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Concentration of liquid sample for gamma-ray spectroscopy by using ultrasonic nebulizing

Jungsoon Kim[1†], Jihyang Kim[2], Jihee Jung[2], Moojoon Kim[2], and Kanglyeol Ha[2]
([1]Department of Media Engineering, Tongmyong University, Busan, Korea;
[2]Department of Physics, Pukyong National University, Busan, Korea)

1. Introduction

Generally, to analyze the radionuclides in sea water, a large amount of sea water, about 40~60 L, needs to be concentrated because the radionuclides are infinitesimal[1]. The liquid radioactive wastes from nuclear facilities contain various radionuclides, and the radionuclides, except for ^{137}Cs, are extracted by precipitation method with MnO_2 powder[2]. However, this method requires more than 24 hours for precipitation to achieve the high collection rate of sediment, and this makes it difficult to get fast results in emergencies. In this study, we propose a new concentration method, which can to remove the moisture from the liquid sample within a short time by ultrasonic nebulizing in a short time, and then the validity of this method is verified.

2. Theory

When a small particle is in a droplet from an ultrasonic nebulizer, the particle has an acceleration, a, because the droplet is catapulted out of the ultrasonic capillary wave. The force acting on the particle is as follows[3]:
1)External force from the ultrasonic nebulizer:

$$F_e = \frac{4}{3}\pi r_s^3 \rho_s a_e , \quad (1)$$

where ρ_s and r_s are the density and the radius of the sphere, respectively.
2) The total surface tension force:

$$F_s = 2\pi r_s \tau \cos\alpha \cos(\pi - \theta - \alpha), \quad (2)$$

where τ is the surface tension of the droplet, θ is the angle of contact between the droplet and the surface of the sphere and α is the angle between a tengent line to the sphere at the contact point to the droplet and a vertical line.
3) The buoyancy force:

$$F_b = \rho_l \frac{\pi}{3} r_s^3 (1-\sin\alpha)^2 (2+\sin\alpha)(a_e - g). \quad (3)$$

where ρ_l is the density of the liquid, g the gravitational acceleration. If the acceleration is sufficient to satisfy the condition,

$$F_e + F_b \geq F_s , \quad (4)$$

the particles would be separated from the droplet. From Eqs. (1)~(4), the critical radius to separate the particles from the droplet is determined by

$$r_s > \sqrt{\frac{6\tau\cos\alpha\cos(\pi-\theta-\alpha)}{4\rho_s a_e + \rho_l(1-\sin\alpha)^2(2+\sin\alpha)(a_e-g)}} . \quad (5)$$

3. Experiment

The ultrasonic nebulizer consists of 10 piezoelectric vibrators, and each vibrator is 1.25 mm in thickness and 19.85 mm in diameter, as shown in **Fig. 1**. The resonant characteristics of the vibrator were measured and presented in **Fig. 2(a)**, and according to this the resonant frequency is 1.7 MHz. The admittance loci were measured with an impedance analyzer to obtain the electro-acoustic efficiency, and are presented in Fig. 2(b).

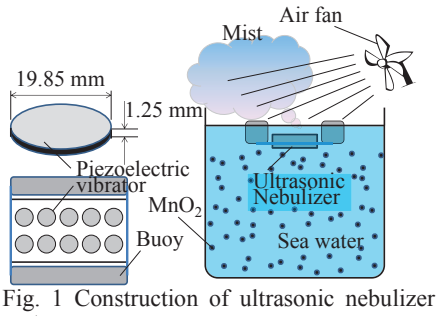

Fig. 1 Construction of ultrasonic nebulizer and experimental system.

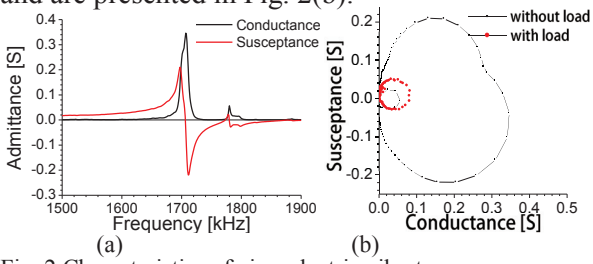

Fig. 2 Characteristics of piezoelectric vibrator.
(a) Resonant characteristics (b) Admittance loci

From this result, the efficiency was found to be 76 %. Consequently, the acoustic power was 2387.7 W when the input electric power was 3141.7 W. **Figure 3** demonstrates the process of concentrating sea water to extract the radionuclides.

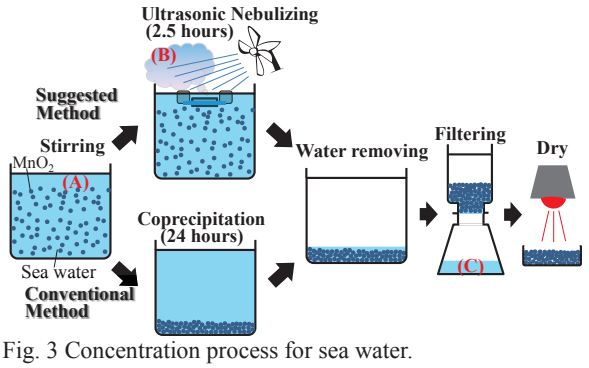

Fig. 3 Concentration process for sea water.

A plastic container is filled with 15 L of sea water, and for a precipitant 30 g of MnO_2 is added to the water. The pH value of the sea water is adjusted to be 8 using ammonia to make it easy to displace

kimmj@pknu.ac.kr

metal ions, and then the sea water is stirred for 1 hour. The buoy is carefully adjusted so that the surface of the vibrators can be situated at 40 mm below the water surface. An air fan assists the process of nebulizing by removing the mist produced by the ultrasonic nebulizer. The velocity of the wind from the fan is 2 m/s. After the nebulizer removes 10 L of moisture for 2.5 hours, the residual moisture is completely removed through the filtering and infrared drying process. To compare the results from this method with the ones from the conventional method, another sea water sample is prepared as follows. First, MnO_2 is added to sea water and stirred according to the conventional method. Next, the upper clear water is removed after being precipitated for 24 hours. After that, the sample was treated in a manner same as that described above. The gamma-ray spectrums of each dried sample from the two different methods are measured with High Pure Ge Detector (CPVDS30).

4. Result

Figure 4(a) shows the distribution of the catchable particles in the droplets calculated by Eq. (5).

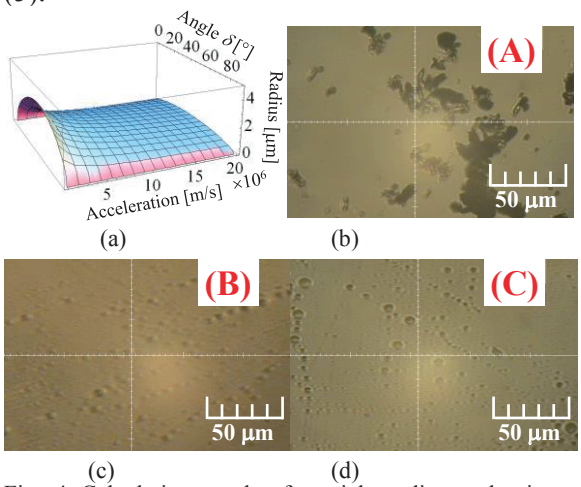

Fig. 4 Calculation result of particle radius and microscope images of particles. (a) Calculated radius of catchable particle, (b) Particles in the stirred sea water, (c) Particles in the mist, (d) Particles in the filtered water.

This result shows the radius of the catchable particle as a function of the acceleration due to the external force over all the angle α. Figure 4(b) shows the optical microscopic image of sample (A) in Fig. 3. In the image, there are relatively large particles due to the agglomerated particles of MnO_2 and ones displaced with metal ions. Figure 4(c) shows the image of the collected particles from sample (B), after the ultrasonic nebulizing. This result shows the agglomerated particles under 5μm in diameters were captured in the droplets, and were scattered into air. This result agrees with the calculated one in Fig. 4(a). Figure 4(d) shows the image of the particles in the water which was removed by filtering, sample (C), in Fig. 3. The result shows that the particles in the water shown in Fig. 4(d) are bigger than those in Fig. 4(c). Therefore, it becomes clear that the particles that are lost due to nebulizing do not affect the analysis of the sample. The mass of the dried sample was measured, and was compared with that of MnO_2 powder put in the sea water to obtain the collection rate.

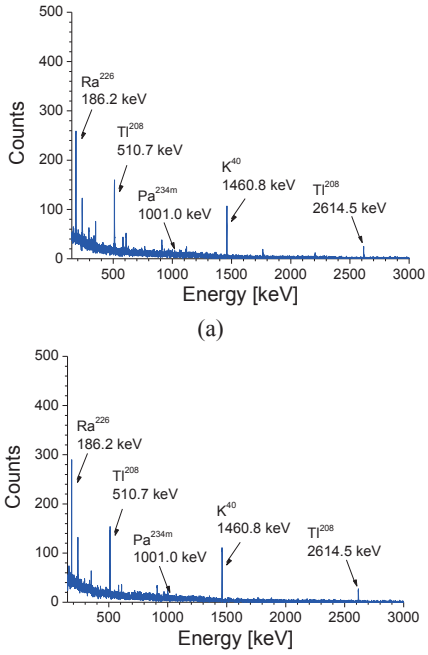

Fig. 5 Gamma-ray spectrum of samples.
(a) Proposed method, (b) Conventional method

The collection rate in the case of the proposed method was 116 % when that of the conventional method was 106 %. It was rather higher than that of the conventional method. The gamma-ray spectrums of the dried samples were measured, as shown in **Fig. 5**. The result of the sample concentrated by the proposed method is shown in Fig. 5(a), and each radionuclide is marked at the corresponding peak. This result is not significantly different in terms of each peak compared with that from the conventional method shown in Fig. 5(b). Therefore, it is confirmed that the proposed method could reduce the time dramatically in the process of sea water concentration.

5. Summary

In the process of sea water concentration for the analysis of environmental radioactivity, a water removing method using an ultrasonic nebulizer was proposed instead of the conventional process that requires long precipitation time. It took only 2.5 hours to remove the moisture from 15 L of sea water with the proposed method, whereas the conventional method requires at least 24 hours for precipitation. From the results of the gamma-ray spectrums of the concentrated samples, the validity of the proposed method was verified.

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