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Trapping of iodine compounds by pool scrubbing: Hydrodynamic aspects

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Résumé :

Dans l'industrie nucléaire, le barbotage est l'un des systèmes de rétention capables de réduire autant que possible le relâchement des produits de fission, lors d'un accident nucléaire. Il s'appuie sur un ensemble de processus physiques et chimiques régis par le comportement des bulles. L'efficacité de ce processus d'épuration dépendra des différentes conditions d'écoulement et des paramètres hydrodynamiques. Des travaux expérimentaux ont été réalisés, pour lesquels une technique de mesure optique a été mise en place afin de caractériser différents paramètres liés à l'hydrodynamique des bulles tels que la fréquence, la forme, le volume des bulles et la fraction de vide. Deux approches pour caractériser la formation des bulles sont mises en œuvre. Une approche classique a été menée, basée sur les hypothèses usuellement considérées dans les principaux modèles de la littérature, à savoir un régime unique de bulles et une forme sphérique. La comparaison des caractéristiques des bulles (volume et diamètre), qui sont obtenues après avoir déterminé la fréquence de détachement des bulles montre un bon accord avec ces modèles. Cependant, la comparaison à la morphologie des bulles observées (coalescence, non sphéricité) montre les limites de ces hypothèses. Cela nous a conduits à introduire, via une approche davantage phénoménologique, le concept de globule, à savoir une structure de volume plus important, générée après coalescence de plusieurs bulles dans la zone d'injection. Cette approche apparaît plus pertinente pour des études ultérieures. Ces volumes sont généralement supérieurs à ceux prédits par l'approche usuelle, mettant en évidence les limites des modèles existants pour des nombres de Weber élevés. Cette approche phénoménologique est confortée en comparant le débit calculé expérimentalement (grâce aux volumes et fréquences déterminés des globules formés) au débit réel injecté. A terme, cela permettra d'améliorer la modélisation de tels phénomènes, notamment dans le code de calcul ASTEC développé par l'IRSN pour simuler un accident nucléaire grave et ses conséquences.

Abstract:

In the nuclear industry, pool scrubbing is one of the retention mechanisms capable of reducing the release of fission products during a severe nuclear accident. It relies on a set of physical and chemical processes that are governed by the behavior of bubbles. The efficiency of this filtration process will depend on the different flow conditions and hydrodynamic parameters. Experimental work has been carried out, for which an optical measurement technique has been implemented in order to characterize different parameters related to bubble hydrodynamics such as frequency, shape, bubble volume and void fraction. Two approaches to characterize bubble formation are implemented. A classical approach has been conducted, based on the assumptions usually considered in the main models of the literature, namely a unique regime of bubbles and a spherical shape. The comparison of the bubble characteristics (volume and diameter), which are obtained after determining the frequency of bubble detachment shows good agreement with these models. However, the comparison with the observed bubble morphology (coalescence, non-sphericity) shows the limits of these assumptions. This led us to introduce, via a more phenomenological approach, the concept of globule, i.e. a structure of larger volume, generated after coalescence of several bubbles in the injection zone. This approach appears more relevant for further studies. These volumes are generally higher than those predicted by the usual approaches, highlighting the limits of existing models for high Weber numbers. This phenomenological approach is confirmed by comparing the flow rate calculated experimentally (thanks to the determined volumes and frequencies of the globules formed) with the actual flow rate injected. In the long term, this will make it possible to improve the modelling of such phenomena, in particular in the ASTEC calculation code developed by the IRSN to simulate a serious nuclear accident and its consequences.

Mots clefs: Barbotage, Globules, Coalescence, Volume de bulles

Keywords: Pool scrubbing, globules, coalescence, volume of bubbles

1 Introduction

In nuclear industry, the injection of a carrier gas transporting gaseous or aerosol fission products through a water pool leads to the formation of a bubble-liquid flow. During bubbles rise along the pool, these fission products such as iodine can reach the bubble surface due to mass transfer for gaseous species and aerosols, these latter being notably concerned by deposition mechanisms. This retention mechanism is referred as pool scrubbing, which is a mean of filtration comprising a set of physical and chemical processes to trap fission products in different nuclear accident scenarios [1, 2]. It is considered to be a significant filtration mechanism that is presented widely in different nuclear installations, such as Filtered Containment Venting Systems (FCVS) [1]. Pool scrubbing may also occur in pressurizer or steam generator tube rupture (SGTR) [2]. Studies of iodine behaviour in containment reported that it could exist in volatile forms such as elemental iodine I_2 or gaseous organic iodine CH_3I , but also in form of aerosols such as Caesium iodide CsI [3]. Depending on the iodine species and among the trapping mechanisms, each one has its own significant impact considering the hydrodynamics' aspects of the accident. In fact, the retention of aerosols is mainly based on the physical behavior of the gas flow in

the liquid, taking into account the present interactions and phenomena [4]. Therefore, bubble hydrodynamics are highly regarded in determining the efficiency of filtration systems such as pool scrubbing. Within this concept, Clift et al. [5] considered that the shape of a bubble is one of the most important factors affecting bubble motion and heat and mass transfer from the bubble. More specifically, it is common that the key parameter for the mass transfer is the interfacial area, which depends on the inertial deformation, shape, size distribution, and velocity of bubbles. The systematic analysis of pool scrubbing is still not well maintained scientifically, because the hydrodynamics' aspects are poorly characterized experimentally, since the data existing in the literature are very fragmentary or not representative [6]. There are still some experimental aspects that are not considered within pool scrubbing codes such as BUSCA, SPARC, and SUPRA. It is reported the latter codes did not achieve satisfactory results regarding the decontamination factor measurements (DF) [6]. This parameter refers to the retention efficiency of fission products through injecting contaminated air into a water pool and is calculated through the following equation:

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

Where \dot{m}_{in} refers to the fission product mass flow rate entering the pool and \dot{m}_{out} refers to the fission product mass flow rate leaving the pool.

The knowledge of bubble dynamics is essential for determining the efficiency of filtration systems that depend on injecting contaminated gas, from a submerged orifice, through liquid column or facility. Since massive research have been conducted to characterize the behaviour and dynamics of bubble formation [7-9], however few of them aimed to characterize the chaotic behaviour of bubbling [10, 11]. That is notably because in high flowrate injection systems, bubble-bubble interactions is considered to be key of nonlinear behaviour of bubbles, as inertial effects over dominate the interfacial – viscous forces. In Farhat et al. [11], it is reported that classical approaches (assumptions of spherical shape of bubbles, single bubbling regime) to characterize the bubble dynamics are not consistent with the flow morphology. That is why it is important to introduce adequate approaches in order to better characterize the bubble hydrodynamics in the different conditions.

The trajectory of bubbles in the pool scrubbing facility is divided into three hydrodynamic zones. Each zone has its specific contribution depending on different removal mechanisms. There is an injection zone where there is the globule bubble formation, then the transition zone where globule breaks up during its rising and finally there the bubble rise zone which is identified as a horizontally expanding bubble swarm that consists of tiny equal sized bubbles. Regarding the different bubbling regimes, and through literature, it is suggested Weber number defined as $We = \frac{D_o U_{inj}^2 \rho_g}{\sigma}$ is pertinent for describing the flow morphologies [8, 11, 12].

In this paper, we present the experimental work carried on the bubble hydrodynamics facility in IUSTI - Marseille. We have implemented an optical technique that will be discussed later, in order to characterize the bubble hydrodynamics such as frequency of bubbling, shape, volume, and void fraction. Different bubble's morphologies have been observed, where we classified different bubble regimes and different phenomena that have not been modelled yet within the concept of the pool scrubbing. We provided a phenomenological approach of characterizing bubble sizes and compare it with classical approaches that seems to unsatisfactorily characterize the bubbles size.

2 Materials and methods

This experimental setup is dedicated for hydrodynamic tests, which have been already conducted. The transparent cube permits us to visualize the flow anywhere, which facilitates the optical technique's measurements.

2.1 Experimental setup

The experimental setup used to study the bubble formation process is shown in Figure 1. Air is injected from single nozzle submerged into stagnant liquid in a cubic tank. It is made of Plexiglas plates forming a 0.34 m^3 cube ($0.7\text{ m} \times 0.7\text{ m} \times 0.7\text{ m}$) and is filled with tap water to a height of 50 cm. The inner-diameter of the nozzle is 12 mm, and the air is supplied to the tank at constant flow conditions, by a compressor through a manometer at ambient temperature. The injected flowrate Q_{inj} is regulated from $10^{-4}\text{ m}^3/\text{s}$ to $10^{-3}\text{ m}^3/\text{s}$ with a volumetric flowmeter.

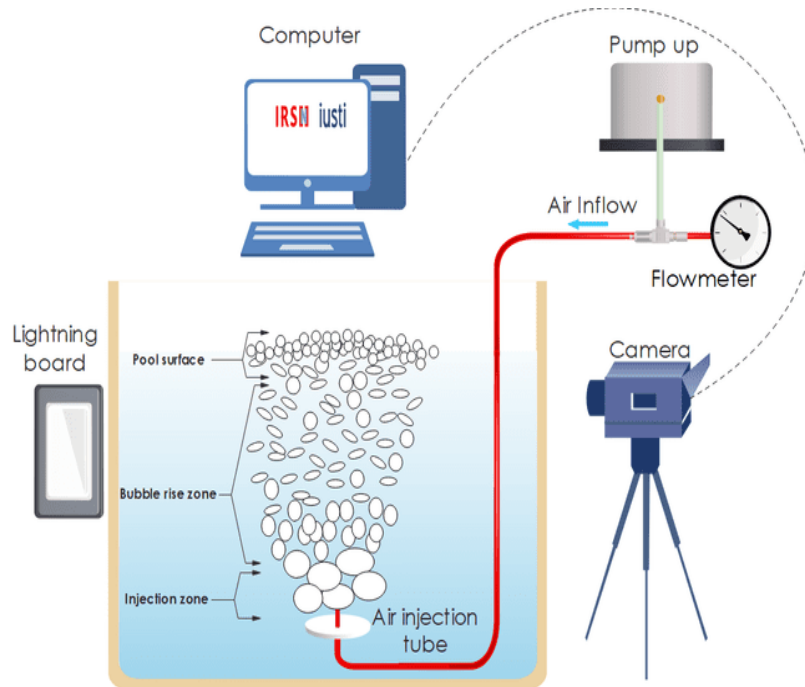


Figure 1 – Schema of the experimental setup

2.2 Data acquisition and treatment

The measurement system for bubble hydrodynamics is composed of a high-speed camera, a LED light source with a diffuser, and associated to the CAM-WARE software. The position of the high-speed video camera is fixed at 40 cm from the cell, and it records the bubble formation process with a frame grabbing of 500 frames/s. Each acquisition of images (1280×1024 pixels) lasts for 10 s, corresponding to 5000 images. The contrast of the raw images are adjusted and blurred to reduce background noise as shown in Figure 2. This is followed by removing noise from images through filtering the grey-scale images using a pixel-wise adapting low-pass wiener filter.

Thereafter, the contour of air bubbles (hereafter referred as structures) is detected using the edge-detection algorithm, which is followed by returning a binarized image. Holes that could appear inside the bubbles due to light refraction or the alignment of the light source are filled in order to have homogeneous binarized structure.

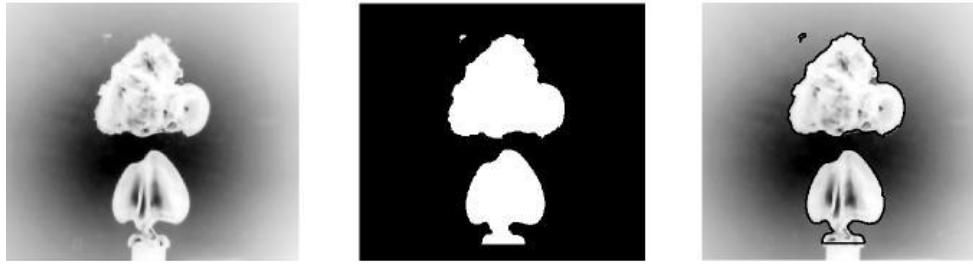


Figure 2 Image processing; (a): Sobel filter, (b): Binarization, (c): Implementing of contours

3 Results and Discussion

Generally and to summarize the morphological description above, when air is injected at intermediate or high flow rates through the orifice into the pool, an air bubble starts to form at the level of the orifice (Figure 3-a). The bubble growth continues as it rises, however it is still connected to the orifice through a neck (Figure 3-b). This channeling effect is also observed as in [13], where $0.1 < We < 200$. Prior to the bubble detachment, the width of the neck increases in a form of trailing bubble, which penetrates the leading bubble (Figure 3-c, d). Eventually, the two bubbles coalesce into large bubble (globule), which is detached finally from the orifice (Figure 3-e). Therefore, in order to characterize the bubble hydrodynamics within this non-periodic behavior of bubbles formation, it is vital to track the formation, departure, and the coalescence of bubbles.

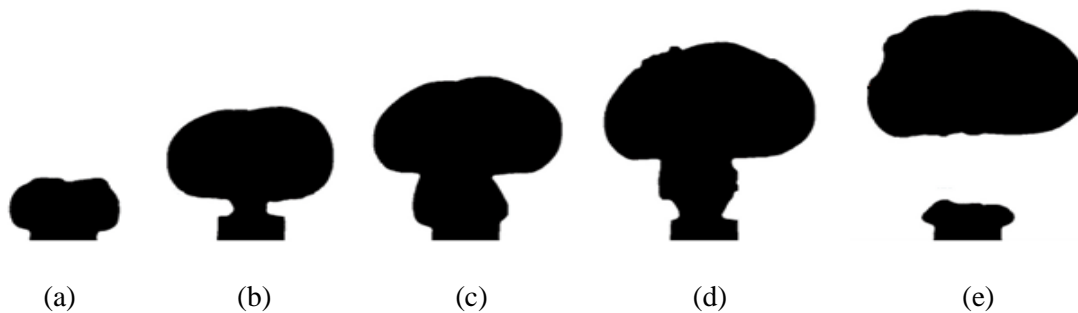


Figure 3- Departure of bubbles resulting in globule formation

3.1 Classical approach

3.1.1 Bubbles departure frequency

Through literature review, we have found that most of the developed models, within the single bubbling regime, established force balance approaches, which neglected bubbles interaction, and made assumptions of spherical shape of the bubble. In this paragraph, we computed the characteristic bubble size by mean of determining bubble formation time t_b , such that $t_b = 1/f_b$.

For the departure frequency, an event is recorded at the instant where there is a structure detachment from the orifice. In case this bubble is re-attached to the orifice by mean of coalescence, the following departure of merged structure is considered as second. The variation of departure frequency for different flowrates is depicted in Figure 4. There is a considerable increase from low to intermediate flowrates until reaching a maximum value at $We = 1.4$ ($Q_{inj} = 18$ L/min). After that, with a further increase of the flowrate, the frequency decreases until it fluctuates around a constant value.

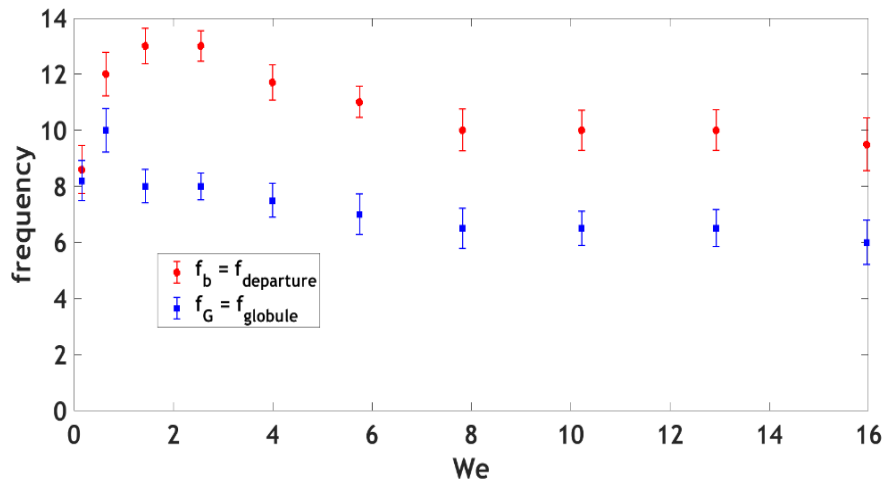


Figure 4 - Variation of departure and globule frequencies as function of Weber number

3.1.2 Bubbles classical volume V_b

Based on the bubble formation time $t_b = V_b/Q_{inj}$ and under the constant gas inflow, we computed $V_b = Q_{inj} \times t_b = Q_{inj}/f_b$. This volume corresponds to the bubble formed without exhibiting any coalescence, supposing a single bubbling regime. We show the good agreement of the characteristic bubble volume for the different correlations [7-9], as shown in Figure 5. As mentioned before, these correlations have made the assumption of spherical shape of the bubbles and single bubbling regime.

On the basis of the morphological description, the spherical assumption is not consistent with the bubbles shape observed experimentally. It is considered a critical limitation especially in aperiodic formation of bubbles (non-single bubbling); amid bubble-bubble interactions, that deforms considerably the shape of bubble (non-spherical shape). This inaccurate characterization of bubbles has a negative impact on the modelling of bubble dynamics in industrial applications, where air is injected at high flowrates. The latter is relevant to pool scrubbing, where the precision is of key importance. Based on what stated, we investigated in the following paragraphs, the volume large bubbles, globules, resulting from the bubbles coalescence in the injection zone.

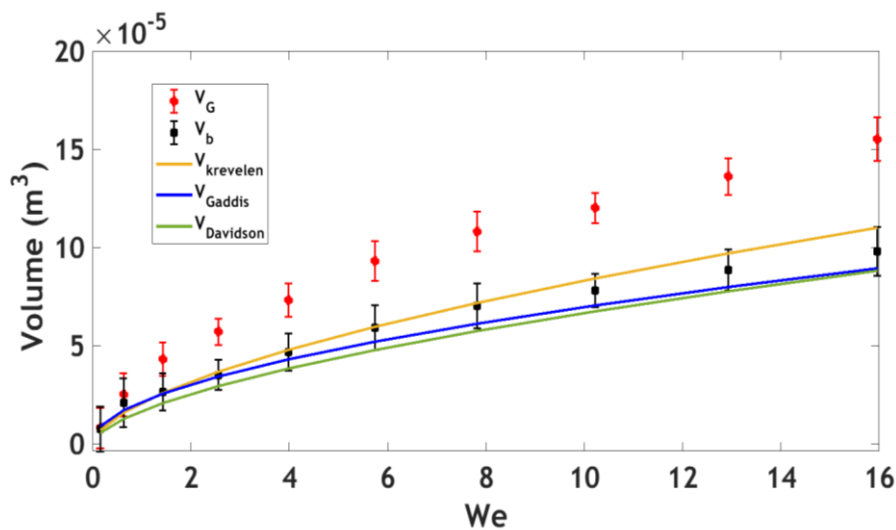


Figure 5 - Comparison of Classical bubbles volume and globules volume obtained experimentally

3.2 Phenomenological approach

3.2.1 Globules frequency

The detection of globule formation is recorded by tracking the growth and the rise of bubbles that have detached from the orifice [11]. Therefore, an event is recorded when a complete formation of a globule is achieved, which is a result of coalescence of two or more bubbles, when it is detached completely and finally from the orifice. Figure 4 shows the variation of globules frequency as function of Weber number.

3.2.1 Globules volume

Beyond the tracking of each globule at the instant of detachment from the orifice, its volume is computed numerically by considering a unitary volume corresponding to a pixel in a binarized image [11]. We supposed a cylindrical revolution of the pixels around an equidistant line, which bisects the structure radially into two equal parts. Then, one part is rotated at 360° around the equidistant line with axisymmetric assumption (the two volumes induced by each part are equal in this case). Thus, the unitary volume is computed for each pixel. Therefore, the volume of the structure is the total volume of the unitary volumes constituting this structure. In the Figure 5, the average globules volume is presented as function of the Weber number.

This phenomenological approach is confirmed by comparing the experimentally calculated flow rate (thanks to the determined volumes and frequencies of globules) with the real injected flow rate, as reported in Farhat et al. [11].

4 Conclusion

Flow structure in pool scrubbing conditions, where for example air is injected at high momentum, exhibits different patterns than in conventional experiments. That recalls for approaches that better describe the bubble hydrodynamics besides the different phenomena. Within this concept and its relevance in nuclear safety, experimental work is carried out to characterize the bubble hydrodynamics in the injection zone, where flow is induced by injection of air from a submerged orifice, and then the experimental results are provided in a systematic analysis. Through this technical paper, we examined the temporal variation of bubbling in terms of bubbling frequency, and described its spatial evolution in terms of bubbles volume and void fraction.

Through the temporal description of the bubble flow, we defined the frequency of departure, which makes it possible to quantify a characteristic bubble volume. The comparisons between bubble volume and literature models have shown a good agreement. Nevertheless, the assumption of spherical bubbles being unsatisfactory regarding to images, it was important to carry out the classification of the bubble morphologies in order to introduce the concept of globule formation. Thus, the frequency of globule formation and the volume of these globules have been determined and seem to be more accurate than existing models to characterize the main gas structure formed in the injection zone.

In parallel, concerning the quantification of the decontamination factor of iodine species, the knowledge of other dynamics such as surface area and velocity of bubbles is of key importance. Respectively these dynamics are linked to the mass transfer and the residence time of bubbles. An extensive experimental database is going to be achieved, through coupling hydrodynamic experiments with decontamination factor measurements for several iodine compounds: CsI aerosols and volatile CH_3I and I_2 . For that, we aim to investigate several flow conditions, comprising different physical and chemical aspects, in order to qualify or improve existing models. Then, we may finally develop a new-coupled hydrodynamic and chemical models, which could contribute in accurate determination of the decontamination factor.

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