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Removal of Radioactive Gas by Zeolite Filter from Nuclear Power Plants

Research Article

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Abstract

During the normal operation of nuclear power plants, some radioactive wastes are produced in the form of particles or aerosol gas in the reactor building. Particulate radioactive aerosols can be produced in a wide variety of particle sizes, possibly in combination with non-radioactive aerosols. Emission of corrosion products and fission products that are activated by the effect of nuclear rays generate aerosols from two sources. These; are created by the adsorption of gases generated by radioactive decay and volatile radionuclides formed during the fission process on the present suspended material. The most important volatile radionuclides that form the gaseous radioactive waste produced during the normal operation of nuclear power plants are halogens, noble gases, tritium, and carbon-14. The composition and amount of radioactivity present in the various airborne waste streams depend largely on the reactor type and release path. All gaseous waste from nuclear power plants must be treated before discharging into the atmosphere. In this paper, radioactive radon gas was used to represent the radioactive gases generated from nuclear power plants and natural zeolite was used as adsorbent material for radon removal. A series of experiments were conducted to measure the performance of the filter made in the zeolite. First of all, an approximate particle distribution size in the range of 1 to 3 mm was obtained by grinding natural zeolite. The ground material was then compressed in cylindrical adsorbent moulds of 35 mm diameter and 10 mm height. After the moulds were filled with the material, they were dried by heating to 110 ° C for 24 hours. At the end of the heat treatment, the adsorbent beds were cooled and connected to the test apparatus. RAD7 radon test device was used in the experiments. The RAD7 is a portable instrument that uses a solid-state alpha detector to measure radon gas concentrations in the range of 4.0-750,000 Bq / m3. The sampler of the RAD7 device works by drawing an air sample from an inlet filter into a 0.7 L sample cell covered with an electrical conductor. At the centre of the hemisphere, the cell is a planar silicon detector implanted with an ion to measure radioactivity. As result of the experiments, it shows that the zeolite filter absorbs 85% radioactive radon gas and can be used as an air filter in nuclear power plants.

Keywords: Radioactive, radon, gaseous, zeolite, filter.

Özet

Nükleer santrallerin normal çalışması sırasında, reaktör binasında partikül veya aerosol gazı şeklinde bazı radyoaktif atıklar üretilir. Parçacık radyoaktif aerosoller, çok çeşitli parçacık boyutlarında, muhtemelen radyoaktif olmayan aerosollerle kombinasyon halinde üretilebilir. Nükleer ışınların etkisiyle harekete geçen korozyon ürünleri ve fisyon ürünlerinin emisyonu iki kaynaktan aerosol üretir. Bunlar; radyoaktif bozunma tarafından üretilen gazların ve fisyon işlemi sırasında oluşan uçucu radyonüklidlerin mevcut asılı malzeme üzerinde adsorpsiyonu ile oluşmaktadır. Nükleer santrallerin normal çalışması sırasında üretilen gaz halindeki radyoaktif atıkları oluşturan en önemli uçucu radyonüklitler halojenler, soy gazlar, trityum ve karbon-14'tür. Havadaki çeşitli atık akışlarında bulunan radyoaktivite bileşimi ve miktarı, büyük ölçüde reaktör tipine ve salınım yoluna bağlıdır. Nükleer santrallerden çıkan tüm gazlı atıklar atmosfere deşarj edilmeden önce arıtılmalıdır. Bu yazıda, nükleer santrallerden üretilen radyoaktif gazları temsil etmek için radyoaktif radon gazı kullanılmış ve radon giderimi için adsorban malzeme olarak doğal zeolit kullanılmıştır. Zeolit içinde yapılan filtrenin performansını ölçmek için bir dizi deney yapılmıştır. Öncelikle, doğal zeolitin öğütülmesiyle 1 ila 3 mm aralığında yaklaşık bir parçacık dağılım boyutu elde edildi. Öğütülmüş malzeme daha sonra 35 mm çapında ve 10 mm yüksekliğinde silindirik adsorban kalıplarda sıkıştırılmıştır. Kalıplar malzeme ile doldurulduktan sonra 110 ° C'de 24 saat ısıtılarak kurutuldu. Isıl işlemin sonunda adsorban yatakları soğutulmuş ve test aparatına bağlanmıştır. Deneylerde RAD7 radon test cihazı kullanılmıştır. RAD7 cihazı, 4.0-750.000 Bq / m3 aralığındaki radon gazı konsantrasyonlarını ölçmek için katı hal alfa dedektörü kullanan taşınabilir bir cihazdır. RAD7 cihazının örnekleyicisi, bir giriş filtresinden bir hava örneğini bir elektrik iletkeni ile kaplı 0,7 L'lik bir örnek hücreye çekerek çalışır. Yarım kürenin merkezinde, hücre, radyoaktiviteyi ölçmek için bir iyon implante edilmiş düzlemsel bir silikon detektördür. Deneyler sonucunda zeolit filtrenin % 85 oranında radyoaktif radon gazını tutabildiği ve nükleer santrallerde hava filtresi olarak kullanılabileceğini göstermektedir.

Anahtar Kelimeler: Radyoaktif, radon, gaz, zeolit, filtre.

1. INTRODUCTION

Fission and activation gases are also produced during the normal operation of nuclear power reactors. Radioactive gases are transported to various parts of the building where the facility is located as a result of their leakage in the reactor system. This is because the gases formed during the reaction are completely dissolved in the reactor cooler. As a result, if a suitable filter is not used, fission gases and radioactive particles spread around the facility. By integrating suitable filters into the plant ventilation system, these gases are kept in the filter environment and not spread to the atmosphere. In nuclear power reactors, radioactive gases are trapped with filters in order to minimize the radioactivity released into the environment. In addition, radionuclides in the form of particles can also be transported in the air in nuclear power reactors. The emission points and size of radioactive gases and airborne particles depend on the reactor type (PWR / BWR) and plant design. Typical gaseous and airborne particle source terms are defined in Table 1. The main sources of gaseous waste generation are presented in Table 2.

Radioactive gas waste systems from nuclear power reactors are designed to keep the fission and activation gases at extremely low levels for radioactive decay before they are released. Containment methods include storage in pressurized tanks, long delay lines, and holding by absorption in charcoal beds. The gas retention system, consisting of filters to keep radioactive gases free from the environment, is integrated with radiation monitors that are capable of monitoring emissions and also detect leaks, spikes, or other unintentional releases.

Table 1. PWR Gaseous Source Terms Prior to Treatment.

Gaseous Radionuclide	Ci/year
Kr-85m	2.7
Kr-85	2.6
Kr-87	1.1
Kr-88	3.3
Xe-131m	6.5×10^3
Xe-133m	1.1×10^2
Xe-133	7.7×10^3
Xe-135m	3.0
Xe-135	2.8×10^2
Xe-137	<1
Xe-138	4.0
	Total: 1.7×10^4

The main gaseous fission products released in nuclear power reactors consist of various noble gases such as xenon (Xe) and krypton (Kr). Many fission products are produced during the nuclear fission process in reactors. Radioactive Gaseous fission products are released into the reactor cooling system due to defects in the fuel coating and transport through the fuel coating. Major emitted noble gases include (Kr, Xe) and radioiodine. Radioiodine can exist in a variety of chemical forms such as I2, IO3, HOI, and CH3I.

Activation products as well as gaseous fission products are released as a result of the nuclear fission process. Major released activation gases include nitrogen, oxygen, argon, and tritium. A large number of particles are also activated in the reactor environment with these radioactive gases. These particles include normal radionuclides found in both solid waste and liquid waste streams. A list of specific activation products that generally occur in PWRs is presented in Table 2.

Table 2. PWR Radioactive Particulate Release.

Radionuclide Particulates	Total Release Rate Ci/year
Cr-51	9.7×10^{-5}
Mn-54	5.8×10^{-5}
Co-57	8.2×10^{-6}
Co-58	4.1×10^{-4}
Co-60	5.9×10^{-5}
Fe-59	2.7×10^{-5}
Sr-89	1.6×10^{-4}
Sr-90	6.3×10^{-5}
Zb-95	9.1×10^{-6}
Nb-95	5.0×10^{-5}
Ru-105	1.7×10^{-5}
Ru-106	7.8×10^{-7}
Sb-125	1.7×10^{-6}
Cs-134	4.8×10^{-5}
Cs-136	3.3×10^{-5}
Cs-137	1.1×10^{-4}
Cs-138	2.9×10^{-6}
Ce-144	1.3×10^{-5}
Total:	1.2×10^{-3}

Since much more dangerous emissions are taken into account in nuclear accidents, radon gas emission is not usually included in this type of scenarios. But in some nuclear reactors, radium is used to provide alpha particles that can interact with beryllium to produce neutrons through the reaction and by this way radon gas is emitted. In scope of the nuclear decay chain, radon gas is the decay product of radium, and radium is the decay product of uranium. Since radon gas is the main decay product of radium, radon gas is one of the radioactive gases in a nuclear reactor.

PWRs have a relatively small volume of gas (15 cfm) enabling batch processing; Whereas, BWRs have high gas volumes (250 cfm), which necessitates a continuous processing system was defined by Moghissi, Godbee, and Hobart (1986, pp.2-3). Gaseous radioactive wastes are managed in three ways; a) Particle filtration, b) Gas adsorption, and c) Decay in holding tanks. Various filtering units have been designed to remove both gaseous radioactive waste and airborne particles. Commonly used systems include High-Efficiency Particulate Air Filters, HEPA filters, and coal adsorption. These systems include integrated holding tanks for degradation and final release.

2. METHODS AND MATERIALS

In this research, zeolite-based mineral composite filter was prepared and experiments were made to remove radioactive gaseous waste. Zeolite filters are prepared in 10 mm thickness and 45 mm diameter. Radon gas is used to represent radioactive gases. Radon gas retention performance of the mineral-based composite radon filters were determined. Laboratory tests were performed to determine effects of the additive ratio on the filter performance. Composite zeolite filter samples were prepared for this research (Figure 1).

Figure 1. Zeolite Filter Radon Test Device RAD7.

The trend in the applications to EPO was similar during this time period (**Figure 2**); composite, polymeric and implantable biomaterials are also the leading sub-groups [19]. Furthermore, applications for especially "coating materials" are seen in the EPO database, which was not grouped for USPTO. In other words, the range of biomaterial types are expanded in EPO. However, the main in-demand groups or types of biomaterials in USPTO as well as in EPO are equal.

Radon is a naturally occurring radioactive noble gas. It is part of the natural decay series of uranium (U) and thorium (Th) found in all soils and rocks to a varying concentration. There are three radioisotopes of radon naturally present in the environment: Radon-222 from the uranium-238 decay series, radon-220 from thorium-232 decay series and radon-219 from uranium-235 decay series. Radon-219 is of low radiological significance because of its short half-life of 4 seconds and uranium-235 represents a small percentage (0.3%) of the activity of naturally occurring uranium. Inhalation of indoor radon (222Rn) and thoron (220Rn) decay products is the most important source of exposure to ionizing radiation for the human respiratory tract suggested by Lubin et.al (1997, 126-134), Chau et al (1997, 69-74) and Yuan et al (2019, 204-205). In this study, the radon measuring device (Rad7) and integrated into the radon accumulation cell were used in the experiment system (Fig.1). CAPTURE software was used to evaluate the measured values. The regular gas flow was maintained via one-way gas valves to the radon measuring device (RAD7). The gas drying column was added to the test system as an intermediate stage in order to prevent the measurement system of the device from being affected by humidity. One-way valves are also used to prevent radon gas recirculation. The test system is integrated into the radon meter and the results are transferred to the computer via CAPTURE (Figure 2). In the beginning, unfiltered radon gas test measurements were taken in the sniffing mode and unfiltered radon gas activity values were determined. After the initial values were determined, the zeolite composite filter prepared for this study was used for performance measurements.

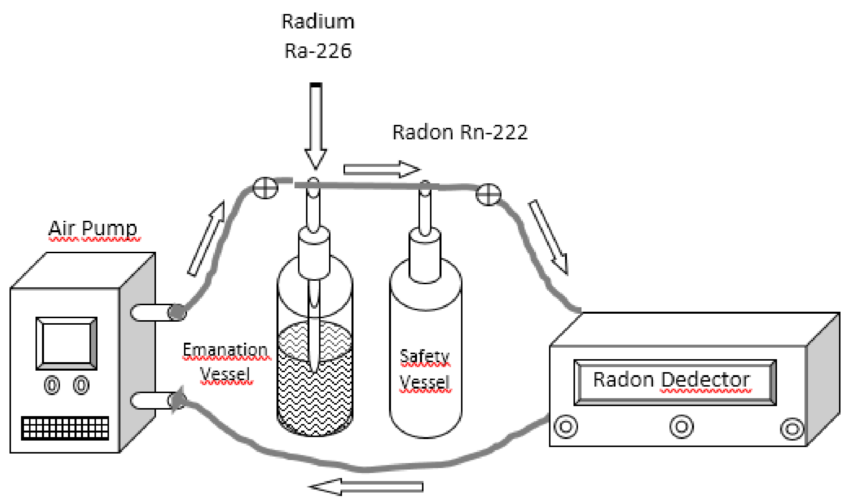

Figure 2. Radon Measurement System.

3. RESULTS

Zeolite filter tests were performed when the radon accumulation in the radon deposition chamber reached equilibrium. Initially, the reference material prepared only as binder material was tested as reference material. Then, each filter sample was placed in the filtration system and their adsorption capacities were determined. Comparison of additive amounts in terms of radon gas filtering and determination of filtering performance of composites is determined and results were presented in Figure 3.

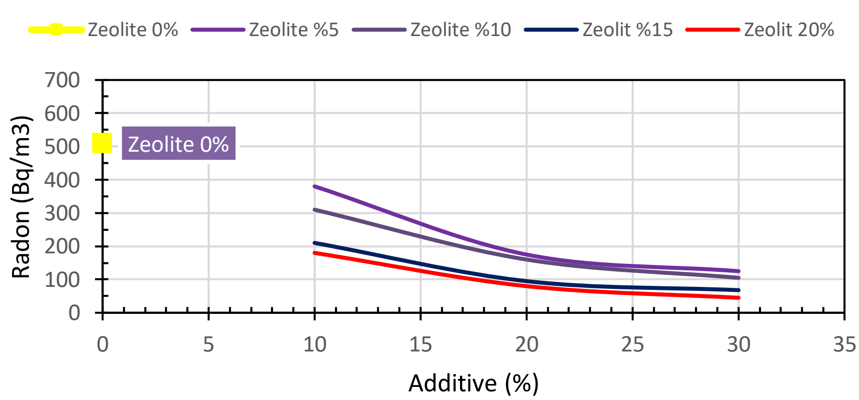

Figure 3. Filter Performance Test Results.

4. RESULTS

The performance tests of zeolite-added composite filters for removing radioactive gaseous wastes were successfully performed and their values were recorded. It was determined that the zeolite filters can hold 70% radioactive radon gas. In the case of comparison of test materials, the best performance was recorded for the 20% zeolite doped filter composite sample.

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