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Research on the additives to reduce radioactive pollutants in the building materials containing fly ash

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ABSTRACT

Several kinds of functional additives such as barite, zeolite, ferric oxide, gypsum, and high alumina cement were introduced to prepare a low-radiation cement-based composite to reduce radioactive pollutants contained in fly ash. The effect of content and granularity of the functional additives on the release of radioactive pollutants were investigated. Composites were characterized by X-ray diffraction, Scan electron microscopy. The results indicate that the radioactive pollutants contained in the fly ash can be reduced by adding a proper amount of zeolite, ferric oxide, gypsum, and high alumina cement. The release of radon from fly ash decreases with a decrease in the granularity of additives. Compared with traditional cement-based composite containing fly ash, the release of radon can be reduced 64.8% in these composites, and the release of γ-ray is decreased 45%. Based on the microstructure and phase analysis, we think that by added functional additives, there are favorable to form self-absorption of radioactivity in the interior composites. This cement-based composite will conducive to fly ash are large-scale applied in the field of building materials.

1. Introduction

1.1. Radon and its carcinogenesis

Radon disintegrated from radioactive radium is a colorless, odorless, and inert radioactive gas. It is transformed from radium in the decay chain of uranium, an element found at varying amounts in all rocks, soil, ores and some mines all over the world. Radon gas escapes readily from ground or rocks into the air and disintegrates into short-lived decay products called radon daughters or radon progeny. The short-lived progeny, which decays by emitting heavily ionizing radiation called alpha particles, can be electrically charged and attach to aerosols, dust and other particles in the air that humans can inhale into their body. Radon is highly dissolvable in fat and inhalable. Having a strong affinity to human fat, radon can be easily absorbed into fatty tissues, nerve systems, the reticuloendothelial system, and blood causing cell damage and cancer. The decay products of the radon, such as ^{218}Po, ^{214}Bi, ^{210}Pb, and metal particles, mainly cause the damage. They deposit on the breathing system, and produce alpha particles continuously. As a result, the body tissues and cells are suffered from long-term radiation attacks [1]. This causes damage to DNA and RNA molecule structure through electrophoresis, therefore affecting the information carried on them, cell reproduction, and finally causing mutation of the chromosomes. Ultimately, it can result in lung cancer. According to the data provided by the Environmental Protection of America, there are 21,000 people died of radon pollution or radon annually, exceeding the number of death caused by AIDS every year in the United States of America (http://www.house.sina.com.cn). There are about 550,000 people died of cancer caused by radon or its decay products in Mainland China. In Hong Kong, 30% of the cancer sufferers are victims of radon and its progeny. The World Health Organization (WHO) has announced that radon is one of the 19 main cancer-causing materials in the environment, which is also included in the list of the indoor cancer-causing substances by the international cancer research organization. Radon will not only increase the chance of contracting cancers (in particular lung cancer), septicemia, but also descend the possible damage to the second and even the third generations due to the systemic damage to human cells [2]. All these cell damages may remain undetectable for quite some time.

1.2. Indoor radon contaminations

With the rapid economic development, a large number of industrial waste were discharge into circumstance in our world, mainly fly ash, slag, blast furnace slag, steel slag, copper slag, coal gangue,

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Table 1
The activity of natural radionuclide in fly ash samples Bq/kg.

Sample origin	^{238}U	^{232}Th	^{236}Ra	^{40}K
Hangzhou, Zhejiang	181	103	173	230
Quzhou, Zhejiang	201	164	179	347
Taizhou, Zhejiang	132	143	140	197
Zhenhai, Zhejiang	284	96	148	183
Xiamen, Fujian	–	77	73	378
Zhanping, Fujian	–	92	90	481
Dezhou, Shandong	113	250	111	92
Laiwu, Shandong	98	87	99	293
Zhibo, Shandong	147	112	148	382
Yantai, Shandong	180	134	176	170
Shanrao, Jiangxi	–	80	65	738
Guixi, Jiangxi	–	98	95	423
Benbu, Anhui	75	96	93	225
Hefei, Anhui	133	97	156	120
Jinling, Jiangsu	–	109	104	317
Xuzhou, Jiangsu	–	60	65	299
Average value	154	103	120	315

The data origin from Shijianzhun, etc., Institute of Nuclear Agricultural Sciences; Zhejiang University.

special metallurgical slag, calcium carbide residue, lithium residue, caustic sludge, phosphorus gypsum, etc. at present, these solid waste mainly were used to prepare to building materials. Fly ash coming from coal combustion is the one of most common waste residue, which is bound to have enrichment radioactive elements. Some building materials with fly ash, which was used in building such as bricks, can release amount of radioactive substances such as radon that is harmful to human body. γ-Ray spectrum analysis of fly ash sample coming from Zhejiang, Fujian, Shandong, Jiangxi, Anhui and Jiangsu Province showed that the main radioactive component is ^{238}U, ^{232}Th, ^{236}Ra, and ^{40}K (Table 1).

Radioactive pollutants in the fly ash mainly originate from coal which contain a lots of crude radioelement including ^{226}Ra, ^{232}Tu, ^{40}K, ^{238}U, etc. After a series of disintegration, there are some radioactive pollutants such as γ-ray, X-ray and radioactive gas-radon can be released, as a result the radioactive pollution of building materials containing fly ash may come into being a great harm to human body [3]. The symptoms after contacted with fly ash include muzzy, thorax depression and hair-losing, etc. Because of long half live of radioelement' disintegration (even the shortest ^{226}Ra has 1620 years), radioactivity long-term effects just like domino will last for several generations and even more on the certain parts of body and gene [4]. Therefore, some actions need to be taken to reduce radioactivity in the building materials contained fly ash.

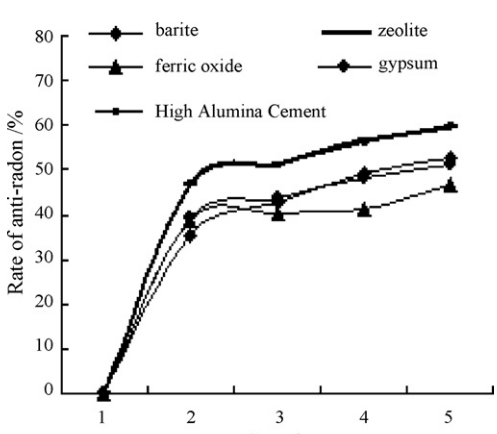


Fig. 1. Effect of content of additives on release of radon.

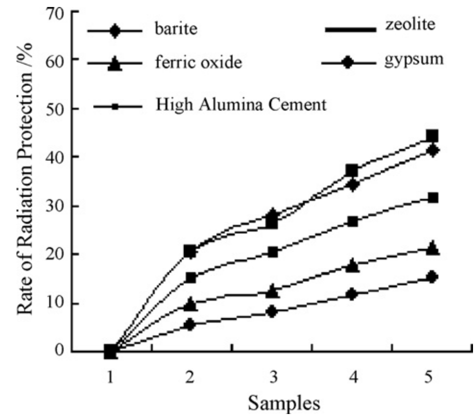


Fig. 2. Effect of content of additives on radial.

1.3. Previous studies

There have been traces of radon-mitigation research in China and other countries. In Egypt, it is proven that using natural fiber material can mitigate radon radiation; for example, applying furnace dust, cement onto the inner wall surface of a building can effectively reduce radon radiation [5]. Davis et al. used molecular

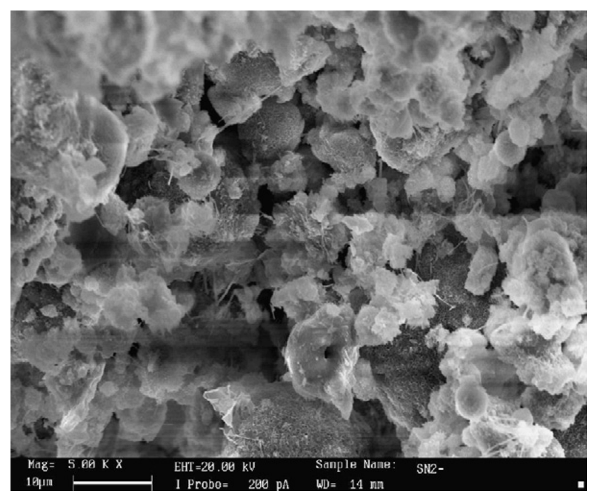


Fig. 3. SEM photographs of common building material.

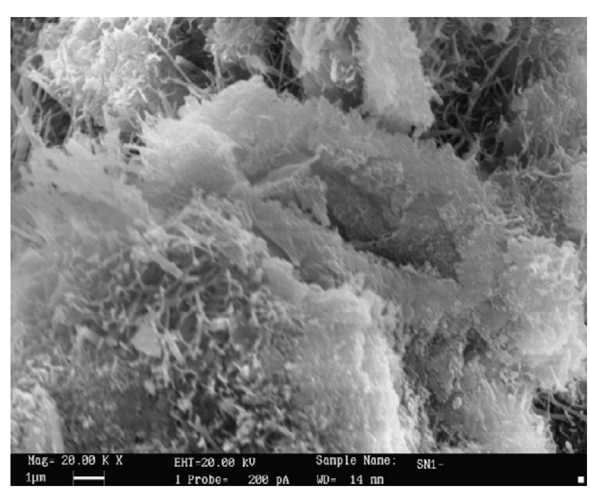


Fig. 4. SEM photographs of hydrate production in composites.

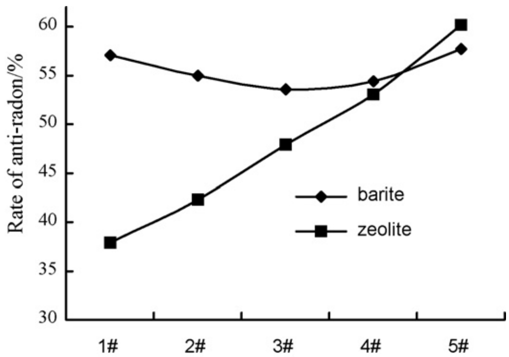


Fig. 5. Effect of additives granularity on release of radon.

sieve containing zeolite to filter out radon [6]. Wilhelm proposed the use of anti-radon board made of polyolefin with more than 20% carbon powder to absorb radon [7]. Hesco used a water solution composed of a solvent ethylene X-chlorine-acrylic ester copolymer disperse system to reduce indoor radon pollution [8]. Winder developed an anti-radon system composed of damp proof architectural compound. Matson used polymer membrane, silicate and porous membranes to mitigate radon radiation [9]. Takriti et al. suggested applying a layer of film on the cement wall to reduce radon backscatter radiation [10]. Yu et al. applied silica fume into Portland cement to stop radon backscatter radiation [11]. Kant et al. added metakaolin into cement as anti-radon additive, which could reduce 30% of radiation compared to normal cement [12]. Gao invented an anti-radon mortar made from cement, sand and water, etc., which was composed of top and bottom layers. The top layer is made of ARM-3, cement, common clean sand, and water. The bottom layer is made of ARM-4 and water [13–15]. These two layers form an interpenetrating polymer networks (IPN) microstructure in the mortar. This IPN microstructure can prevent radon passing through, and reduce the indoor radon concentration by 85–90%. However, not all the above-mentioned studies can achieve high radon-mitigation efficiency as compared to below-mentioned newly developed anti-radon coating system.

Therefore, some additives, which can absorb and screen radioactive pollutants contained in fly ash, be introduce to prepare a kind of building materials, it may be an effective method for large-scale application of solid waste fly ash in the field of building materials.

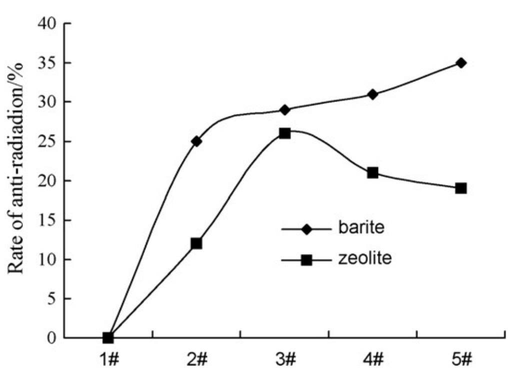


Fig. 6. Effect of additives granularity on radial.

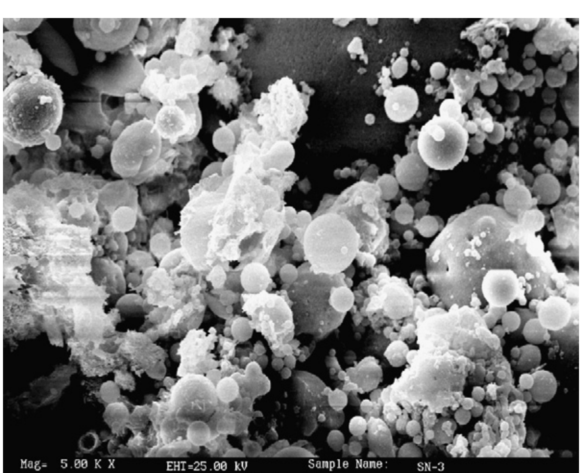


Fig. 7. SEM photographs of common building material containing fly ash.

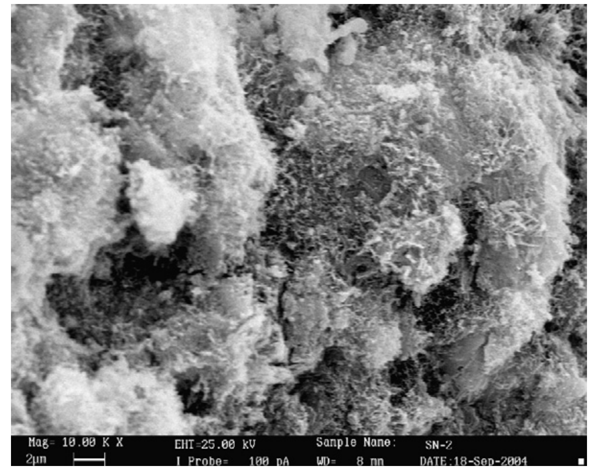


Fig. 8. SEM photographs of ultra-fine zeolite containing fly ash.

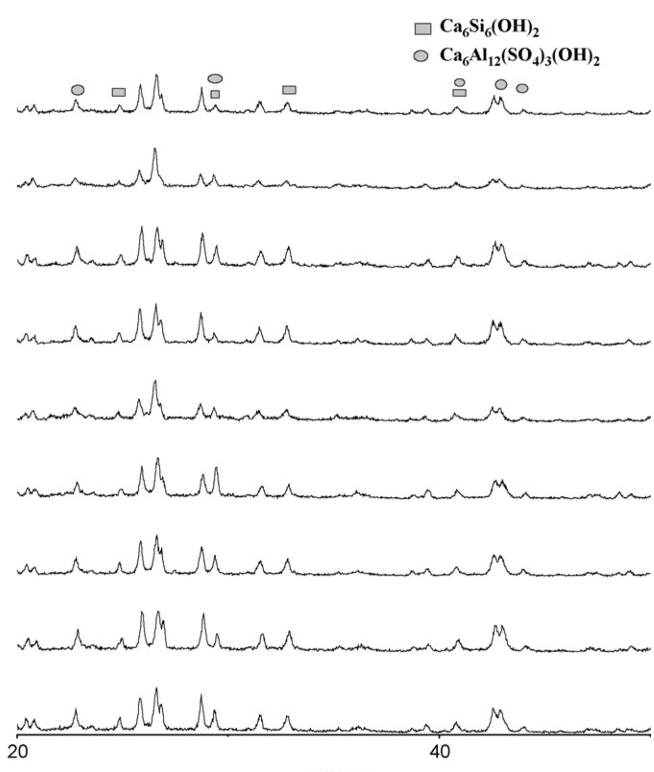


Fig. 9. XRD phase analysis of composites containing fly ash.

Table 2
Radon-preventing performance of composites.

Samples	Background value (Bq/m^3)	Release amount (Bq/m^3)	Shielded volume (Bq/m^3)	Anti-radon rate (%)
1#	192	105	87	45.3
2#	192	112	80	41.7
3#	192	123	69	35.9
4#	192	91	101	52.6
5#	192	56	136	70.8
6#	192	76	116	60.4
7#	192	79	113	58.9
8#	192	86	106	55.2
9#	192	66	126	65.6

Table 3
Irradiation index of γ-ray testing of composites.

Samples	(Bq/Kg)				Internal exposure index	External exposure index	
	Ra		Th	K			
0#	68.74	17.50	11.57	150.55	0.34	0.26	0.29
1#	46.37	20.88	11.57	25.74	0.23	0.21	0.21
2#	49.31	9.55	110.32	152.82	0.25	0.20	0.21
3#	57.78	22.15	56.24	298.53	0.29	0.25	0.31
4#	20.76	16.53	74.47	21.31	0.10	0.14	0.12
5#	68.94	0.33	36.69	156.10	0.34	0.20	0.22
6#	58.45	9.01	38.56	11.57	0.29	0.20	0.20
7#	80.55	17.29	76.62	261.21	0.40	0.30	0.35
8#	58.33	9.63	69.82	279.55	0.29	0.21	0.26
9#	83.08	3.77	77.22	292.84	0.42	0.26	0.31

2. Experiment

2.1. Materials

Barite, fly ash, ferric oxide, gypsum and cement were obtained from Szechwan Province, China. Zeolite was obtained from Henan Province, China. Criteria sand was obtained from Fujian Province, China.

2.2. Prepared methods

Cement-based composites were prepared with two kinds of specification, one is a cube with 300 mm × 300 mm × 3 mm, and another is cylinder with diameter 44 mm and height 50 mm. Criteria sand were replaced, respectively, by barite (A), zeolites (B), ferric oxide (C), high alumina cement (D), and gypsum (E), proportion of additives is as follows: A1–A5: 0%, 10%, 15%, 20%, 25%; B1–B5: 0%, 5%, 8%, 10%, 12%; C1–C4: 5%, 8%, 10%, 12%; D1–D4: 3%, 5%, 7%, 9%; E1–E2: 10%, 15%, 20%, 25%. The amount of cement was 20% in these composites, the content of fly ash in these composites were 55%.

2.3. Property test and characterization

Radon measurement equipment called the DURRIDGE RAD-7 Professional Radon Detector was used to measure the radon concentration. The measurement method is called surface separate out method, a certain surface of a sample was sealed by a plastic box so that radon can be accumulated continually in obturator, once radon was enriched enough to test its concentration, using RAD-7 Emanometer we can detect the release of radon in composites. The samples were sealed in plumbic barrel to test irradiation index of γ-ray, once the disintegration of radioelement was on balance, then using low-background γ and NaI crystal spectroscopy to detect the activity of ^{226}Ra, ^{232}Tu, ^{40}K and irradiation index of γ-ray under the same condition.

The phases were determined by X-ray diffraction analysis (X' Pert PRO, Panalytical, Holland) with filtered Cu Kα radiation operated at 40 kV and 40 mA. The XRD pattern were recorded from 5° to 80° of 2θ with a scanning speed of 0.01° of 2θ per second; microstructure was measured with SEM (LEOS400, Britain).

3. Results and discussion

3.1. Effects of content of additives on release of radioactive pollutants

Effects of content of additives on release of radioactivity pollutants are given in Figs. 1 and 2. The experiment results indicate that the barite, zeolite, ferric oxide, gypsum, and high alumina cement all can reduce effectively the release of radon contained fly ash (Fig. 1). Effects of zeolite content on radon are obviously, when proportion of zeolite reaches to 12%, the release of radon is less than 100 Bq. Furthermore, these additives can cut down irradiation index of γ-ray. High alumina cement and gypsum had also an obvious impact on; the release of radon can be reduced 50%. However, the amount of high alumina cement and gypsum should be reduce, because it may led composites to curdle rapidly, therefore the amount of high alumina cement and gypsum should be control to 5–7%. As can be seen from Figs. 3 and 4,

Table 4
Orthogonal analysis on Radon-preventing of composites.

Samples	Factor				Anti-radon rate (%)
	Barite	Zeolite	High alumina	Gypsum	
K_1	122.9	156.8	160.9	181.7	E = 486.4
K_2	183.8	167.7	159.9	161.0	
K_3	179.7	161.9	165.6	143.7	
K_1	41.0	52.3	53.6	60.6	
K_2	61.3	55.9	53.3	53.7	
K_3	59.9	54.0	55.0	47.9	
Z	20.3	3.6	1.4	12.7	

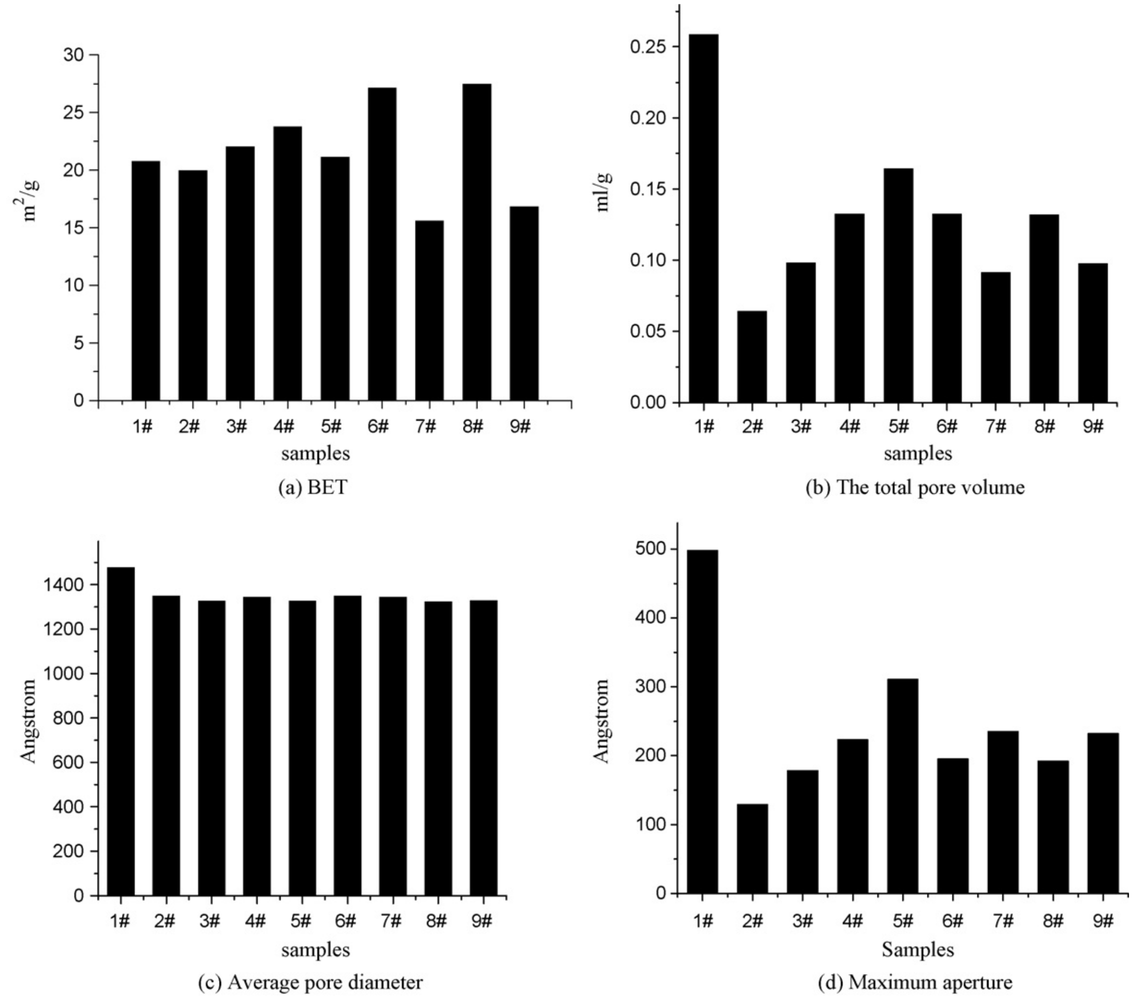


Fig. 10. Surface area and pore of composites containing fly ash. (a) BET. (b) The total pore volume. (c) Average pore diameter. (d) Maximum aperture.

in cement-based composites, hydration products (calcium silicate hydrates CSH gel) was accounted for more than 50%, is significantly greater than normal fly ash slurry generated by the hydration products.

3.2. Effects of granularity of additives on release of radioactive pollutants

Effects of ultra-fine barite on radon escape are weakly, but ultra-fine zeolite powders can effectively inhibit the release of radon (Fig. 5). Irradiation index of γ-ray from cement-based composites containing fly ash can be block by super-pulverization of barite (Fig. 6).

Compared with common building material, ultra-fine zeolites can come into being more hydration products in cement-based composites. These Layer of hydration products parceled fly ash in the composites, which may be CH-CSH double membrane, that is, the outside is a layer of velvet-like CSH gel, inside is a continuous film layer. At same time some fibrous CSH gel directly grow on the surface of fly ash, which well formed a tight sealed layer (Figs. 7 and 8), thus preventing the fly ash from releasing radioactive pollutants to the environment.

3.3. Orthogonal experiment results

In this section, using orthogonal experiment of four factors and three levels to obtain the best ratio. Four factors, respectively, were the percentage of the barite (Factor A), zeolite content (Factor B), high alumina cement (Factor C), and gypsum (Factor D). Content of each factor is follows: A1 = 10%, A2 = 15%, A3 = 20%; B1 = 4%, B2 = 8%, B3 = 12%; C1 = 1%, C2 = 3%, C3 = 5%; D1 = 1%, D2 = 3%, D3 = 5%.

Radon-preventing performance of composites was given in Table 2. The background value (0#) is 192 Bq/m^3, the release of radon from 1# to 9# sample, respectively, is 87 Bq/m^3, 80 Bq/m^3, 69 Bq/m^3, 101 Bq/m^3, 136 Bq/m^3, 116 Bq/m^3, 113 Bq/m^3, 106 Bq/m^3 and 126 Bq/m^3. Irradiation index of γ-ray of composites was given in Table 3, internal exposure index of composites (0# to 9#), respectively, is 0.34, 0.23, 0.25, 0.29, 0.10, 0.34, 0.29, 0.40, 0.29, and 0.42. External exposure index, respectively, is 0.26, 0.29, 0.21 and 0.21, 0.20, 0.21, 0.25, 0.31, 0.14, 0.12, 0.20, 0.22, 0.30,

Table 5
Testing of Radon-preventing performance of composites.

Samples	Testing value (Bq/m^3)	The release amount of radon (Bq/m^3)	Anti-radon ratio (%)
Common building containing fly ash	1130	1120	0
Composites	398.3	383.3	65.8

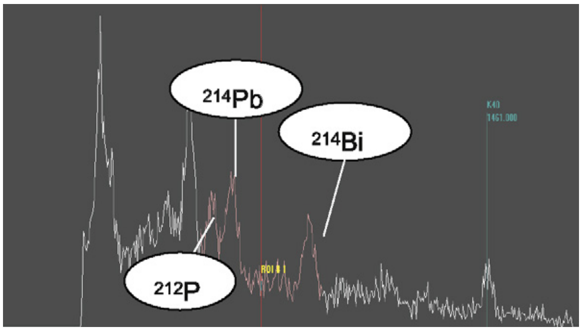


Fig. 11. γ pattern of cement-based composite containing fly ash.

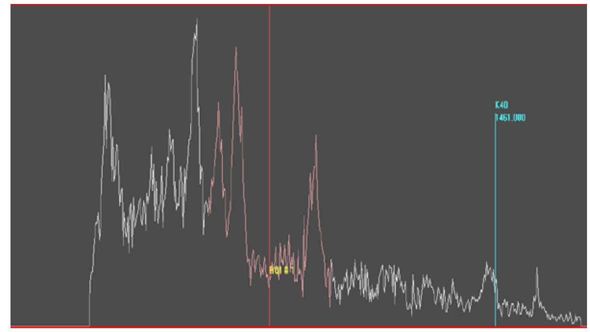


Fig. 12. γ pattern of common building material containing fly ash.

0.35, 0.21, 0.26, 0.21, 0.26, 0.26, and 0.31. Orthogonal analysis on Radon-preventing performance of composites was given in Table 4.

The results show that the importance of each effect factors on the release of radon is barite > gypsum > zeolite > high alumina cement. The best percentage of barite, zeolite, gypsum and high alumina cement in the composites should be 15%, 8%, 5% and 1%.

Phase analyses were determined with X-ray diffraction (Fig. 9). From the pattern, we can see that hydration products are similar with in the different composites; an only differences is in the quantity of hydration products. The main chemical component in fly ash is SiO_2, Al_2O_3, Fe_2O_3, and CaO; it is the same as the main component of cement. Therefore, it has little effect on the type of cement hydration products, only α-quartz and mullite diffraction peaks appeared in the XRD patterns; when additives were joined in the composite, the content of CSH will be increased gradually.

Specific surface area and porosity test used by the high-speed automatic specific surface area and porosity analyzer (U.S. NOVA3000), the test results are shown in Fig. 10. We can see a maximum value of the surface area is 27.5025 m^2/g (8#), a minimum value is 15.5680 m^2/g (9#). The largest value of the total pore volume is 0.25866 mL/g (1#), a minimum value is 0.09775 ml/g (9#). The largest value of maximum aperture is 1477.841 Å (1#), A minimum value is 1322.592 Å (8#). The largest value of average pore size is 498.315 Å (2#), a minimum value is 129.187 Å (2#). The experimental results show that ultra-fine can greatly increased density of the pore, so rays during from the radioactive source to the external space would be refracted more times. Similarly, a dense microstructure can confined radon gas in-house, so that radon gas does not diffuse into space through the tiny holes.

Based on the orthogonal experiment results, cement-based composites were prepared using barite, zeolite, ferric oxide, gypsum, and high alumina cement. Radon-preventing performance of composite was given in Table 5. From Figs. 11 and 12, we can see that the emissive rate of γ-ray is about 45%, and it is equal to the data measured by CIT-2000F building materials radioactivity meter. Irradiation index of γ-ray of composite was given in Table 6.

3.4. The mechanism of self-absorption of radioactive pollutants

(1) Zeolite is a natural pozzolana mineral, containing a certain amount of active silica and active alumina, which can come into being absorption effect in cement-based composites as a kind of excellent sorbent for radon, the main reason is that there are some even cavity and powerful static in zeolite (Fig. 13); and distributing positive ions which maybe form powerful electromagnetic field in crystal lattice cavity. Once fissile production of radwaste was adsorbed by cavity of zeolite, it also could be exchanged into crude zeolite crystal [14]. On the other hand, the ultra-fine zeolite has large specific surface area, which can absorb a large number of water molecules and gases, so Rn may be react with H_2O to prod-

Table 6
Testing of irradiation index of γ-ray of composites.

Samples	^{226}Rnradiation equivalent activity (Bq/kg)	Interior irradiation index IRa	Exterior irradiation index IRa		Rate of prevention (%)
Common building containing fly ash	105.01	0.53	0.32	0.29	–
Composites	55.74	0.28	0.23	0.18	46.9%

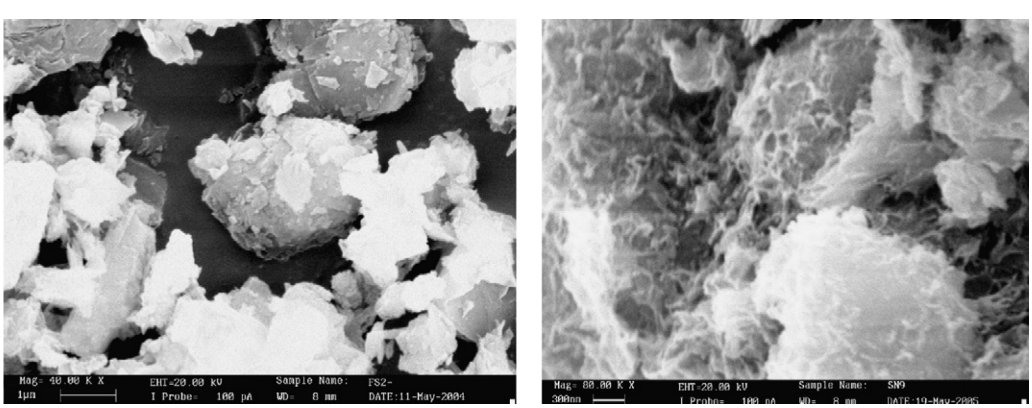
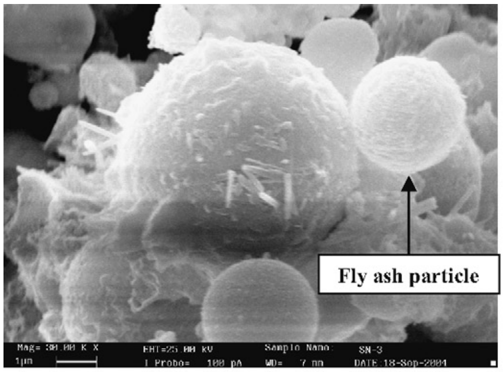
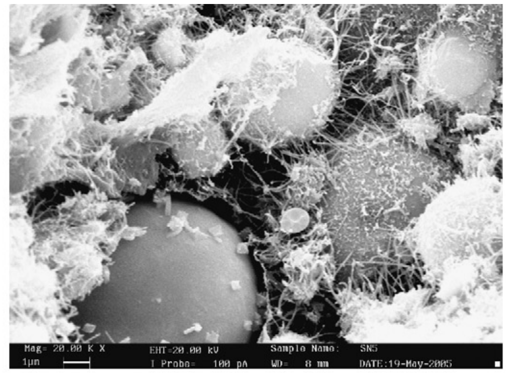


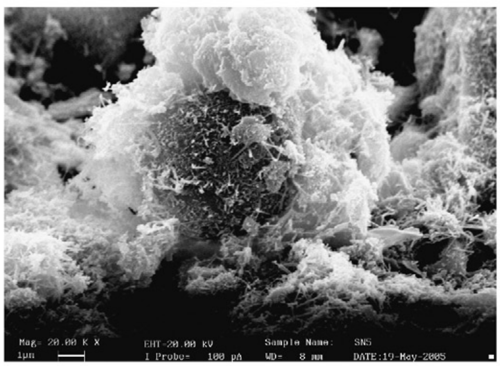
Fig. 13. SEM photographs of zeolite in the composites.



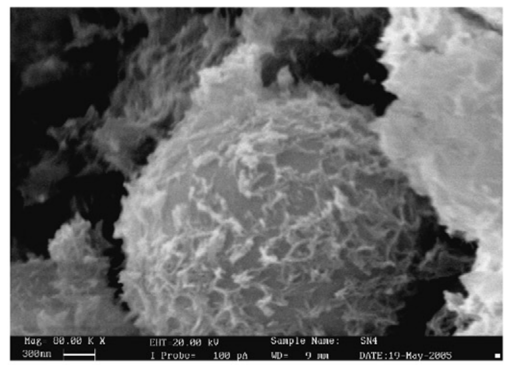
(a) SEM photograph of common building material containing fly ash



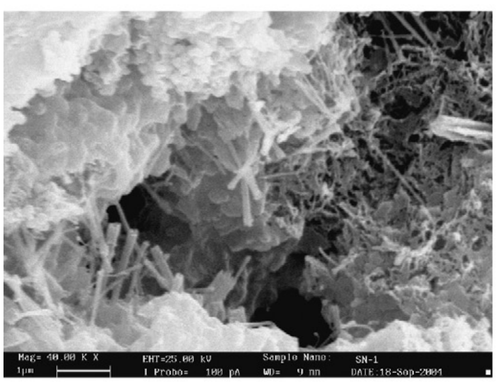
(b) SEM photograph of common building material containing fly ash



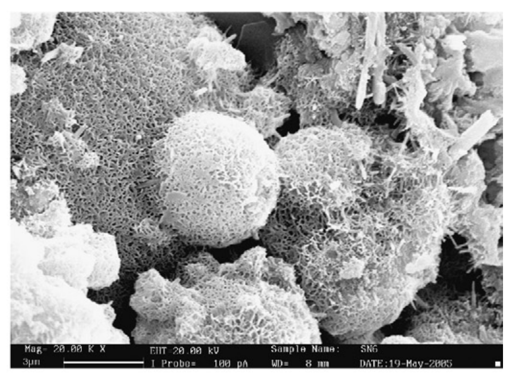
(c) Products of hydrate on the surface of fly ash in the composites



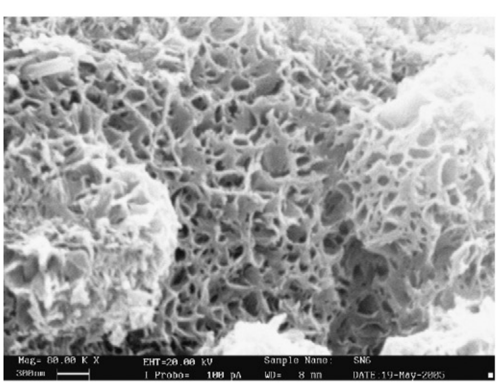
(d) Fly ash was enwrapped by Products of hydrate in the composites



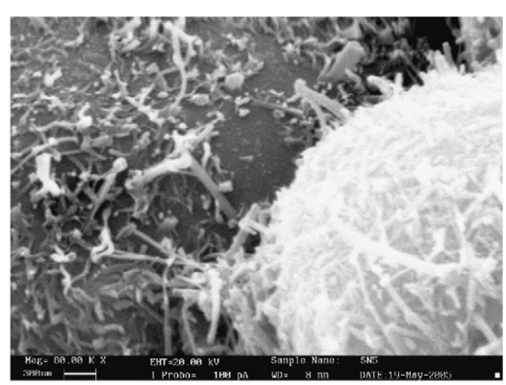
(e) Production of stick shape in composites



(f) Alveolate products of hydrate in the composites



(g) Alveolate products of hydrate in the composites



(h) Acerate products of hydrate in the composites

Fig. 14. SEM photographs of composites. (a) SEM photographs of common building material containing fly ash. (b) SEM photographs of common building material containing fly ash. (c) Products of hydrate on the surface of fly ash in the composites. (d) Fly ash was enwrapped by products of hydrate in the composites. (e) Production of stick shape in composites. (f) Alveolate products of hydrate in the composites.

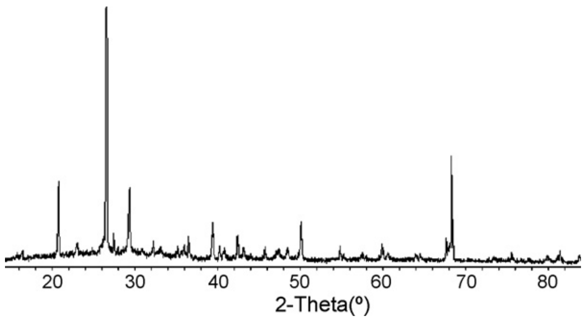


Fig. 15. XRD pattern of common building material.

uct some crystal compounds, then going into the crystal lattice of zeolite [15–17].

(2) C–S–H gelatin is help to enhance the intensity of composites, however a few calcium hydroxide which may form an unsubstantial layer in adjacent to interface of material might depress the intensity of composites. In the alkaline condition, aluminum and silicon released from additives can react with $Ca(OH)_2$, it is help to produce more hydrated calcium silicate. At some time, a large number of C–S–H gelatin can be produced in the composites by ultra-fine additives, it may consume more calcium hydroxide in composites, which is facilitate to reduce the release of radioactivity pollutants.

The products of hydrate $Ca(OH)_2$ reaction with aluminosilicate to produce $CaSiO_3$ in common cement building material, but the degree of hydrate be under 10%, then a weakness layer was formed round fly ash (Fig. 14a and b). By joining, activate aluminosilicate, $Ca(OH)_2$ maybe fully react with aluminosilicate contained in additives so that it can produce more C–S–H gelatin, and the degree of covering with fly ash is more tightness (Fig. 14c and d). From XRD pattern of composites (Figs. 15 and 16), we can see that a great deal of hydrate products such as fiber and stick shape comes forth in composites, their longness is about 1 μm (Fig. 14e and h). In addition, the hydration products deposit directly on the surface of fly ash to form a hydrate layer in the hydrated course, the hydration layer may be CH-CSH double membrane (Figs. 14f and 16g), which can restrain the release of the radioactive pollutants.

(3) γ-Ray from radioelement including Ra, Th, K, and radon can interact with barium element and cause the photoelectron effect, Compton effects and electron effects [18], which can absorbed the energy or alter the movement orientation of γ-ray. Therefore, composites containing barite can effectively screen the ray coming from radioelement. At some time, the smaller the granularity of additives is, the more tightly the size of aperture is. Figs. 17 and 18 show that the ultra-fine additives is tighter than common additives, so compact microstructure can make the ray produce repetitious refraction in the course of entering into exterior space, the mostly energy of ray can be absorbed during this course.

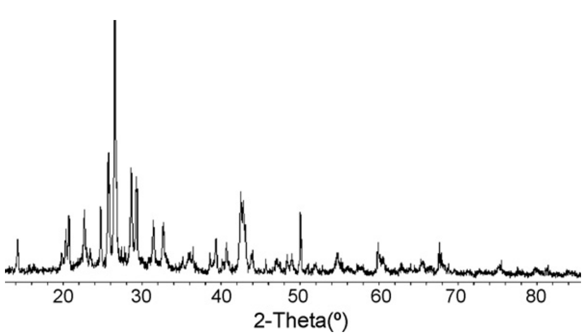


Fig. 16. XRD pattern of self-absorption of radioactivity cement-based functional composites.

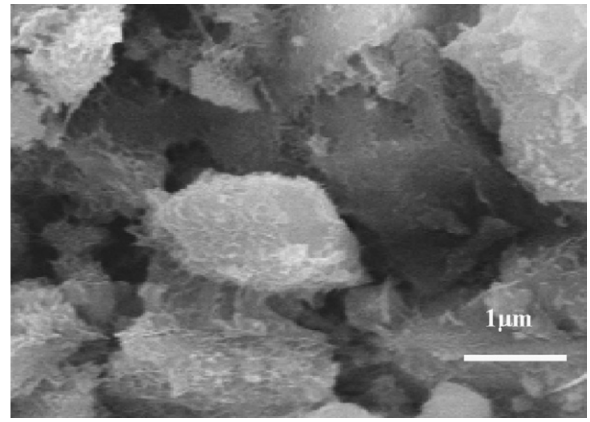


Fig. 17. SEM photographs of common functional additives.

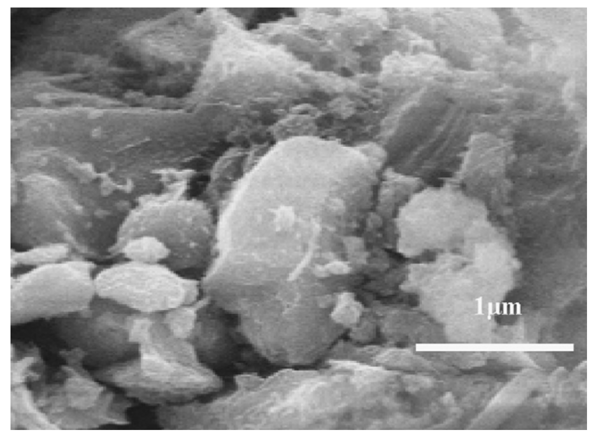


Fig. 18. SEM photographs of super-pulverization functional additives.

4. Conclusions

By adding a proper amount of additives such as braite, zeolite, ferric oxide, gypsum, and high alumina to form self-absorption of radioactivity in the composites, the radioactive pollutants contained in fly ash can be absorbed. Compared with traditional building materials containing fly ash, the release ratio of radioactive pollutants can be restrained successfully in these composites. This cement-based composites will help to solid waste fly ash are large-scale applied in the field of building materials.

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