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Recovery of Tantalum Sintered Compact from Used Tantalum Condenser Using Steam Gasification with Sodium Hydroxide

Shigeyuki Katano[a,*], Takaaki Wajima[a] and Hideki Nakagome[a]

[a]*Graduate School of Engineering, Chiba University, 1-33, Yayoi, Inage, Chiba, 236-8522, Japan*

Abstract

Recovery of tantalum resource from used tantalum capacitor in electric equipment is important because the production of tantalum have not been stable for the price and the quantity. However, recovery of tantalum sintered compact from tantalum capacitor is difficult since the compact strongly covered with the flame retardant resin made of halogenated compounds (mold resin). In this study, steam gasification with sodium hydroxide was applied for recovery of tantalum sintered compact by destroying mold resin and stabilization of halogenated compounds in sodium hydroxide to prevent exhausting halogenated gas. Mold resin can be decomposed by steam gasification with NaOH to recover the sintered compact of tantalum. Furthermore, most halogen gas generated from decomposition of mold resin can be trapped in sodium hydroxide not to exhaust halogen gas. These results suggested that recovery process of tantalum sintered compact from the used condenser using steam gasification with sodium hydroxide is expected as a feasible way to recycle the rare metal in electric equipment.

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Keywords: Tantalum recovery, mold resin, stream gasification, sodium hydroxide, halogen capture

1. Introduction

Rare metals are indispensable resources to various electronic devices, and the demand is increasing, but the price and output are not stability due to the uneven distribution of rare metal resources. Tantalum metal is one of the rare metals used for the tantalum capacitor in an electronic device. Tantalum capacitors with high

* Corresponding author. Tel.: +81-43-290-3466; fax: +81-43-290-3466.
E-mail address:z9t0210@students.chiba-u.jp.

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concentration tantalum is manufactured in smaller sizes and higher performance than other types of capacitors, and are used in many products such as PCs and servers, and small-sized home appliances. The Japanese government selected tantalum as one of 14 priority minerals for recycling and as one of five most critical minerals for intensive recycling [1]. Based on these factors, the development of recycling technology from used tantalum capacitors is required from the point of view of "urban mines" [2]. The structure of a tantalum condenser is shown in Fig. 1 [3]. Tantalum condenser consists of the sintered compact which sintered the tantalum powder used as an electrode, mold resin used as the cover of a sintered compact, and the terminal connected with an electrode. In order to collect tantalum resources from a tantalum condenser, recovery of the sintered compacts with high concentration in stable mold resin is important. In the processing method to decompose mold resin, combustion [4] and solubilization [5] have been proposed previously, but these methods has some problems, such as high combustion temperature, high pressure to use pressure reactor, and generation of harmful halogen compounds to need another treatment.

Steam gasification processing is possible at reaction temperature lower than combustion and under normal pressure. The subject of cost or safety can be solved. As for what used sodium hydroxide, molten salt is cheap and can use it by ordinary pressure as a widespread reaction medium, and this is because the resolution of the organic halide can use it [6].

In this study, we investigated recovery of tantalum metal from used tantalum capacitor by using the steam gasification with sodium hydroxide for the recovery of tantalum compact by decomposition of mold resin and stabilization of halogenated compounds in the sodium hydroxide.

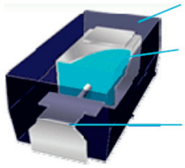

Fig. 1. Structure of tantalum capacitor

2. Materials and Methods

The tantalum capacitor (size: 6.0 × 3.2 × 2.5 mm, model number: ESVC1V475M) was obtained from NEC /TOKIN Co., Ltd. The capacitor approximately composed of tantalum sintered compact (40 %), mold resin (44 %) and others (16 %). The composition of the mold resin is molten silica (70 %) [7, 8] OCN (o-Cresol novolac)-type epoxy resin (15 %), phenolic novolac resin (7 %) and flame-retardants (10 %).

The experimental apparatus is shown in Fig. 2. Sodium hydroxide put into the reactor, and heated with an electric furnace while flowing nitrogen (160 mL/min) and steam (1 mL/min) warmed by the heater. After heating to setting temperature (530 ºC), tantalum capacitor (about 3 g) was thrown in the reactor, heated for 5 min, and then naturally cooled to room temperature. Since the capacitor was included in fused salt solid, it dissolves in distilled water, and filtrates to obtain the residual substance of the capacitor. The residual substance sieved under 500 μm to recover the sintered compact of tantalum in the condenser by destroying the mold resin. The contents of carbon and hydrogen in raw mold resin and sieved residue were analyzed by CHN elemental analyzer (Perkin-Elmer 2400).

During the experiment, the gas generated in the react pass through the water bubbling bottle to capture the halogen content in the gas, then the passing gas was analyzed by a gas chromatograph (GC-2014ATF, SHIMADZU). After the experiment, halogen contents in the filtrate and the solution in bubbling bottle were analyzed by ion chromatography (DX-120) to estimate the capture of halogen content in NaOH.

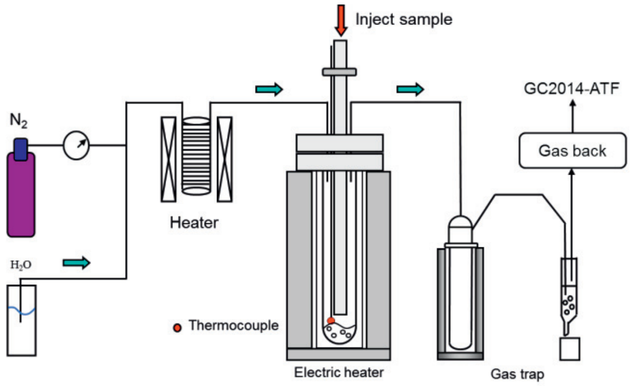

Fig. 2. Experiment apparatus of this experiment

3. Result and Discussion

Figure 3 shows the photos of the residue on the sieve after heating. Without addition of NaOH, molded resin was strongly covered with tantalum compact, and could not recover the compact from the capacitor (Fig. 3(a)), while, with addition of NaOH, mold resin could remove completely to recover the compact from the condenser and collapse of a tantalum sintered compact was not observed. These results suggested that tantalum compact can be recovered from the condenser by destroying mold resin using steam gasification with NaOH (1 - 3 g for 3 g of capacitor).

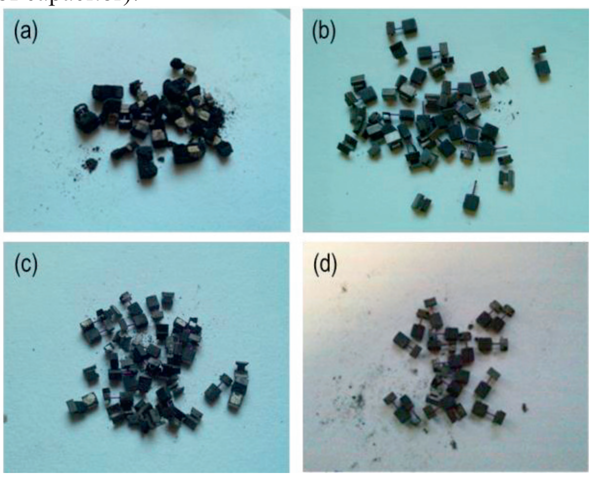

Fig. 3. The residue after the experiment with addition of NaOH of (a) 0 g, (b) 1 g, (c) 2 g, (d) 3 g

Figure 4 shows the product gas from condenser using steam gasification with sodium hydroxide. With increasing addition of NaOH, the amount of hydrogen production was significantly increases.

Figure 5 shows the contents of carbon and hydrogen in raw mold resin and sieved reidue after the experiment using 1 g of NaOH. While raw resin contains 16 % carbon and 2 % hydrogen, the residue after experiment contains 10 % carbon and 2 % hydrogen, which means that the carbon content of mold resin was reacted to release from the resin and the resin was decomposed to collaspe.

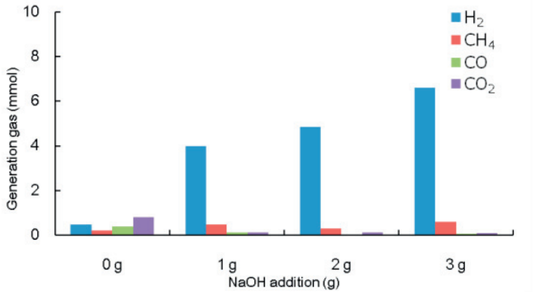

Fig. 4. The product gas from the condenser using steam gasification with sodium hydroxide

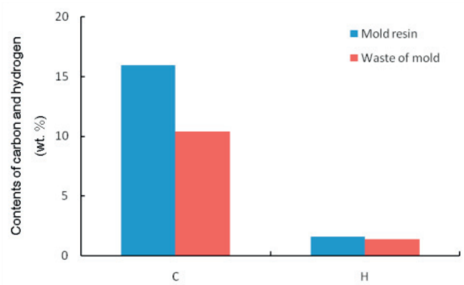

Fig. 5. Composition of resin before and after the experiment

Kamo, et al. reported that polyvinyl chloride (PVC) was gasified in the precense of sodium hydroxide at 600 ºC for 150 min at a partial steam pressure of 1.7 MPa to produce hydrogen and sodium carbonate [9]. We supposed that mold resin of the capacitor was collapsed by the water gas reaction *(1),* water gas shift reaction *(2)* and steam gasification reaction with sodium hydroxide bath *(3)* [10]. In addition, steam is supplied by sodium hydroxide, and CO_2 gas is absorbed in NaOH to promote H_2 gas generation; *(4), (5)* and *(6)*.

$$C + H_2O \rightarrow CO + H_2 \quad (1)$$

$$CO + H_2O \rightarrow CO_2 + H_2 \quad (2)$$

$$C + H_2O + 2NaOH \rightarrow Na_2CO_3 + 2H_2 \quad (3)$$

$$2NaOH \rightarrow Na_2O + H_2O \quad (4)$$

$$C + 2H_2O \rightarrow CO_2 + 2H_2 \quad (5)$$

$$2NaOH + CO_2 \rightarrow Na_2CO_3 + H_2O \quad (6)$$

Figure 6 shows the halogen contents in NaOH and gas phases. Sodium hydroxide in the reactor react with mold resin and dehalogenation from halogenated contents (Flame retardant) in the resin occured. Sodium ion in sodium hydroxide in the molten salt at 530 ºC is combined with halogen included in the mold resin by nucleophilic substitution, and became inorganic salts, such as NaF and NaCl fixed in residual substances *(7),(8)*.

$$Na^+ + F^- \rightarrow NaF \quad (7)$$

$$Na^+ + Cl^- \rightarrow NaCl \quad (8)$$

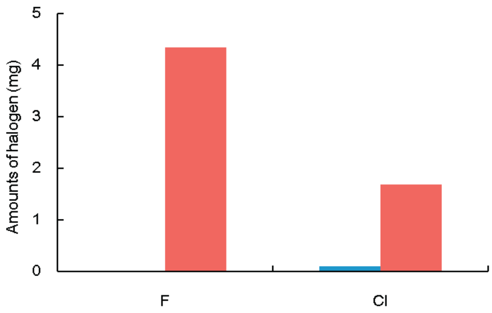

Fig. 6. Halogen contents in NaOH and gas phases

4. Conclusions

The mold resin was decomposed by using steam gasification with sodium hydroxide to recover the tantalum sintered compact in a tantalum condenser. Although the compact cannot be recovered from the condenser by steam gasification without NaOH because the decomposition of mold resin was not enough to collapse, the compact can be recovered from the condenser using steam gasification using sodium hydroxide. In the steam gasification using sodium hydroxide, the exhaust of halogen gas can be inhibited by trapping in NaOH in the reactor. These results suggested that steam gasification with NaOH can be applied for the recovery of tantalum sintered compact from the condenser.

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