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Potential Effect of Saline Solution on Fission Products Mitigation by Pool Scrubbing

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ABSTRACT

The potential release of fission products (FPs) to the atmosphere during an hypothetical severe accidents on nuclear facilities is a main issue in nuclear safety. Accurate estimation of these releases is crucial for risk assessment and implementation of appropriate measures. To this end, IRSN studies the mitigation of FPs transported in a carrier gas injected through a liquid pool. This process, referred to 'pool scrubbing', can occur in various accident situations in Pressurized Water Reactors (PWRs), such as Filtered Containment Venting Systems (FCVS) or with the Steam Generator Tube Rupture (SGTR), as well as in nuclear-powered submarines or in new Small Modular Reactors (SMRs) with pressurized water. Experiments dealing with characterization of bubbles hydrodynamics and trapping of iodine compounds (decontamination factors measurements) are currently conducted. In this objective, a dedicated facility named TYFON (Trapping and hYdrodynamics for FissiON products behaviour in pool scrubbing) at IRSN enables to perform hydrodynamics and iodine retention tests for different carrier gas injection rates (characterized by Weber number). In this context, advanced results have been obtained on hydrodynamics in the injection zone and CsI aerosol retention. In line, a new study focused on the impact of a saline solution on these phenomena and on bubble plume modelling has been launched. In the long term, all the experimental data collected, including hydrodynamics and the decontamination factor (DF), will be used to improve the modelling of pool scrubbing phenomenon in the ASTEC code developed by IRSN.

1 INTRODUCTION

Accidents like those which occurred at Three Mile Island (USA, 1979), Chernobyl (Ukraine, 1986), and Fukushima Daiichi (Japan, 2011) have led the relevant authorities to focus on the mitigation of fission products that are most likely to be disseminated, such as those containing iodine. Aerosols such as CsI or CsOH, or volatile products like I₂ or CH₃I, could have a significant impact on public health and the environment in the near term. Indeed, iodine compounds are inhaled or ingested very quickly by the human body. The French "Institut de

Radioprotection et de Sûreté Nucléaire” (IRSN) conducted several studies to assess the impact of these contaminants in order to be predictive in the case of an accident. In this context, one of their studies focuses on the efficiency of iodine compound mitigation by pool scrubbing. This phenomenon can occur in different accident scenarios such as:

- Steam Generator Tube Rupture (SGTR) in Pressurized Water Reactors (PWR) or Boiling Water Reactors (BWR).
- Filtered Containment Venting System (FCVS), an accident management facility used in PWRs.
- Pressure Suppression Pool of BWRs.
- Submarines, where one proposed solution is to immerse the engine when it is docked.

The first accident mentioned above occurs when a breach is located in the steam generator, creating a flashing effect due to the pressure difference. The second scenario happens when the containment building is saturated with pressure in case of a breach in the primary circuit. The solution involves a depressurization line leading all the containment building atmosphere into a pool called a wet FCVS [1], generating a bubble plume. Thus, the concept of pool scrubbing is when contaminated gas is transported through a pool, to decontaminate the flux by generating bubbles. The phenomenon of pool scrubbing is divided into three zones: the injection zone, where the first bubble forms, the transition zone where coalescence and breakup occur in chaotic behavior, and finally, the bubble rise zone, where a bubble plume is formed. During their rise along the pool, FPs can reach the bubble surface due to aerosol transport and deposition mechanisms or mass transfer for volatile compounds, allowing them to be trapped in water. To investigate the retention of these contaminants, the decontamination factor is evaluated by comparing the FPs mass flowrate entering the pool versus the one exiting it. The ratio is named the Decontamination Factor (DF).

In the literature, many studies on pool scrubbing focus on the trapping of contaminants in clear or alkaline water. Indeed, the trapping of iodine products is more efficient in a pool where $\text{pH} > 8$ [2], [3]. International programs, such as the IPRESCA project[4], were launched to gather international research related to pool scrubbing, with a special focus on modeling and experimental studies. Calculation modules have been developed, such as SPARC, BUSCA, ASTEC, etc. The latter, which stands for Accident Source Term Evaluation Code, is developed by IRSN. This software package is designed for the simulation of events that occur during a reactor core meltdown accident. The software is capable of reproducing the full spectrum of phenomena, from the initial triggering incident to the subsequent release of radioactive products. For example, the evolution of a bubble plume in wet FCVS represents a key element in understanding the decontamination of FPs. A dedicated module attached to ASTEC is in the process of being improved and validated, with the objective of achieving predictive capabilities. A more precise study of the theory of bubble hydrodynamics, particularly the bubble plume, is required to inform the development of the new ASTEC pool scrubbing module. This will be one of the contributions of this PhD research.

Another aim of this study is to understand the pool scrubbing behavior in a different pool matrix. As mentioned before, current studies in the literature focus on clear or alkaline water. However, accidents might occur in submarines. If the engine is immersed, the idea is to evaluate whether seawater can naturally mitigate the FPs and thus avoid their release into the environment.

Thus, in the present study, experimental work is carried out to characterize bubble dynamics in the presence of salt under different conditions. First, a study of bubble volume, frequency,

and velocity was performed, and second, DF measurements were performed for I₂ and CsI. Finally, the potential impact of a saline solution will be discussed, and further experiments will be presented.

2 STATE OF THE ART ON SALINE ENVIRONMENT

We will present an overview of the literature concerning two-phase flows in the presence of saline solutions.

2.1 Retention of iodine in a saline context

Very few studies in the literature have focused on iodine retention in seawater. However, experiments conducted at IRSN have shown that the presence of salt could improve the retention of volatile species such as I₂. [5]

Table 1: Retention of I₂ in seawater vs in clear water[5]

Contaminant	Pool Matrix	Regime	DF
I ₂	Clear water	Jetting (We = 205)	$DF < 10$
I ₂	Seawater	Jetting (We = 199)	$100 < DF < 1000$

According to Table 1, an increase in the DF in the jetting regime was observed. This might be due to the composition of the water (seawater/clear water), which significantly influences the trapping of I₂, with relatively low retention in clear water and higher retention in seawater. This effect could be attributed to the increased chemical reactivity between iodine and the elements present in seawater.

2.2 Bubbles behaviour in presence of salt

2.2.1 In the Injection Zone

As mentioned in the introduction, pool scrubbing is a phenomenon that involves three zones of interest. In the literature, it is noted that different hydrodynamic phenomena can occur depending on the zone of the pool being studied. Farhat et al. studied the retention of various iodine compounds such as, CsI, I₂ or CH₃I,[2], [6]. For example, for the first contaminant, he noted that retention is favoured at low flowrates and tends to decrease with increasing flowrate. Then he observed a minimum of retention in between the bubbly and the jetting regime and then an increase in retention with higher flowrate[2].The flowrate was controlled using an adimensionnal number, the Weber number, which is defined as the ratio of inertia to capillary forces. It can be written as :

$$We = \frac{\rho D U_{inj}^2}{\sigma} \quad (1)$$

Another study was the PASSAM project [7] conducted several experiments using seawater to investigate bubble behavior in pool scrubbing. In the injection zone, they evaluated the distribution of bubble size under different flow rates and in different pool matrices. To study the behavior of bubbles from bubbling to the jetting regime, they evaluated various We numbers as well.

They compared bubble behavior in three matrix: saltwater using the ASTM D1141 standard [8], demineralized water, and water containing surfactants. Their results indicated that, in the

injection zone, there was no impact on bubble size in the bubbling regime across the different matrices [7]. However, in the jetting regime, a different behavior was observed between seawater and clear water. Specifically, they observed a bimodal bubble size distribution in the presence of salt or surfactant.

2.2.2 In the transition and bubble rise zone

In these two upper zones, it was observed that the presence of electrolytes changes the dynamics of the bubbles. Indeed, the surface tension tends to increase in seawater. Nayar et al. observed this phenomenon with different types of salts at varying salinities [9]. Figure 1 shows the evolution of surface tension as a function of temperature and salinity. These tests were carried out using the ASTM D1141 standard, without heavy metals. As mentioned above, the first observation is that salt content affects surface tension: the higher the salinity, the higher the surface tension. Furthermore, an important point emphasized by Jamialahmadi et al. is that this surface tension evolves linearly with the concentration of dissolved salts. This observation was made using potassium chloride (KCl), as shown in Figure 1, [9],[10].

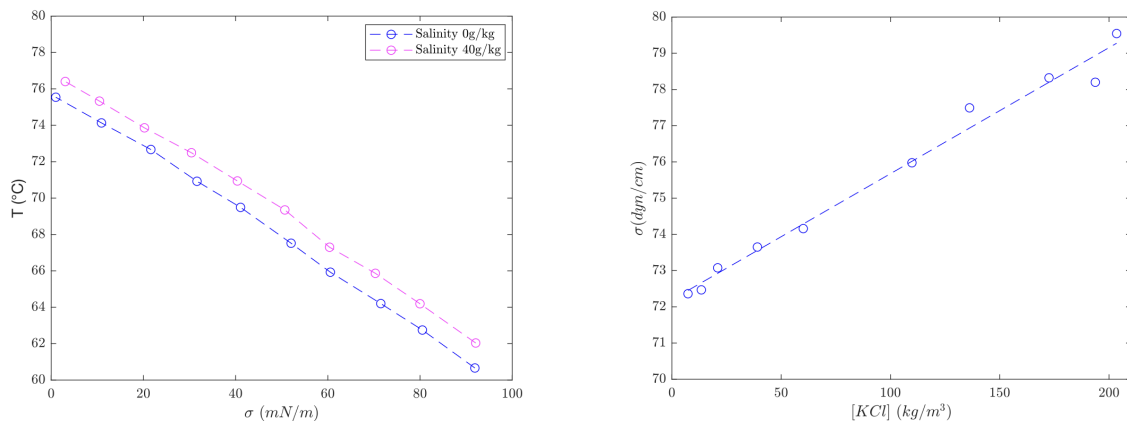


Figure 1: In the left, Surface tension vs salinity and temperature inspired by Nayar et al. [9]. In the right, Surface tension vs KCl concentration inspired by Jamialahmadi et al. [10].

3 EXPERIMENTAL WORK

In order to perform experiment at IRSN, we used the facility TYFON (Trapping and hYdrodynamic for FissiON product behaviour in pool scrubbing). In this section we will present the set up.

3.1 TYFON facility

TYFON is a pool scrubbing set up that allow various studies. Indeed, with this set up we can controll different parameters such as the injection flowrate, the pool and gas temperature, the contaminant that is carried out through the pool, nozzles diameter etc... The facility is depicted in figure 2 with all its features.

Air is introduced through nozzles of varying diameters (2 mm, 5 mm, 10 mm, 20 mm, and 50 mm) into the pool. The bubble column has an internal diameter of 500 mm and a pool depth of 1.3 meters, resulting in a total volume of 250 liters. TYFON's walls are constructed from stainless steel with a mirror polished finish to minimize iodine compound deposition. The

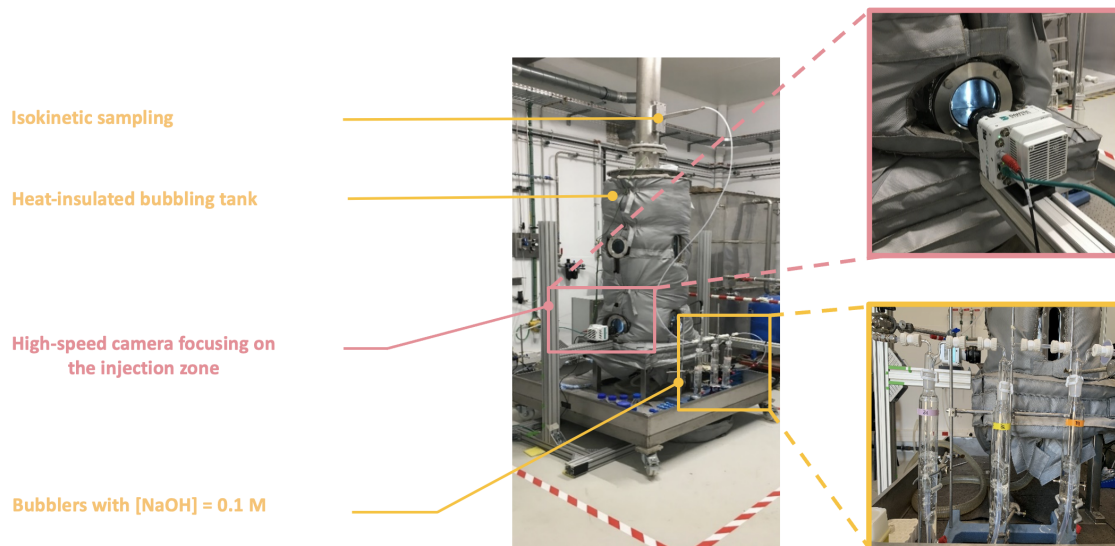


Figure 2: TYFON facility

system is equipped with several pressure and temperature sensors. Additionally, the column features flat windows in order to follow bubbles dynamics. An isokinetic sampling cane is placed downstream of the installation to collect residual iodine in any physico-chemical form, depending on the study. This cane feeds 2 or 3 bubblers, each containing a known amount of NaOH with a concentration of 0.1M. This concentration has been chosen in order to trap iodine compounds efficiently. Finally, at the outlet we place a HEPA filter to collect the remaining iodine that has not been trapped upstream and to include it into our mass balance to obtain the most accurate DF.

3.2 Analysis method

This section will present the different mean used to perform the hydrodynamic tests and to evaluate the retention rate.

3.2.1 Retention of iodine

Inductively Coupled Plasma - Mass Spectrometry (ICP-MS) is an analysis instrument used to measure iodine compounds (figure 3).

ICP-MS procedure involves analyzing a sample by separating, identifying, and quantifying its constituent elements based on their mass. It combines an ionizing plasma torch with a mass spectrometer (quadrupole) to distinguish ions based on their mass-to-charge ratios. Its measurement technique allows the determination of low concentrations in the range of ppb (parts per billion) in aqueous solutions. This instrument was used to measure the concentration of liquid samples taken from the I₂ or CsI experiments.

After analyzing each sample of a test, we obtain the iodine rate. For I₂, we will have the total amount of I obtained and for CsI, we will have the amount of the total amount of cesium and the total amount of iodine. The ratio between the two should be approximately 1. This will allow us to evaluate the retention efficiency by calculating the DF ratio as follows :

$$DF = \frac{\dot{m}_{in}}{\dot{m}_{out}} \quad (2)$$

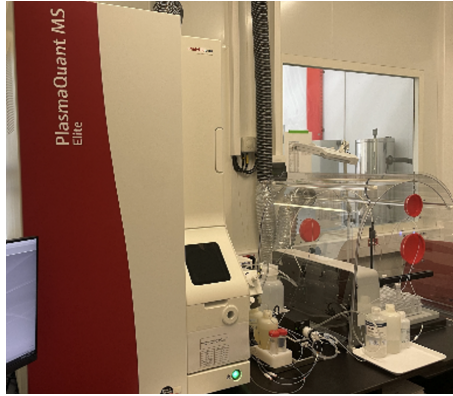


Figure 3: Inductively Coupled Plasma - Mass Spectrometry (ICP-MS) used to measure iodine compounds

3.2.2 Hydrodynamics measurements

To acquire images, we are using a high-speed Dantec camera named “Phantom Speed sense VEO-E”. The camera is depicted in figure 4. The maximum frequency of the camera is 3500 frames per second at 1280×800 pixels. In the background we added a continuous light with a matte sheet in order to diffuse the light. Finally, for the image treatment, we will use mainly MatLab, using a method developed by Farhat et al. [6] in order to estimate the globule volume. The globule is by definition the first bubble forming and completely detached from the nozzle.



Figure 4: High Speed Camera Dantec Dynamics

3.2.3 Globule Volume V_G

Figure 5 shows an example of the visualisation of a globule. The aim is to determine the volume of this object and its frequency. The volume of the structure is calculated numerically once it has reached a stable size.

The volume is calculated by assuming axisymmetry and a cylindrical rotation of the pixels around an equidistant line (see figure 6, the green line) which divides the structure radially into two equal parts. The volume of each cylindrical slice of the structure is the sum of all 1-pixel torus volumes of the corresponding slice. Finally, the volume of the structure is the sum of all cylindrical volumes calculated along the vertical equidistant line [6].

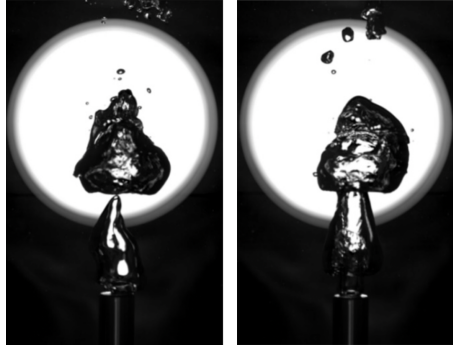


Figure 5: Image taken on the TYFON installation before coalescence (left) and during coalescence (right)

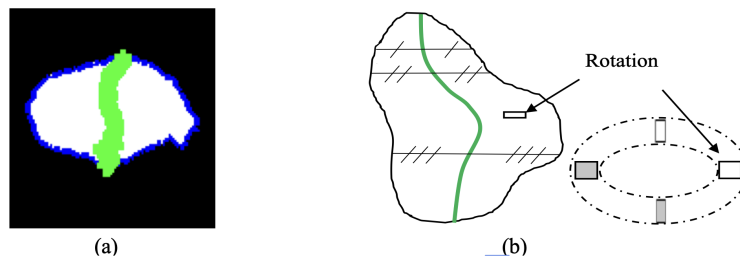


Figure 6: Tracking and computation of globule volume V_G ; (a): Tracked globule, (b): Schema of unitary volume of a pixel [6]

3.2.4 Globule Frequency f_G

The appearance of a globule is detected by observing its rise immediately after its release from the injector. As a reminder, we only examine the globule after it has undergone one or more coalescences, so that we can study it in its most stable form. To carry out these globule frequency measurements, we are going to use image post-processing techniques. The numerical quantification of the globules' formation will be done by analyzing a chronograph of bubbles evolution as presented in Farhat et al. [6] did. This chronograph is efficient for differentiating between the events of bubbles formation, detachment, and coalescences.

4 RESULTS AND DISCUSSION

To recall, our aim is to evaluate the retention of iodine in a saline environment. To optimize the composition of the solution for future saline solution tests on TYFON, we have developed a test grid. The goal of this grid is to simplify the seawater composition used in all our saline solution tests, regardless of the contaminant (CsI or I_2). Seawater is composed of at least 60%w NaCl. Therefore, we will start with the ASTM D1141 standard, as RSE did in the PASSAM project [7]], and then attempt to simplify the solution by removing minority salts that have no significant impact on the results. The final composition (whether the standard or a simplified version) will be used in the test grid to compare with results previously obtained in clear water [2], [6] to assess the influence of the saline environment on the hydrodynamics and retention of iodine compounds. The results confirmed the tests performed at IRSN presented in Section 2.1. The presence of salts tends to molecular iodine compounds more efficiently, with few modification of hydrodynamics in the injection zone.

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