Applied Radiation and Isotopes 114 (2016) 40–44

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

# Experimental investigation on feasible bioreactor using mechanism of hydrogen oxidation of natural soil for detritiation system

Yuki Edao*, Yasunori Iwai, Katsumi Sato, Takumi Hayashi

*Japan Atomic Energy Agency, 2-4 Shirane, Shirakata, Tokai-mura, Naka-gun, Ibaraki 319–1195, Japan*

## HIGHLIGHTS

- A bioreactor for tritium oxidation by soil at room temperature was studied.
- The efficiency of a bioreactor was evaluated by kinetics.
- A bioreactor was superior to a catalytic reactor generally used in tritium oxidation.
- A bioreactor with soil bacteria has the feasibility for a tritium oxidation reactor.

## ARTICLE INFO

*Article history:*
Received 26 January 2016
Received in revised form
18 April 2016
Accepted 3 May 2016
Available online 4 May 2016

*Keywords:*
Bioreactor
Tritium
Hydrogen oxidation
Soil
Hydrogenoxidizing
Bacteria
Detritiation

## ABSTRACT

A passive reactor for tritium oxidation at room temperature has been widely studied in nuclear engineering especially for a detritiation system (DS) of a tritium process facility taking possible extraordinary situation severely into consideration. We have focused on bacterial oxidation of tritium by hydrogen-oxidizing bacteria in natural soil to realize the passive oxidation reactor. The purpose of this study was to examine the feasibility of a bioreactor with hydrogen-oxidizing bacteria in soil from a point of view of engineering. The efficiency of the bioreactor was evaluated by kinetics. The bioreactor packed with natural soil shows a relative high conversion rate of tritium under the saturated moisture condition at room temperature, which is obviously superior to that of a $Pt/Al_2O_3$ catalyst generally used for tritium oxidation in the existing tritium handling facilities. The order of reaction for tritium oxidation with soil was the pseudo-first order as assessed with Michaelis-Menten kinetics model. Our engineering suggestion to increase the reaction rate is the intentional addition of hydrogen at a small concentration in the feed gas on condition that the oxidation of tritium with soil is expressed by the Michaelis-Menten kinetics model.



## 1. Introduction

The multiple confinement concept has been applied to existing tritium process facilities. In an event of tritium leakage into the facilities, leaked tritium will be removed by a detritiation system (DS) to avoid the release of tritium into the environment. A nuclear fusion reactor has been researched under the international collaboration for a future energy source, where a large amount of tritium will be mandatorily processed over the facility. In order to gain public acceptance of the nuclear fusion reactor facility, the DS will play an extremely important role. The existing DS consists of catalytic reactors to combust leaked tritium to tritiated vapor and adsorption columns to remove tritiated water vapor (Naruse et al., 1990). The catalytic reactors for the existing tritium facilities have been operated at an elevated temperature to ensure the best efficiency. In contrast the passive catalytic reactor which can combust tritium at room temperature should be developed for the DS of a future nuclear fusion reactor facility, because the passive catalytic reactor without requiring heating will considerably contribute to enhancing safety of the facility. The future nuclear fusion reactor facility will handle a kilogram level of tritium as a fuel. Hence the DS should be designed carefully taking the possible extraordinary situations into consideration.

It is a well-known issue that a passive reactor to oxidize tritium at room temperature in the presence of moisture is hopeless in a case using commercial platinum catalyst. It has been well demonstrated that oxidation efficiency strongly depends on the concentration of hydrogen at room temperature (Iwai et al., 2011). The overall reaction rate constant decreases with a decrease in

* Corresponding author.
*E-mail address:* edao.yuki@jaea.go.jp (Y. Edao).

http://dx.doi.org/10.1016/j.apradiso.2016.05.003
0969-8043/

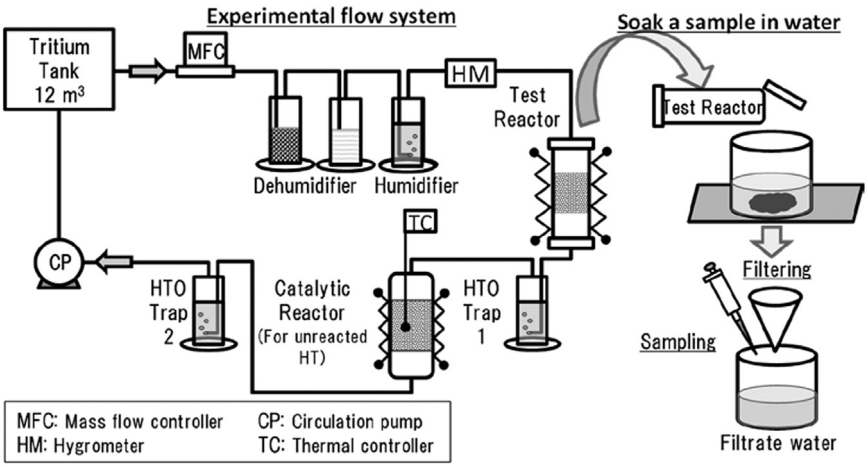


**Fig. 1.** Experimental flow system for measurement of the efficiency of tritium oxidation in an oxidation reactor packed soil.

hydrogen concentration. In addition, the overall reaction rate constant decreases with an increase in moisture concentration. The water layer formed on the active sites of the catalyst and the layer considerably blocks the transfer of hydrogen to the active sites. To enhance the efficiency under a very small concentration of tritium and a large concentration of moisture, the catalytic reactor for the existing tritium facility has been obliged to heat up to an elevated temperature around 473 K. In recent years, a lot of efforts have been put into developing a special catalyst to improve the efficiency of tritium oxidation at room temperature in the presence of moisture (Iwai et al., 2011).

Looking at the issue from a different point of view, we have focused on bacterial oxidation of tritium by hydrogen-oxidizing bacteria in natural soil. It is interpreted that hydrogenase acts the enzyme to oxidize tritium in hydrogen-oxidizing bacteria. One can safely state that the best condition for hydrogen-oxidizing bacteria is humid at room temperature. As demonstrated previously by other researchers, various kinds of natural soil can efficiently oxidize a small concentration of tritium (Conrad and Seiler, 1981; Klüber et al., 1995; Komuro et al., 2002; Wilson et al., 1952). From a point of view of biology and environmental science, the difference in hydrogen oxidation efficiency among natural soils has been investigated (Ichimasa et al., 1988, 1999; Komuro et al., 2002; McFarlane et al., 1978; Momoshima et al., 1990; Ota et al., 2008; Sweet and Murphy, 1981). The dependencies of pH, temperature and moisture concentration on oxidation efficiency have been discussed (Schuler and Conrad, 1991; Ota et al., 2008). The previous investigations were mainly conducted as studies on behavior of tritium in the environment. The literature concerning on feasible bioreactor for the DS is limited (Ichimasa et al., 2005).

The purpose of this study was to examine the feasibility of a bioreactor as a passive reactor for the DS. We focused on the oxidative capacity of soil bacteria. The tritium oxidation experiments were carried out using a bioreactor packed with natural soil. The efficiency of the bioreactor was evaluated from a point of view of kinetics.

## 2. Experimental

### 2.1. Selection of natural soil

The natural soil having a high activity of bacterial oxidation of tritium was applied to investigate the feasibility of a passive bioreactor. In order to make clear the superiority of the bioreactor as a passive reactor, (1) a commercial $Pt/Al_2O_3$ manufactured by N. E. Chemcat, Japan was prepared as a comparison. In this experiment, the samples of soil were collected from (2) a grassy pine forest, (3) a dense grassy field, (4) a sparse grassy field. Previous research has suggested that hydrogen-oxidizing bacteria inhabits everywhere in natural soil, not in a special environment (Komuro et al., 2002). It has been extensively reported that soil accumulated with fallen leaves and growing plants has high activity of hydrogen oxidation because the soil was habitable to soil bacteria (Ichimasa et al., 1988, 1999). Since earlier studies have shown that tritium oxidation activity was quite high in the surface soil, all the soil were sampled 0–5 cm in depth from the surface at Ibaraki, Japan in this study (Ichimasa et al., 1999). It is known that the soil itself has a capacity to exchange hydrogen isotopes between tritium and moisture (Häring and Conrad, 1994). To distinguish bacterial oxidation of tritium from isotope exchange, (5) compost artificially manufactured was prepared. The compost had been treated with heat, therefore hydrogen-oxidizing bacteria were completely killed. By using the compost, the effect of isotope exchange was evaluated.

### 2.2. Tritium oxidation test by an oxidation reactor packed soil

The conversion rate of tritium by a bioreactor packed with soil was measured by the experimental flow system schematically shown in Fig. 1. Air loaded with molecular tritium (HT) was supplied from a tank of 12 $m^3$ to the experimental flow system. The volume is large enough that the tritium concentration in the tank negligibly decreased in an experiment. We applied the circulation flow system to minimize the decrease in tritium concentration in the tank as much as possible by returning the unreacted tritium into the tank again. The concentration of the tritium in air was approximately 0.37 GBq/$m^3$. It is true that a certain concentration of hydrogen is naturally contained in air. The concentration is around 0.5 ppm. We have not removed the natural hydrogen from the air in the tank. Although the radioactivity of tritium used in the experiments was large, the hydrogen concentration of tritium as a hydrogen isotope was negligibly small against natural concentration in the air. In the experiments, we have evaluated the kinetics of bioreactor for the natural hydrogen concentration using tritium tracer. Moisture in the air was removed by two dehumidifiers packed with silica gel beads and molecular sieves 5 A pellets, respectively. In the case of addition of water vapor to check the effect of moisture, saturated water vapor was added in the test reactor through a humidifier of a water bubbler installed after the dehumidifier. The gas flow rates were controlled with a mass flow controller (8500MC, KOFLOC, Japan) to be 100 – 1200 $cm^3$/min.