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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_2). 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_{in} the scrubber to the mass leaving it M_{out}:

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

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 lasts 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 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:

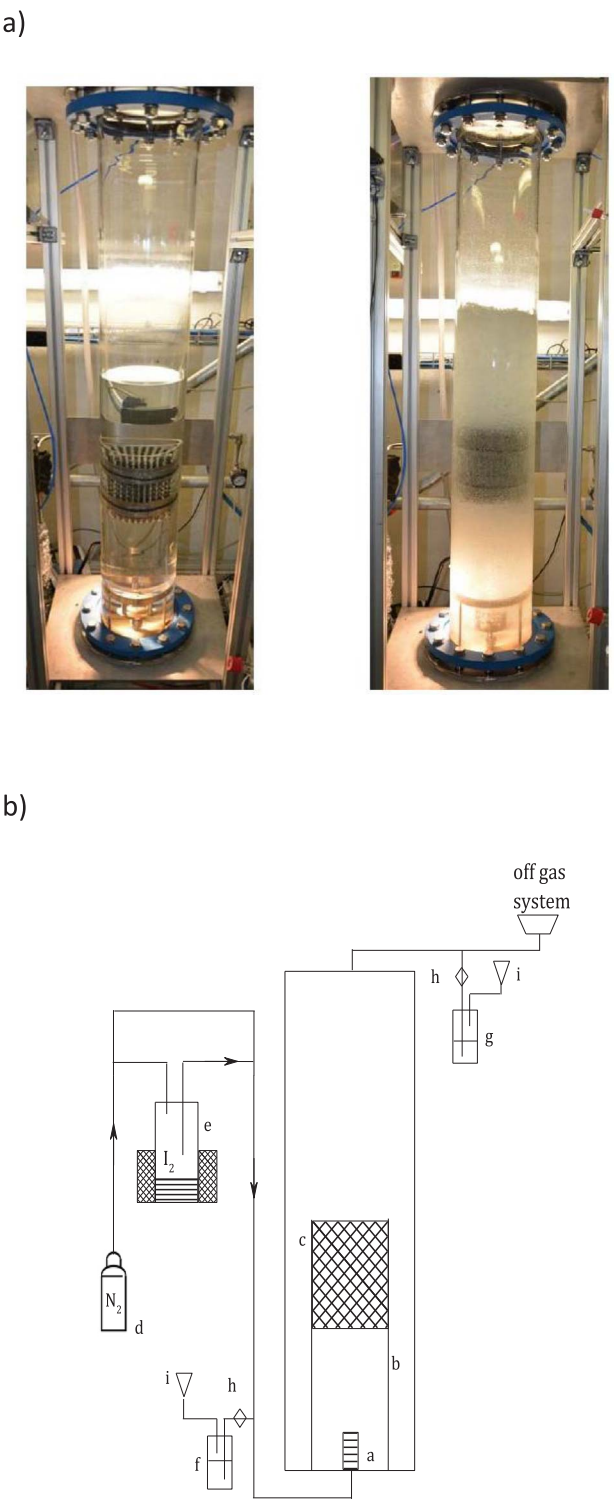

Fig. 1. Mini-VEFITA a) Set-up: Bubble breaker and riser holder are visible at stagnant condition (left) in the lower part of the scrubber while riser support and nozzle can be seen with gas flow (right), and b) Simplified schematic diagram of the facility showing iodine feed system and sampling lines.