Optimization of wet oxidation process for the determination of ^{129}I in simulated radioactive wastes

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This study has been focused on the improvement of wet oxidation procedure including the separation of iodine by distillation for the characterization of ^{129}I in radwastes. In particular, under optimal conditions, we have identified a notable enhancement in the recovery rate of iodine along with the reduction of reaction time. Considering several factors affecting the recovery rate, it has been confirmed that the amount of oxidant, leading to the oxidation of iodide ion to volatile iodine, plays a critical role in maximizing the recovery of ^{129}I. Furthermore, in terms of the minimum detectable activity (MDA) of ^{129}I for the specific method, the optimized wet oxidation process resulted in a more than 10-fold reduction in MDA value compared to other well-known methods that show the limitation on the available amount of samples.

I. INTRODUCTION

According to the regulation of radioactive waste discharges and disposals, it requires the radiochemical waste characterization that determines the concentration of major radionuclides such as ^{14}C, ^{3}H, and ^{129}I. To establish the need for further treatment of radwastes or their suitability for storage, numerous analytical procedures have been developed for radionuclides of interest. Among those radionuclides, the determination of radioactive ^{129}I in low- and intermediate-level radwastes (LILW) has been getting more and more attention due to concerns about the radiological impacts to human health and the environment. Even if ^{129}I in most LILW generated from nuclear power plants would be found at a lower concentration than MDA, it is essential to know an accurate radioactivity of ^{129}I while optimizing reaction parameters and lowering the MDA value. In this respect, simulated dry active wastes (DAW) with a different concentration of ^{129}I were prepared to mimic LILW. The recovery rate of ^{129}I based on the non-radioactive iodine carrier of ^{127}I was evaluated at different conditions by optimizing several parameters including reaction temperature, reaction time, and the amount of oxidant. The aim of this study is to provide a reliable analytical technique by reducing any possible source of error or uncertainty in the measurement of ^{129}I in LILW.

II. EXPERIMENTAL

The effect of oxidant on the recovery rate of ^{129}I was investigated with different amounts of 30 % H_2O_2 ranging from 1.0 mL to 8.0 mL. As an example, the determination of recovery rate on 5.0 mL of H_2O_2 was carried out as follows. To the filtrate from simulated DAWs containing a mixture of ^{129}I and iodine carrier (KI) was added 3.0 M H_2SO_4. 5.0 mL of 30 % H_2O_2 was then added to the reaction mixture at room temperature. Subsequent distillation to vaporize iodine was performed at 97 ^{o}C for 5 h while blowing Ar as a carrier gas at 0.1 mL / min. After collecting iodine in chloroform solution, 5.0 mL of 0.2 M $NaHSO_3$ as a reducing agent for back-extraction was added into the solution followed by shaking for 5 min. Additionally, to convert iodide ions into AgI precipitate, 3.5 mL of 0.2 M $AgNO_3$ was slowly added to the corresponding aqueous solution. The precipitate separated by centrifugation was finally transferred to a planchet and its activity was measured for 10000 sec with the low energy gamma-ray spectrometry. As part of the optimization for a wet oxidation-distillation process, other factors related to reaction temperature and time was also investigated in the same manner as above.

III. RESULTS

Figure 1 shows the recovery of iodine carrier as a function of the amount of oxidant added to the leaching solution. Interestingly, it was observed that the recovery of carrier was remarkably increased from 46.5 to 87.5 % while increasing the amount of 30 % H_2O_2 from 1.0 to 6.5 mL. However, above 8.0 mL of oxidant, a gradual decrease in recovery was identified (84.5 %). In addition, we found that the reaction time was significantly influenced by the distillation temperature under the same amount of oxidant. For example, based on the maximum recovery of iodine, the distillation at 110 °C provided a notable result that indicates a 3.3-fold faster reaction time compared to that at 97 °C. Under this optimal condition, more than 80 % of iodide could be isolated from the leaching solution and the corresponding recovery rate of ^{129}I was increased up to 85.0 ± 5 %.

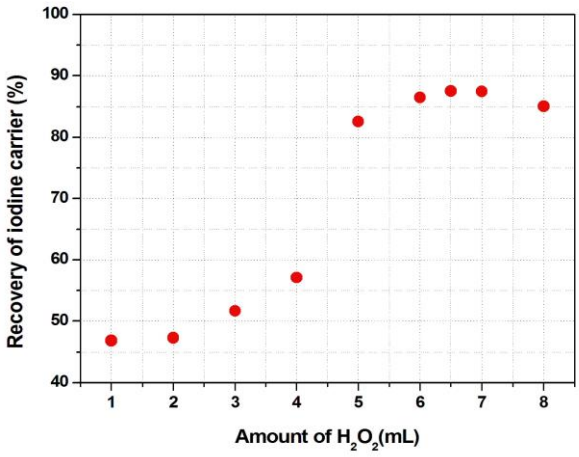

Fig. 1. The effect of oxidant on the recovery of iodine carrier

IV. CONCLUSIONS

We optimized a wet oxidation-distillation technique for the determination of ^{129}I in simulated radioactive wastes. To measure the accurate concentration of ^{129}I in radwastes, iodide was first leached from the simulated radioactive waste with a diluted sulfuric acid, and oxidized to iodine using 30 % H_2O_2 followed by the separation through distillation. As an effort to optimize the reaction conditions, the recovery rate of iodine was evaluated at different conditions as a function of the amount of oxidant, reaction temperature, and distillation time. The highest recovery was 85.0 ± 5.0 %, which was optimal for a distillation time of 1.5 h using 6.5 mL of 30 % H_2O_2 at 110 °C. Furthermore, our current condition is capable of lowering the MDA value more than 10 times compared to the other well-known methods that show the limitation on the available amount of samples.

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