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## Ruthenium Release from Thermally Overheated Nitric Acid Solution Containing Ruthenium Nitrosyl Nitrate and Sodium Nitrate to Solidify

Kayo Sawada<sup>a</sup>, Yasuyuki Ueda<sup>a</sup>, and Youichi Enokida<sup>a\*</sup>

<sup>a</sup> Nuclear Chemical Engineering Laboratory, Nagoya University, Nagoya 4648603, Japan

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### Abstract

Radioactive ruthenium (Ru) is one of the dominant elemental species released into the environment from a fuel reprocessing plant in a hypothetical design accident due to its relatively higher fission yield and longer half-life. After the hypothetical accident assuming the loss of all electric power and cooling functions, high-level liquid waste (HLLW) may be overheated by the energetic decays of many fission products in it, and Ru may be oxidized to the volatile tetroxide, RuO<sub>4</sub>, which is released through the off-gas pathway. At a reprocessing plant in Japan, alkaline solution from the solvent scrubbing stream is sometimes mixed with the HLLW followed by vitrification, which can be influenced by the addition of sodium nitrate to a simulated HLLW containing ruthenium nitrosyl nitrate that was experimentally evaluated on a small scale using the overheated nitric acid solution of 2 mol dm<sup>-3</sup>, which was kept at 180 °C in a glass evaporator placed in a thermostatic bath. The release fraction of Ru increased by approximately 30% by the addition of sodium nitrate. This may be partially explained by the existence of relatively highly concentrated nitrate ions in the liquid phase that oxidize the ruthenium species to RuO<sub>4</sub> during the drying process.

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**Keywords:** ruthenium release; hypothetical nuclear accident; high level liquid waste; over-heated nitric acid solution; accident management.

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\* Corresponding author. Tel. +81-52-789-5937; fax +81 52-789-5935.  
E-mail address: [yenokida@nagoya-u.jp](mailto:yenokida@nagoya-u.jp)

## 1. Introduction

### 1.1. Background and scope

The huge earthquake followed by tidal waves on March 11, 2011 triggered the severe nuclear accident in the northeastern part of Japan due to station blackout at the Fukushima Dai-ichi Power Plant owned and operated by Tokyo Electric Power Company, Japan. Additionally, one of the afterquakes in the same area hit the Rokkasho Reprocessing Plant (RRP) on April 7, 2011; The plant experienced a station blackout, and managed to normally recover by relying on backup power generators. Eighty percent of the blackout area had recovered within three days, and 94% within 8 days.<sup>1</sup> As a lesson learned from this disaster, we should assume a longer period of station blackout even for the reprocessing plant in Japan.

Radioactive ruthenium (Ru) is one of the dominant elemental species released into the environment from a fuel reprocessing plant in a hypothetical design accident due to a relatively higher fission yield and longer half-life. After the hypothetical accident by assuming the loss of all electric power and cooling functions, high-level liquid waste (HLLW) may be overheated by the energetic decays of many fission products in it, and Ru may be oxidized to the volatile tetroxide, RuO<sub>4</sub>, which is released through the off-gas pathway.

In this paper, we discuss the hypothetical Ru release from the thermally overheated HLLW, which may be caused by the loss of the cooling functions due to the longer station blackout.

A few previous investigations that have appeared in open literature, such as Ref. 2, have discussed the release of Ru from the overheated HLLW based on the hypothetical condition of the loss of the cooling system, and the Ru release has been very well defined based upon experimental measurements of the Ru release rate for the actual liquid waste and correlated with the temperature and enrichment of the nitric acid.

At the RRP, according to a process sequence, alkaline solution containing sodium species from the solvent scrubbing stream is sometimes mixed with the HLLW followed by vitrification.

### 1.2. Goal of the study

Since at a reprocessing plant in Japan, significant concentration of sodium nitrate is not always, but sometimes may be present in HLLW followed by vitrification, we propose to investigate the impact of adding small amount of sodium nitrate to a simulated Japanese HLLW containing ruthenium nitrosyl nitrate. The experiment will be performed under the overheated nitric acid solution which was kept at constant temperature in a vacuum evaporator of glass placed in a thermostatic bath in a small scale. The experiment will clarify if the release rate of Ru may increase or not by the addition of sodium nitrate and increase our knowledge of the process design of RRP.

The current study was performed at Nagoya University, Japan, as a third party independent of the nuclear industries, utility companies, and the nuclear regulatory authority in Japan to discuss the hypothetical Ru release from the RRP. Apart from a large project, the study was actually a small one at a national university, whose research budget was provided for fundamental research in Japan, which is described in the **Acknowledgements** section.

## 2. Experimental

### 2.1. Materials

Since the reference process specification of the HLLW in the RRP is described in open literature including the concentration of sodium, 3.4 mol dm<sup>-3</sup> when the solvent scrubbing stream is mixed,<sup>3</sup> we prepared the simulated nitric acid solution of 2.0 mol dm<sup>-3</sup> for the HLLW in the RRP in a chemical laboratory using non-radioactive materials, which contained the same concentrations of the constituents described in Ref 3. We added equivalent amount of neodymium for the actinide elements. All chemicals used, except for Ag, Ce and Ru, were of reagent grade and purchased from Kanto Chemical Co., Inc., Japan, Ag and Ce were of reagent grade from Wako Pure Chemical Industries, Ltd., Japan, and the reagent grade Ru from Rare Metallic Co., Ltd., Japan, as ruthenium nitrosyl nitrate. The acidity of the simulated HLLW was adjusted 2.0 mol dm<sup>-3</sup> using concentrated nitric acid. In

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