Performance of Chemical Trapping Method for Radon

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Abstract. Radon is one of the most important radioactive substances contained in the atmosphere and about half of the natural radiation dose to humans is due to radon and its decay products. Since radon is a noble gas, it is very difficult to trap stably the gas for long time. Using activated charcoal traps as a common method, it has some disadvantages; e.g. a lack of stability over the long-term and a low trap efficiency per unit volume. In order to obtain an effective and stable trapping method for radon gas, new trapping technique utilizing a chemical reaction between radon and fluorine has been studied and developed. The trapping phenomenon of radon gas was observed by a discharge treatment of a mixed gas of radon and fluorine introduced to the corona discharged area. CF₄, the chemically stable fluorine gas, was used for the treatment. In the previous study, it was confirmed that the concentration of radon was reduced from approximate 1,000 Bq/m³ to the back ground level of 10-20 Bq/m³. For conducting the advanced study of the chemical trapping method against the high concentrated radon gas, an experiment using high concentrated radon gas of approximate 3 x 10⁵ Bq/m³ was carried out. From the experiment, it was observed that more than 99% of radon concentration was trapped using CF₄ of 5%. It was also observed that radon gas trapped during the discharge treatment was reemitted by shutting off the discharge treatment, and the amount of the radon gas emitted was consistent with the amount of radon gas trapped. The technique developed is adaptive method to trap radon gas with high efficiency over 99%, and also it is expected that the technique can be applied to the enrichment and recovery of radon.

1.Introduction

By our study, the trapping phenomenon of radon gas was observed by a discharge treatment of a mixed gas of radon and fluorine introduced to the corona discharged area. From the previous experimental results using tetra-fluoric carbon excited by the discharge as a reactant, it has been observed that the phenomena of trapping radon gas reduced from approximate 1,000 Bq/m³ to the back ground level of 10-20 Bq/m³ during the discharge and releasing trapped radon gas in a split second at shutting off the discharge treatment were conducted, and the radon trapping efficiency was affected with CF₄ concentration. By the discharge treatments individually carried out with the CF₄ concentrations of 0 %, 2.5 %, 5 % and 7.5 %, it was confirmed that the discharge treatment with CF₄ free had no effect on the radon concentration, and the radon concentration was reduced by accompanied with increasing CF₄ concentration. These chemical trapping phenomenon of radon reacted with excited CF₄ were presented at the last IRPA 10 [1,2]. From the conclusion of our previous study, it is assumed by the experimental results that the trapping and reemission phenomenon of radon gas might be occurred because radon gas was chemically and unstably trapped by the discharge treatment.

According to the chemical trapping and releasing phenomena of radon gas, we made an assumption as shown in Figure 1 and as follows. In the condition of coexisting radon and fluorine gases in the
discharged area, radical radon (\(^{222}\text{Rn}\)) and fluorine (\(^{19}\text{F}\)) with highly reactive properties are chemically bonding during discharge and form any fluoric radon compound; e.g. \(\text{RnFx}\), then that may be immediately decomposed at discharge off.

![Chemical reaction image of radon and fluorine compound gas](image)

**FIG.1. Chemical reaction image of radon and fluorine compound gas**

### 2. Experiment

From our previous experiments, the chemical trapping behavior and efficiency of radon were not exactly cleared and confirmed yet. In order to elucidate the trapping and releasing behaviors of radon, a testing apparatus enabling the control of experiment conditions such as reaction time, gas flow rate and form of the discharged area was fabricated by modifying the current testing apparatus using highly concentrated radon gas utilizing a new ceramic source impregnated with a high level \(^{226}\text{Ra}\).

The experimental apparatus is mainly consisted of a gas flow system for radon and CF4 gas, a corona reactor and a radon monitoring system as shown in Figure 2. The volume of the reaction chamber equipped at the discharged area was designed smaller than that of the current apparatus to increase the contacting efficiency between radon and CF4 gases excited at the discharging surface. By the reason, the corona reactor was the surface discharge type (Model OC002, Masuda Lab.) set in a rectangular aluminum cylinder with the effective volume of 7.9 cm\(^3\).

The air containing CF4 maintained to be a slightly reduced atmospheric pressure (720±20 torr) for stabilizing the discharge was introduced through the testing system with flow rate of 100 cc/min and the average residence time of sample air in the reactor was calculated to be 4.7 sec. The concentration of CF4 gas was set at 5\%, which was the most effective condition observed from our previous experiments, and the gas was introduced through inside of a radon source (\(^{226}\text{Rn}\) of 3.3 \(\times\) \(10^4\) Bq) vessel for a long time to obtain the high concentrated radon gas, and the discharge treatment was performed. Radon monitors were set at each of the downstream and upstream of the discharge treatment apparatus. The radon concentrations were continuously measured with two scintillation cell type monitors (AB-5, Pylon Co., Canada) located at upstream and downstream of the reactor. By using two radon monitors, it was possible to correct the measurement errors of the radon removal efficiency.

Experiment with CF4 concentration of 5\% was carried out utilizing the radon source with 1.85 \(\times\) \(10^6\) Bq and the trapped radon amount was measured during the discharge treatment and released radon amount after shutting off the discharge. Discharge treatment was continued for 6 h, and radon concentration trends after the discharge treatment were measured also for 6 h.
3. Results and Discussion

Discharge treatment was firstly carried out for 140 min as a pre-experiment. Figure 3 shows the trap and release behavior of radon introduced after 30 min of treatment start. In the case of the experiment without using CF4, radon gas was not entirely trapped, and the concentration was measured $1.9 \times 10^5$ Bq/m$^3$. There was no change in behavior after the treatment. On the other hand, in the case of the experiment using 5% CF4, the radon concentration was gradually reduced to $1.4 \times 10^5$ Bq/m$^3$ during the treatment, and the trapped radon was released immediately after the treatment.

Figure 4 shows the trapping and releasing behaviors that the concentration of radon gas contained in the introduced air was reduced from $3 \times 10^5$ Bq/m$^3$ to $1.2 \times 10^3$ Bq/m$^3$, and it shows that the trapped radon was released immediately at shutting off the discharge. As a result of the advanced experiments, it is assumed that the chemical trapping method is effective to trap the highly concentrated radon gas. Moreover, according to the comparison measurement of the trapped radon amount during 6 h discharge treatment and the released amount for 6 h after shutting off the discharge, 55% of the trapped radon was released within 10 minutes after shutting off the discharge and 88% for 6 h. The phenomenon indicated that the highly concentrated radon gas
which was enriched by the discharge treatment is possibly recovered within short time by controlling the treatment time of discharge. It was assumed that the mass balance of radon with 88% should be contributed by the measurement errors because of such excessive changes in radon concentration of after-treatment from $1.2 \times 10^3$ Bq/m$^3$ to $6.2 \times 10^6$ Bq/m$^3$.

It was clarified by the experimental results that radon is trapped in the discharged area by the discharge treatment, and trapped radon was released immediately at shutting off the discharge treatment. From the behavior of the trapping and releasing of radon, it is expected that radon can easily be trapped or released by the discharge treatment controlled the discharge time.

Physical adsorption method currently applied for radon trap utilizing the activated charcoal requires huge system and it is difficult to be shielded against gamma radiation from the trapped radon and its decay products. However, the new chemical trap method described above requires compact system and it is easy to be shielded.

![FIG.4. The mass balance (Ratio) between trapped and released radon](image)

**FIG.4. The mass balance (Ratio) between trapped and released radon**

**4.Conclusion**

It was cleared that the radon trapping efficiency utilizing the chemically reactive discharge treatment was over 99% and the trapped amount of radon could be mostly released and recovered by shutting off the discharge. This new chemical trapping method is suitable for removal of radon with either low or high concentrations. And the trapping and releasing process of radon can be conducted by the comparatively simple process needed just the operation discharging on and off. Moreover, the new method is effective in an aspect of radiation protection because the application of the new method leads to decrease in external exposure dose.

**REFERENCES**