CHARACTERIZATION OF RADIOACTIVE AEROSOLS IN TEHRAN RESEARCH REACTOR CONTAINMENT

by

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The objectives of this research were to determine the levels of radioactivity in the Tehran research reactor containment and to investigate the mass-size distribution, composition, and concentration of radionuclides during operation of the reactor.
A cascade impactor sampler was used to determine the size-activity distributions of radioactive aerosols in each of the sampling stations. Levels of α and β activities were determined based on a counting method using a liquid scintillation counter and smear tests. The total average mass fractions of fine particles (particle diameter $dp < 1$ μm) in all of the sampling stations were approximately 26.75 %, with the mean and standard deviation of 52.15 19.75 μg/m³. The total average mass fractions of coarse particles were approximately 73.2%, with the mean and standard deviation of 71.34 24.57 μg/m³. In addition to natural radionuclides, artificial radionuclides, such as ^{24}Na, ^{91}Sr, ^{131}I, ^{133}I, ^{103}Ru, ^{82}Br, and ^{140}La, may be released into the reactor containment structure. Maximum activity was associated with accumulation-mode particles with diameters less than 400 nm. The results obtained from liquid scintillation counting suggested that the mean specific activity of alpha particles in fine and coarse-modes were 89.7 % and 10.26 %, respectively. The mean specific activity of beta particles in fine and coarse-modes were 81.15 % and 18.51 %, respectively. A large fraction of the radionuclides' mass concentration in the Tehran research reactor containment was associated with coarse-mode particles, in addition, a large fraction of the activity in the aerosol particles was associated with accumulation-mode particles.

Key words: radionuclide, mass-size distribution, particle, activity, research reactor

INTRODUCTION

Recent developments in nuclear science and technology have heightened the need to develop new nuclear power plants and research reactors for production of radioisotopes and radiopharmaceuticals for the treatment of patients, and also for electricity production. There are around 84 research reactors in developed countries and nearly two-thirds of these are more than 30 years old [1].

The main concerns about the construction and commissioning of new nuclear power plants and research reactors are potential health problems and the need for protection of workers and the surrounding population from nuclear reactor hazards, such as the emission of radioactive substances from the reactors. In general, emission control strategies of radioactive substances into the environment, protection of workers and researchers against radiation exposure, the establishment of negative pressure inside reactor buildings to mitigate the health hazards of radioactive emissions, and the provision of a safe working environment for employees, are important components of nuclear reactor safety programs [2]. The International Atomic Energy Agency (IAEA) has established a number of standards and safety requirements for assessing the safety conditions of research reactors. The main variable that determines the types of radionuclides and isotopes released from research reactors is integrated power level [3]. Throughput levels of research reactors vary considerably from a few watts to over 100 MW, which are lower than the throughput levels of power reactors (3000 MW). The risk of research reactors harming surrounding residents is lower than the risk of damage from power reactors. However, it is thought that the risks of releasing radioactive materials from research reactors are higher than from power reactors [4]. Some gas, liquid, and solid wastes with low to moderate activ-

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ity levels, are emitted from the surfaces of reactor cladding or reactor fuel, during normal operation of nuclear reactors. The type and quantity of generated wastes depend on various factors, including; type of reactor, design features, operation conditions, and integrity of the reactor fuel. The main sources of airborne radioactive aerosol emissions are; fission products, non-volatile elements emitted from the decay of radioactive gases and adsorption of volatile radionuclides, produced during the fission process onto suspended aerosols in the air, especially on accumulation-mode aerosols in the size range of 0.1 to 1 μm [3, 5]. Reineking *et al.* [6] demonstrated that more than 50 % of activity distributions in different particles are associated with accumulation-mode particles. The results also showed that 10-40 % of this activity is related to particles in Aitken-mode and only 10 % of the activity is associated with particles in coarse-mode [6]. The results of some studies have suggested that more than 80 % of ^{131}I activity is viewed in fine aerosol particles (with diameter d = 0.1-1 μm), while less than 20 % of activity is related to coarse aerosol particles (d >10 μm) [7]. The behavior of atmospheric aerosols has effects on the behavior of airborne radionuclide particles, which can be absorbed on atmospheric particles and create radionuclide particles. Deposition of bonded and unbonded radioactive particles in the human respiratory system is primarily dependent on the particle's size. Since it is difficult to measure the activity of radioactive particles in the human respiratory tract, activity-size distribution is the leading parameter used to estimate radiation dose in cases of inhalation exposure [8]. Although surveys such as that, conducted by Anvari and Safarzadeh [2], have identified and assessed the level of radionuclides released into the environment and evaluated the total effective dose equivalent for accidental release from the Tehran research reactor (TRR) [2], one major drawback in this area is that there has been a little discussion about the behavior of radionuclides in the internal environment of the reactors.

The objectives of this research were to determine the level of radioactivity in the TTR and to investigate the radionuclide's size distribution, composition, and concentration levels during operation of the research reactor. This study was designed to identify radionuclides and to determine the size distribution and activity of radionuclides, in the TRR containment, in accordance with the International Organization for Standardization (ISO) 2898 method [9].

MATERIALS AND METHODS

The Tehran research reactor is a light water open pool-type research reactor. Iran's 5 MW research reactor, Materials Testing Reactor (MTR) has been operating since 1967, and it uses highly enriched uranium as a fuel [2]. The sampling of radioactive aerosols was carried out according to procedures of the ISO 2898 standard method [9]. The sampling stations were selected on the basis of their distance from radionuclide producing sources and vents providing fresh air into the research reactor containment, to ensure the accurate assessment of the concentrations and activities of the aerosols. In order to measure the activities of the radioactive aerosols, three sampling stations were selected as pilot stations. To identify the range of radionuclides produced during the operation of the TRR, a high-volume sampler equipped with a PVC filter, using an airflow rate of 0.05 m^3/min was applied, and a gamma ray spectrometer (High-purity germanium (HPGe) detector) was used to identify the radionuclides. A collection efficiency of 99.99 % for 0.3 μm aerosols was reported for the PVC filter [10]. Furthermore, an Andersen cascade impactor with an effective cutoff diameter in the micron size range, can be used to separate particles from the air according to their aerodynamic diameters [9, 10]. The cascade impactor sampler (Andersen 1-ACFM aerosol cascade impactor) was used to determine the distribution of radioactive particles according to their size-activity, in each sampling station. An eight-stage cascade impactor was used to collect the radioactive particles and the effective cut off diameter for stage 1 to 7 of the impactor and filter were 11.7, 4.7, 3.3, 2.1, 1.1, 0.7, and 0.4 μm. The last stage of the impactor consisted of a collision plate and a high performance filter (E 99.99). Radioactive particles less than 0.4 μm in diameter were collected on glass fiber filters. The cascade impactor flow rate was 28.3 l/min, according to the manufacturer's recommended procedure. In order to weigh the aerosol mass, each stage of the impactor was measured before and after collecting aerosols. Levels of α and β activities were determined based on a counting method using a liquid scintillation instrument and smear tests. The smear filter paper results were counted using liquid scintillation counting, after preparation in polyethylene vials. Five milliliters of 0.1 M nitric acid (HNO_3) were added to the vials in order to wash contaminants from the smear papers. Vials were placed for 10 minutes in an ultrasonic bath, then the smear papers were taken from the vials and 12 ml of Hi-Safe 3 scintillation fluid were added to the vials. Sample preparation and radioactivity measurements of the filters used in the impactor were conducted in a similar way with the smear papers, but instead of 0.1 M HNO_3, hydrofluoric acid solution was added to the vials to wash the filter.

RESULTS

Radioactive aerosol samples were collected continuously using a high-volume sampler (approximately 100 cubic meters). In this study, measurements of the radioactive aerosols were performed at fixed lo-

Table 1. Aerosol mass concentrations and sampling conditions at the three sampling stations

Sampling station	Number of samples	Average concentration [μgm^{-3}]	*P*-value	Temperature [°C]	Relative humidity [%]	Air velocity [m min^{-1}]
1 (the first quarter of the year)	5	130	0.345	27.22	38	13.25
2 (the second quarter of the year)	5	110		28.16	39	16.27
3 (the third quarter of the year)	5	87		25.55	44.4	13.60

cations in the TRR containment and during the operation time of the reactor, thus, the spatial variables had a minimum effect. Sampling stations selection criteria were based on the distance from the radionuclide producing sources and vents for providing fresh air into the research reactor containment, to ensure the accurate assessment of concentrations and activities of the aerosols. The results obtained from the statistical parameters associated with the measured values, during three seasons from April to December 2012, are shown in tab. 1. Considering the fact that the measurements were performed during the operation time of the reactor, in different months, the aerosol-size distributions showed fluctuations across the measurements. Total aerosol mass concentration, temperature, relative humidity (RH) and air velocity at each workstation, are shown in tab. 1.

Figure 1 presents the results obtained from aerosol mass-size distribution measurements in the TRR containment, at three different sampling stations. It is apparent from this figure that the majority of aerosol mass-size distribution measurements were related to coarse-mode particles ($dp > 1\mu m$).

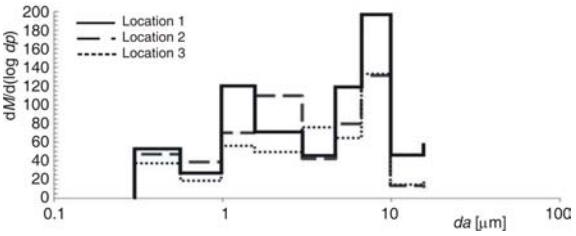

Figure 1. Aerosols mass size distribution in TRR containment at three sampling stations
da is the aerodynamic diameter, dp – the particle diameter, and M – the particle mass concentration

The total average mass fractions of fine-mode particles ($dp < 1\mu m$), in all of the sampling stations, were approximately 26.75 %, with the mean and standard deviation of 52.15 19.75 μg/m^3. The total average mass fraction of the coarse-mode particles was approximately 73.2 %, with the mean and standard deviation of 71.34 24.57 μg/m^3.

The results of gamma-spectrometry for determination of natural and artificial radionuclides, in airborne particles collected onto PVC filters, during the operation time of the reactor, are presented in tab. 2. Most of the radionuclides showed negative beta decay. Artificial radionuclides produced during fission process consisted of ^{91}Sr, ^{131}I, ^{133}I, and ^{103}Ru. ^{24}Na radionuclides were neutron activation products of ^{23}Na. ^{24}Na nuclides are activation products and this is not related to fission products [9-11].

The current study found that the average activity median aerodynamic diameters (AMAD), at the three sampling stations, were 2.4, 3.1, and 3 μm, with geometric standard deviations (GSD) of 2.30, 2.12, and 2 μm, respectively.

Figure 2 shows the mass-size distribution and specific activity for the alpha and beta particles, according to diameter distribution, at the three sampling stations. The results obtained from liquid scintillation counting suggested that the mean specific activity of alpha particles, in fine ($dp < 1\mu m$) and coarse-modes ($dp > 1$ μm), at the three sampling stations, were 89.7 % and 10.26 %, respectively. The mean specific activity of the beta particles, in fine ($dp < 1\mu m$) and coarse-modes ($dp > 1\mu m$), at the three sampling stations, were 81.15 % and 18.51 %, respectively.

As shown in fig. 2, there was no clear relationship between particle mass concentration and the specific activity for alpha and beta particles. Moreover, no significant differences were found between the activities of particles at the three sampling stations.

Table 2. Released radionuclides into TRR containment

Natural radionuclides	Type of decay	Half life	Artificial radionuclides	Type of decay	Half life
^{40}K	β^-	1.24 year	^{24}Na	β^-	897 min
^{226}Ra	β^-/α	1601 years	^{82}Br	β^-	2116 min
^{232}Th	α	1.4 10^{10} years	^{91}Sr	β^-	578 min
^{212}Pb	β^-	638 min	^{131}I	β^-	11548 min
^{214}Pb	β^-	26.8 min	^{133}I	β^-	1248 min
^{212}Bi	β^-/α	60.55 min	^{103}Ru	β^-	56500 min
^{208}Tl	α	3.07 min	^{140}La	β^-	2416 min
^{228}Ac	β^-/α	367 min	–	–	–

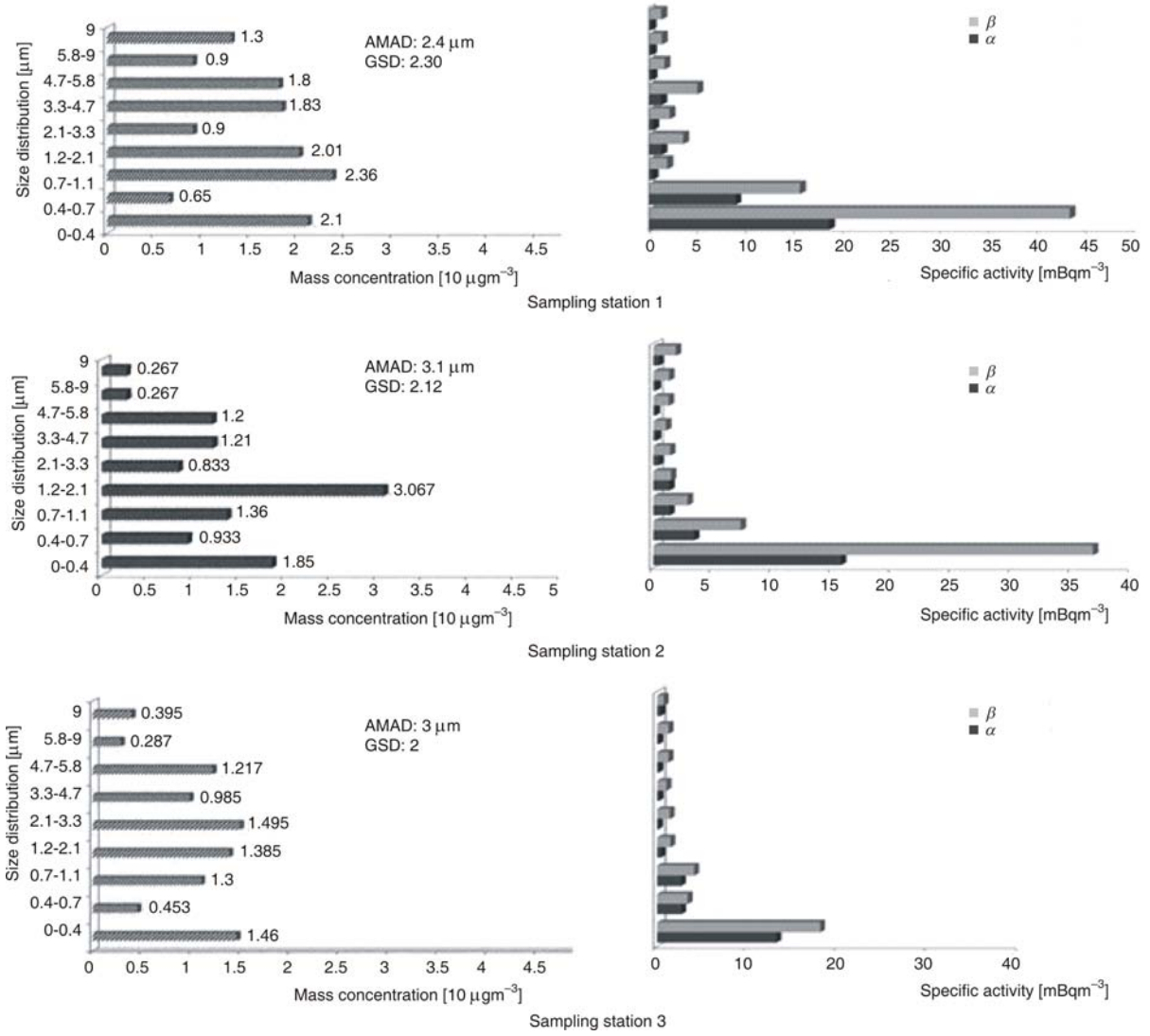

Figure 2. Mass concentration (left) and specific activity (right) for alpha and beta particles at three sampling stations
GSD is the geometric standard deviation, AMAD – the average activity median aerodynamic diameters

DISCUSSIONS

This study set out to determine the levels of radioactivity in the TRR containment and to investigate the radionuclide's size distribution, composition and concentration, during operation of the research reactor. This study endeavored to identify the radionuclides produced and to determine the size distribution and activity of the radionuclides in the TRR containment.

There was no significant difference between aerosol mass concentrations at the three sampling stations in the TRR containment (P-value = 0.345, 95 % confidence interval). The results obtained from a coefficient of variation (CV) method indicated that the distribution of measured aerosols in sampling station 1 (near the heart of the reactor), was more uniform, while the distribution of the measured aerosols in sampling stations 2 and 3, were less uniform. Particle size distributions observed, were in the range of 4.6 to 9.8 μm, furthermore, large fractions were associated with coarse-mode particles. The distribution of particle mass-size depends on the nature of the production process and also on corresponding psychrometric conditions [12]. Due to the higher rate of airflow in the TRR containment (15.23 m/min), the particle size distribution tended to shift toward the larger particle sizes.

Analysis of filter samples, using gamma spectrometry, showed that natural and artificial radionuclides were released into the TRR containment. Most of the radionuclides showed negative beta decay. There are two forms of iodine, gas phase and particle phase iodine. The particle forms of iodine were released from the studied research reactor. These ^{131}I radionuclides may be released into the environment from nuclear reactors during nuclear accidents [13-15]. It was found that ^{131}I had a rather different activity-size distribution from other fission products such as ^{103}Ru and ^{137}Cs. ^{131}I with diameters less than 0.47 showed a higher fraction on the back-up filter of

the cascade impactor, whereas, ^{103}Ru and ^{137}Cs showed a more pronounced maximum at 0.93 μm geometric mean particle diameter [7, 9, 13].

The activity and mass concentrations of particles, at the three sampling stations, were identified on the basis of particle sizes, AMAD, and geometric standard deviation. The current study found that the average AMAD at the three sampling stations were 2.4, 3.1, and 3 μm, respectively. This study produced results which corroborate the findings of a number of previous studies conducted in this field [16].

The differences in activity size distributions may be due to differences in the effective cut off diameter obtained by collecting the particles using different stages of the Andersen cascade impactor [9]. The results obtained from investigating the specific activity, at the three sampling stations, indicated that more than 59 % of the beta particles' activities were related to particles with diameters less than 0.4 μm, 14.3 % of activities of beta particles were associated with particles with diameters in the range of 0.4 μm to 0.7 μm and more than 6 % of the beta particles' activities were related to particles with diameters in the range of 3.3 μm to 7.4 μm. The current study found that more than 66 % of the alpha particles' activities were related to particles with diameters less than 0.4 μm, more than 18 % of activities of alpha particles were associated with particles with diameters in the range of 0.4 μm to 0.7 μm and more than 7 % of activities of alpha particles were related to particles with diameters in the range of 3.3 to 7.4 μm. Maximum activity at the three sampling stations was associated with accumulation-mode particles (dp < 1μm) with diameters less than 400 nm. Activity-size distributions in particle-bounded radionuclides are affected by surface distribution [17-19]. Maximum activity was related to accumulation-mode particles, which are important in terms of surface area distribution [18]. The results suggested that a large fraction of the activity in the aerosol particles was associated with accumulation-mode particles in the range of 100 nm to 1000 nm and only a small fraction was related to nuclei mode in the range of 10 nm to100 nm. Wiever's study [20] found that approximately 74 % of the absorption activity of the particles was associated with accumulation-mode particles (0.1 μm-1 μm). The activity of coarse and fine-mode particles was 2 % and 24 %, respectively. These findings were in agreement with previous findings, which showed that a large fraction of activity in the aerosol particles was associated with accumulation-mode particles (dp < 1μm) [21-23].

No significant differences were found between the activities of alpha particles (P-value <0.607) and beta particles (P-value <0.334), at the three sampling stations. Although, a large fraction of mass concentration, at three sampling stations in the TRR containment, was associated with coarse-mode particles (dp > 1μm), the specific activity for alpha and beta particles was higher in the fine-mode particles (dp < 1μm), than in the coarse-mode particles. The settling velocity of accumulation-mode particles is about 10^{-2} cm/s. Taking into consideration this settling velocity, particles are slowly removed from the particle-laden air, as these are suspended in the air for a relatively long time. The settling velocity of coarse-mode particles is about 20 cm/s, which is higher than the settling velocity of accumulation-mode particles. In regards to this settling velocity, coarse-mode particles have a short residence time in the air and radionuclides cannot adhere to coarse-mode particles [24]. Size, distribution and the number of particles are also important factors in the adsorption of radionuclides onto aerosol particles. High surface to volume ratio, a higher number of fine-mode particles, and more interactions between fine-mode particles, could be predicted to produce greater activity in these particles.

CONCLUSION

This study was designed to determine the levels of radioactivity in the TRR containment and to investigate the radionuclide's size distribution, composition, and concentration levels during the operation of the research reactor. Maximum activity was associated with accumulation-mode particles with diameters less than 400 nm. No significant differences were found between the activities of alpha particles and beta particles at the sampling stations. A large fraction of the mass concentration of radionuclides in the TRR containment was associated with coarse-mode particles and a significant amount of activity in aerosol particles was associated with accumulation-mode particles.

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AUTHORS' CONTRIBUTIONS

The study concept and design was performed by G. Moradi, A. Sadighzadeh, and R. Yarahmadi. Data acquisition was done by G. Moradi and B. Rezaei Fard. The interpretations of results were performed by R. Yarahmadi, G. Moradi, A. Sadighzadeh, and A. A. Farshad. The manuscript was written by G. Moradi and L. Omidi and was reviewed by R. Yarahmadi.

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КАРАКТЕРИЗАЦИЈА РАДИОАКТИВНИХ АЕРОСОЛА У ЗАШТИТНОМ СУДУ ТЕХЕРАНСКОГ ИСТРАЖИВАЧКОГ РЕАКТОРА

Циљ овог истраживања је утврђивање нивоа радиоактивности у заштитном суду Техеранског истраживачког реактора и испитивање расподеле маса и димензија, састава и концентрације радионуклида током рада реактора. Коришћен је каскадни сударач за узорковање како би се одредиле расподеле димензија и активност радиоактивних аеросола на свим локацијама узорковања. Нивои алфа и бета активности одређени су употребом бројача са течним сцинтилатором и тестова са брисевима. Укупни просечни масени удео финих честица ($dp < 1\ \mu m$) на свим локацијама узорковања био је приближно 26,75 %, са средњом вредношћу и стандардном девијацијом 52,15 19,75 $\mu g/m^3$. Укупни просечни масени удео грубих честица био је приближно 73,2 %, са средњом вредношћу и стандардном девијацијом 71,34 24,57 $\mu g/m^3$. Поред природних радионуклида, унутар заштитног суда могу се ослободити и вештачки радионуклиди: ^{24}Na, ^{91}Sr, ^{131}I, ^{133}I, ^{103}Ru, ^{82}Br и ^{140}La. Максимална активност је приписана прикупљајућим честицама пречника мањег од 400 nm. Резултати добијени помоћу бројача са течним сцинтилатором упућују да је средња специфична активност финих и грубих алфа честица 89,7 % и 10,26 %, респективно, а средња специфична активност финих и грубих бета честица 81,15 % и 18,51 %, респективно. Велики део концентрације масе радионуклида у заштитном суду Техеранског истраживачког реактора потиче од грубих честица, при чему већи део активности аеросола потиче од прикупљених честица.

Кључне речи: радионуклид, расподела маса и димензија, честица, активност, истраживачки реактор