy. This might result in non-conservative results regarding the collection efficiency when compared to realistic plant conditions.

- The iodine retention in the bubble-rise zone in the scrubber pool is well described by the two film theory as experimental data confirm a clear exponential decrease in iodine content with increasing residence time.

Further work is planned to better characterize the wet scrubber hydrodynamics and its effect on the iodine retention, as well as to describe the effect of iodine concentration on the retention in the injection zone.

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