Experimental study of methyl iodide absorption by zeolite dry filtration

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1. Introduction

An organic iodide, especially, methyl iodide (CH_3I) would generated non-negligibly from a severe accident in a nuclear power plant. In order to consider a generation of organic iodide, a new reference source term, NUREG-1465, includes an organic iodide, CH_3I. This CH_3I will be dangerous for human when it was inhaled, it is highly toxic and causes a serious nerve disorder. Even it is a major contributor to a thyroid cancer. In order to prevent its environmental release, it is required to decontaminate using a filtration system. There are two kinds of filtration methods, one is a wet-type using a pool scrubbing and the other is a dry-type using absorbents like zeolite, sand, charcoal, etc. For the removal of CH_3I from the release gases, wet-type is not ideal due to a high re-volatile characteristics of CH_3I. It may become volatile after dissolving in a pool and forms CH_3I again. Therefore, a dry-filtration should be installed to remove the CH_3I. In this study, we preliminary investigate the characteristics of zeolite filtration methods for the removal of CH_3I. We used both silver ion exchanged Y-zeolite and proton exchanged Y-zeolite to study the effect of silver ion for the removal of iodide from CH_3I.

2. Experimental details

In this study, the commercial Y-zeolite was used and ion-exchanged by silver and proton, respectively. These zeolite was packed in the teflon tube of 10 cm length. The gas chromatography with a FID detector was used to measure a concentration of CH_3I in the flowing gas that passed the teflon tube.

3. Results and Discussions

To comparison between chemical absorption and physical absorption capacity and characteristics of methyl iodide, two experiments was conducted by different zeolite materials. For identifying the chemical absorption performance and characteristics of methyl iodide, Y-zeolite exchanged Ag^+ (Ag^+-Y) was used. On the other hands, for identifying the physical absorption performance and characteristics of methyl iodide, Y zeolite exchanged H^+ (H^+-Y) was used. Both of experiments was conducted with methyl iodide flow rate 50ccm/min and room temperature (about 25℃) condition. The Fig. 1 showed the change of methyl iodide concentration as a function of time when Ag^+-Y was used. The observed methyl iodide concentration was not detected or had negligible value observed until just before 1000min (16.7hour). After that time, the concentration was increased with drastic slope until about 1500min (25hour) and observed maximum concentration 1070ppm at that time. After reaching the maximum concentration, the methyl iodide concentration was monotonically decreased and observed 154ppm when the experiment was finished.

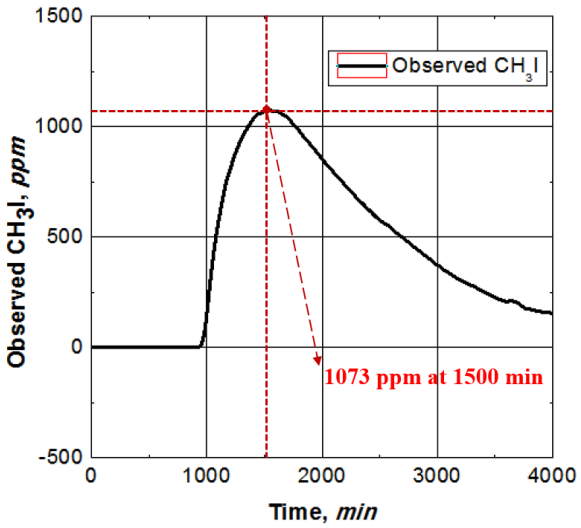

Fig. 1 Observed methyl iodide concentration as a function of the time when Ag^+-Y zeolite was used.

The Fig. 2 showed the color change of Ag^+-Y zeolite as time passed. The color was changed from white to yellow, which can be considered the yellow reaction sediments (AgI) were generated as methyl iodide and Ag^+-Y zeolite were reacted. The color change was progressed from right end of the loop to middle of loop. The direction of color change was dominant until around 1200min, after that time, the direction of color change (left end of the loop to middle of loop) became dominant. The visual material color change rate was similar with the methyl iodide concentration change rate. The largest materials color change rate was identified from 0 to 180min, among the pictures. It was understood well because during that period, the observed methyl iodide concentration was negligible, which means the almost all of input methyl iodide was reacted with Ag^+ ions. On the other hands, the smallest materials color change rate was also identified from 1440 to 1680min. It was also understood because during that period, the observed methyl iodide concentration was comparative highest value during the experiment, which means comparative much quantitative of methyl iodide was not reacted with Ag^+ ions. There was an interesting point to see both of

Fig. 1 and Fig. 2. After the color change rate from left to middle of loop became dominant, the concentration of methyl iodide was started to decrease. These phenomenon can be considered the yellow reactant made methyl iodide gas hard to flow through loop, therefore, the observed methyl iodide concentration was started to decrease, but it needs more experiment data to explain this phenomenon clearly.

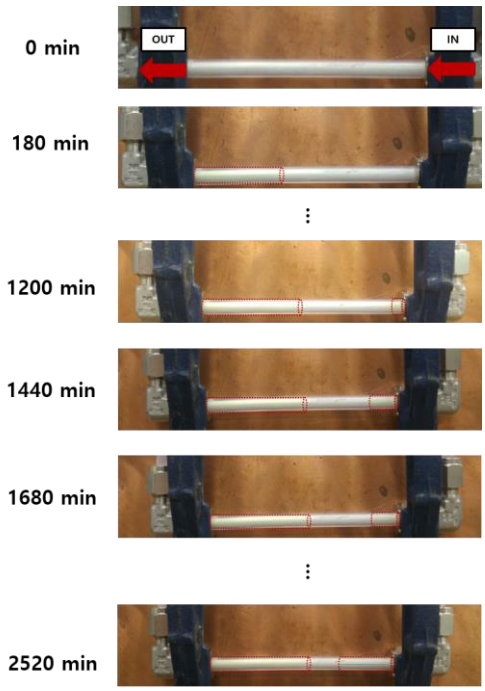

Fig. 2 The color change of Ag^+-Y as time passed

The Fig. 3 also showed the change of methyl iodide concentration as a function of time when H^+- Y zeolite was used. The methyl iodide concentration was not detected or had negligible value observed until just before 1300min (21.6hour). After that time, the concentration was monotonically increased until reached maximum value 1493ppm when the experiment was finished. There was no color changes of H^+-Y zeolite because physical absorption just filtered out the methyl iodide with their unique conformation.

The Fig. 4 compared absorption capacity and progress characteristics between Ag^+-Y and H^+-Y zeolite as a function of the time. In Ag^+-Y zeolite case, the time duration until the noticeable increment about concentration of methyl iodide can be detected was about 960min. On the other hands, in H^+-Y zeolite case, the time was about 1300min. Therefore, H^+-Y zeolite can be considered that has better performance to delay the methyl iodide passed through the loop compared with Ag^+-Y zeolite. However, in respect to detected maximum value of methyl iodide concentration, Ag^+-Y zeolite can be considered that has better performance compared with H^+-Y zeolite.

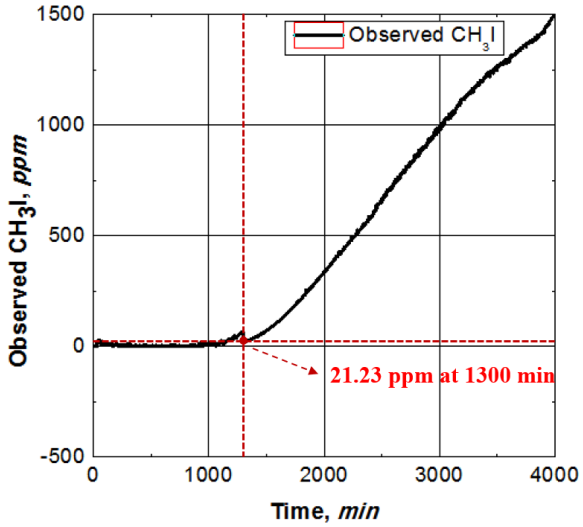

Fig. 3 Observed methyl iodide concentration as a function of the time when H^+-Y zeolite was used.

But it is need to acquire the experimental data about value of inlet and outlet pressure of loop, because the decrease of methyl iodide concentration phenomenon might be occurred because of blocking the input gas, if so, as pressure accumulated, the system can become unstable.

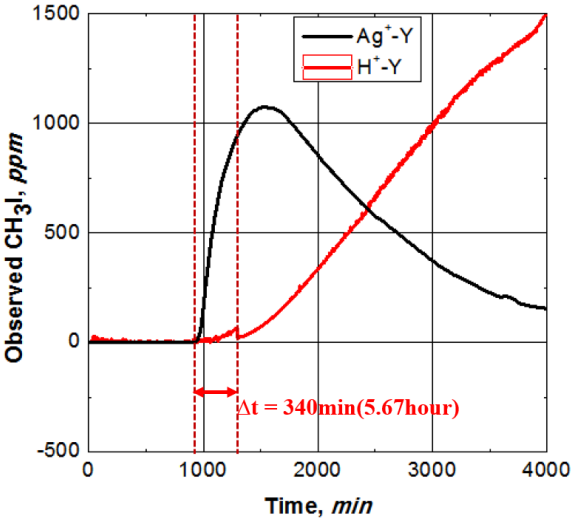

Fig. 4 Comparison between Ag^+-Y and H^+-Y zeolite about methyl iodide concentration as a function of the time.

4. Conclusions

To compare between chemical and physical absorption performance and characteristics of methyl iodide, Ag^+-Y and H^+-Y zeolites were used. The experiments were conducted by teflon tube packed with each zeolites and constant methyl iodide flow rate at room temperature. There were three conclusion through this study.

1. The characteristics and performance of chemical absorption, which was identified by Ag^+-Y zeolite, were that the delayed time until

observed the remarkable change concentration of methyl iodide was around 1000min and the concentration was started to increase rapidly and reached the maximum concentration 1073ppm at about 1500min. After reached that point, it was decreased monotonically and observed 154ppm when the experiment was finished. The visual observation, which can identify the color change rate, also showed a good agreement with the observed methyl iodide concentration as a function of time.

2. The characteristics and performance of chemical absorption, which was identified by H^+-Y zeolite, were that the delayed time until observed the remarkable change concentration of methyl iodide was around 1300min and the concentration was started to increase rapidly without any decreasing phenomenon and reached the maximum concentration 1493ppm when the experiment was finished.
3. In respect of delaying the observed remarkable concentration change of methyl iodide, H^+-Y zeolite showed more good performance compared with Ag^+-Y zeolite(Ag^+-Y: around 1000min, H^+-Y: around 1300min). However, in terms of reducing the final maximum observed concentration, Ag^+-Y zeolite overwhelmed H^+-Y zeolite(Ag^+-Y: around 1073ppm, H^+-Y: around 1493min). But it should be checked the concentration decreasing phenomenon, occurred during Ag^+-Y zeolite experiment, for evaluating each zeolites performance properly.

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