

CATALYTIC PROCESS FOR TRITIUM EXCHANGE REACTION

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Abstract

The catalytic activities for a hydrogen isotope exchange were measured through the reaction of a vapor and gas mixture. The catalytic activity showed to be comparable with the published data. Since the gas velocity is relatively low, the deactivation was not found clearly during the 5-hour experiment. Hydrogen isotope transfer experiments were also conducted through the liquid phase catalytic exchange reaction column that consisted of a catalytic bed and a hydrophilic bed. The efficiencies of both the catalytic and hydrophilic beds were higher than 0.9, implying that the column performance was excellent.

1. INTRODUCTION

Tritium concentrations in the moderator and the heat transport systems of a CANDU reactor have been increased due to the neutron irradiation of heavy water. This leads to additional public exposure, which subsequently requires the tritium removal facility (TRF) at the PHWR site to reduce the tritium concentration to the desired extent.

The liquid phase catalytic exchange (LPCE) process combined with cryogenic distillation (CD) is one of the available processes for practical application. A hydrophobic catalyst is essential in the LPCE process for preventing the dramatic reduction of the catalytic activity during a reaction, and it has been improved by Atomic Energy of Canada Limited (AECL) since the 1970s [1]. Recently, KAERI/KEPRI have also developed a hydrophobic catalyst consisting of polystyrene support and platinum. The catalyst has enough surface area and water expelling property for forwarding the gas-vapor-water reaction adequately.

The overall reaction for transferring a hydrogen isotope from the liquid phase to the gas phase consists of two steps: a catalytic reaction at the catalytic bed in gas-vapor phases, and a mass transfer reaction at the hydrophilic bed in vapor and liquid. The efficiency of the LPCE column is, therefore, dependent on both the catalyst and the hydrophilic packing material.

This study describes the performance of the catalyst and the LPCE column. The catalytic activity was measured by the vapor-gas reaction in separately designed reaction column. The overall reaction efficiency was also measured through a column in which each stage was comprised of a catalytic bed for the gas-vapor reaction and a hydrophilic bed for the mass transfer between vapor and liquid.

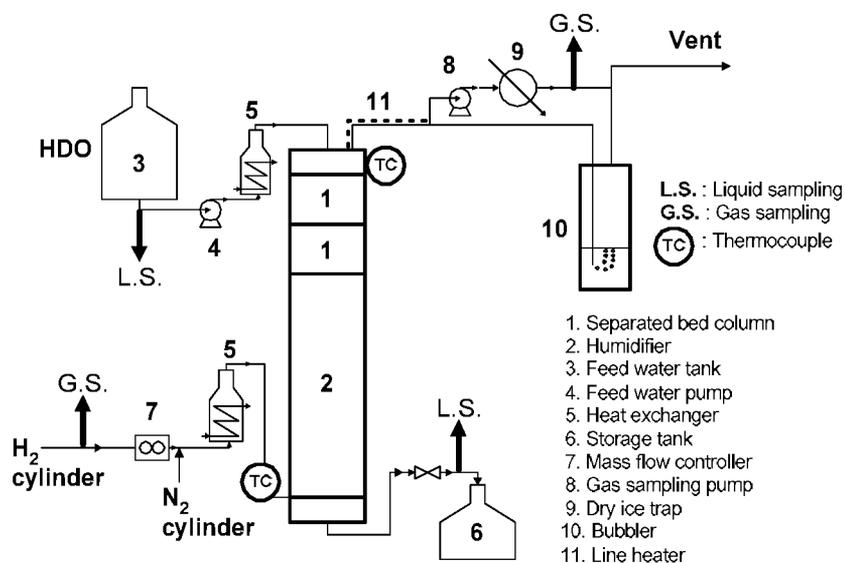


FIG.1. Schematic view of experimental apparatus for hydrogen isotope exchange reaction.

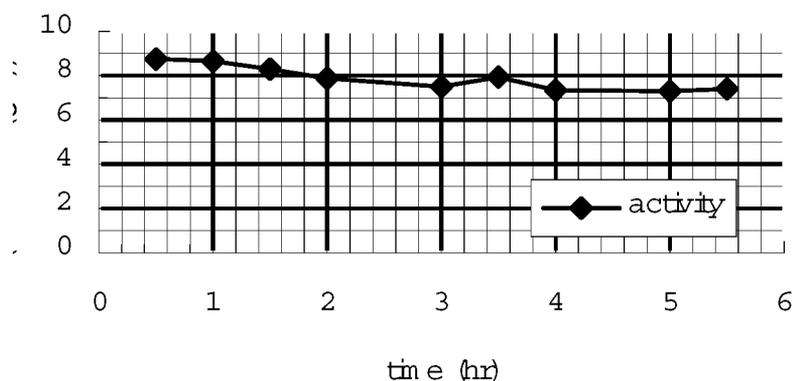


FIG.2. The catalytic activity change with time.

2. EXPERIMENTAL

The experimental apparatus for the LPCE column is shown in Fig. 1. The column included a humidifier to saturate the inlet gas, and two stages in which both the catalyst bed and the hydrophilic bed were configured. The deuterium in heavy water streamed in from the top of the column and the hydrogen gas flowed up from the bottom of the column, resulting in the transfer of deuterium from the heavy water to the hydrogen gas. The outlet concentration of deuterium in the hydrogen gas was measured by a gas chromatograph to analyze the column performance.

Catalytic activity was measured in a similar column. It contains a catalytic bed and a saturator. The water was vaporized at the saturator under the catalyst bed, then mixed with the hydrogen-deuterium gas, and the mixture flowed up to the catalyst bed. The outlet deuterium concentration in the hydrogen gas mixture was measured to determine the catalytic activity.

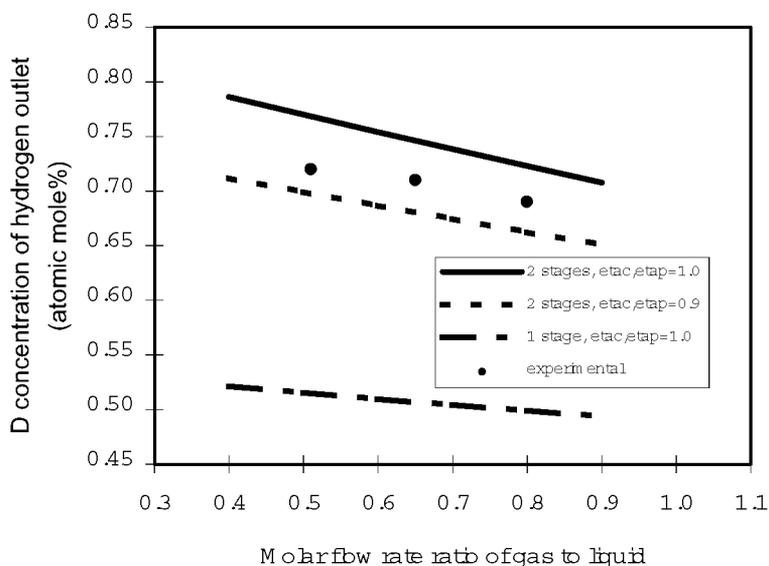


FIG. 3. Experimental results for hydrogen isotope exchange reaction.

3. RESULTS AND DISCUSSION

The catalytic activity, as shown in Fig. 2, kept its activity invariant for about 5 hours. The measured results are comparable with other published data [2] even though the experimental conditions are a little different. The hydrogen velocity in this experiment is 0.1 m/s that is relatively slower than the data [2] where the velocity is higher than at least 0.15 m/s. This explains that the catalytic activity in this figure has not significantly changed with time, while the data [2] showed remarkable catalytic deactivation with time due to a more severe experimental condition for catalytic deactivation.

Model calculations for the outlet concentration of deuterium in hydrogen providing the efficiencies of the catalytic bed, η_c and the hydrophilic bed, η_p are available elsewhere [3,4]. Fig. 3 shows the experimental results of the outlet concentration of deuterium, and compares them with the computed results carried out with assumed η_c and η_p values. The column efficiency increased with the number of stages, as shown in the figure. The increase of the molar ratio of gas to liquid also raised the column efficiency. The experimental results lay between the computed results with $\eta_c=1.0$ and $\eta_p=1.0$ and the results with $\eta_c=0.9$ and $\eta_p=0.9$. This implied that the column performance with respect to the internal design configuration was excellent.

4. CONCLUSION

Activity of the catalyst manufactured by KAERI/KEPRI was measured and compared with the published data. The results were satisfactory. The catalytic activity has not been significantly changed for 5 hours, for the gas velocity is relatively low. LPCE column experiments were conducted and the model calculation was used to compare experimental results and calculation results. The comparison showed that both the efficiencies of the catalytic bed and hydrophilic bed are higher than 0.9, implying the column performance was fair.

REFERENCES

- [1] BUTLER, J.P., et al., AECL-6055, Canada (1978).
- [2] SHON, S.H., et al., KEPRI Technical Report, TR.96NJ18.J1998.45, Korea (1998).
- [3] SHIMIZU, M., et al., J. Nucl. Sci. and Tech., 17, 448(1980).
- [4] SHIMIZU, M., et al., J. Nucl. Sci. and Tech., 20, 36(1983).