

## Adsorption behaviors of organic iodide in the silver ion exchanged zeolite

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### 1. Introduction

An organic iodide, especially, methyl iodide ( $\text{CH}_3\text{I}$ ) would be generated non-negligibly from a severe accident in a nuclear power plant. This  $\text{CH}_3\text{I}$  would be dangerous for human when it is inhaled for 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  $\text{CH}_3\text{I}$  from the release gases, wet-type is not ideal due to high re-volatile characteristics of  $\text{CH}_3\text{I}$ . It may become volatile after dissolving in a pool and forms  $\text{CH}_3\text{I}$  again at the surface of water pool. Therefore, a dry-filtration should be installed to remove the  $\text{CH}_3\text{I}$ . In this study, we investigate the characteristics of zeolite filtration methods for the removal of  $\text{CH}_3\text{I}$ . We used both silver ion exchanged ZSM-5-zeolite ( $\text{Ag}^+$ -ZSM-5) to study the effect of silver ion for the removal of iodine from  $\text{CH}_3\text{I}$ . This work is to draw a better understanding of organic iodide removal mechanism based on the previous work [1-2]. The details of test procedures and results are reported in [1-2]. In this paper, the newly investigated mechanism is provided to improve the previous analysis in [2].

### 2. Discussion on the mechanism

The organic iodide is captured in the pores of zeolite in either chemically or physically. This kind of capture method could be explained by a host-guest chemistry as shown in Fig. 1.

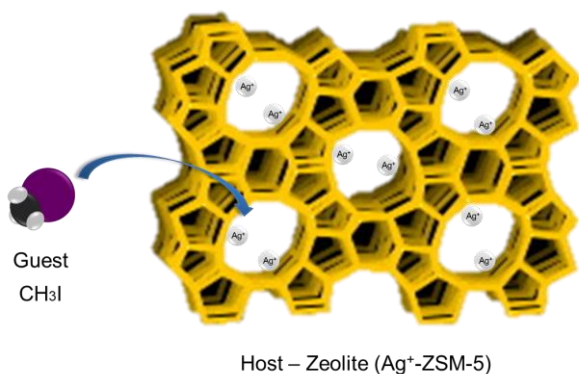


Fig 1. Host-guest chemistry of organic iodide capture

As shown in Fig.1, the pore structure of zeolite acts as a host for an incoming organic iodide whose role is a guest. To accommodate the “guest” in the “host”, the host zeolite should have an acceptable space in its pore structure. The size of zeolite pore and organic iodide are given in Fig. 2.

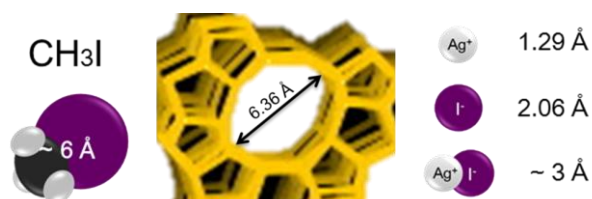


Fig. 2 Size of zeolite pore and organic iodide

The size of organic iodide is around 6 Å and the size of zeolite pore is slightly bigger than the size of organic iodide. However, the zeolite pore is partially filled by a silver ion whose size is 1.29 Å as shown in Fig. 2. The most puzzling question to understand the adsorption behavior of iodide would be how many silver ions participate in the chemisorption reaction by forming a silver iodide ( $\text{AgI}$ ) with the iodide from  $\text{CH}_3\text{I}$ .

Azambre and Chebbi found that two silver ions react with one iodide by forming a silver iodide in their organic iodide capture test with a silver ion exchanged zeolite as shown in Fig. 3.

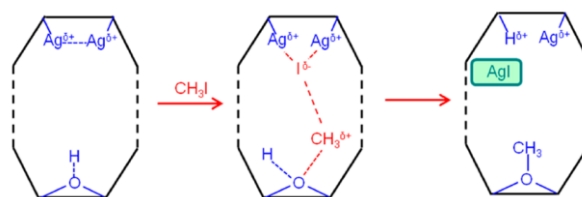


Fig. 3 AgI formation in the zeolite pore [3]

However, it is still puzzling why only half of silver ion reacts with iodide and the other half remains in the zeolite pore without reaction. No explain for the left half silver ion was provided in [3]. It seems that the left half silver ion might be in an inaccessible site for the iodide capture.

The previous assumption in [2] by authors was that one silver ion reacts with one iodide, and we calculated the absorbed iodide mass in the zeolite based on the mass balance equation of  $\text{Ag}^+ + \text{I}^- \rightarrow \text{AgI}$ . In this assumption, we obtained a higher mass gain of iodide than the actual mass gain of iodide, and it led to a negative value of physisorption amount for 90, 120, 150°C of tests. In the previous analysis in [2], this negative value was understood as the desorption amount in rather elevated temperature. However, if iodide is chemically bound with silver ion, it cannot be easily broken to be fallen. For this reason, the negative amount of physisorption amount seems not to be in physical, and this needs to be corrected by a new assumption based on the finding in [3] which is binuclear reaction between silver ion and iodide.

As provided in [2], the 20 mg of  $\text{Ag}^+$ -ZSM-5 was used in each test and the 7.2wt% of silver ion is exchanged in the total mass of sample. Therefore, 1440  $\mu\text{g}$  of silver ion is loaded in the initial sample. If half of silver ion (720  $\mu\text{g}$ ) reacts with iodide and forms the silver iodide, 847.07  $\mu\text{g}$  of iodide should be captured by forming the silver iodide. Therefore, the chemisorption mass gain of iodide by forming a silver iodide would be 847.07  $\mu\text{g}$  for all temperature performed in this test as shown in Fig. 4.

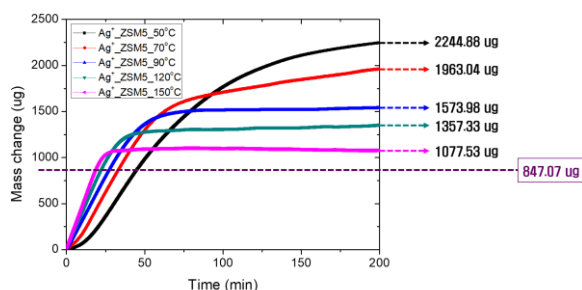


Fig. 4 Mass change curves of organic iodide capture test

In the assumption of binuclear reaction between  $\text{Ag}^+$  and  $\text{I}^-$ , the physisorption amount can be obtained by subtracting the chemisorption amount of 847.07  $\mu\text{g}$  from the actual mass gain for each temperature. The physisorption amount for each temperature is 1397.81 (50°C), 1115.97 (70°C), 726.91 (90°C), 510.26 (120°C), and 230.46  $\mu\text{g}$  (150°C). The lower temperature, the higher physisorption of iodide in the zeolite is obtained.

### 3. Conclusions

In summary, the revised chemisorption amount and physisorption amount are calculated from the previous work [2] to understand the adsorption behavior of organic iodide in the silver ion exchanged zeolite. The

modeling of organic iodide adsorption is ongoing based on the improved understanding of silver and iodide reaction.

### ACKNOWLEDGEMENT

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### REFERENCES

- [1] Sanggil Park, Joonyoung Sung, Ki-ppeum Kim, Jaeyoung Lee, Organic iodide capture using a zeolite dry filtration, Transaction of the Korean Nuclear Society Spring Meeting, Jeju, Korea, May 18-19, 2017.
- [2] Sanggil Park, Ki-ppeum Kim, Jaeyoung Lee, A study of organic iodide removal mechanism using a silver ion exchanged zeolite, Transaction of the Korean Nuclear Society Autumn Meeting, Jeju, Korea, October 26-27, 2017.
- [3] Bruno Azambre, Mouheb Chebbi, Evaluation of silver zeolites sorbents towards their ability to promote stable  $\text{CH}_3\text{I}$  storage as  $\text{AgI}$  precipitates, Applied Materials and Interface 9 (2017) 25194-25203.