Removal of Rn-222 from Contaminated Tap Water by Micro/Nano Bubble Aeration

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Abstract - Recently, the micro/nano bubble (MNB) technologies are applied in many versatile fields in the world, such as environmental fields, industrial fields, engineering, food industry, agriculture, and even medical fields. In environmental fields, since radon (Rn-222) in nature is recognized to be a group 1 carcinogen by IARC to induce cancer, an effective method to remove Rn-222 is strongly required. In this work, air FB aeration technique was applied for removal of Rn-222 radioactivity from tap water and compared with the conventional aeration process (milli-bubble), collected in Mo.11 Sum Sao sub-district, Phen district, Udon Thani province, Thailand. The results showed up to 97% removal of Rn-222 radioactivity in the tap water by the FB aeration technique and higher efficiency than the conventional aeration about 20%.

Keywords – radon (Rn-222) radioactivity, fine bubble aeration (FB aeration), RAD7, removal of Rn-222 radioactivity, tap water

I. Introduction

Radon (Rn-222) is a radioactive gas which is produced from the decay of Radium (Ra-226) in rocks and soils throughout the Earth's crust [1]. Radon sources are separated as soil (69.3%), outdoor (9.2%), building materials (2.5%), and groundwater (19%), respectively [2]. Released Rn-222s from rocks and soils are preferentially dissolved in the ground water and eventually low concentration diffused in air as represented [3,4].

Most people's activities in the World use groundwater for drinking, bathing, washing and so on. When the Rn-222 is much ingested, it can induce diseases especially cancers according to the International Agency for Research on Cancer (IARC) [5]. From the literature reviews, the alpha ($4He^{++}$) particles are produced from the decaying Rn-222 chain, consequently they hit DNA and tissues and damage. This phenomenon is the key factor of DNA and tissues damaged, and unfortunately it can induce internal cancer [6].

According to the United State Environment Protection Agency (USEPA) concern about the Rn-222 concentration in community water supplies, a drinking water should be no higher than 148,000 Bq/m^3 (148Bq/L, 4,000 pCi/L) and 11,000 Bq/m^3 (300 pCi/L), respectively [7,8].

Our group has measured the radon concentration of the groundwater samples in Khon Kaen province area, Thailand (this work has been collecting a data and preparing a manuscript for publication). The radon concentrations in the groundwater samples are in ranges of 0.01 – 28.98 Bq/m^3. Whereas some area in Nong Ruea district, a high radon concentration has been detected about 4,000 Bq/m^3. In present day hasn't informed a limitation of radon concentration in an environment for inducing a cancer disease [6,7]. However, the radon exposure had been investigated in cases of indoor high radon concentration and indoor low radon concentration [9], and studied with Canadian lung cancer of radon exposure mixed with smokers and non-smokers conditions [10]. The result found that the lifetime relative risk of radon induced lung cancer increases with the radon concentration in an environment [9, 10]. The radon concentration is recommended as bellow 200 Bq/m^3 guideline value [10]. In Thailand, a radon guideline or regular rules were not prepared by a guideline report.

The major mechanism of a removal Rn-222 from water is Rn-222 attached bubbles travelling to the water surface (or floatation process) [1,2]. Oxygen ion species (OH radical, OH^-, O_2^-, and O_3^-) are great simultaneously generated with MNB generation [17,18]. In our assumption, negatively charged air microbubbles attract Rn-222 on their surfaces, and float up to the water surface to release to the atmosphere.

Depth and surface area of a water are thus important factors for a removal of Rn-222 from water [1, 5,22].

Many research groups in the World have published the results of removal of Rn-222 from contaminated drinking water and community water supplies [11-13].

There are two type of processes of removal of Rn-222 from contaminated water by an aeration system; 1) diffused bubble aeration system, and 2) packed lower aeration system [1].

The conventional technique for removal of Rn-222 from water is the aeration technique with bubbles of an average diameter of millimeters. The mechanism is based on an activate strong convection in the water.

Jastaniah and et al. [1] and K.C. Cristina et al. had applied the aeration technique for removal of Rn-222 from a water sample and can be removed up to 97%. However, the aeration process was very long time (4 days, [12]). In our work, the MNB aeration technique had been applied for the removal of Rn-222 from a tap water sample.

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In case of the MNB technologies recently emerging rapidly, on the other hands, they have been wildly applied in wastewater or water treatment in relatively simple manner [12-16]. Therefore, we have chosen to apply the MNB technology for removal of Rn-222 contaminated community water supplies.

In this work, we applied the air FB (MNB) aeration technique for increasing removal efficiency of Rn-222 radioactivity under conditions of aiming at shorter aeration time, and a simple aeration system. Finally, we will discuss details of the reduction of Rn-222 radioacivity by FB aeration process in results and discussions. Also recently, definition of Micro/Nano bubbles are set by diameters by ISO TC/281, as follows; Fine bubbles (FB) means microbubbles (MB) (1 ~ 100μm) and ultrafine bubbles (UFB) nanobubbles (NB) (<1μm), respectively.

II. Experiment

A. Sample preparation

Tap water samples were collected at Non Sa-art village, Mo. 11, Sum Sao sub-district, Phen district, Udon Thani, Thailand (the GPS position of 17.5912725, 102.9264031), known as relatively high Rn-222 concentration in Thailand.

A Tap water was released for about 10 minutes and then collected in a plastic bottle. 1,000 mL-tap water is collected and kept in a 1,500 mL-plastic bottle with a plastic film covered and also by tightly closed cap for avoiding Rn-222 leakage. The tap water in the plastic bottle was stored at an air-condition control room and temperature of 26.5 – 27.5 ^{0}C for 12 hours.

B. Conventional Aeration, FB Aeration Process and Measurement of Rn-222 Concentration by RAD7 H_2O Electronic Radon Detector

The 1,000 mL-tap water in a 2,000 mL-glass beaker (the tap water depth and surface area of 8.25 cm and 122.26 cm2, respectively) was used for removal of Rn-222 radioactivity by the conventional process and the air FB aeration process. The conventional process (milli-bubble) consists of an air generator (model of BT6500 and an air maximum outlet of 2.5 L/min) and a sand ball head and sand cylindrical head for generating milli-bubles in conventional aeration process, as shown in Fig. 1. The FB generator has an external nozzle (model: RMUTL-VKM-01), as shown in Fig. 2, and a water flow rate of 1,000 mL/min, supported by the Center of Excellence of High-voltage Plasma & Micro/Nano bubble to Agriculture and Aquaculture, Faculty of Engineering, RMUTL, Chiang Mai Campus, Chiang Mai, Thailand [17]. Nozzle pressures for the FB aeration process are 0.25, 0.40, 0.60, and 0.70 MPa, respectively. The air conventional and FB aeration process was made to the Rn-222 contaminated tap water for 10 minutes. Whereas, the air flow rate was not controlled, and used just for ventilation.

Waiting for 10 minutes after the 10 min. The conventional and FB aeration process, the milli-bubbles and FB in the tap water samples has become in a stable state (bubbles did not collapse or decrease bubble sizes). The 250 mL of before treatment (original tap water), milli-bubbles and FB aerated tap water was then measured by the RAD7 H_2O electronic radon detector (DURRIDGE

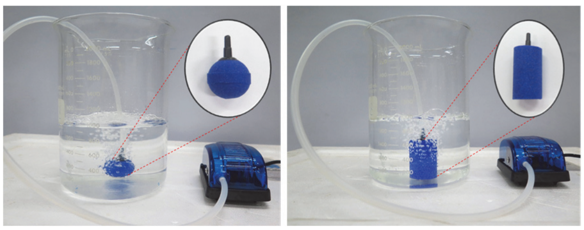


Fig. 1. Conventional aeration process consists of the air generator and a sand ball head and a sand cylindrical head, respectively.

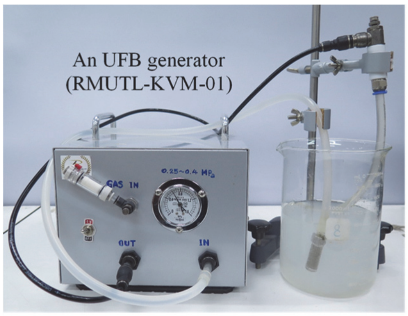
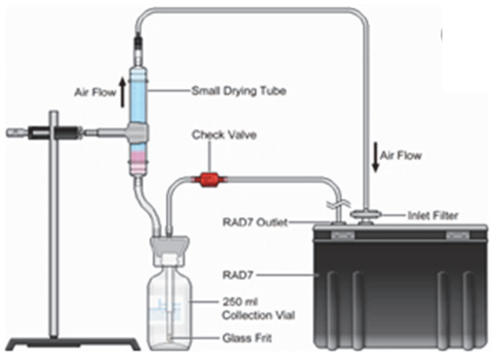


Fig. 2. Photo shows the FB generator and the external nozzle [17].

(a)



(b)

Fig. 3. Fig. 3 (a) Photo and (b) schematic of a measurement system of Rn-222 radioactivity in a tap water sample by RAD7 H_2O electronic radon detector [16].

Company Inc. USA) [18]. An operation mode of the RAD7 H_2O was Wat250, replication times for measurement of 4, and time for each replication time was 4 minutes. The photo and schematics of a measurement system of Rn-222 by the RAD7 detector are shown in Fig. 3 (a) and Fig. 3 (b), respectively.

C. Measurement of FB Size and Concentration Before and After Detection of Rn-222 by RAD7 H_2O

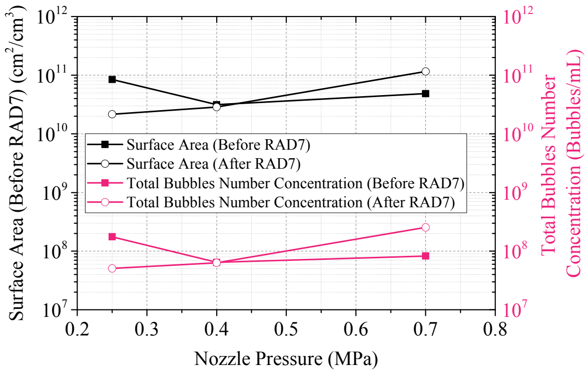
FB size and concentration before and after the measurement by the RAD7 H_2O were measured by the laser diffraction particle size analyzer (HORIBA LA-960A) at RMUTL, Chiang Mai campus, Chiang Mai, Thailand.

III. Results and Discussions

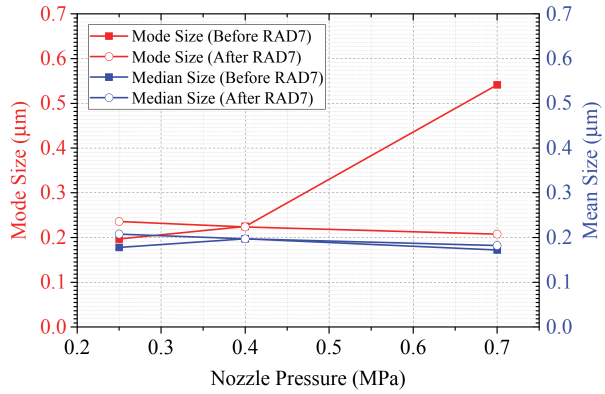
A. Measurement FB Size and Concentration Before and After Detection of Rn-222 by RAD7 H_2O

Persistence of the FBs which were generated by the FB generator does not disturb FBs by the process of a bubbles generation of RAD7 H_2O system. The results are presented in Table I and Fig. 4.

Results of bubble properties; a surface area, mode size, median size, and total bubbles number concentration dependence on the nozzle pressure are listed in Table I, where (A) and (B) before and after the Rn-222 measurement by the RAD7 H_2O, respectively. Graphs of the FB properties are also shown in Fig. 4, as a function of the nozzle pressures in the event of before and after measurement by the RAD7 H_2O. It is seen that the results show that the process of Rn-222 measurement by RAD7 H_2O does not affect the FB size and total number concentrations.



(a)



(b)

Fig. 4. Surface areas and total particle number concentrations are presented in Fig. 4 (a); black line is the surface area; pink line is the total particle number concentration; solid and circle symbol are the case of before and after bubble generation by the RAD7 H_2O system, respectively. Fig. 4 (b), red and blue line are the mode and median MNB bubble size, respectively. Whereas solid and circle symbol are the case of before and after bubble generation by RAD7 H_2O system, respectively.

TABLE I

Parameters of Surface Area, Mode Size, Median Size, and Total Bubbles Number Concentration of the Aerated FB Process Depend on the Nozzle Pressure in Case of (A) Before the Bubble Generation and, (B) After the Bubble Generation of the RAD7 H_2O System, Respectively.

(A)

Nozzle pressure (MPa)	Surface Area (cm^2/cm^3)	Mode size (µm)	Median size (µm)	Total bubbles number concentration (bubbles/mL)
0.25	8.47E+10	0.19672	0.17774	1.76E+8
0.40	3.15E+10	0.22461	0.19712	6.46E+7
0.70	4.85E+10	0.54117	0.17210	8.25E+7

(B)

Nozzle pressure (MPa)	Surface Area (cm^2/cm^3)	Mode size (µm)	Median size (µm)	Total bubbles number concentration (bubbles/mL)
0.25	2.16E+10	0.23554	0.207504	5.08E+7
0.40	2.86E+10	0.22374	0.197120	6.32E+7
0.70	1.16E+11	0.20734	0.182108	2.54E+8

B. Removal of Rn-222 Radioactivity by the Conventional and FB Aeration Process

The purpose of this work is to increase mitigating efficiency of Rn-222 radioactivity in the tap water sample by the FB aeration process when compared with the conventional process. Four nozzle pressures which control FB properties were chosen for investigating their effects on the removal efficiency of Rn-222 in the tap water samples. Tap water qualities, Rn-222 radioactivity and a removal Rn-222 of the tap water by conventional aeration process and there are two shapes of sand head for aeration (ball or spherical and cylindrical shape), as shown in Table II. Results of Rn-222 radioactivity, dissolved oxygen (DO), FB surface area and electrical conductivity and other parameters on the nozzle pressures are shown in Tables III and IV, where all these were measured at 10 minutes later after conventional and FB aeration process were initiated. Also the evolution of these parameters are shown in Figs. 5-7, respectively. And the before treatment

TABLE II

TAP WATER QUALITIES, RN-222 RADIOACTIVITY AND A REMOVAL RN-222 OF THE TAP WATER BY THE CONVENTIONAL AERATION PROCESS.

Parameter/ Sand model	Before treatment	Sand ball head	Sand cylindrical head
Electrical conductivity (mS/cm)	0.584	0.558	0.558
Temperature (C°)	26.7	25.8	25.5
PH	7.12	8.03	7.98
Salinity (ppt)	0.41	0.39	0.39
TDS (ppm)	390	382	385
Rn-222 (Bq/m^3)	2876	674	655
Removal of Rn-222 (%)	0	76.56	77.23

TABLE III

RN-222 RADIOACTIVITY, ELECTRICAL CONDUCTIVITY, DISSOLVED OXYGEN (DO) IN WATER, AND MNB SURFACE AREA DEPEND ON THE NOZZLE PRESSURE. WHEN A BEFORE TREATMENT CONDITION IS AN ORIGINAL TAP WATER.

Nozzle pressure (MPa)	Rn-222 (Bq/m^3)	Electrical conductivity (mS/cm)	DO (mg/mL)	MNB surface Area (cm^2/cm^3)
Before treatment	2,420	0.554	4.44	0
0.25	143	0.437	6.03	8.47E+10
0.40	179	0.420	5.80	3.15E+10
0.60	503	0.503	6.38	1.79E+11
0.70	71.8	0.532	5.61	4.85E+10

TABLE IV

RN-222 RADIOACTIVITY, FB SURFACE AREA, FB MODE SIZE, FB MEDIAN SIZE, AND TOTAL FB PARTICLE NUMBER CONCENTRATION DEPEND ON THE NOZZLE PRESSURE.

Nozzle pressure (MPa)	Rn-222 (Bq/m^3)	Surface Area (cm^2/cm^3)	Mode Size (µm)	Median Size (µm)	Total Bubbles number concentration (Bubbles/mL)
Before treatment	2,420	0	-	-	-
0.25	143	8.47E+10	0.19672	0.17774	1.76E+8
0.40	179	3.15E+10	0.22461	0.19712	6.46E+7
0.60	503	1.79E+11	0.24140	0.12453	2.50E+8
0.70	71.8	4.85E+10	0.54117	0.17210	8.25E+7

of the tap water means no conventional and FB aeration process made or the original tap water, as presented in Table II-IV.

In case of conventional aeration process (milli-bubble), values of electrical conductivity, temperature, PH, salinity and TDS of the tap water aren't changed by the convention aeration process with the sand ball and sand cylindrical head when compared with the before treatment tap water (original tap water), as shown in Table II. Whereas, a removal of Rn-222 radioactivity is mitigated of 76.56% (674 Bq/m^3) and 77.23% (655 Bq/m^3) for the sand ball head and sand cylindrical head, respectively, when compared with the before treatment tap water (original tap water) (2,876 Bq/m^3).

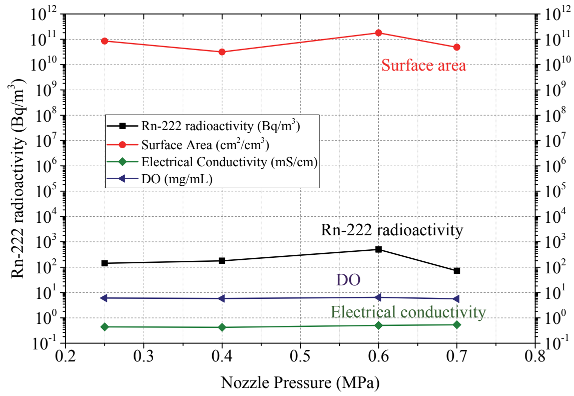


Fig. 5. Graph of Rn-222 radioactivity, electrical conductivity, dissolved oxygen (DO) in water, and FB surface area depends on the nozzle pressure. Black-, red-, blue-, and green-line are the Rn-222 radioactivity, FB surface area, DO, and electrical conductivity, respectively.

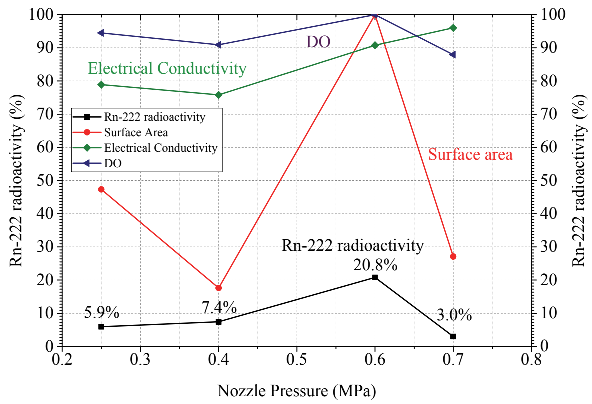


Fig. 6. Graph of Rn-222 radioactivity, electrical conductivity, dissolved oxygen (DO) in water, and FB surface area depends on the nozzle pressure.

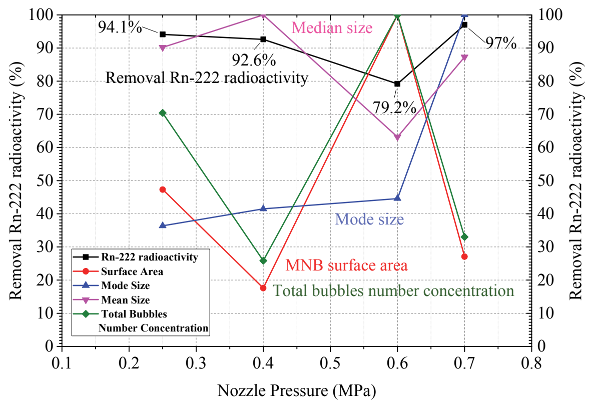


Fig. 7 Graph of removal Rn-222 radioactivity, FB surface area, FB mode size, FB median size, and total FB particle number concentration depends on the nozzle pressure. Black-, red-, green-, blue-, and pink-line are the removal Rn-222 radioactivity, FB surface area, total FB particle number concentration, FB mode size, and FB median size respectively.

In case of FB aeration process, it is seen clearly that all parameter change depends strongly on nozzle pressures, i.e., FB properties. Also it is seen that the Rn-222 concentrations dramatically decrease from 2,420 (100%) Bq/m^3 to 143 (5.9%), 179 (7.4%), 503 (20.8%), and 71.8 (3.0%) Bq/m^3 for the nozzle pressure of the MNB aeration process at 0.25, 0.40, 0.60, and 0.70, respectively.

The MNB aeration technique was compared with the previous works which were based on the conventional aeration technique (the average bubble diameter size of millimeter). Advantages of the present FB aeration technique are;

1) in a same time of aeration process, the FB aeration is higher efficiency removal Rn-222 than the conventional aeration process, 2) a shorter treatment time (10 minutes) compared with the conventional aeration (2-4 days) in the same flow rate of water [1,12], and 3) a compact system compared with the aeration system of Al-Jaseen et al., [22].

Dissolved oxygen (DO) in the tap water is seen to increase by about 30% when compared with the original tap water due to air MNB injection (4.44 mg/mL --> 6.38 mg/mL). And the electrical conductivity decreases to minimum by about 20% for the nozzle pressure of 0.40 MPa and then recovers to about 95% for the nozzle pressure of 0.70 MPa when compared with the original tap water. The PH values were found to range in 7.2 - 8.3. When the PH value was higher than 4, the Zeta potential of bubbles was found negative [23, 24]. We thus expected that the surface charge of bubbles is the main point of a mechanism of a removal of Rn-222 from water. MBs (or FBs) attached with Rn-222, then floating and rising to the surface, and finally evaporation into atmosphere. Our assumption, the MNB aeration technique is highly effective to removal of Rn-222 than the conventional aeration process because of much more number of MNB bubbles than the conventional aeration in the same volume.

Whereas the total MNB surface area is an ambiguous explanation about a relation of the nozzle pressure and Rn-222 concentrations. From Fig. 7, it is found that tendencies of the removal Rn-222 concentration and MNB median size are similar and a higher median size seems to be effective to removal of Rn-222 from the tap water than smaller median size. We assume that the bubble size is one of key parameters concerning the removal of Rn-222 from water. In the future work. this will be deeply and carefully considered.

The Rn-222 can be removed more than 90% for the nozzle pressures of 0.25, 0.40, and 0.70 MPa, although the RAD7 electronic radon detector is not precise enough for measurement of a low Rn-222 concentration [Manual Rn-222].

In future work, the MNB sizes, gas species (such as air, oxygen, nitrogen, and argon) and ratio of gas flow rate to water flow rate will be considered for further increase of removable efficiency of Rn-222 from community water supplies. In case of mechanisms of the removal Rn-222 from a water, a system for measuring a Rn-222 radioactivity in air after a FB aeration process will be designed, constructed, and measured, respectively.

IV. Conclusions

Air MNB was generated by the UFB generator (RMUTL-KVM-01), and MNB size and concentration were analyzed by the laser diffraction particle size analyzer (HORIBA LA-960A) at RMUTL, Chiang Mai, Thailand.

Rn-222 radioactivity in the 1,000mL-tap water samples could be removed about 76% by the conventional aeration process for 10 minutes, whereas more than 95% with the water flow rate of 1,000 mL/min and the FB aeration process. The FB aeration process is a very effective method for Rn-222 removal compared with the conventional aeration process, as we expected. From the results, the nozzle pressure at 0.25 MPa of the FB aeration process is a proper condition for a removal Rn-222 from a tap water.

Parameters of the tap water (electrical conductivity, temperature, PH, salinity and TDS) aren't changed by the conventional aeration process when compared with the before treatment tap water condition. Whereas, FB parameters (total FB surface area, electrical conductivity, DO, FB mode size, FB mean size, and total FB number concentration) were investigated with dependence on the nozzle pressures and Rn-222 concentrations in the tap water samples. Tendencies of the parameters of tap water are difficult clarified with the nozzle pressure dependence.

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