

## Removal of hydrogen produced from fission process using $\text{CuO}_x$ impregnated in porous alumina ball

Myunggoo Kang, Taewoon Kim, Seung-Kon Lee, Suseng Lee, and Jun Sig Lee \*

Division of Radioisotope Research, Korea Atomic Energy Research Institute, 989-111 Daedok-daero, Yseonggu, Daejeon 34057, Republic of Korea

\*Corresponding author: jlee15@kaeri.re.kr

### 1. Introduction

$^{99m}\text{Tc}$  radioisotope, the daughter of  $^{99}\text{Mo}$ , is the most widely used medical radioisotope in the world.  $^{99}\text{Mo}$  has been almost generally produced from the fission of high-enriched uranium (HEU).<sup>1, 2</sup> However, the increasing concern over nuclear proliferation is leading to new research in order to use low-enriched uranium (LEU) instead of HEU.<sup>2</sup>

Currently, most commercial producers of  $^{99}\text{Mo}$  use highly concentrated alkaline digestion of uranium target.<sup>3</sup> From this process, various gases such as xenon, iodine, hydrogen, etc. are released during the dissolution of the irradiated target.<sup>4</sup> Therefore, all the off-gas produced from the dissolver unit should be collected in the gas storage tank during the decaying period of radioactive gases. The total volume of off-gas is significantly large. To decrease the volume of all off-gas, the produced hydrogen was oxidized to water on copper oxide under the heating condition.

Generally, the copper oxide acted as hydrogen-oxidation catalyst was used in bulk form.<sup>5</sup> Practically, the conversion of hydrogen was induced on the oxygen of surface. In this regard, we designed the nanosize of copper oxide impregnated in the porous alumina ball (PAB) in order to maximally capitalize on the effect of catalyst. Also, the hydrogen removal test was performed by using the handmade column furnace.

### 2. Methods and Results

The copper oxide impregnated in porous alumina ball ( $\text{CuO}_x\text{-PAB}$ ) was synthesized by simply annealing method (Fig.1). The hydrogen removal test of the obtained  $\text{CuO}_x\text{-PAB}$  was performed by using the handmade column furnace.

#### 2.1 Synthesis of $\text{CuO}_x\text{-PAB}$

In order to remove the water absorbed on surface of PAB, the PAB was dried by oven at 140 °C for overnight. In a typical synthesis process, 400 mL of Cu aqueous solution (4.47 M) was completely absorbed in the 1.0 kg of the dried PAB. The as-prepared sample was calcinated in furnace at 500 °C for 1 hour under ambient condition. The above processes were performed twice more. At this time, the volumes of Cu aqueous solution were adjusted to 350 mL and 300 mL, respectively.

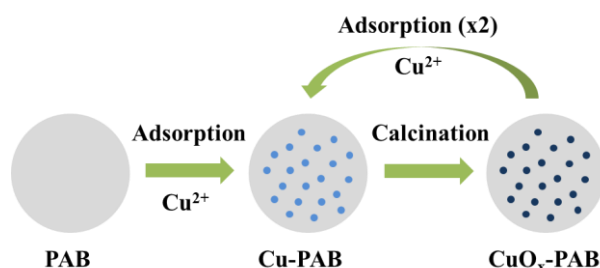


Fig. 1. Schematic diagram for preparing  $\text{CuO}_x\text{-PAB}$ .

#### 2.2 Hydrogen removal test of $\text{CuO}_x\text{-PAB}$

The removal test of hydrogen was set and conducted by handmade column furnace. Fig. 2 shows the set-up of the equipment for the test. Firstly, we filled the 8.2 kg of the obtained sample (the weight portion of Cu is 21.77%) in the column. Before the conversion test, the  $\text{CuO}_x\text{-PAB}$  in column was subjected to pre-annealing under 400 mL  $\text{min}^{-1}$  flow of He gas and 10.0 L  $\text{min}^{-1}$  flow of  $\text{O}_2$  gas at 300 °C for 2 h to completely oxidize the remainders of Cu ion or metal. Then, the cylinder of  $\text{O}_2$  gas was turned off and the flow of He gas was maintained. The oxidation process of hydrogen in column was facilitated by introducing 5.0 L  $\text{min}^{-1}$  flow of  $\text{H}_2$  gas under 400 mL  $\text{min}^{-1}$  flow of He gas as the carrier gas. The temperature of each area was measured by the thermocouple.

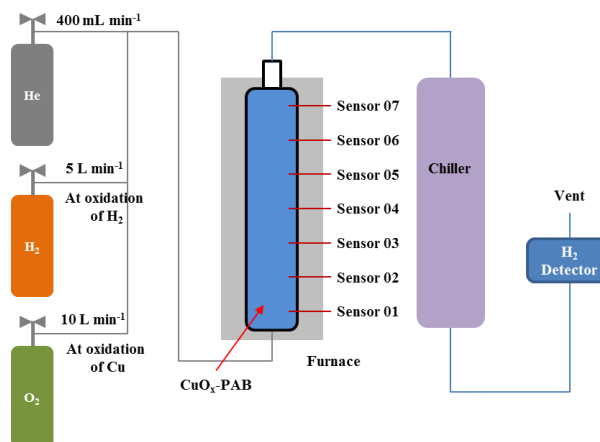


Fig. 2. Schematic diagram of the set-up for removal test of hydrogen.

Fig. 3 shows that the temperature of each part was varied with the passage of time. The oxidation of

hydrogen is an exothermic reaction. Thus, we determine the area which the hydrogen's oxidation was produced through the increment of temperature. The sequential change of temperature from sensor 01 to 07 suggests that the  $\text{CuO}_x$  in sample was gradually converted from beneath.

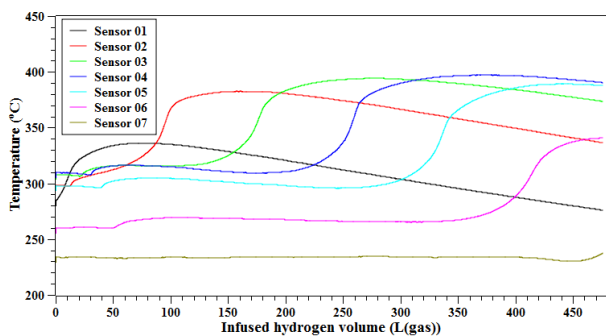


Fig. 3. The temperature curves of each sensor while infusing  $\text{H}_2$  gas at  $5.0 \text{ L min}^{-1}$  flow.

We determined the breakthrough of the loaded sample, when the  $\text{H}_2$  level of vent gas was passed over at 1.0 %. Consequentially, totally volume of the removed hydrogen was approximately 455 L. This value presents 72% of the calculated value (approximately 629 L). From these results, we concluded that the synthesized  $\text{CuO}_x$ -PAB has potential as new catalyst in order to remove excess hydrogen during the process of target dissolution.

### 2.3 Stability test of $\text{CuO}_x$ -PAB

To investigate its capacitive stability for the hydrogen removal, the test was frequently examined by alternating between oxidation and reduction of copper. To decrease the time of measurement, also, the test was conducted by reducing the scale of column and increasing flow rate of  $\text{H}_2$  gas.

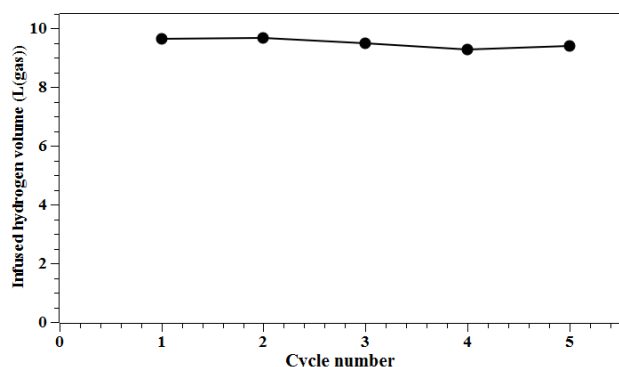


Fig. 4. The cycle capacity of  $\text{CuO}_x$ -PAB.

Fig. 4 shows each volume of removed hydrogen until 5 times. Prolonging the cycle count only slightly changed the capacity of sample, indicating that the

catalyst effect of  $\text{CuO}_x$ -PAB still maintains. We believe that this result was induced by the phenomenon, which the aggregation of  $\text{CuO}_x$  was prevented by the porous structure of alumina ball during oxidation and reduction.

### 3. Conclusions

In conclusion,  $\text{CuO}_x$ -PAB with interpenetrating porous networks and impregnated  $\text{CuO}_x$  were successfully synthesized by the calcination method. The obtained  $\text{CuO}_x$ -PAB showed good catalytic performance for the conversion of from hydrogen to water. Also, this result was maintained in spite of several repeated tests. Based on these results, we believe that the  $\text{CuO}_x$ -PAB has potential as new commercial material for hydrogen removal through more optimization.

### REFERENCES

- [1] Management of Radioactive Waste from  $^{99}\text{Mo}$  Production, IAEA-TECDOC-1051, International Atomic Energy Agency, Vienna, Austria, 1998.
- [2] Fission Molybdenum for Medical Use, IAEA-TECDOC-515, International Atomic Energy Agency, Vienna, Austria, 1989.
- [3] S.-K. Lee, G. J. Beyer, J. S. Lee, Development of Industrial-Scale Fission  $^{99}\text{Mo}$  Production Process Using Low Enriched Uranium Target, Nuclear Engineering and Technology, Vol. 48, p. 613, 2016.
- [4] V.A. Andrade, J.L. Gross, A.L. Maia, The effect of methimazole pretreatment on the efficacy of radioactive iodine therapy in Graves' hyperthyroidism: One-year follow-up of a prospective, randomized study, Journal of Clinical Endocrinology and Metabolism, Vol. 86, p. 3499, 2001.
- [5] A.-H. A. Sameh, Production Cycle for Large Scale Fission  $\text{Mo-99}$  Separation by the Processing of Irradiated LEU Uranium Silicide Fuel Element Targets, Science and Technology of Nuclear Installations, Vol. 2013, p. 14, 2013.