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Environmental Radioactivity Comparison Study for the Glaze-Clay Surface of Ceramic Tiles by Tracks Technique

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TRACKS DENSITY, radon concentration, radon exhalation rates and radium concentration were measured from ceramic tiles for both of glaze and clay by using the track technique, containing CR-39, to estimate the radiation exposure in the vicinity of ceramic tile. For ceramic tiles of wall, the average of tracks density, the radon concentration, radon exhalation rates and radium concentration were found in the range 230-356 tracks.cm^{-2}, 389-600 Bq.m^{-3}, 21-31 mBq.m^{-2}.h^{-1}, 16-25 Bq.kg^{-1}, respectively. While for ceramic tiles of floor, the average of tracks density, the radon concentration, radon exhalation rates and radium concentration were found in the range 274-509 tracks.cm^{-2}, 463-860 Bq.m^{-3}, 25-46 mBq.m^{-2}.h^{-1}, 19-46 Bq.kg^{-1}, respectively. The average level of radon concentrations caused by these ceramic tiles for Egyptian companies covering both of wall, floor, glaze and clay giving an annual exposure dose 22±2 mSv.y^{-1} which is higher than internationally recommended range.

Keywords: *Track density, CR-39, ceramic tiles, annual effective dose, radon exhalation rate.*

Materials obtained from the earth's crust, such as ceramic tiles, may contain traces of ^{238}U and ^{226}Ra. These radionuclides decay to radon (^{222}Rn), which is a radioactive gas with a half-life of 3.82 days. For over a century and a half, soils containing naturally occurring radioactive materials (NORM) have been used in manufacturing glass and ceramic products, as base material (Bruzzi *et al.*, 1993). Recognition of the hazards inherent in such applications led to regulations governing incorporation of byproduct radioactive materials (EC 1999). Raw zirconium sand is one of the substances, NORM which is widely used in the ceramic industry (Verita *et al.,* 2009 and Luisa *et al.,* 2008).

Prolonged exposure to radon may increase the risk of lung cancer (ICRP, 1979, ICRP, 2007, Maged *et al.,* 2000, Popovic *et al.,* 1996, Todorović *et al.,* 1999 and Saad, 2008) because it delivers 55% of the total dose to the cells of the respiratory system UNSCEAR (2006). In some, but not all, studies of groups of people either occupationally exposed to, or resident in areas of, high natural radiation, including elevated levels of radon and its decay products, an increased incidence of chromosomal aberrations has been observed. Radon and its decay products did not induce chromosomal aberrations *in vivo* in rabbits in one laboratory experiment but did induce chromosomal aberrations in human cells *in vitro* and sex-linked recessive lethal mutations in *Drosophila* (WHO, 1988).

There were numerous studies confirming indoor radon as a cause of over 20,000 lethal lung cancer cases in EU every year, making about 9% of all lethal lung cancer cases per year in EU and about 2% of lethal cancer cases per year in EU in general. The majority of European countries, as well as Canada and USA, proclaimed intervention levels for exposures from radon in closed space and concentrations of radionuclides in building materials. These issues were also regulated by numerous publications and recommendations by international organizations: International Commission for Radiation Protection (ICRP), Radon Indoor Concentrations and Activity of Radionuclides in Building Materials in Serbia, (WHO, 2009), United Nation Commission for Atomic Radiation Research, (UNCEAR, 1989), etc. According to these recommendations the general population should not be exposed to more than 0.7 mSv of radiation from building materials, therefore the total gamma radiation index for radionuclides ^{40}K, ^{226}Ra and ^{232}Th should not be higher than 1.

Due to the long half-life of radon gas, it can reach from the earth's crust or from the walls and floors of buildings into both outdoor and indoor air. In the case of indoor air, the risk of exposure to radon is higher, especially for buildings with poor ventilation systems, which may lead to a higher indoor concentration of radon. In the last 20 years, more attention has been paid to the measurement of radon exhalation from building materials in many countries worldwide, including Great Britain, Finland, (Mustonen, 1984), Taiwan, (Ching-Jiang *et al.,* 1993), Greece, (Savidou *et al.,* 1996), Turkey, (Turhan, 2008) and Egypt, (Maged *et al.,* 2005). In light of this information, and due to the lack of radon exhalation studies from construction materials in Egypt, it is important to begin this preliminary study to measure the track density, the radon concentration, the radon exhalation rates, radium concentration and annual effective doses in situations where construction materials are a significant source of radon in houses. This particular study will be carried out in the National Centre for Radiation Research and Technology to help in development database for all building materials.

Materials and Methods

CR-39 detector

The CR-39 plastic solid-state nuclear track detectors, SSNTDs are a $C_{12}H_{18}O_7$ polymer with density 1.3 g.cm^{-3}. In this work we used CR-39 produced by Page Mouldings (England). A study of tracks density from wall-floor ceramic tiles for both of glaze and clay surface was performed. Measurement of the time-integrated radon exhalation rate was carried out by placing an inverted hollow holder on the ceramic tiles surface for wall and floor. The contact between the hollow holder and those sampled ceramic tiles was covered with filter paper to prevent any alpha particles enter the hollow holder and then sealed with silicone sealant as shown in Fig. 1.

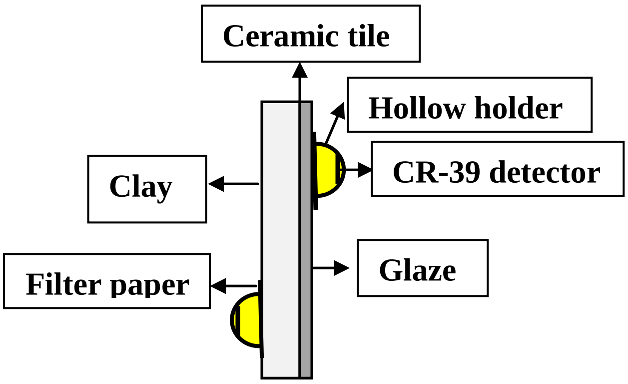

Fig. 1. The setup diagram for measuring radon exhalation rate of ceramic tiles.

The detection system involves the use of SSNTDs of type CR-39, which were cut into small pieces, 1.5 cm × 1.5 cm. Each CR-39 detector was fixed onto the top center of the inverted hollow holder by means of adhesive tag. Once equipped with CR-39 detectors, the hollow holder were properly labeled and mounted for about two weeks in equilibrium. Three detectors were used for each sample. During this exposure time, the tracks of alpha particles from the decay of radon and its daughters that had entered the air volume of the hollow holder were registered in the CR-39 plastic detector. The CR-39 plastic detector were removed from the hollow holder and etched chemically in a 6 M NaOH solution at 70 °C for 18 h to display and enlarge the latent alpha tracks due to radon decay. The etched tracks on the detectors were counted manually, using an optical microscope with objective lens 4× magnification. The average area of one hundred fields of view was calculated by using image analyses system and the track density was calculated in terms of tracks per cm^{-2}. The background track density was determined by processing a virgin detector under the same etching conditions.

The background was subtracted from the measured track density. In order to obtain realistic statistics of the tracks, 100 fields of view were scanned continually to cover the detector surface. The calibration factor obtained from an earlier calibration experiment for the CR-39 track detector (Maged *et al.*, 1993) was used to compute the radon activity from the track density.

Results and Discussion

The results showed that the ceramic tiles of floor yielded the highest average tracks density values, while the ceramic tiles of wall produced the lowest values. However, the average values of radon concentrations from the floor tiles are higher than the wall tiles and this may be due to the thickness of floor tiles is higher. For the glaze of wall ceramic tiles, the tracks density, the radon concentration, the exhalation rate and radium concentration according to Culot *et al.* (976) were found 230±36 $tracks.cm^{-3}$, 289±61$Bq.m^{-3}$, 21±3 $mBq.m^{-2}.h^{-1}$, and 16±3 $Bq.kg^{-1}$, respectively. While for clay of wall ceramic tiles, the tracks density, the radon concentration, the exhalation rate and radium concentration were found 356±31 $tracks.cm^{-3}$, 600±53 $Bq.m^{-3}$, 32±3 $mBq.m^{-2}.h^{-1}$, and 25±2 $Bq.kg^{-1}$, respectively as shown in Table 1.

The glaze of floor ceramic tiles, the tracks density, the radon concentration, the exhalation rate and radium concentration were found 274±35 $tracks.cm^{-3}$, 463±58 $Bq.m^{-3}$, 25±3 $mBq.m^{-2}.h^{-1}$, and 19±2 $Bq.kg^{-1}$, respectively. While for the clay of floor ceramic tiles, the tracks density, the radon concentration, the exhalation rate and radium concentration were found 509±41 tracks.cm^{-3}, 860±69 Bq.m^{-3}, 46±4 mBq.m^{-2}.h^{-1}, and 36±3 Bq.kg^{-1}, respectively as shown in Table 2.

TABLE 1. Represents the ceramic tiles wall parameters as the track density (ρ), the radon concentration (C), the exhalation rate (E) and the radium concentration (Ra) for glaze and clay surface.

Ceramic tile wall								
Glaze					Clay			
Code No.	**ρ tracks. cm^{-2}**	**C Bq. m^{-3}**	**E mBq. m^{-2}.h^{-1}**	**Ra Bq.kg^{-1}**	**ρ tracks. cm^{-2}**	**C Bq.m^{-3}**	**E mBq. m^{-2}.h^{-1}**	**Ra Bq.kg^{-1}**
1	220	372	20	15	318	537	29	23
2	290	490	26	22	380	642	34	26
3	260	439	24	19	390	659	35	27
4	240	406	22	17	350	591	32	25
5	175	296	16	12	370	625	33	26
6	280	473	25	19	360	608	32	25
7	220	372	20	16	365	617	34	26
8	250	422	23	18	370	625	33	25
9	230	389	21	16	370	623	32	25
10	255	431	23	18	400	676	36	28
11	190	321	17	13	320	541	29	22
12	190	321	17	14	290	490	26	20
13	195	329	18	14	340	574	31	24
Mean	230	389	21	16	356	600	32	25
SD	36	61	3	3	31	53	3	2
Max	290	490	26	22	400	676	36	28
Min	175	296	16	12	350	591	32	25

The average value for all ceramic tiles for the different companies were found for the tracks density, the radon concentration, the exhalation rate and equivalent dose according to *(UNSCEAR, 2000)* were found 390±46 tracks.cm^{-3}, 661±68 Bq.m^{-3}, 39±5 mBq.m^{-2}.h^{-1}, and , 22±2 mSv.y^{-1}, respectively as shown in Table 3.

The radium concentration in Egyptian ceramic is still less than others (EC, 1999, Verita *et al.*, 2009 and Luisa *et al.*, 2008). The alpha tracks density (ρ) due to alpha particles which emitted from radon comes out from ceramic tiles appeared in one field (from 100 fields) in CR-39 detector by using objective magnification 20X for Egyptian ceramic companies is shown in Fig. 2.

TABLE 2. Represents the ceramic tile floor parameters as the track density (ρ), the radon concentration (C), the exhalation rate (E) and the radium concentration (Ra) for glaze and clay surface.

Ceramic tile wall								
Glaze					Clay			
Code No.	ρ tracks.cm^{-2}	C Bq.m^{-3}	E mBq. m^{-2}.h^{-1}	Ra Bq.kg^{-1}	ρ tracks.cm^{-2}	C Bq.m^{-3}	EmBq. m^{-2}.h^{-1}	Ra Bq.kg^{-1}
١	260	٤٣٩	٢٥	١9	٤٧٠	٧٩٤	٤٣	٣3
٢	٣٢٠	٥٤١	٣٠	٢٣	٤٥٠	٧٦٠	٤١	٣2
٣	٢٦٥	٤٤٨	٢٤	١9	٥٢٠	٨٧٩	٤٧	٣7
٤	٢٥٠	٤٢٤	٢٤	١٨	٤٦٠	٧٧٧	٤٢	٣3
٥	٢٣٠	٣٨٩	٢١	١٦	٥٠٠	٨٤٥	٤٦	٣٦
٦	٣١٠	٥٢٤	٢٨	٢٢	٥١٠	٨٦٢	٤٦	٣6
٧	٢٥٠	٤٢٢	٢٣	١8	٥٤٥	٩٢١	٥٠	٣9
٨	٢٨٠	٤٧٣	٢٥	١٩	٥٤٠	٩١٢	٤٩	٣8
٩	٣٣٠	٥٥٨	٣١	٢٣	٥٢٠	٨٧٩	٤٧	٣٦
١٠	٢٤٥	٤١٤	٢٢	١٧	٥٨٠	٩٨٠	٥٣	٤١
Mean	٢٧٤	٤٦٣	٢٥	١٩	٥٠٩	٨٦٠	٤٦	٣6
SD	٣5	٥٨	٣	٢	٤١	٦٩	4	3
Max	٣٣٠	٥٥٨	٣١	٢٣	٥٨٠	٩٨٠	٥٣	٤١
Min	٢٣٠	٣٨٩	٢١	١٦	٤٥٠	٧٦٠	٤١	٣2

TABLE 3. The average value of all ceramic tiles for all the companies.

Code	Company trade mark	Track density (tracks.cm^{-2})	Radon conce ntration (Bq.m^{-3})	Exhalation rate (mBq.m^{-2}.h^{-1})	Equivalent dose (mSv.y^{-1})
1	Cleopatra	377	637	37	21
2	Aracemco	431	728	43	24
3	Lecico	430	727	43	24
4	Venecia	387	654	37	21
5	Pharaoho	381	644	43	21
6	El-Gawhara	430	728	42	24
7	Misr	416	703	41	23
8	Alfa	434	733	43	24
9	Alamir	444	751	44	25
10	Royal	394	673	38	22
11	ND	331	559	33	18
12	ND	288	486	28	16
13	ND	337	569	33	18
Mean		390	661	39	22
Max		444	751	44	25
Min		288	486	28	16
SD		46	68	5	2

[a] E.E.C= Equilibrium Equivalent concentration. ND= No data.

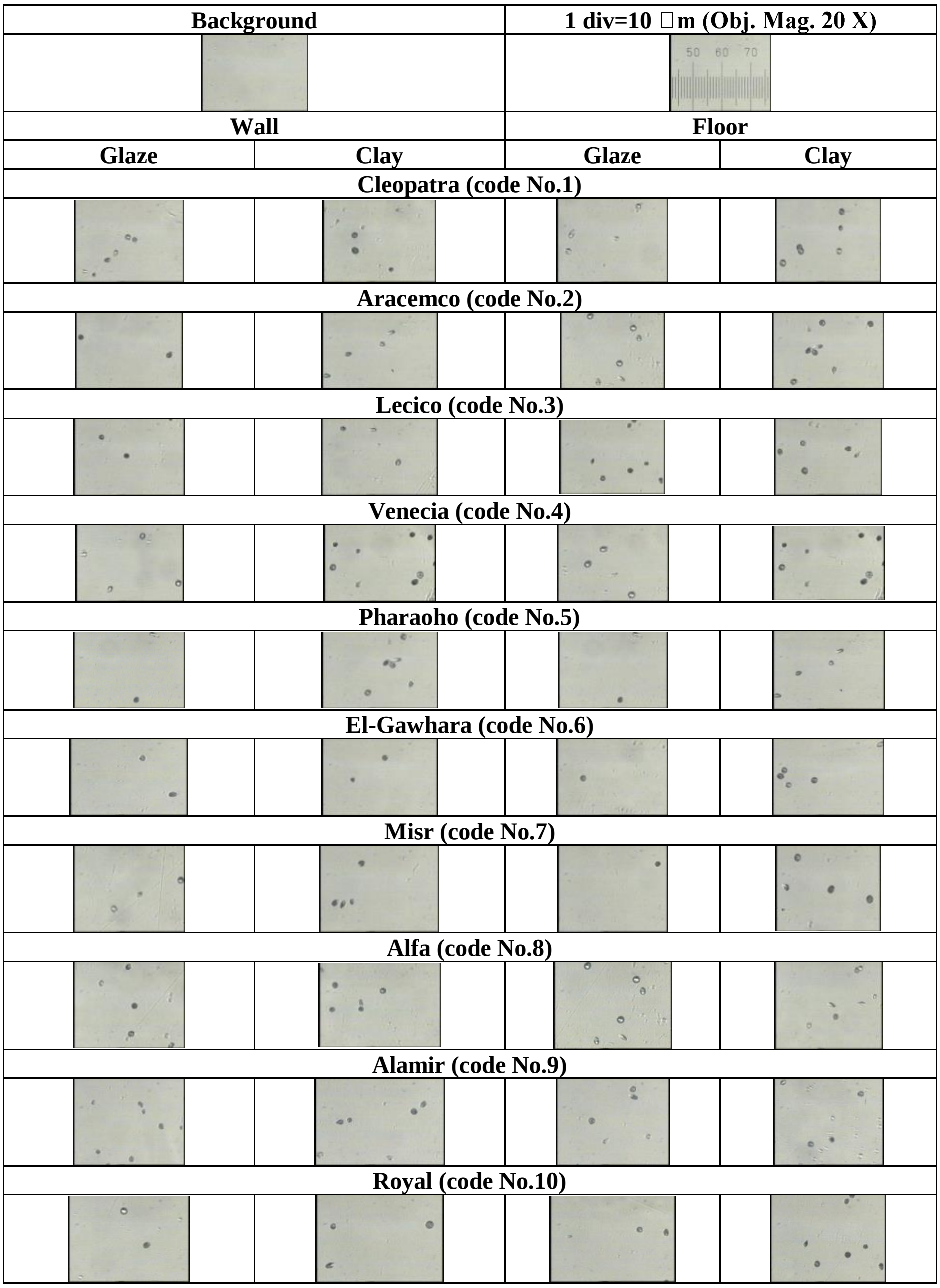

Fig. 2. Photographs showing the alpha tracks density (ρ) appeared in one field (from 100 fields) in CR-39 detector by using objective magnification 20X for Egyptian ceramic companies.

Conclusion

The range of ceramic tiles for the tracks density, the radon concentration, the exhalation rate and radium concentration were found 175-589 tracks.cm^{-3}, 296-980 Bq.m^{-3}, 16-53 mBq.m^{-2}.h^{-1}, and 12-41 Bq.kg^{-1}, respectively. All the measuring parameters for the floor ceramic tiles were higher than the wall ceramic tiles. The average level of radon concentrations caused by these ceramic tiles for Egyptian companies covering both of wall, floor, glaze and clay giving an annual exposure dose 22±2 mSv.y^{-1} which is higher than internationally recommended range.

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دراسه مقارنه للنشاط الأشعاعى البيئي لكل من السطح الخزفى و الطمي لبلاط السيراميك بواسطه تقنيه الاثر النووى

ماجد علي فهمي و ندا لطفي عبد الرحيم

قسم الجوامد و المعجلات الالكترونية ، المركز القومي لبحوث وتكنولوجيا الإشعاع ، ص. ب: ٢٩ مدينة نصر ، مصر.

لقد تم دراسه كل من الاثر النووى و تركيز الرادون و معدل زفير الرادون و تركيز عنصر الراديوم فى بلاط السيراميك لكل من الطبقه الخزفيه و الطبقه الطميه و ذلك باستخدام طريقه عد الاثر النووى. لقد وجد ان كثافه الاثر النووى و تركيز الرادون و معدل الزفير للرادون و تركيز عنصر الراديوم لبلاط السيراميك الحائطى يقع فى المدى ٢٣٠-٣٥٦ تراك.سم-٢ و ٣٨٩-٦٠٠ بيكريل.م-٣ و ٢١ -٣١ مللى بيكريل. م-٢.ساعه و ١٦ -٢٥ بيكريل.كجم-١ على التوالى. بينما بلاط السيراميك الارضى وجد ان كل من معدل كثافه الاثر النووى و تركيز الرادون و معدل الزفير للرادون و تركيز الراديوم يقع فى المدى ٢٧٤ -٥٠٩ تراك.سم-٢ و ٤٦٣ -٨٦٠ بيكريل.م-٣ و ٢٥ -٤٦ مللى بيكريل. م-٢.ساعه و ١٩ -٤٦ بيكريل.كجم-١ على التوالى. لقد وجد ان متوسط مستوى تركيز الرادون المنبعث من بلاط السيراميك للمصانع المصريه تشمل كل من الحائطى و الارضى سواء الطبقه الخزفيه و الطميه يعطى جرعه اشعاعيه سنويه قدرها ٢٢ مللي سيفرت/عام و هى تعتبر اعلى من المعدل العالمى.