Developmental study of chemical trapping method for Radon

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ABSTRACT

Chemical trapping method for radon (Rn) gas utilizing a chemical reaction between Rn and fluorine was developed. By the corona discharge treatment of an air including Rn and CF4 gas of maximum 7.5 vol%, it is assumed that non-volatile and chemically stable radon difluoride (RnF2) suitable for trapping can be generated by combining excited Rn with radical fluorine. The reaction between Rn and radical fluorine may be proceeded by Rn + CF4 --discharge--> excited Rn and radical-F ---> RnF2 (trapped in charcoal) under the conditions of ambient temperature and atmospheric pressure. We tested to confirm the reaction and measured the trapping efficiency of Rn generated from the developed radium (226Ra) source with a high release efficiency of Rn. From the results, Rn concentration of about 1,000 Bq/m3 in a normal air was reduced to background level (10-20 Bq/m3) after trapped by a small charcoal filter at room temperature. The newly developed method of chemical trapping for Rn can make a trap or adsorbent to a small size, and does not need a cooling system of adsorbent in a general and a current charcoal adsorption system. Furthermore, the chemical trapping method can convert directly radioactive noble gases (eg. Rn, Xe, Kr) to solid wastes. For trapping radioactive noble gases of Rn, Xe and Kr, we are developing a chemical trapping method except introducing fluoric-compound gas, and are examining to apply it as a new trapping method instead of an usual gas treatment system of nuclear sites.

INTRODUCTION

In order to trap the noble gases which are radon (222Rn), radioactive xenon (133Xe) and krypton (85mKr, 85Kr-88Kr) with comparably long radioactive-life, several adsorbents of charcoal, zeolite and other porous materials have been used for temporary trapping [1-4]. However, the efficiency of the physical adsorption method for trapping the noble gases are generally very low at an ambient temperature. By the reason, to obtain high efficiency for trapping and removal of them, the column of adsorbent has been cooled at a very low temperature or constructed to a huge size. However, the off-gas including the radioactive noble gases (222Rn, 133Xe, 85Kr) with long radioactive-life, which are generated from the fuel reprocessing plants, nuclear power plants, nuclear medical facilities and nuclear research laboratories, have been exhausted to the atmosphere. It is difficult to trap them with a high efficiency by the reason of their unreactive properties, and low melting points and low boiling points as shown in Table 1 [5].

Commonly, the noble gas does not have a chemical reactivity to combine other elements. As shown in Table 1, Rn, Xe and Kr form several compounds of fluoride, oxy-fluoride, oxide and complexed fluoride [5]. Utilizing the reactivity of noble gases with fluorine, we tried to develop a new trapping method using chemical reaction. If the chemical trapping method for noble gases utilizing a chemical reaction can be developed, many problems of nuclear sites may be solved.

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To proceed the reaction between the noble gases and fluorine, we used the corona surface discharge [6, 7] treatment to excite them and to generate radical fluorine produced from CF4 gas (tetrafluoric carbon; as fluoric-compound). As shown the assumed principle in Fig. 1, by the corona discharge treatment of an air including Rn and CF4 gas, the reaction between excited Rn and radical fluorine may be conducted, and non-volatile and chemically stable radon difluoride (RnF2) [8] suitable for trapping may be generated. The reaction between Rn and radical fluorine may be proceeded by Rn + CF4 --discharge--> excited Rn and radical-F ---> RnF2 (trapped in charcoal) under the conditions of ambient temperature and atmospheric pressure.

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EXPERIMENT

Apparatus

As shown in Fig. 2, the experimental apparatus consists of the gas introducing system with mass flow controllers, a developed Rn source mentioned below (chamber volume: about 100 cm3), a corona reactor of surface discharge type (chamber volume: 250 cm3, the discharge area: about 30 cm2 on a small cylindrical alumina) with a heat-pump cooling system, trap packed with a small sized charcoal (volume: about 5 cm3), a Rn

monitor of gas flow type (type AB-5 made by Pylon Co., monitoring cell volume: about 200 cm3), a suction pump (100-760 Torr, maximum exhaustion speed: 2 L/min), pressure gauges (capacitance manometer; Baratron) with maximum measuring range of 1,000 Torr, backup filter packed by a large sized charcoal (volume: about 500 cm3), and pipe (stainless steel and teflon; outer diameter 0.25 inch), and valves.

To generate conventionally a Rn gas for test, we have developed Rn source made from predetermined amounts of radium (226Ra) sintered on cylindrical porous silicone-carbide (SiC) [9]. The Rn source has a property to generate constantly Rn gas with a higher release efficiency than approximately 30-40 %, and Rn gas can be easily and stably supplied by flowing of an air as a carrier gas.

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Procedure

To generate Rn gas, a mixture gas of air and CF4 (0-7.5 %) of total 200ml/min was introduced into the chamber of Rn source. The mixture gas including Rn gas was introduced into the corona reactor chamber. At the condition of non-discharge, the stable Rn concentration in the mixture gas was measured by the Rn monitor through the charcoal trap. In the case of gas flow rate of 200 ml/min, the Rn concentration was about 1,000 and 3,600 Bq/m3 using 13.61 and 20.64 kBq of 226Ra, respectively. By the control of suction pump, the gas in the corona reactor and Rn monitor were controlled to a little reduced atmospheric pressure using a capacitance manometer (Baratron). After measured the stable concentration of Rn, the discharge treatment by the corona reactor was started. The discharge of corona reactor was performed by high frequency of 8-10 kV of peak to peak. During flowing the test gas, the concentration of Rn was continuously measured. From the tendency of Rn concentration during the test, the chemical reaction of Rn was occured or not, and the trapping efficiency of Rn was calculated by compared with Rn concentration at discharge and non-discharge. All tests were conducted at ambient temperature without cooling and heating.

RESULTS and DISCUSSIONS

1) Test-1 (Confirmation of a chemical reaction and the effects of CF4 concentration)

As shown in a) of Fig. 3, by the discharge treatment, the Rn feeded about 1,000 Bq/m3 was reduced to background level (10-20 Bq/m3) after 4-5 h treated by the corona reactor, but after corona reactor be powered off, the Rn concentration was spontaneously and promptly increased. On the other hand, in the case of CF4 0 % as shown in b) of Fig. 3, the Rn concentration was constant with and without discharge treatment. From the results, the reaction between Rn and fluorine generated from discharged CF4 may be occured. Furthermore, increasing CF4 concentration to maximum 7.5 % effected a gradually higher removal efficiency of Rn.

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2) Test-2 (Reappearance of repeated treatments)

The test of the repeated treatments of a corona reactor powered on and off every 1 h was conducted. From the result, the concentration of Rn was observed as equal as before about 1,000 Bq/m3 at powered off, and the concentration at powered on was gradually reduced for 1 h. After the corona reactor every powered off, the Rn release increased spontaneously and promptly were observed each.

3) Test-3 (Chemical trapping behavior of Rn)

Using a higher Rn source (3,600 Bq/m3), the test to observe the chemical trapping behavior was conducted. After the discharge treatment for 3-3.5 h, the mixture gas was flowed by-passing the Rn source. At the same time the discharge treatment was stopped. Then, to determine the Rn release position, each Rn concentration in the gases through the corona reactor and through the trap of activated carbon were measured. As shown in Fig. 4, the same Rn release behavior at the discharge off mentioned above was also observed, and the main release position was corona reactor rather than charcoal filter. From the result, Rn may be chemically trapped onto corona reactor as an unstable compound; this trapping behavior cannot be understand and should be elcidate in a future.

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SUMMARY

From the experimental result based on a new concept and principle of chemical trapping for the noble gases, the chemical reaction of Rn may be occured. The experimental results were obtained as following.

(1) By the discharge treatment of mixture gas (air+Rn+CF4), Rn about 1,000 Bq/m3 was reduced to background level (10-20 Bq/m3).

(2) Increasing CF4 concentration to maximum 7.5 % effected a gradually higher removal efficiency of Rn.

(3) The reappearance of Rn chemical trapping behavior was observed by the repeated treatments of corona

reactor powered on and off.
(4) After the corona reactor powered off, the Rn concentration was spontaneously and promptly increased.
(5) From the result observed the release of Rn trapped, the main trapped position was in the chamber of corona reactor rather than charcoal filter. Rn may be chemically trapped onto corona reactor as an unstable compound; this trapping behavior cannot be understand.
Such being the case, it can be expected that the chemical trapping method has an ability to recover the noble gases under an ordinary conditions using a plasma treatment instead of the current methods using a cooling at a very low temperature, or a huge adsorbent and a large system.
The newly developed method of chemical trapping for Rn can make a trap or adsorbent to a small size, and does not need a cooling system of adsorbent in a general and a current charcoal adsorption system. Furthermore, the chemical trapping method has an ability to convert directly radioactive noble gases (eg. Rn, Xe, Kr) to solid wastes, and also to recover directly the trace resources of noble gases in an air, e.g. Xe, Kr or others. For trapping radioactive noble gases of Rn, Xe and Kr, we are developing a chemical trapping method of exciting surface fluoric-ceramics layer using surface discharge except introducing fluoric-compound gas, and be examining to apply the new trapping system instead of an usual gas treatment system of nuclear sites.

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