Transactions of the Korean Nuclear Society Spring Meeting
Jeju, Korea, May 12-13, 2016

Review on Organic Iodide Retention in FCVS

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1. Introduction

Various researches which are capable of coping with Severe Accident (SA) have proceeded after the Fukushima nuclear accident. Especially, the CH_3I retention efficiency has been a noted issue in FCVS.

An organic iodide, especially CH_3I, has been recognized as one of the most hazardous fission products (FPs) for its high volatility and contribution to a thyroid cancer [1]. A design requirement for domestic Filtered Containment Venting System (FCVS) is determined as a decontamination factor (DF) of radioactive fission product and a passive operation time. For example, a required DF of aerosol and molecular iodine is relatively high, more than 1000. However, DF of methyl iodide (CH_3I) is relatively low, around 50 for its difficulty in separating iodine ion from CH_3I and in preventing further recombination and re-volatilization. For this reason, there have been numerous attempts to achieve a high DF of CH_3I by using various methodologies. Several institutes have been studying and developing their own technology by participating some projects such as ICHMM (Iodine Chemistry and Mitigation Mechanisms) and PASSAM (Passive and Active Systems on Severe Accident source term Mitigation) [5-7, 9].

A retention efficiency is directly connected with DF. The retention efficiency of organic iodide CH_3I is closely related with many parameters: temperature, pH control, chemical additive, radiolysis, etc. This paper aims to review a current state of the art of a retention efficiency of CH_3I by investigating an effect of parameters mentioned. In addition, several retention technologies are also introduced to understand better how these are applied and used for CH_3I retention.

2. Retention Technology

2.1. Sorbents retention stages using Zeolite

Organic iodide retention efficiency of zeolite depends on various parameters. For example, sorbent material, geometry and flow dynamics should be considered in this field. Additionally, heat repartition, residence time and relative humidity, gas composition are also important elements [6].

Retention efficiency of methyl iodide by using various silver ion-exchanged zeolites (AgX) is changing along the temperature and silver ion-exchanged level. The maximum adsorption capacity of AgX increases in the temperature range of 150 to 250 ℃ and when the silver ion-amount becomes higher [9].

Retention efficiency depends on the residence time. Thus the filter area and bed depth of the zeolite are noted issues to fulfill the requirements for organic iodide retention [10].

2.2. TEDA (Triethylene-Diamine)

The process of physical and chemical adsorption causes interactions between TEDA and organic iodide forming stable ammonium salts. That maintains the fixed porous structures of adsorbent, and increases the retention efficiencies of organic iodide up to 98.1% [14].

2.3. Pool Scrubbing

Pool scrubbing is a decontamination method using scrubber pool. The injection parts have various options such as a venturi scrubber, vents, and orifice, etc. In this field, understanding of phenomenon is needed to analyze CH_3I retention effect. Bubble hydrodynamics and chemical knowledge are required to enhance the DF of organic iodide. In addition, understanding of interfacial mass transport phenomenon is needed to improve the retention performance of organic iodide. Retention of organic iodides by the pool scrubbings has been a technological challenge [12].

2.4. Combined method EFADS

The EFADS (emergency filtered air discharge systems) is made for removing most of fission products from containment. This system includes several filtration methods such as charcoal filters, HEPA filters. Undergoing this filtration process, most of FPs including CH_3I can be retained by passing the system. Thus the organic iodide retention efficiency is over 99.8% [12].

3. Influence of Parameters

3.1. Temperature

As temperature increases, retention efficiency becomes high. Since the bonding between CH_3^+ and I^- could be broken easily at high temperature and I^- dissolves in an aqueous solution. For example, a degree of hydrolysis of CH_3I as a function of temperature of an aqueous solution was reported in [2] as follows: 0.028 %/hr at 22℃, 0.27%/hr at 50℃ and 17.33 %/hr at 70℃. This hydrolysis rate (in s^{-1}) was given in temperature (in K).

Zhou et al. obtained a result on the effect of temperature on retention efficiency. While the gas flow rate is constant of $0.2m^3/h$, CH_3I retention efficiency is rising significantly with increasing temperature range from 10°C to 80°C [11].

The system applying zeolite filter has a similar trend between temperature and retention efficiency. In other words, high retention efficiency occurs with increasing temperature [9].

3.2. pH control

It was reported that iodine retention was strongly dependent on the water pH. Higher pH of an aqueous solution produces a higher retention efficiency of CH_3I. For this reason, it might be critical to maintain a high pH to achieve a high DF of CH_3I. Equation (1) shows a formation of CH_3OH and a deformation of CH_3I in alkaline solution.

$$CH_3I + OH^- \rightarrow CH_3OH + I^- \quad (1)$$

Although the pH effect is not significant within the range pH 5 to pH 9 [2], it is obvious that higher pH level allows scrubber pool to maintain high CH_3I retention efficiency.

3.3. Chemical additives

One of the most commonly used chemical additives is thiosulphate ($Na_2S_2O_3$) containing Sulphur, hydrazine hydrate (N_2H_5OH) containing Nitrogen, hydroxylamine (NH_2OH) and the tertiary amine trioctyl methyl ammonium chloride ($CH_3N[(CH_2)_7CH_3]_3Cl$). Additives like these materials that produce other reductants from OH^- radicals will increase CH_3I retention efficiency [2].

Especially, Aliquat 336 is acting as a catalyst to increase the retention efficiency between aqueous solution and methyl iodide in pool. Furthermore, it is used as an anionic exchanger [2].

3.4. Relative humidity

As a relative humidity increases, the amount of organic iodide and volatilization decreases in FCVS. In addition, the volatilization of molecular iodine decreases at the same time. Since a polarized iodine by water vapor allows an enhanced interaction of chemisorption that leads to a low volatilization [1]. It was reported that when the relative humidity was increased from 20 to 60%, the volatilization of CH_3I decreased [1]. When relative humidity increases, the fraction of organic iodide volatilized and molecular iodine volatilized decreased in the experiments of IRSN [8].

The distance to the dew point makes the difference of retention efficiency. Having the high dew point means that relative humidity is high. Therefore, higher dew point, higher organic iodide retention efficiency can be obtained [10].

In the experiment of AREVA using JAVA facility, the data between containment pressure and two parameters was obtained. When the pressure increase in a containment, residence time reduce. On the other hand, the relative humidity increases at that time. This phenomenon keeps the CH_3I retention efficiency on the steady state [7].

3.5. Pressure

As pool pressure increased, superheating was promoted, but organic iodide retention time in a pool decreased. Longer retention time allowed more chance to decompose of CH_3I in a pool. However, a promoted superheating may increase a relative humidity in a system. For this reason, an effect of pool pressure on a retention effect of CH_3I was not significant in that case [3]. However, the effect of system pressure on CH_3I retention is valid in general. Zhou et al. acquired data to represent the relation between system pressure and retention efficiency. The retention efficiency of CH_3I is enhanced with increasing the partial pressure, and the data have linearity between two parameters [11].

3.6. Flow dynamics

As flow rate decreased, a residence time of bubble in a pool increased and a decomposition of CH_3I in a bubble would be enhanced with an increased residence time [4].

Zhou et al. also draws a similar conclusion indicating that the organic iodide retention efficiency decreases significantly with increasing gas flow rate. As gas flow rate increased, the gas-liquid contact time in a pool decreased, which resulted in the reduced retention efficiency [11].

If a filtering system is not in pool but in air, the CH_3I penetration increases with high air velocity [13].

The relationship between retention efficiency and flow rate is found to be represented by a quadratic equation [15].

3.7. Water submergence level

The research on the retention efficiency related with liquid pool level is conducted under pressure of 0.1MPa and temperature of 80°C. High water level allows gas-liquid contact time to increase [11].

4. Acknowledgement

This work was supported by the Nuclear Safety Research Program through the Korea Foundation Of Nuclear Safety (KOFONS), granted financial resource from the Nuclear Safety and Security Commission(NSSC), Republic of Korea (No. 1305008-0315-SB130)

5. Conclusions

Various researches related to the CH_3I retention efficiency, a noted issue in FCVS, have been being conducted. Several technologies and parameters intimately related with methyl iodide retention were reviewed in this paper. A decomposition of CH_3I was promoted with an increase of temperature and pH of an aqueous solution, a relative humidity of FCVS system and a residence time in a pool. However, it decreased with an increase of flow rate of bubble in a pool and a pressure of pool. More detailed investigation on retention of CH_3I is ongoing, and an improved venturi scrubber design is being developed to achieve a high DF of CH_3I. In addition, the understanding of dry filtration enhancing the retention effect is needed. For example, the effect of zeolite filtration is closely related to many phenomena: chemical and physical characteristics, aerodynamics, thermodynamics and various relationships including some climatology.

Each of wet and dry scrubbing technologies taking a main function of FCVS would be required to be enhanced technically and phenomenologically for organic iodide retention.

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