Original research article

Effects of Micro/Nano Bubble-Aeration Time on Removal of Rn-222 Contamination in Tap Water

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ABSTRACT

        In recent years, scientists and people have been concerned about an effect of radon (Rn-222) contamination in water and air which can induce the lung and stomach cancer. The radon contamination can be removed by two techniques: granular activated carbon (GAC) filters and aeration techniques, respectively. Whereas, micro/nano bubble (MNB) technologies have been studied in mechanisms and applied for many fields in the world such as the environment, industry, engineering, food processing, agriculture, and medicine. In this work, we have focused on the removal of radon contamination in tap water (community water supplies) through the air-MNB aeration. Tap water samples were collected at Non Sa-art, Sum Sao sub-district, Phen district, Udon Thani province, Thailand (GPS position of 17.5912725, 102.9264031). Each 1000 mL-tap water had been made the air-MNB aeration as 2, 4, 6, 8, 10, 15 and 20 minutes by the FB generator (model: RMUTL-KVM-01). After 10 minutes, 12 hours, and 24 hours of the air-MNB aeration process, 250 mL-aerated tap water had been measured the Rn-222 contamination by the RAD7 detector in operation mode of Wat250. Results show that about 92% of the Rn-222 is removed by the air-MNB aeration technique in a short aeration time. A major key of the removal of Rn-222 from the tap water is the time in making the MNB, which depends on an interfacial surface area, a bubble size and a concentration of MNB. Mechanisms of the removal Rn-222 contamination in the tap water will be presented and discussed in this paper.

Keywords: Radon; Micro/Nano bubble (MNB) Aeration; RAD7

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1. Introduction

Radon (Rn-222) is a radioactive noble gas, colorless, odorless, tasteless, which occurs naturally through decay of uranium and thorium series [1-3]. The normal isotope of Rn-222 is decayed from the decay chain of U-238 with half-life 3.82 days. An original source of Rn-222 are released from soil, rock, and the earth's crust, and then can be released/dissolved to underground water and atmosphere [4-6] which come from the decay of radium (Ra-226). Alpha product is directly released from Rn-222 as an energy of 5.49 MeV as shown in Fig. 1 (a) and (b) [4]. Alpha decayed from Rn-222 interacts with bio-organisms and can induce cell killing, chromosome damage, DNA strand breakage, and cause cancer.
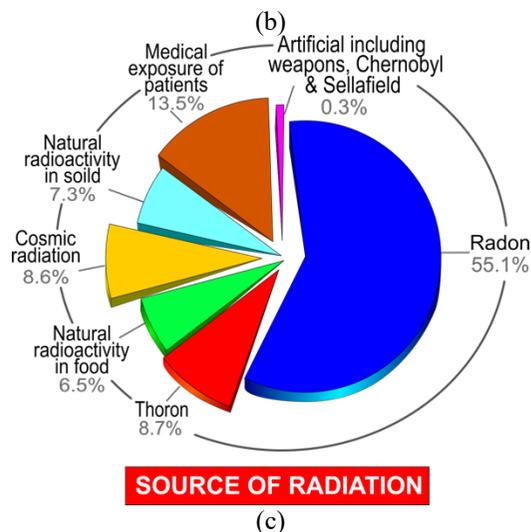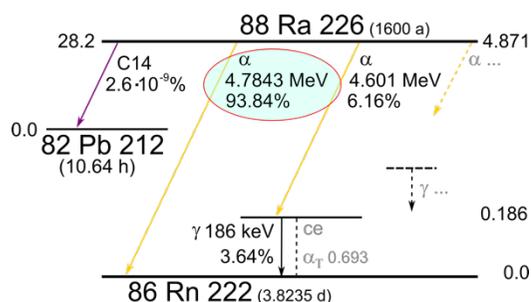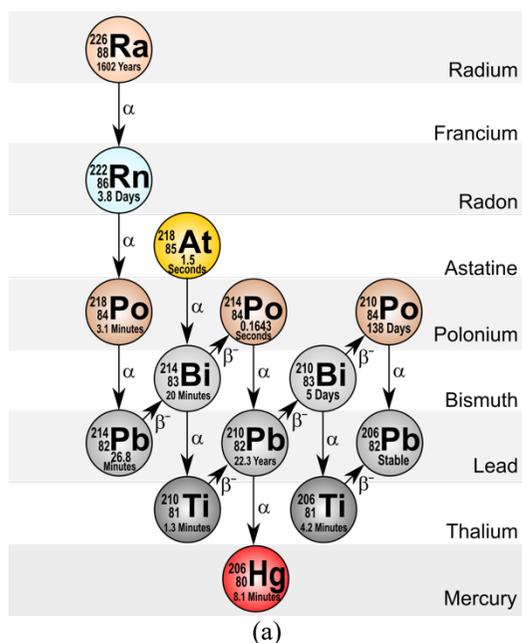

(a)

SOURCE OF RADIATION

(c)

Fig. 1. (a), (b) Radon decay chain and (c) source of radiation [4].

Rn-222 is the largest radiation source in the environment (55.1%), followed by thoron (8.7%), natural radioactivity in food (6.5%), cosmic radiation (8.6%), natural radioactivity in soil (6.3%), medical exposure of patients (13.5%), and artificial sources, including weapons Chernobyl and Sellafield (0.3%), as shown in Fig. 1 (c) [7].

Groundwater and tap water (called "community water supplies") have been used in every life activity e.g., drinking, bathing, washing, as well as inspiration/inhalation. Thus, Rn-222 can be accumulated in life activities through two paths: inhalation and direct ingestion. Long term Rn-222 exposure can have effects on health. If Rn-222 and its progeny are permitted to enter an internal body, they can go to the gastrointestinal tract, and from there to liver, lung and body tissues [8, 9]. Absorbed Rn-222 and its progeny can be a cause of lung and stomach cancer [9]. Consequently, organizations concerned with radon protection propose a limitation of the radon concentration. The guidelines of Rn-222 concentration for the drinking water and community water supplies by United State Environmental Protection Agency (USEPA)

are less than 11,100 Bq/m3 and 148,000 Bq/m3, respectively, [10].

A Micro/Nano bubble (MNB) is a tiny bubble with a diameter between 10-50 µm and less than 200 nm, respectively [11-13]. However, for a generated MNB a specific bubble's size cannot be controlled. Whereas, bubbles familiarly seen in a daily life have a diameter in the order of millimeters and are called conventional bubbles (CB). The behaviors of CB and MNB are different; the former rapidly rises and bursts to the surface, whereas the MNB remains stable in a solution for a few months because of the higher internal bubble pressure explained by the Young-Laplace's equation [11]. An MNB has higher dissolved oxygen (DO) because of its higher interfacial surface area, while the DO is the important factor for bioactivity.

The Rn-222 elimination techniques can be main classified in 3 categories: 1) aeration, 2) granular activated carbon (GAC) and 3) combination process, respectively. Aeration techniques have been widely investigated and applied in this field because of their simple preparation and green technology (without chemical reagent in the process). Our research group focuses on aeration techniques combined with micro/nanobubbles technology for the removal of Rn-222 contamination from water. Removal of Rn-222 from the community water supplies was studied by Jaseem, et al. [14] and Jastaniah, et al. [15] using aeration techniques (a conventional bubble aeration process). They could remove about 98% of Rn-222 through a continuous [14] and long-term aeration process [15]. Whereas, P. Thopan, et. al applied the MNBs technology for removal of Rn-222 contamination in tap water and found that the MNBs technology has higher potential than the conventional bubble aeration [16]. MNB has been applied for groundwater remediation by Hu et al. [17], Li et al [18] and Menendez et al. [19]. An aerobic bioremediation of the groundwater

is improved by MNB aeration process because the DO is greatly increased when compared with the original water [17-19].

As presented by Thopan et.al [16], the micro/nanobubbles technology can be applied for the removal of Rn-222 contamination in water by aeration process and shows higher efficiency than the conventional aeration (conventional bubble or macro bubble aeration). Therefore, this work studies the removal of Rn-222 from the tap water using the aeration technique combined with the micro/nanobubbles technology, which is called micro/nanobubbles aeration (MNB-aeration). Time, a parameter of MNB-aeration, is a major factor for considering and discussing the removal Rn-222 in tap water samples.

2. Materials and Methods

Tap water samples were collected at Non-Sa-art, Sum Sao sub-district, Phen district, Udon Thani province, Thailand as the GPS positioning of 17.5912725, 102.9264031. 1,000 mL-tap water samples were kept in 1,500 mL-plastic bottles.
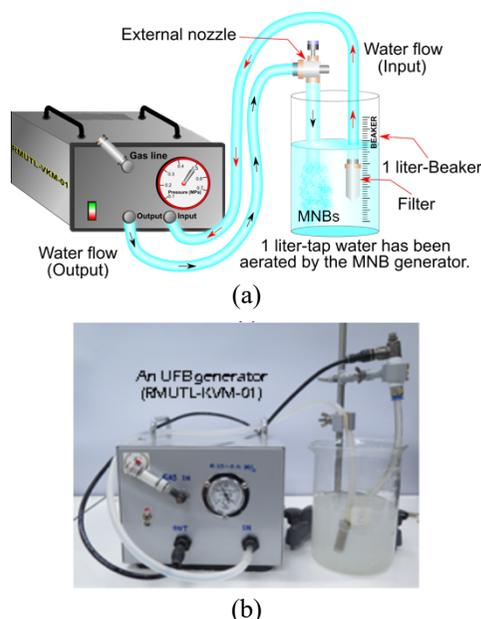

(a)

(b)

Fig. 2. (a) Schematic and (b) photo of 1,000 mL-tap water sample in 2,000 mL-beaker operated by the FB generator (model: RMUTL-KVM-01).

The 1,000 mL-tap water sample in a 2,000 mL-beaker (the depth and surface area of the tap water are 8.25 cm and 122.66 cm2, respectively) was aerated (air-MNB aeration process) by the FB generator (model: RMUTL-KVM-01) with an external cavitation nozzle, as show in Fig. 2. The FB generator was handmade by the Center of Excellence of High voltage Plasma & Micro/Nano bubble to Agriculture and Aquaculture, Faculty of Engineering, Rajamangala University of Technology Lanna, Chaing Mai, Thailand. The external nozzle pressure and water flow rate of the FB generator are 0.25 MPa and 1 L/min, respectively. Whereas, the air flow rate is not controlled, but used for ventilation. Under this condition, the FB generator generates air-MNBs of about 30 μm in diameter after 5-40 minutes, about 1 μm in diameter after 50 minutes-3 days, and about 300 nm in diameter after 4-5 days, respectively [20]. A concentration of air-MNBs is about 1×10^{12} particles/mL. Properties of the air-MNBs generated by the FB generator were measured by the HORIBA LA-960A analyzer. MNB-aeration was operated for 0 (non-aerated process/control condition), 2, 4, 6, 8, 10, 15, and 20 minutes, respectively (total 8 conditions).

(a)
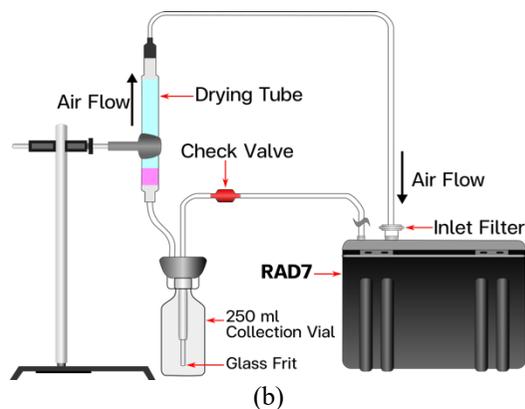
(b)
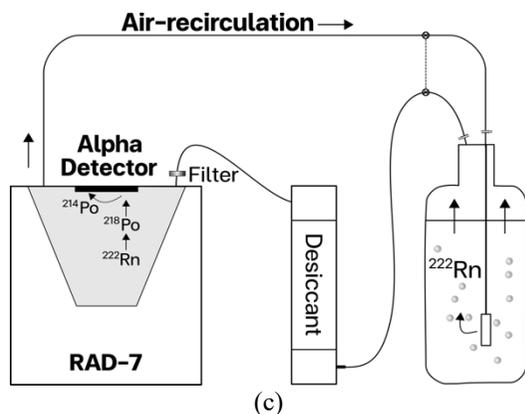
(c)

Fig. 3. (a) Photo and schematic (b), (c) of RAD7 system for measuring Rn-222 concentration in a water sample.

After 10 minutes of the MNBs aeration process (in order to achieve a stable state of MNBs), 250 mL-aerated MNBs tap water sample is gathered in a 250 mL-glass bottle. Thereafter, Rn-222 contamination in the tap water sample was measured using a RAD7 electronic radon detector of DURRIDGE Company Inc. USA [4]. A photo and schematic of the RAD7 system for measuring Rn-222 are shown in Fig. 3. RAD7 operation for measuring Rn-222 is a mode of Wat250 with replication measurement of 4 cycles and a measurement time per cycle of 4 minutes for detecting the alpha particles [22]. For each condition, the Rn-222 concentration was measured in triplicate (number of measurements is 3). Whereupon, the tap water samples were kept in an 1,500 mL-plastic bottle in an

46

ambient temperature. A plastic-cap bottle is tightly closed to avoid Rn-222 leakage.

Twelve hours and twenty-four hours later, the tap water samples were measured for Rn-222 concentration again with the RAD7 system. A layout of the experiment is presented in Fig. 4.
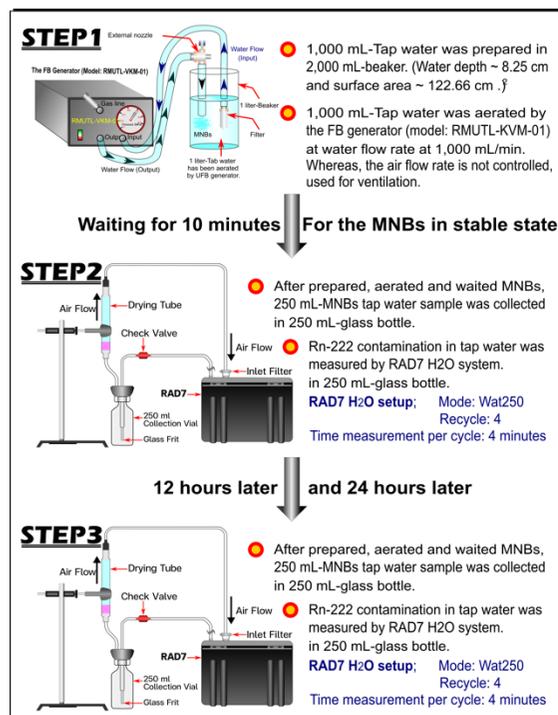

Fig. 4. A layout of the experiment.

3. Results and Discussion

In this work, we have focused on the removal Rn-222 from tap water by the MNBs aeration process. The key parameter is the aerated time of the aeration process by the micro/nanobubbles technology. Electrical conductivity (EC), temperature, pH, salinity, and total dissolved solids (TDS) in water are presented. The parameters of water were detected using the HANNA HI-9829 Multiparameter with optional GPS (HANNA instrument, Singapore).

Results of Rn-222 contamination and the concerned parameters are presented in

Tables. 1 – 3 for after 10 minutes, 12 hours later, and 24 hours later of the air-MNB aeration processes, respectively.

In Fig. 5, the solid line, dash line, and dot line are the Rn-222 concentrations measurement after the air-MNBs aeration process for 10 minutes, 12 hours, and 24 hours, respectively. From the experiment, in the case of the 10 minutes after air-MNBs aeration process, the Rn-222 concentration shows linear mitigation for 10 minutes of aerated air-MNB process. Whereas the Rn-222 concentration of the air-MNBs aeration process time from 10 minutes to 20 minutes is almost constant. For 12 hours later of air-MNB-aeration process, the Rn-222 concentration suddenly decrease for 2 minutes of the air-MNBs aeration process. After 2 minutes to 10 minutes of the air-MNBs aeration process, the tendency seems to be fluctuation, and the air-MNBs aeration time from 10 minutes to 20 minutes has an almost constant tendency. Finally, in the event of 24 hours later of air-MNBs aeration process, the tendency of Rn-222 concentrations seems to be constant. However, a poor Rn-222 has a problem of measurement by the RAD7 (a proper range measurement by RAD7 in medium to high Rn-222 concentration).

Electrical conductivity, temperature, pH, salinity, and TDS after 10 minutes, 12 hours, and 24 hours of MNB-aeration process are presented in table 1, table 2 and table 3, respectively. The results show that the parameters of EC, pH and TDS are obvious constant and independent from the removal Rn-222 contamination. However, in the case of the 12 hours-MNBs aeration process, the temperature of water increases from about 27.8 oC to about 30.3 oC. While, tendencies temperature parameter of the 10 minutes and 12 hours of air-MNBs aeration process look like same as decreased from about 31.7 oC down to 27.0 oC.

Table 1. An air-MNB aeration time dependence by MNBs aeration process. Rn-222 measurement after 10 minutes of the air-MNB aeration process.

Aeration Time (Min)	Rn-222 (Bq/m^3)	EC (μS/cm)	Temp (ºC)	pH	Salinity (ppt)	TDS (ppm)
0	$1,560 \pm 658$	547	31.8	7.42	0.24	271
2	940 ± 343	539	30.3	7.62	0.24	259
4	760 ± 277	532	29.5	7.76	0.24	265
6	687 ± 297	529	28.8	7.76	0.24	265
8	471 ± 300	530	28.3	7.74	0.23	264
10	217 ± 84.7	523	27.8	7.84	0.23	261
15	361 ± 85.6	525	27.3	7.86	0.23	265
20	253 ± 140	525	27.0	7.89	0.23	261

Table. 2. An air-MNB aeration time dependence by air-MNBs aeration process. Rn-222 measurement for 12 hours later of the air-MNB aeration process.

Aeration Time (Min)	Rn-222 (Bq/m^3)	EC (μS/cm)	Temp (ºC)	pH	Salinity (ppt)	TDS (ppm)
0	$1,410 \pm 329$	534	28.0	7.74	0.24	266
2	361 ± 85.6	514	27.7	7.86	0.23	258
4	543 ± 323	515	27.8	7.90	0.23	256
6	144 ± 118	511	27.5	7.94	0.23	255
8	543 ± 299	530	30.1	7.92	0.23	264
10	252 ± 138	505	30.1	7.95	0.23	252
15	108 ± 137	511	30.4	7.95	0.23	255
20	180 ± 137	508	30.5	7.99	0.22	254

Table. 3. An air-MNB aeration time dependence by air-MNBs aeration process. Rn-222 measurement for 24 hours later of the air-MNB aeration process.

Aeration Time (Min)	Rn-222 (Bq/m^3)	EC (μS/cm)	Temp (ºC)	pH	Salinity (ppt)	TDS (ppm)
0	325 ± 247	531	31.7	7.82	0.23	266
2	108 ± 138	550	32.4	7.95	0.23	273
4	144 ± 166	520	32.0	7.92	0.23	258
6	36.1 ± 72.2	537	31.3	8.04	0.23	268
8	180 ± 182	532	30.3	8.05	0.23	266
10	71.8 ± 82.9	528	29.9	8.06	0.23	262
15	108 ± 71.9	536	29.0	8.05	0.23	266
20	180 ± 273	527	29.0	8.08	0.23	262

Note of Table1, Table 2 and Table 3: All parameters are measured for 3 replications, *n*=3. And the parameters of EC, temperature, pH, salinity and TDS are presented of an average value.

An average normalized removal Rn-222 is shown is shown in Fig. 6 as solid-, dash- and dot-line are the removal Rn-222 after air-MNBs aeration process for 10 minutes, 12 hours and 24 hours, respectively.
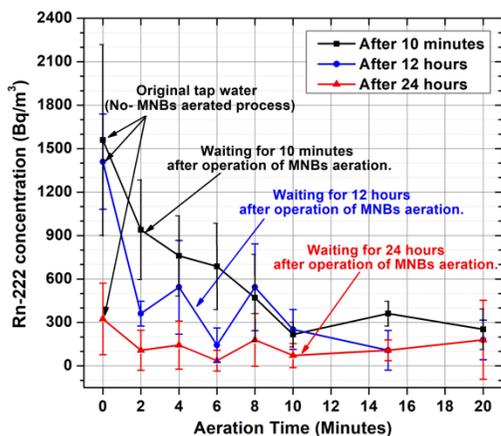

Fig. 5. The Rn-222 concentration in the tap water when measured after air-MNBs aeration process for 10 minutes, 12 hours and 24 hours represented by the solid line, dash line, and dot line, respectively.
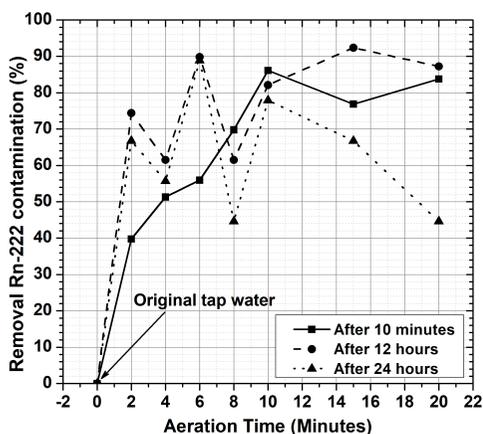

Fig. 6. An average normalized removal Rn-222 contamination in the tap water sample by the air-MNBs aeration process. Solid-line, dash- and dot-line are the removal Rn-222 after air-MNBs aeration process for 10 minutes, 12 hours and 24 hours, respectively.

The removal of Rn-222 is similar for the air-MNBs aeration time from 8 minutes to 20 minutes. And the removal Rn-222 is possibly reaches saturation as the air-MNB-aeration time increases from 10 minutes. The removal of Rn-222 from the tap water is eliminated up to 86% at an aeration time of 10 minutes (1,000 mL-tap water and water flow rate of 1,000 mL/minute). After 12 hours, the Rn-222 in the tap water sample was measured again (no MNB aeration process, the air-MNB aeration is generated only once) and it was found that the Rn-222 is removed more than 92% (lack of RAD7 is a detecting low Rn-222 concentration, then a low Rn-222 concentration is detected with fluctuated values). The results in Fig. 5 show that the Rn-222 concentration in the tap water sample is about 250 Bq/m3 after the air-MNB aeration time of 10 minutes of the all case measurements (10 minutes, 12 hours and 24 hours of air-MNB aeration process). This condition of the air-MNB aeration times is the proper condition which optimizes cost process together with the removal of Rn-222 contamination.

Mechanisms of a mitigated/removed Rn-222 from water have been presented by many research groups [14-18]. A brief summary of the mechanism is that the oxygen (unclear that OH$^\bullet$ radical, O^+, O_2^+ or O_3^+ is high effectiveness) brings Rn-222 to the water surface where it evaporates to the atmosphere. And the research groups [8, 23] propose that the removal of Rn-222 contamination by aeration process depends on parameters of: 1) temperature of water and air, 2) physicochemical characteristics of Rn-222, 3) Rn-222 concentration in the air and water 4) air to water ratio 5) residence period (flow rate/residence period of bubbles rise to the top surface water) and 6) accessible area for mass transfer (bubbles size/smaller size for higher interfacial surface area and higher mass transfer), respectively. From the assumptions of the mechanism above, the micro/nanobubbles

technology has advantages for the aeration process since small bubbles (in this work, the bubbles size range from 1 μm down to 300 nm) have properties of: 1) negative charge on surface of MNBs (negative zeta potential), 2) increasing interfacial area of the MNBs contact with water solution (higher absorbed/interaction with an ionic ion or chemical composition in water solution), 3) increasing a mass transfer and 4) more resident in the water following the equation of Stokes law [24]. The first 3 parameters are controlled by properties of a water solution, whereas the last 3 parameters are controlled by the device and working situation. In this work, we guest that a key effect of removal of Rn-222 contamination from the progeny of Rn-222 (alpha particle-an ionic/positive charge, the RAD7 detects alpha particles) attached/absorbed at the interfacial surface MNBs (very high interfacial surface interaction when compare with the conventional bubble or conventional aeration) then slowly rises to a top surface water and finally, evaporation to the atmosphere. Another minor mechanism, MNBs resident in the water solution can be changed. Therefore, we found that the bubble size and the gas species inside the bubbles are important parameters of a removal of Rn-222 concentration. Thereby, in our future work, bubble sizes and gas species inside the MNBs will be studied and reported. Whereas depth and surface area of water sample can't be ignored.

Advantages of the air-MNB aeration (an average of diameter bubble size is in order of μm or less than μm) compared with a former aeration technique (an average of diameter bubble size is in order of mm) are high effective in the removal of Rn-222 contamination, short time aeration, compact MNBs aeration process system, and no-chemical contamination in process.

4. Conclusion

The novel technique is the air-MNBs aeration process for removal Rn-222 contamination in tap water samples.

For 2 minutes of the MNBs aeration process, the Rn-222 concentration can be removed about 40%. For 2 minutes to 10 minutes of the air-MNB aeration process, the removal Rn-222 concentration tendencies of both conditions increase until to about 90%. Finally, for 10 minutes to 20 minutes of the air-MNB aeration process, the removed Rn-222 concentration is a saturation.

Measurement of the Rn-222 concentration at 12 hours and 24 hours later, shows the removed Rn-222 tendencies are similar in the case of the measured Rn-222 after 10 minutes of the MNBs aeration process. However, the tendencies fluctuate because of a limitation of the RAD7 electronic detector (very low Rn-222 concentration in the tap water, the best detected range of the RAD7 in medium to high Rn-222 concentration).

Finally, the relation of the electrical conductivity (EC), pH, temperature, salinity and total dissolved solid (TDS) with the removed Rn-222 are difficult explanations concerning the mechanism of removal of Rn-222.

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