Synthesis of Porous MoO_3 by Chemical Precipitation

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1. Introduction

Molybdenum-99 (Mo-99) and technetium-99m (Tc-99m) are used in about 80 percent of all nuclear medicine procedures worldwide. Not only using for diagnostic imaging, Tc-99m is used for the detection of disease and for the study of organ structure and function. Tc-99m is especially useful for nuclear medicine procedures because it can be chemically incorporated into small molecule ligands and proteins that concentrate in specific organs or tissues when injected into the body.

Almost all of the Tc-99m in nuclear medicine today is produced by radioactive decay of Mo-99. About 88 percent of the Mo-99 decays (red line) produce Tc-99m via the pathway depicted in the Figure 1.

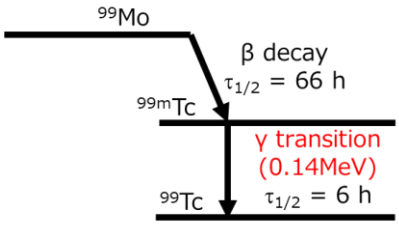


Figure 1. Graphical depiction of the Mo-99 decay chain.

One technique to produce Mo-99 which is used in Tc-99 generators is based on neutron activation of a stable nuclide Mo-98 through the neutron capture reaction:

$$^{98}_{42}Mo + ^{1}_{0}n \rightarrow ^{99}_{42}Mo$$

In our preliminary work, the water solubility of MoO_3 was found to be increased after the neutron irradiation. This phenomenon can lead the reacted Mo-99 / Tc-99m separated and recovered by water. Besides, if MoO_3 powder is more micronized to increase the surface area, the solubility of Mo-99 and Tc-99m in water may be increased. From this point, if a porous MoO_3 target can be produced, it is not necessary to dissolve all the target, and it is possible to recover Mo-99 and Tc-99m simply by pouring water. Furthermore, if it is possible to keep the target in the reactor, a new method can be proposed to recover the reacted Mo-99/Tc-99m simply by circulating water.

In order to produce porous ceramics, there is a known a method of mixing and sintering ceramic powder with NaCl powder and dissolving NaCl in water. In this study, the preparation of porous MoO_3 targets that are water-permeable are described.

2. Experimental procedure

MoO_3 powder (Taiyo Koko Co., LTD, 99.9%) was mixed with NaCl (SIGMA Life Science, 99.9%) at a ratio of 20, 50 and 70 vol %. The mixed powder then was filled into a die with 10 × 10 × 3 mm in size and were pressed with punches at with 20 MPa. Then the die was turned over and the same pressure was applied for 3 minutes for even compaction. After that, heating was performed in using a electric furnace (Nitto Kagaku Co., Ltd., NHK-170) shown in Fig 2, for 3 hours at 500°C with heating rate is 4°C/min. The atmosphere of heating was in the air. The evaluation was carried out by phase identification by X-ray diffraction (RINT-2500HF+PC manufactured by Rigaku) and microstructure observation by a table-top SEM (TM 3000 type Miniscope manufactured by Hitachi High-Technologies). Microstructure observation was made into two places of the target surface and the section divided.

Figure 2 Electric furnace image

3. Results

Figure 3 shows the appearance of the MoO_3 target after heating. The microstructure of samples was observed on the surface and cross section (Figure 4). It can be seen that NaCl was uniformly dispersed on the surface. Figure 5 shows the cross section of 50 vol% NaCl sample with higher magnification. It can be seen that MoO_3 extends columnar around NaCl as a shell.

NaCl amount	20 vol.%	50 vol.%	70 vol.%
Appearance			

Figure 3 MoO_3 appearance after heating

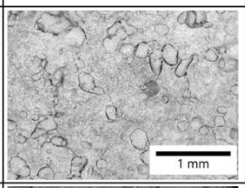
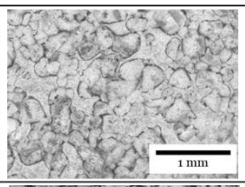
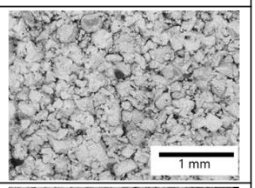
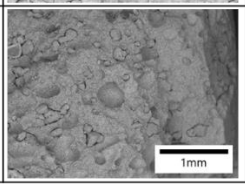
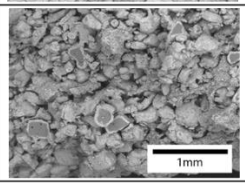
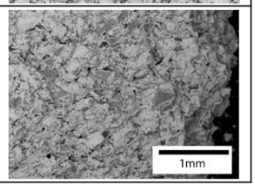
NaCl amount	20 vol.%	50 vol.%	70 vol.%
Surface			
Cross-section			

Figure 4 Microstructure observation

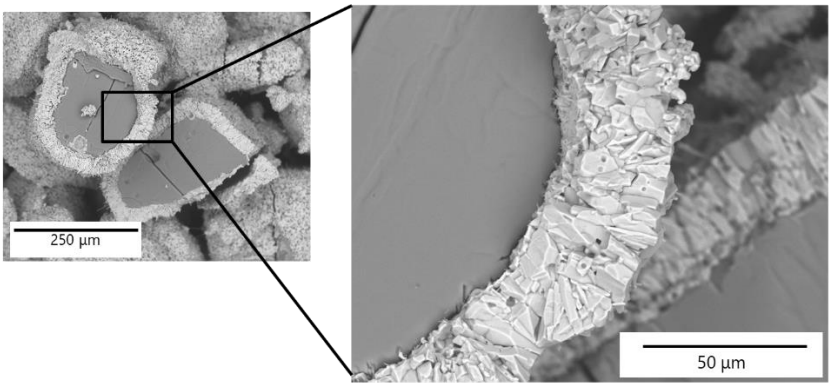
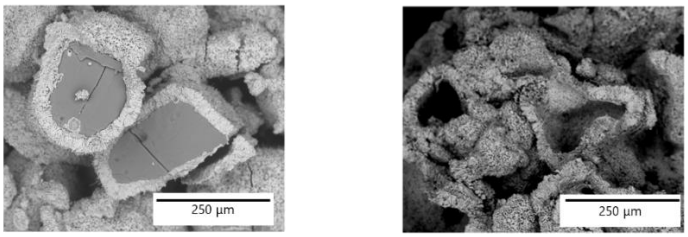


Figure 5 Microstructure image of cross-section with 50% vol.% NaCl

Water was applied to confirm water permeability. Figure 6 shows the result of microstructure observation of the sample after immersing into water. NaCl inside the MoO_3 shell has almost disappeared. From this, it was found that the shell of MoO_3 is permeable to water and water can enter and exit through the shell. Furthermore, it was found that the shell maintained its shape even when it touched water.



a) Before immersing into water b) After immersing into water

Figure 6 Microstructure images a) before and b) after immersing into water

Figure 7 shows the X-ray diffraction results. MoO_3 and NaCl as the main components in 20 vol.% and 50 vol.% samples while Na_2MoO_4Cl was found in 70 vol.% sample. It was suggested that the addition amount of NaCl is preferably less than 70 vol.%. Since the large surface area of the shell be in contacting water is desirable, the addition amount of NaCl is preferably 50 vol.% to 70 vol.%.

Since the particles are grown in columnar form surrounding NaCl, precipitation of MoO_3 form a gas or liquid phase can be considered. The phenomenon such as chemical vapor deposition (CVD) is occurring as an example of this. CVD is a method to deposit the gas containing an element forming a layer on a substrate surface. Here, the reaction process using tungsten (W) oxide of the same group 6 as Mo and NaCl is quoted. Saeki et al. [1] are examining chlorination of tungsten trioxide (WO_3), and intentionally increase the chlorine partial pressure in the experimental environment, tungsten deoxy dichloride (WO_2Cl_2) generated. It has also been reported that $Na_2W_2O_7$ at 140°C, $Na_2W_4O_{13}$ at 350°C, and WO_3 at 480°C from sodium tungstate (Na_2WO_4) [2]. This report also describes that $WOCl_4$ is produced from WO_3 and NaCl. From these reports, a similar reaction occurs in MoO_3, in which MoO_3 reacted with NaCl, and the Mo-Cl compound is precipitated on the surface of NaCl thank for Cl_2 gas and then oxidized to form MoO_3.

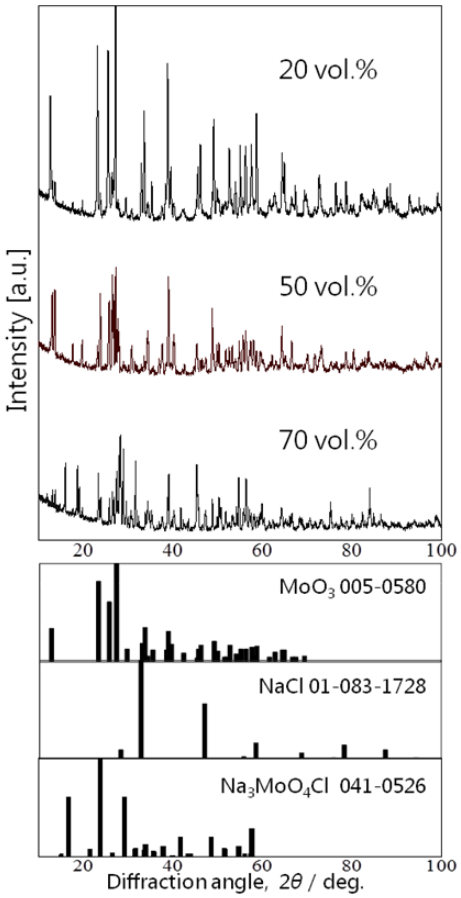


Figure 7 XRD pattern of MoO_3 targets after heating

4. Conclusion

The process produces MoO_3 shells on the surface of NaCl were studied. The reaction between MoO_3 and NaCl was took place, and Mo-Cl compound precipitated and then Mo was oxidized into MoO_3.

References

[1] Yuzuru Saeki, Takashi Kasama, Yoshihiro Kaburagi, “Production of tungsten hexachloride by chlorination of tungsten trioxide”, 工業化学雑誌 73 巻(1970)6 号、pp.1093-1096 (in Japanese)

[2] Yuzo Saeki, Akemi Kato and Ryoko Matsuzaki, “Reaction process between sodium tungstate and chlorine”, Chemistry Letters (1975), Chemical society of Japan, pp. 935-938.