Study of Wet Oxidation Process for C-14 Separation of Cellulose-Based Radioactive Waste

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1. Introduction

The decommissioning of nuclear power plants generates substantial amounts of radioactive waste, necessitating disposal methods based on radioactivity levels. Radioactive waste exceeding regulatory limits must be managed in designated disposal facilities, requiring precise characterization of key radionuclides. The primary radionuclides targeted for analysis include tritium (^{3}H), carbon-14 (^{14}C), and iodine-129 (^{129}I). Among these, ^{14}C accounts for a significant proportion of the key radionuclides present in radioactive waste [1].

^{14}C is a long-lived radionuclide with a half-life of approximately 5,730 years, raising concerns about its potential long-term environmental impact. Moreover, as ^{14}C is a pure beta emitter releasing low-energy beta particles with a maximum energy of 156 keV, it is difficult to distinguish from other beta-emitting radionuclides. Therefore, a chemical separation procedure is required to achieve accurate quantification of ^{14}C in low-level radioactive waste.

Currently, the combustion and wet oxidation methods are primarily employed for the separation and quantification of ^{14}C. The combustion method involves high-temperature oxidation (~900°C) of the sample, converting ^{14}C into carbon dioxide ($^{14}CO_2$) for subsequent analysis. While this method offers high efficiency, it has drawbacks due to the high initial costs associated with equipment setup and operation. In contrast, the wet oxidation method, which utilizes chemical oxidants, is relatively cost-effective and simple to implement. This method has been widely applied for the analysis of environmental samples. However, their applicability to cellulose-based radioactive waste has yet to be thoroughly investigated.

Cellulose is a major component of various forms of radioactive waste, such as laboratory-generated wipes, paper waste, and other organic materials. Nevertheless, cellulose can exhibit resistance to oxidation under specific conditions, potentially resulting in incomplete decomposition. This resistance may compromise the accurate recovery and quantification of radionuclides such as ^{14}C, reducing the reliability of the analytical results. Accordingly, this study aims to evaluate the applicability of the wet oxidation method to cellulose-based radioactive waste and to propose an optimized oxidation protocol that enhances the accuracy and reliability of radionuclide analysis.

2. Experimental

Experiments were conducted using ^{14}C-labeled sodium carbonate ($Na_2{}^{14}CO_3$) and ^{14}C-labeled formaldehyde ($H^{14}CHO$) as representative carbon species commonly found in various types of radioactive waste [2]. The oxidation process was performed using sulfuric acid and potassium persulfate as oxidizing agents, with silver nitrate as a catalyst. After oxidation, the $^{14}CO_2$ produced from the samples was trapped in Carbo-Sorb E under a continuous flow of nitrogen carrier gas and measured using a liquid scintillation counter.

3. Results and Discussion

3.1. Comparison of $Na_2{}^{14}CO_3$ Recovery on Cellulose

The $Na_2{}^{14}CO_3$ exhibited high recovery rates regardless of cellulose. The recovery rate was 93.5 % in the absence of cellulose and 93.9% when cellulose was present under identical experimental conditions. These results indicate that $Na_2{}^{14}CO_3$ remains stably immobilized on the cellulose matrix, with negligible loss during the separation and recovery process.

3.2. Comparison of $H^{14}CHO$ Recovery on Cellulose

The $H^{14}CHO$ demonstrated distinctly different recovery rates depending on the presence of cellulose. In the absence of cellulose, the recovery was 105.3 %; however, in the presence of cellulose, the recovery significantly decreased to 77.4 %. This suggests the possibility that $H^{14}CHO$ forms acetal linkages within the cellulose matrix, promoting immobilization and subsequently reducing extraction and recovery efficiency.

4. Conclusions

This study assessed the effect of cellulose presence on the recovery of $Na_2{}^{14}CO_3$ and $H^{14}CHO$. The experimental results confirmed that $Na_2{}^{14}CO_3$ maintained high recovery in both aqueous solution and

cellulose matrix, demonstrating stable and reliable analytical performance under the given conditions. Conversely, $H^{14}CHO$ showed a tendency toward reduced recovery when applied to cellulose, which is attributed to the formation of acetal linkages within the matrix and its inherent volatility.

Future research will focus on clarifying the chemical interaction mechanisms between $H^{14}CHO$ and cellulose, as well as conducting additional experiments to optimize recovery conditions.

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