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Radiotracer study for quantitative evaluation of gas-hydrate based radioactive wastewater purification system

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1. Introduction

This study aimed to explore the feasibility of optimizing the gas hydrate-based decontamination process. The gas hydrate process can eliminate all contaminants except pure water in a single step without generating secondary pollutants. To evaluate this process, experiments using open radioactive isotopes as tracers were conducted [1].

2. Methods and Results

2.1 Gas hydrate reactor and radiotracer

A batch-type reactor with an impeller (4 L) was used, and a solution was prepared to form gas hydrates under conditions of 3°C and 2.0 atm. HFC-125a (1,1,1,2,2-pentafluoroethane) was used as the guest gas for hydrate formation [2]. About 30 minutes after the reaction started, hydrate slurry resembling ice slurry was visually observed through a reactor window. Before the reaction, radioactive isotopes (^{99m}Tc and ^{131}I) were introduced into the reactor with a NaCl 3.5 wt% solution.

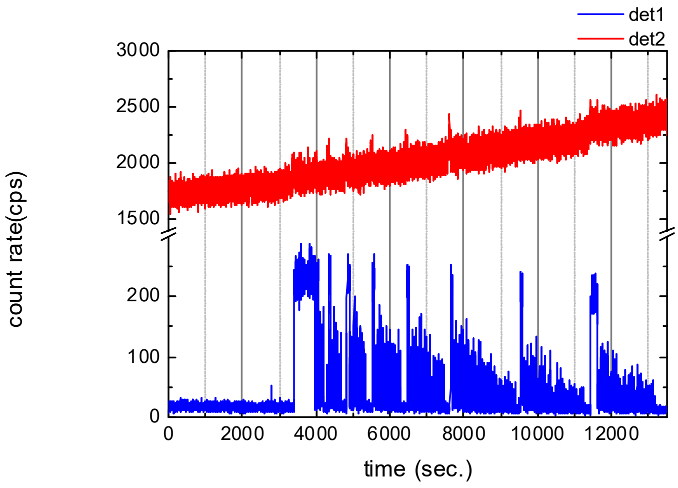

Fig. 1. Radiotracer response curves from the NaI detectors installed on the reactor and over the returning water pipeline.

The gamma-ray intensity variations, corresponding to the behavior of the radioactive tracers, were measured using NaI(Tl) detectors installed on the reactor's side, bottom, and pipelines for effluent and recirculation. The final decontamination efficiency was analyzed with a $LaBr_3$ detector housed in a 1-inch-thick Pb shield [3].

2.2 Optimization of palletization

The gas hydrate slurry was compressed at the reactor's bottom to form solid crystalline pellets. Excess liquid with traces of unreacted substances from the compression process was recirculated back into the reactor. The gas hydrate crystals formed as pellets consist solely of pure water molecules (H_2O, host) and gas molecules (guest), naturally excluding impurities during the pellet formation. This enables the effective treatment of highly concentrated wastewater, which conventional methods fail to purify. During the experiment, pure water corresponding to 10% of the original water, with salts and radioactive isotopes removed, was recovered in the form of gas hydrate pellets. The concentration of radioactive tracers increased in the reactor's upper section over time, which was attributed to the repetitive downward movement of the hydrate slurry.

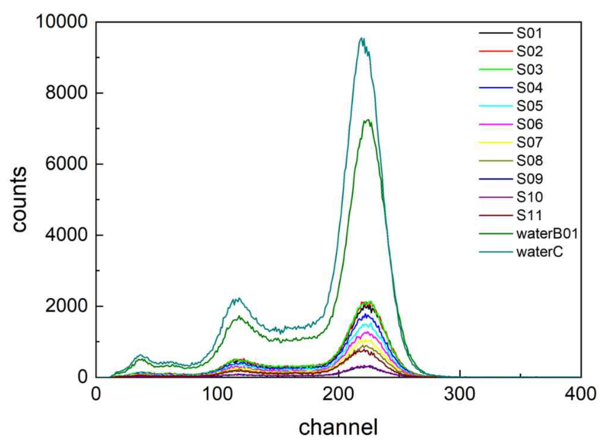

Fig. 2. Variation of radiotracer activities according to hydrate pellet melting percentages.

The experiment achieved a decontamination efficiency of 72 %. The tracer concentration varied in the recirculation pipeline during the compression process. To confirm the impact of impurities trapped within the crystals, a melting step was added, allowing partial melting of the pellets. The analysis showed that the decontamination efficiency increased to 88% at a 50 % melting level. In subsequent experiments, the slurry and impurity separation process was replaced with a micro jet-water wash method, improving the decontamination efficiency to 97.9 %.

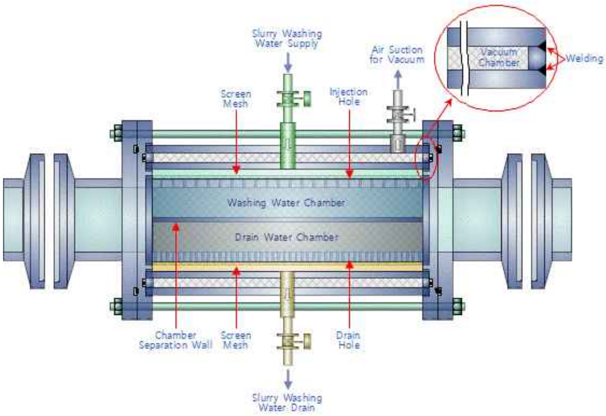

Fig. 3. Slurry washer for continuous gas-hydrate production.

2.3 Effect of guest gases on hydrate purity

Further experiments were conducted to compare the decontamination efficiency of cationic radioactive isotopes using $^{68}Ga^{3+}$. The hydrate experiment with HFC-125a gas yielded less than 50 % decontamination efficiency. However, when HFC-152a (1,1-difluoroethane) gas was used as the guest, similar decontamination efficiencies to those observed with ^{99m}Tc and ^{131}I were achieved. This result suggests that the electronic density differences of the guest gas molecules influence hydrate formation and that hydrate structures also vary depending on the type of guest gas. Further analysis is ongoing, and a pilot-scale demonstration capable of treating 5 tons/day is under development.

3. Conclusions

Radioisotopes such as ^{99m}Tc, ^{131}I, and ^{68}Ga were employed as tracers to evaluate the effectiveness of a gas-hydrate based water purification system. Initially, the purification efficiency was measured at approximately 70%. However, it was found that the compression step resulted in returning water containing significant amounts of radioisotopes. By implementing a hydrate surface melting process, the removal efficiency increased to about 88%. Further enhancement was achieved by introducing a micro jet-water slurry wash method in the continuous purification system, which exhibited a notably higher removal efficiency of 97.9%. The chemical composition of the guest gas was observed to affect the purification of cationic radioisotopes, indicating the need for further research to understand the influence of the guest gas's electron density on the behavior of target ions.

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