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Silica Fume as a Radon Retardant from Concrete

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Radon is a radioactive gas that can be generated from concrete. Its concentration is enhanced in indoor environments, and tracheobronchial deposition of radon progeny can lead to lung cancers. Aggregates (granite) are known to be the radon source in concrete. Recently, silica fume is introduced as a partial substitution in Portland cement to produce high-strength concrete. It can set in the aggregate−cement paste interface and lower the porosity in the interface zone. It can also effectively fill up the voids between cement grains, which suggests its ability to retard radon emission from concrete aggregates. In the present work, radon exhalation rates from concrete cubes with silica fume were measured using charcoal canisters and γ-spectroscopy and were considerably smaller (by $\sim$ 4 mBq m^{-2} s^{-1}) than those from normal concrete ($\sim$ 9 mBq m^{-2} s^{-1}). The indoor radon concentration reduction is estimated as $\sim$17 Bq m^{-3} using a room model, while the radon dose reduction is $\sim$ 1 mSv yr^{-1}. Therefore, silica fume is a simple material to reduce the indoor radon concentration and the radon dose.

Introduction

Radon (^{222}Rn) is a naturally occurring radioactive inert gas with a half-life of 3.82 days and is a decay product of ^{226}Ra, which is present in geological materials (rocks, soil, etc.) and concrete at natural levels. The radon gas concentration can be greatly enhanced in man-made indoor environments, mainly due to poor ventilation. Radon decays to form radon progeny. While most of the radon gas inhaled will be exhaled, the radon progeny will adhere to the respiratory tract. It is now established that the tracheobronchial deposition of radon progeny in the human body can lead to lung cancers (*1, 2*).

There are two common approaches to measuring radon exhalation rates from concrete. The first is to measure enclosed samples giving the exhalation rate per unit of mass (μBq kg^{-1} s^{-1}), while the second one is to measure walls or blocks giving the exhalation rate (mBq m^{-2} s^{-1}) (see, e.g., ref *3*). Mean exhalation rates per unit of mass have ranged from 0.6 to 28 μBq kg^{-1} s^{-1} (*4−7*), while mean exhalation rates have ranged from 0.6 to 8 mBq m^{-2} s^{-1} (*7*). The mean values for concrete in Hong Kong were respectively 21 μBq kg^{-1} s^{-1} (*8*) and 8 mBq m^{-2} s^{-1} (*9, 10*). It is noted that the radon exhalation rate will be affected by the moisture content of the concrete (*4, 6, 10−12*). Nevertheless, for mature building materials, the variation in moisture will not be very large (*13*).

From the universal aim to reduce exposures to ionizing radiations as much as reasonably achievable, it is necessary to identify efficient methods to mitigate the radon hazard. The main source of indoor radon for grounded houses is the soil underneath the building, while that for high rise buildings is the concrete used as a building material. Although it was established that radon from concrete effectively came from the aggregates (granite) (*14*), up to now, relatively few efforts have been devoted to identify ways to reduce the radon exhalation from concrete surfaces.

When a radon atom is formed from radium through α-decay, several possible fates await it within the concrete depending on the position of its formation and the particular characteristic medium through which it traverses. It may stop within the originating grain material, traverse to adjacent grains, or otherwise stop in the pore system and capillary network of the concrete. Only those atoms that stop in the pore system and capillary network can efficiently leave the concrete by diffusion through the pore system or by transportation within the pore fluids, those generally being air or water.

High-strength concrete containing silica fume is increasingly being adapted in design and construction of a variety of important structures (*15*), throughout the world including Hong Kong, because of its improved strength and durability. Silica fume, also known as micro silica, is a byproduct resulting from the reduction of high-purity quartz with coal in electric arc furnaces in the production of silicon and ferrosilicon alloys. Micro silica is a fine powder of spherical particles with an average diameter of about 0.1 μm, which is about 2 orders of magnitude finer than particles of ordinary Portland cement. Micro silica also contains more than 90% silicon dioxide; thus, they are highly effective pozzolanic material to be used in concrete (*16*). A pozzolanic material contains silica in a reactive form which will, in finely divided form and in the presence of moisture, chemically react with calcium hydroxide to form compounds possessing cementitious properties.

Micro silica, when used in concrete, improves the properties of concrete 2-fold. First, due to its extreme fineness, it can effectively set the aggregate−cement paste interface, lower the porosity in the interfacial transition zone, and also effectively fill up the voids between cement grains. Second, because it is a highly effective pozzolanic material, micro silica reacts with calcium hydroxide present in the hydrated Portland cement to produce additional calcium silicate hydrates. As a result, the addition of micro silica is effective in improving the performance of concrete with a considerable improvement in ultimate strength in producing high-strength concrete (*17*) and reduces the porosity of both the matrix and the aggregate−paste transition zone (*18*).

The ability for the silica fume particles to lower the porosity of both the matrix and the aggregate−paste transition zone immediately suggests its ability to retard radon emission from the aggregates and radon exhalation from the concrete. The present work is devoted to study the difference in the radon exhalation rates between normal concrete (NC) and concrete with silica fume (SFC).

Experimental Section

Sixteen (eight pairs) concrete cubes (side length of 150 mm) were cast in the laboratory on the same day (January 5, 1999). Four pairs were cured for 7 days while the other four pairs were cured for 28 days. The date on which curing started

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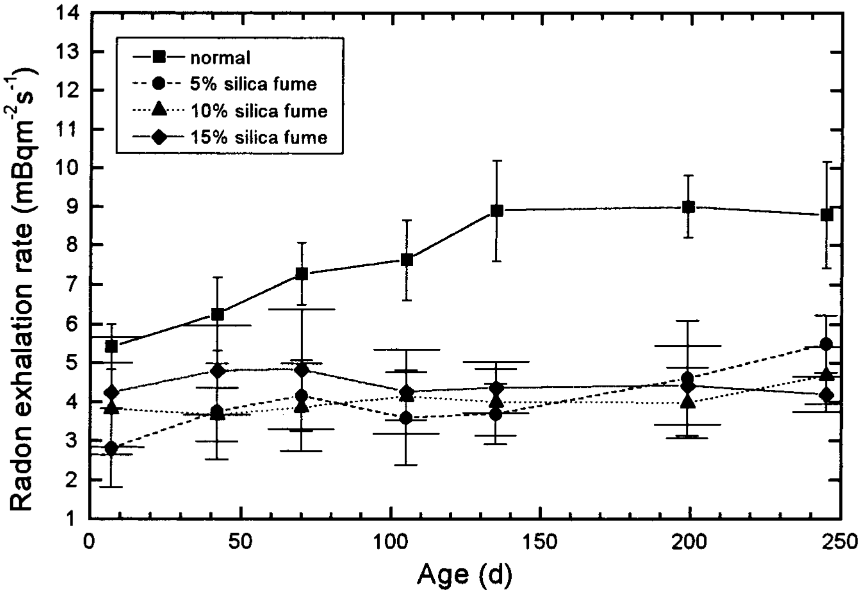

FIGURE 1. Radon exhalation rates from concrete cubes cured for 7 days. (Caps of error bars have increasing width for data from normal concrete to 5%, 10%, and 15% substitution by silica fume.)

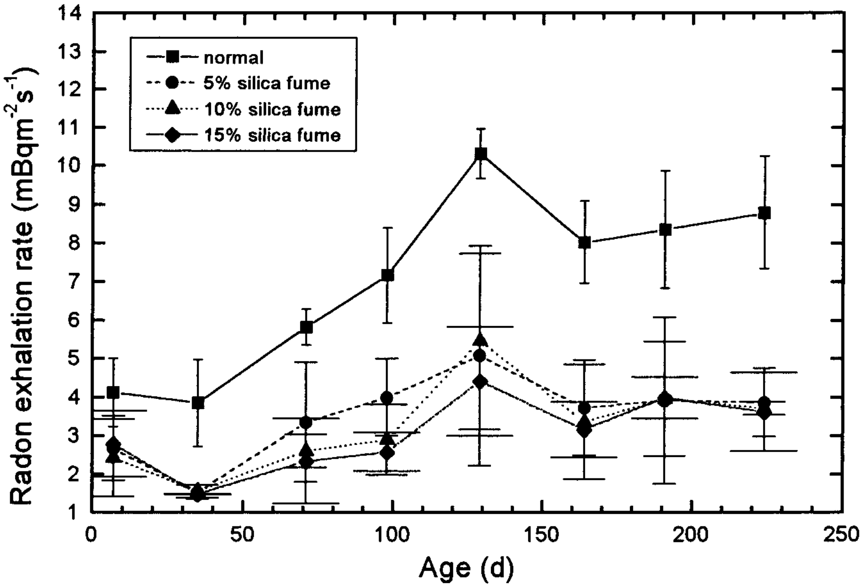

FIGURE 2. Radon exhalation rates from concrete cubes cured for 28 days. (Caps of error bars have increasing width for data from normal concrete to 5%, 10%, and 15% substitution by silica fume.)

coincided with the date of casting. Curing is the process of keeping the concrete saturated with moisture to promote hydration of the concrete until the originally water-filled space in the fresh cement paste has been filled to the desired extent by the products of hydration of cement (*19*). Among the four pairs, one was for NC while three were for SFC (5, 10, and 15 wt % substitution of Portland cement, respectively).

During the course of the experiments, the concrete cubes were kept in a laboratory maintained at a constant temperature of 23 °C and a relative humidity of about 60%. For each of the concrete cubes, three adjacent surfaces were employed for measurement of the radon exhalation rates. The method for measurement of radon exhalation rates was described in detail in ref *20*. Standardized charcoal canisters (with diameters of 10 cm and weights of 70 g) (*21, 22*) were used to collect the radon exhaled for 2–3 days. These were sealed against the concrete surfaces with a silicone sealer to stop air leakage. After collection, the charcoal canisters were removed from the surface, sealed, and stored so that the radon could reach secular equilibrium with its progeny. The radon activities inside the canisters were then determined by counting the γ-ray photons emitted by the radon progeny inside at energies of 295, 352, and 609 keV using a NaI γ-spectrometer for 10 min.

The age of the concrete is defined in this work as the time (d) elapsed after the curing period. The radon exhalation rates from concrete cubes cured for 7 days were measured at their ages of 7, 42, 70, 105, 135, 199, and 245 d, while those from concrete cubes cured for 28 days were measured at their ages of 7, 35, 71, 98, 129, 164, 191, and 224 d. Measurements made on different days for concrete cubes with different curing periods were for experimental convenience only.

Results and Discussion

The results on the radon exhalation rates for concrete cubes cured for 7 and 28 days are shown in Figures 1 and 2 and also in Tables 1 and 2, respectively. From Tables 1 and 2, the

TABLE 1. Radon Exhalation Rates from Concrete Cubes (Normal Concrete and Concrete with 5%, 10%, and 15% Substitution by Silica Fume[a]

	radon exhalation rate (mBq m^{-2} s^{-1})					
age (d)	**normal concrete**	**5% silica fume**	**10% silica fume**	**15% silica fume**	***A* (%)**	***B* (%)**
7	5.42 ± 0.58	2.82 ± 1.01	3.82 ± 1.18	4.25 ± 1.42	91	63
42	6.25 ± 0.93	3.76 ± 1.23	3.67 ± 0.69	4.81 ± 1.15	91	65
70	7.28 ± 0.8	4.16 ± 0.91	3.86 ± 1.12	4.84 ± 1.54	93	46
105	7.63 ± 1.03	3.60 ± 1.22	4.15 ± 0.62	4.27 ± 1.08	95	38
135	8.90 ± 1.31	3.70 ± 0.78	4.00 ± 0.86	4.37 ± 0.66	96	55
199	9.01 ± 0.81	4.62 ± 1.48	3.98 ± 0.91	4.43 ± 1.02	97	35
245	8.79 ± 1.38	5.49 ± 0.74	4.68 ± 0.74	4.20 ± 0.45	97	83

[a] Curing period was 7 days. *A* is the largest probability (for a particular age) that the data for normal concrete are different from the concrete with silica fume substitutions (evaluated using *t*-statistic) while *B* is the largest probability that the data for concrete with silica fume substitution themselves are different (also evaluated using *t*-statistic).

TABLE 2. Radon Exhalation Rates from Concrete Cubes (Normal Concrete and Concrete with 5%, 10%, and 15% Substitution by Silica Fume[a]

	radon exhalation rate (mBq m^{-2} s^{-1})					
age (d)	**normal concrete**	**5% silica fume**	**10% silica fume**	**15% silica fume**	***A* (%)**	***B* (%)**
7	4.12 ± 0.88	2.67 ± 0.84	2.42 ± 1.01	2.79 ± 0.86	79	27
35	3.85 ± 1.12	1.53 ± 0.18	1.55 ± 0.17	1.48 ± 0.01	90	38
71	5.82 ± 0.47	3.35 ± 1.56	2.60 ± 0.43	2.34 ± 1.11	98	47
98	7.17 ± 1.24	4.00 ± 1.00	2.90 ± 0.92	2.58 ± 0.50	96	79
129	10.3 ± 0.65	5.08 ± 2.86	5.46 ± 2.29	4.42 ± 1.41	97	36
164	8.03 ± 1.07	3.73 ± 1.24	3.36 ± 1.49	3.16 ± 0.72	97	37
191	8.36 ± 1.52	3.91 ± 2.16	3.96 ± 1.49	3.99 ± 0.54	94	4
224	8.80 ± 1.45	3.87 ± 0.89	3.71 ± 0.17	3.62 ± 1.01	96	18

[a] Curing period was 28 days. *A* is the largest probability (for a particular age) that the data for normal concrete are different from the concrete with silica fume substitutions (evaluated using *t*-statistic) while *B* is the largest probability that the data for concrete with silica fume substitution themselves are different (also evaluated using *t*-statistic).

results for different amounts of silica fume substitution are essentially the same. This agrees with the argument that once the amount of silica fume is sufficient to cover the surface of all coarse aggregate particles (in our case 5 wt % of the Portland cement), additional silica fume will not be notably beneficial because the excess silica fume can no longer set the aggregate−cement paste interface (*19*). Therefore, we can combine the data for the concrete with a different percentage of silica fume substitution as the data for SFC. Second, the radon exhalation rate saturated around 70 and 130 days for the SFC cubes cured for 7 and 28 days, respectively, while the radon exhalation rate saturated at substantially later dates for NC. This agrees with the observations that strength development for SFC ceases much earlier than NC because of the high early reactivity of silica fume (*23*). Third, considering our data for the longest curing time (245 and 224 d for cubes cured for 7 and 28 d, respectively), the average radon exhalation rates from SFC are 4.8 and 3.7 mBq m^{-2} s^{-1} for curing periods of 7 and 28 days, respectively; for both curing periods, the average radon exhalation rate from NC is 8.8 mBq m^{-2} s^{-1}, which is in excellent agreement with previous results for NC (*9, 10*). In other words, a reduction of 4−5 mBq m^{-2} s^{-1} in the radon exhalation rate from concrete can be achieved by using silica fume.

A room model was employed by Yu (*24*) to assess the indoor radon gas concentration from laboratory radon exhalation experiments. The room model was shown to give results in excellent agreement with real measurements (*25, 26*). Using this room model and assuming that silicon fume only reduces radon exhalation from the walls (i.e., the floor cover is already so efficient that the use of SFC will not further reduce the radon exhalation from the floor, which is the most conservative case), the reduction in the indoor radon concentration ΔC (Bq m^{-3}) by using SFC is $\Delta C = 3.72\Delta\epsilon$ (Bq m^{-3}) where $\Delta\epsilon$ (mBq m^{-2} s^{-1}) is the reduction in the radon exhalation rate from concrete achieved by using silica fume. From the above, $\Delta\epsilon$ is 4−5 mBq m^{-2} s^{-1}, so ΔC is about 15−19 Bq m^{-3}, which is significant as compared to the average indoor radon concentration of ~33 Bq m^{-3} in Hong Kong (*27*). The dose conversion factors for a typical dwelling in Hong Kong (*26*) are 0.072 and 0.049 mSv yr^{-1} (Bq m^{-3})$^{-1}$ for the James model (*28*) and the National Research Council model (*29*), respectively, so the reduction in the radon dose is estimated to be about 1 mSv yr^{-1} (0.7−1.3 mSv yr^{-1}), which is a significant value.

From the above, it is concluded that silica fume is a simple and economical material to reduce the indoor radon concentrations and the corresponding radiation dose from radon. In short, the use of silica fume approximately halved the radon exhalation from concrete and so halved the doses from this source.

Acknowledgments

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