

Trapping of Tritium in Off-gas during Dry-processes

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1. Introduction

Tritium, a weak beta emitter, is produced in nuclear power reactors during the fission of heavy nuclei and by neutron interaction with coolants, moderators and some light elements, such as lithium, beryllium and boron [1]. And it is necessary to be controlled its production at nuclear facilities owing to its relatively long half-life, high residence time in the environment, high isotopic exchange rate and ease of assimilation into living matter.

The amount of tritium produced through neutron reactions in the coolant and moderator is so small that the tritiated effluents from them can be released directly to the environment without additional processing, while the portion of tritium produced in the fuel element is typically ~60%. About 50% of the tritium produced in the fuel element reacts with the zircaloy cladding to form a hydride. The remainder is arrested in the fuel, possibility to be released to environment in the form of HT or T₂ during dry processes such as Voloxidation, OREOX (Oxidation and Reduction) process, Pyrochemical processing and DUPIC (Direct Use of PWR fuel in CANDU Reactors) fuel fabrication process.

In the LWR fuel reprocessing plant, most of the tritium arrested in the fuel and cladding appears in the liquid waste streams in the form of HTO during dissolution in nitric acid, finding 0.5% of them in the dissolver off-gas stream.

Considering processes for recycle of spent nuclear fuel and extending the operation of disposal site are UREX and UREX+ of wet processes and pyrochemical processing of a dry process. The prior subject in the wet processes relating the tritium is treatment of HTO in liquid state. But no appropriate technique is developed for dry processes such as pyrochemical processing.

This paper intends to give a solution for trapping the tritium anticipated to be released during the dry processes such as Voloxidation, OREOX, Pyrochemical processing and DUPIC fuel fabrication processes.

There are two processes for capturing the tritium, one is removal of HT in gaseous state, and the other is removal of HTO in liquid or vapor state.

The removal of HT employs hydrogen getters, these are normally non-oxidative processes. Getters typically consist of zirconium alloys and compounds. However, the selection of the appropriate getter depends greatly on the other constituents of the gas. The fact that most getters will not tolerate oxygen in the feed stream greatly limits the usefulness of getters for most effluent streams.

The removal of HTO is removing tritium from gas streams involves conversion of HT to HTO followed by the removal of the HTO. A number of methods exist for removing HTO vapor from gaseous streams, depending on the degree of removal required.

This study aims removal HTO by molecular sieve after converting HT in the cupric oxide. Few papers are reported on the trapping of tritium during the dry processes except design criteria for the reprocessing plant in JAERI [2].

2. Experiments

To convert H₂ gas into H₂O vapor, 99.9% purity of CuO granule powder (Japan High Pure Chem. Co.) with a size of 2~5mm is used as a catalyst. This powder was filled up as high as needed in the stainless steel column dimensioned 20mm in diameter and 300mm in height. The converting column is manufactured to be heated by 550 °C.

To simulate the tritium gas releasing during voloxidation and/or puncturing, mixed gas of 10,000 ppm H₂ with nitrogen is fed to the converting column through MFC. Voloxidation process is normally carried out at the air condition, the stock gas was fed with air in the concentration ranges of 1000 ppm to 9000 ppm.

Converted H₂O vapor is passed through the Molecular sieve tower to removal the H₂O. The specifications of Molecular sieve tower is as follows.

- Dimension and material: 32 mm I.D. × 320mm length (250 ml), visible Acryl
- Absorbent: molecular sieve 13X (60%) + 4A (40%, indicator)
- Size and type of absorbent: 8-12 mesh (1.68 ~ 2.38 mm), granule type
- Efficiency: (by spec.): 25mL of water in 160g of absorbent

Concentrations of converted H₂O and un-reacted H₂ were monitored on-line by Gas Chromatographys (Multi-GC 2000, Donam Instrument Co. Korea) each. Fig. 1 shows a layout of tritium trapping experimental apparatus in lab-scale.

3. Result and Discussion

The result showed that almost 100% H₂ gas is converted into H₂O in a CuO converting column filled with 100 mm height (about 280g) of CuO at 450 °C, 1 L/min flow rate of hydrogen concentration ranges of 1000 ppm to 9000 ppm, as shown in Fig. 2. Converted

H₂O is perfectly absorbed at the Molecular sieve tower, resulting no H₂O in the effluent.

REFERENCES

[1] McKAY, H.A.C., Tritium immobilization, Eur. Appl. Res. Rep., Nucl. Sci. 1 (1979) 599- 711.

[2] Gunzo Uchiyama et al., Development of Voloxidation Process for Tritium Control in Reprocessing, JAERI-M91-199, 1991.

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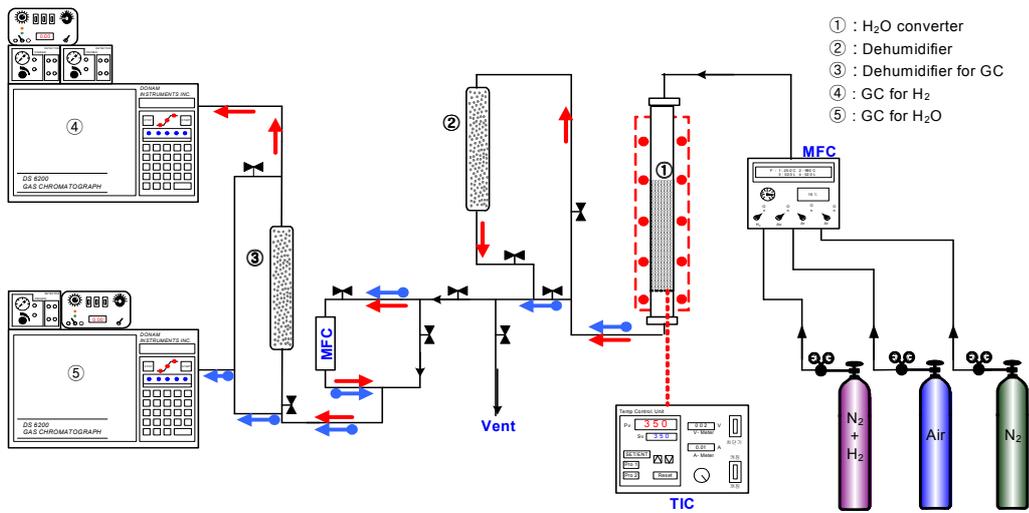


Fig. 1 Layout of tritium trapping experimental apparatus.

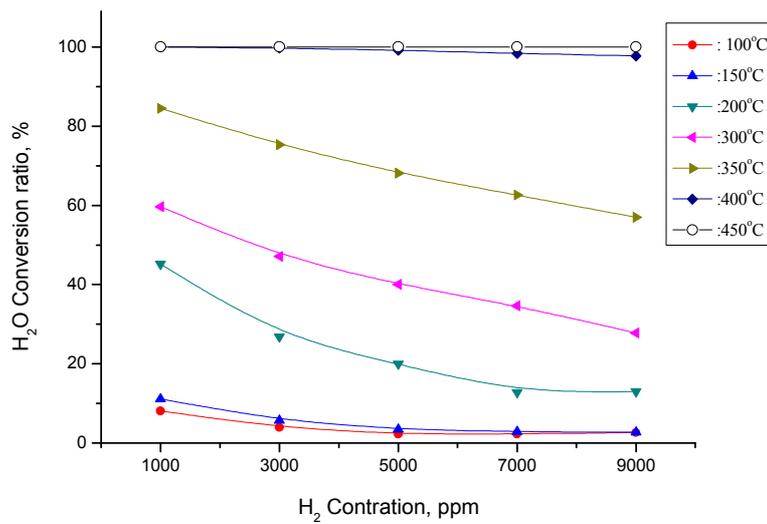


Fig. 2 H₂O converting ratio according to reaction temperatures and hydrogen concentrations.