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Determination of Natural Radioactivity Levels of Some Concretes and Mineral Admixtures in Turkey

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Nine concrete samples with or without mineral admixtures have been analyzed for their naturally occurring radionuclide of ^{226}Ra, ^{232}Th and ^{40}K using HPGe gamma spectrometry. Also, concrete raw materials such as aggregate, cement, fly ash and blast furnace slag have been measured. The radioactivity values of ^{226}Ra, ^{232}Th and ^{40}K in fly ash and those of ^{226}Ra in blast furnace slag are higher than the corresponding world mean specific activities values which are 50, 50 and 500 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The concretes activity concentrations have been found to change with supplementary cementitious materials of the concrete samples. This may be attributed to containing different proportions of different mineral additives such as fly ash and blast furnace slag which were found to contain high natural radionuclide concentrations according to the rest of raw material samples. In addition, radium equivalent activities (Ra_{eq}), γ-index (I_γ), α-index (I_α), absorbed dose rate in air (D) and annual effective dose associated with the natural radionuclide were calculated to assess the radiation hazard of the natural radioactivity in the concrete mixture samples. The concentration of the natural radionuclides and the radium equivalent activity obtained in the present study are compared with the previous results. From these results, it can be seen that these building materials do not pose significant radiation hazards.

Keywords: Natural radioactivity, Concrete, Fly ash, Blast furnace, Cement.

INTRODUCTION

In its most basic form, concrete is a mixture of portland cement, sand, coarse aggregates and water. The principle cementitious material in concrete is portland cement. Today, most concrete mixtures contain supplementary cementitious materials that make up a portion of the cementitious component in concrete. Supplementary cementitious materials such as fly ash and granule blast furnace slag enable the concrete industry to use hundreds of millions of tons of by product materials that would otherwise be used as land-filled as in the west[1]. In recent years fly ash and granule blast furnace have been used as a replacement of sand and cement in premixed concrete, manufacture of blended fly ash portland cement, aerated concrete, fly as clay bricks and blocks and for the filling of underground cavities *etc*.[2]. The building industry requires large quantities of low-cost materials and new products that may be substitutes for the widely used natural products as conventional building materials. However, such new building materials may contain significant quantities of naturally or technologically enhanced levels of radioactivity[3]. The presence of these radioisotopes in materials causes external exposure to people who live in the building[4].

Natural radioactivity in concrete materials comes mainly from uranium (^{238}U) and thorium (^{238}Th) series and the radioactive isotopes of potassium (^{40}K). In the uranium series, the decay chain segment starting from radium (^{226}Ra) is radiological the most important and, therefore, reference is often made to radium instead of uranium. The knowledge of natural radioactivity in these materials is then important for determining the amount of public exposure because people spend most of their time (about 80 %) indoors[5]. This knowledge is important in the assessment of possible radiological hazards to human health and essential for the development of standards and guidelines in using and these materials.

In the recent years, there has been an increasing interest in the study of radioactivity in various building and their raw materials[6-12].

The aim of this paper is to measure the natural activity concentrations of some concrete samples. The natural activity concentrations in two different types of concrete (MF and MB) and their initial raw materials (fly ash and granule blast furnace slag) were also measured in order to assess each material's contribution to total concrete activity. The results obtained are compared with the results available in some other countries of the world.

EXPERIMENTAL

The composition of the mortars all derive from the composition of standardized mortar (concrete) as specified in the TS EN 196-1 standard. All concrete is used in normalized sand CEN EN 196-1 and the ordinary Portland cement, CEM I 42.5 R. This cement was mixed with two mineral additives; fly ash and granule blast furnace slag in some mortars. Concrete samples were prepared by Hobart mixer according to TS EN 196-1. Mortar was placed on the jolting Table is blocked in a mould to be 4 cm × 4 cm × 16 cm. After 24 h of curing 20 °C by 90 % relative humidity, the samples were removed from the molds and placed in standard curing condition in statured lime water at 20 ± 1 °C. Three main different concrete types were prepared. For the basic type, without mineral additives, "ordinary" concrete mixture (MO, 0 %) consists of 450 g cement, 1350 aggregate and 225 g water (proportion 1:3:0.5 respectively). The other two types were produced by adding supplementary cementitious materials to the cement. In case of the fly ash and granule blast furnace slag concretes, respectively, (MF) and (MB) were incorporated in the cementitious materials at proportions of 5, 10, 20 and 30 % by weight. Finally, a total of nine concrete types were obtained. Again, all concrete mixtures have the same weight for total cementitious materials (cement and mineral additives) which equals to 450 g. The compositions of the concretes were listed in Table-1[2].

Radioactivity measurements: Each sample was homogenized and dried in a temperature controlled furnace at 105 °C for 24 h to remove moisture and sieved through a 2 mm mesh. About 120 g of samples were sealed in gas tight, radon impermeable, cylindrical polyethylene plastic containers (5.5 cm diameter and 5 cm height) for γ-activity analysis. Before measurements, the containers were kept sealed for 4 weeks in order to reach the equilibrium of ^{226}Ra and its short lived progeny.

Gamma spectrometry measurements were conducted with a coaxial HPGe detector of 55 % relative efficiency and a resolution of 1.9 keV at the 1332 keV gamma of ^{60}Co (Ortec, GEM55P4-95 model). The detector was shielded in a 10 cm thick lead well internally lined with 2 mm Cu foils. The spectrum analysis was performed using computer software Genie 2000 obtained from Ortec. A performance test using the certified reference samples (IAEA-375, IAEA, Vienna) of known activities was conducted for checking the efficiency calibration of the system. The specific activities of these samples were in accordance with their certified values within errors not exceeding 10 %. The quality assurance of the measurements was carried out by periodical efficiency and energy calibration and repeated sample measurement. The counting time for each sample was selected to be 50,000 to obtain the gamma-spectrum with good statistics. To determine the background distribution in the environment around the detector, an empty container was counted in the same manner and in the same geometry as the samples. The background spectra were used to correct the net peak area of γ-rays of the measured isotopes.

The γ-ray transitions of energies 351.9 keV (^{214}Pb) and 609.3 keV (^{214}Bi) were used to determine the activity concentration of the ^{226}Ra series. The γ-ray lines at 911.1 keV (^{228}Ac) and 583.1 keV (^{208}Tl) were used to determine the activity concentration of the ^{232}Th series. The activity concentrations of ^{40}K were measured directly through the gamma line emission at 1460.8 keV.

The activity concentrations for the natural radionuclides in the measured samples are computed using the following relation:

$$C = \frac{N}{\varepsilon P M t}\left(Bq\ kg^{-1}\right) \quad (1)$$

where N is the net counting rate of the gamma ray, ε is the photo peak efficiency of the used detector, P is the absolute transition of gamma decay, t is the counting time in seconds and M the weight of the dried sample in kilogram.

Radium equivalent activity (Ra_{eq}): The distribution of ^{226}Ra, ^{232}Th and ^{40}K in the concrete materials was not uniform. Uniformity with respect to exposure to radiation was defined in terms of radium equivalent activity (Ra_{eq}) in Bq kg^{-1} to compare the specific activity of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K. The radium equivalent activity is calculated through the following relation[13]:

$$Ra_{eq} = C_{Ra} + 1.43\ C_{Th} + 0.077\ C_K \quad (2)$$

where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively.

γ-Index (I_γ): Number of indices dealing with the assessment of the excess γ-radiation arising from building materials such as external and internal hazard indices and γ-concentration indices has been proposed by several investigators[13-16]. In this study, the γ-index (I_γ) is calculated as proposed by the European commission[14].

$$I_\gamma = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000} \quad (3)$$

where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively.

α-Index (I_α): So far, several α-indices have been proposed to assess the exposure level due to radon inhalation originating from building materials[14]. The α-index is determined by the following formula:

$$I_\alpha = \frac{C_{Ra}}{200\ (Bq\ kg^{-1})} \quad (4)$$

where C_{Ra} (Bq kg^{-1}) is the activity concentration of ^{226}Ra assumed in equilibrium with ^{238}U. The recommended exemption and upper level of ^{226}Ra activity concentrations in building materials are 100 and 200 Bq kg^{-1}, respectively, as suggested by ICRP[17]. These considerations are reflected in the α-index. The recommended upper limit concentration of ^{226}Ra is 200 Bq kg^{-1}, for which $I_\alpha = 1$.

Calculation of air-absorbed dose rates: There is concern that some of the buildings will cause excessive radiation doses to the total body due to γ-rays emitted by ^{214}Pb and ^{214}Bi progeny of ^{226}Ra and ^{232}Th decay chains and ^{40}K also contributes to the total body radiation dose. The absorbed dose rate in indoor air due to γ-ray emission from activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K is estimated using the following formula provided by UNSCEAR[18] and EC[14].

$$D = \alpha C_{Ra} + \beta C_{Th} + \gamma C_K \quad (5)$$

where α, β and γ are the dose rates per unit activity concentrations of Ra, Th and K (nGy h^{-1}/Bq kg^{-1}); C_{Ra}, C_{Th} and C_K are the activity concentrations of Ra, Th and K (Bq kg^{-1}), respectively. The values of α, β and γ were taken to be 0.92, 1.1 and 0.08, respectively[14].

Calculation of the annual effective dose: To estimate the annual effective dose, it must be taken into account: (a) the conversion coefficient (0.7 Sv Gy^{-1}) from absorbed dose in air to effective dose and (b) the indoor occupancy factor (about 80 % for Turkish population). Therefore, the effective dose rate in mSv y^{-1} unit is estimated by the following formula[19]:

$$AED\ (mSv\ y^{-1}) = D\ (nGy\ h^{-1}) \times 8760\ h\ y^{-1} \times 0.7 \times 0.8 \times 10^{-6} \quad (6)$$

RESULTS AND DISCUSSION

The composition (in gram) of the nine concrete mixtures is summarized in Table-1. In Turkey, these types of concretes are commonly used as building material. The results of activity concentrations are given in Table-2 for the natural radionuclides of ^{226}Ra, ^{232}Th and ^{40}K in ordinary concrete MO and concretes loaded with different proportions of different mineral additives (MF and MB). It can also be seen from Table-2 that the results increase with increasing percentage of additions for concrete mixture samples. When the concrete mixture samples were compared in their own right, it was observed that the ^{226}Ra, ^{232}Th and ^{40}K activity concentrations of concrete mixture samples increase with the addition of fly ash and blast furnace slag at percentages of 0, 5, 10, 20 and 30 % as shown Fig. 1. The specific activities values obtained in this study were found to be lower than recommended maximum level for all mixture concrete samples[20].

TABLE-1
COMPOSITION (g) OF THE NINE CONCRETE MIXTURES

Mortars	(%)	Cement	Water	Fly ash	Blast furnace slag	Aggregate
MO	0	450	225	0	0	1350
MF5	5	427.5	225	22.5	0	1350
MF10	10	405	225	45	0	1350
MF20	20	360	225	90	0	1350
MF30	30	315	225	135	0	1350
MB5	5	427.5	225	0	22.5	1350
MB10	10	405	225	0	45	1350
MB20	20	360	225	0	90	1350
MB30	30	315	225	0	135	1350

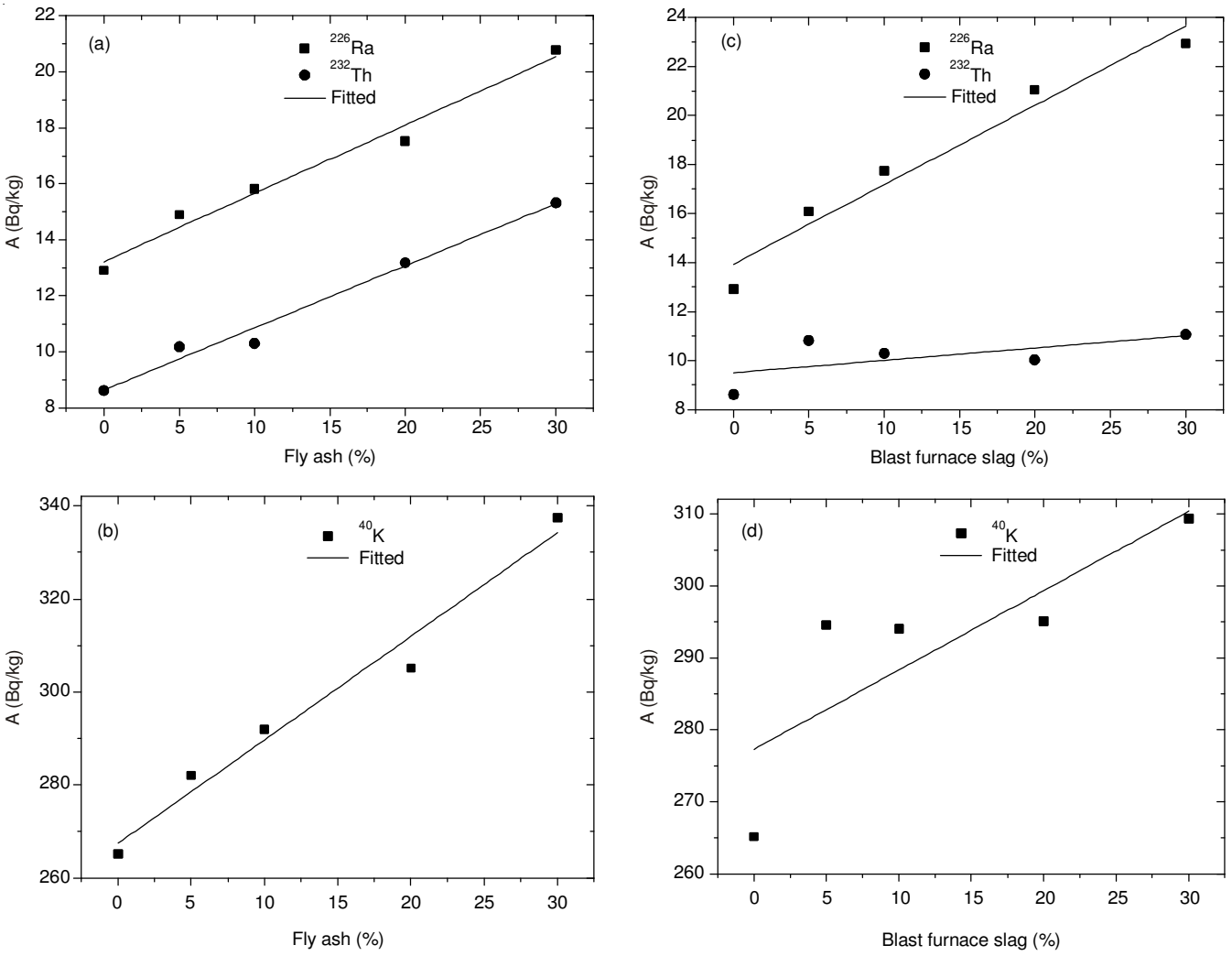

Fig. 1. Activity concentrations in concrete mixture samples *versus* per cent of values of fly ash (a, b) and blast furnace slag (c, d)

TABLE-2
ACTIVITY CONCENTRATIONS (Bq kg^{-1}) OF ^{232}Th, ^{226}Ra AND ^{40}K IN CONCRETE SAMPLES ACCORDING TO THE DIFFERENT PERCENTAGES AND CONCRETE RAW MATERIALS

Samples	Radionuclide concentrations (Bq/kg)		
	^{226}Ra	^{232}Th	^{40}K
MO	12.91 ± 0.59	8.61 ± 0.73	265.18 ± 7.02
MF5	14.90 ± 0.65	10.18 ± 0.82	282.06 ± 7.49
MF10	15.81 ± 0.67	10.30 ± 0.83	291.97 ± 7.68
MF20	17.53 ± 0.70	13.19 ± 0.88	305.16 ± 7.83
MF30	20.78 ± 0.79	15.32 ±1.00	337.45 ± 8.71
MB5	16.08 ± 0.73	10.81 ± 0.89	294.55 ± 8.20
MB10	17.74 ± 0.69	10.29 ± 0.88	294.02 ± 7.92
MB20	21.06 ± 0.75	10.03 ± 0.79	295.05 ± 7.29
MB30	22.95 ± 0.74	11.06 ± 0.87	309.32 ± 8.10
Aggregate	9.51 ± 0.49	8.00 ± 0.65	322.91 ± 7.37
Fly ash	114.67 ± 2.08	87.86 ± 2.71	898.29 ± 16.41
Blast furnace slag	186.69 ± 2.38	35.87 ± 1.67	295.91 ± 9.08
Cement	26.09 ± 0.83	8.40 ± 0.83	168.42 ± 5.76

In addition, the natural radionuclide concentrations of concretes 'raw materials such as cement, fly ash, blast furnace slag and aggregate were listed in Table-2. It is observed that the measured activity concentration values of ^{226}Ra, ^{232}Th and ^{40}K in fly ash, blast furnace slag and ^{40}K in aggregate are higher according to the other raw samples. As can be seen from Table-2, the radioactivity values of ^{226}Ra, ^{232}Th and ^{40}K in fly ash and those of ^{226}Ra in blast furnace slag are higher than the corresponding world mean specific activities values which are 50, 50 and 500 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively[20]. Fly ash and blast furnace seems to be problematic from radionuclides content point of view. However, fly ash is used in Turkish cement industry as an addition to Portland-fly ash cement in an amount not exceeding 35 % (6-35 %)[21]. As a result, the natural activities values of the above-mentioned radionuclides in ordinary concrete (MO), concretes loaded with different proportions of different mineral additives (MF and MB), cement and aggregate are lower than the world mean values. From the point of view radionuclide, the mixing of fly ash and blast furnace slag with concrete components prepared in this work is suitable for the walls of the new buildings.

The mean calculated Ra_{eq} values are shown in Table-3 for the MO, MF and MB and for building raw materials (fly ash, blast furnace, cement and aggregate). The mean Ra_{eq} values varies from 45.64 ± 2.40 to 68.67 ± 2.89 Bq kg^{-1}, 45.64 ± 2.63 to 62.58 ± 2.61 Bq kg^{-1} for MF and MB, respectively. It can be seen that the highest mean values of Ra_{eq} activity were determined in fly ash (309.48 ± 7.22 Bq kg^{-1}) and blast furnace (260.77 ± 5.47 Bq kg^{-1}). However, the obtained results indicate that the mean Ra_{eq} values in this work are less than the recommended maximum levels for building materials to be used for house, which is < 370 Bq kg^{-1} [18,19,21,22]. The results were also shown graphically in Fig. 2 for concrete mixture samples

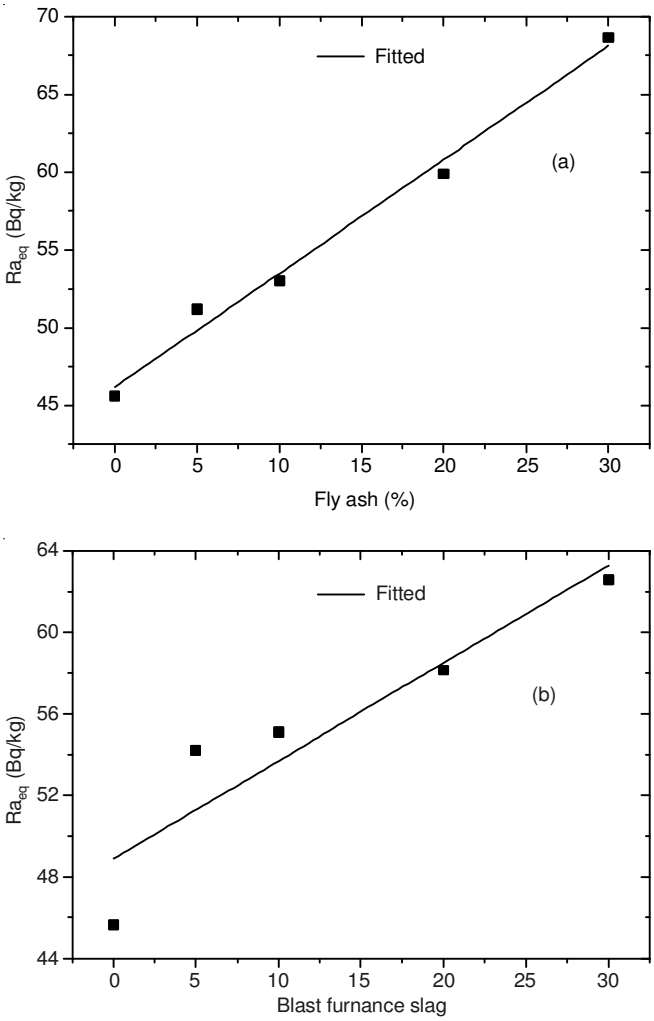

Fig. 2. Radium equivalent (Ra_{eq}) activities in concrete mixture samples *versus* per cent of values of fly ash (a) and blast furnace slag (b)

TABLE 3
RADIUM EQUIVALENT ACTIVITY, γ-INDICES, α-INDICES, THE DOSE RATE AND ANNUAL EFFECTIVE DOSE FOR CONCRETE SAMPLES ACCORDING TO THE DIFFERENT PERCENTAGES AND CONCRETE RAW MATERIALS

Samples	Ra_{eq} (Bq/kg)	I_γ	I_α	D (nGy h^{-1})	AED (mSv y^{-1})
MO	45.64 ± 2.17	0.17	0.06	42.56 ± 1.91	0.21
MF5	51.18 ± 2.40	0.19	0.07	47.47 ± 2.10	0.23
MF10	53.02 ± 2.45	0.20	0.08	49.23 ± 2.14	0.24
MF20	59.89 ± 2.56	0.23	0.09	55.05 ± 2.24	0.27
MF30	68.67 ± 2.89	0.26	0.10	62.97 ± 2.52	0.31
MB5	54.22 ± 2.63	0.21	0.08	50.25 ± 2.31	0.25
MB10	55.09 ± 2.56	0.21	0.09	51.16 ± 2.24	0.25
MB20	58.12 ± 2.44	0.22	0.11	54.01 ± 2.14	0.26
MB30	62.58 ± 2.61	0.23	0.11	58.03 ± 2.29	0.28
Aggregate	45.81 ± 1.99	0.18	0.05	43.38 ± 1.76	0.21
Fly ash	309.48 ± 7.22	1.12	0.57	274.01 ± 6.21	1.34
Blast furnace slag	260.77 ± 5.47	0.90	0.93	234.88 ± 4.75	1.15
Cement	51.07 ± 2.46	0.19	0.13	46.72 ± 2.14	0.23

TABLE-4
COMPARISON OF ACTIVITY CONCENTRATIONS (Bq kg^{-1}) AND THE RADIUM EQUIVALENT (Ra_{eq}) ACTIVITIES WITH PUBLISHED RESULTS IN CEMENT (A), FLY ASH (B), BLAST FURNACE SLAG (C), AGGREGATE (D) AND CONCRETE (E) SAMPLES FROM DIFFERENT COUNTRIES

Countries	Radionuclide concentrations (Bq kg^{-1})			Ra_{eq} (Bq kg^{-1})	References
	^{226}Ra	^{232}Th	^{40}K		
(a) Cement					
Australia	52	48	115	129	[14]
Austria	27	14	210	63	[23]
Algeria	41	27	422	112	[24]
Bangladesh	61	80	1133	262	[25]
China	57	37	173	122	[26]
Cameroon	27[a]	15	277	70.1	[27]
Cuba	23	11	467	74	[28]
Egypt	78	33	337	151	[29]
Finland	40	20	251	88	[30]
Greece	63	24	284	117	[31]
Italy	38	22	218	92	[32]
Isparta, Turkey	26.1	10.4	129.7	51.03	[7]
India	98[a]	81	340	240	[33]
Japan	36	21	139	77	[34]
Malaysia	81	59	203	181	[35]
Netherlands	27	19	230	72	[36]
Pakistan	26	29	273	88	[37]
Turkey	41	26	267	98	[21]
Xianyang, China	51.7	32	207.7	113.5	[11]
Turkey	26.09	8.40	168.42	51.07	Present study
(b) Fly ash					
Greece	1377	65	661	-	[10]
Greece	1041	55	462	-	[8]
India	45.1	39.9	88.4	109.2	[3]
Israel	202.0	62.9	377.7	321.0	[5]
Spain	160	62.3	459.1	-	[38]
Turkey	232.3	117.1	466.2	428.7	[21]
Turkey	114.67	87.86	898.29	309.48	Present study
(c) Blast furnace slag					
Egypt	323	39.8	158	392.3	[39]
India	67.3	77.7	145.1	189.8	[3]
Spain	166.6	47.6	232.3	-	[38]
Turkey	177.9	147.8	242.5	392.95	[21]
Turkey	186.69	35.87	295.91	260.77	Present study
(d) Aggregate					
Algeria	24	10	259	58	[24]
Bangladesh	248	219	389	-	[40]
Bangladesh	25.26[a]	54.71	228.4	121.29	[41]
Brazil	10.3	-	933	82.1	[42]
Cuba	20	13	134	49	[28]
Egypt	9.8[a]	3.5	62.4	19.7	[39]
Greece	11	12	140	38.9	[6]
Israel	15	3	50.4	23.2	[5]
Isparta, Turkey	22.5	6.8	101.7	40.03	[7]
Logbadjeck, Cameroon	24[a]	139	1161	312.51	[27]
Ombe, Cameroon	19.4[a]	26	304	80.12	[27]
Pakistan	33[a]	32	57	-	[43]
Turkey	21.6	25.8	482.2	-	[44]
Xi'an, China	28.5	19.5	286.7	82.6	[45]
Xianyang, China	19.9	22.5	413.0	84.0	[11]
Turkey	9.51	8.00	322.91	45.81	Present study
(e) Concrete					
Brazil	14.8	9.9	361	56.7	[42]
Cuba	25	12	595	87	[28]
Greece	35	17	383	88.8	[6]
Egypt	19.1	8.1	76.4	36.6	[39]
Egypt	11.0	2.8	19.6	16.5	[46]
Ibadan, Nigeria	24.5	37.3	354.4	105.2	[47]
Jos Nigeria	66	126	589	292	[48]
Malaysia	30[a]	24	740	121	[49]
Xi'an, China	46.2	28.4	137.4	98.6	[45]
Xianyang, China	25.8	26.8	553.6	106.7	[11]
Turkey	12.91	8.61	265.18	45.64	Present study

[a] ^{238}U

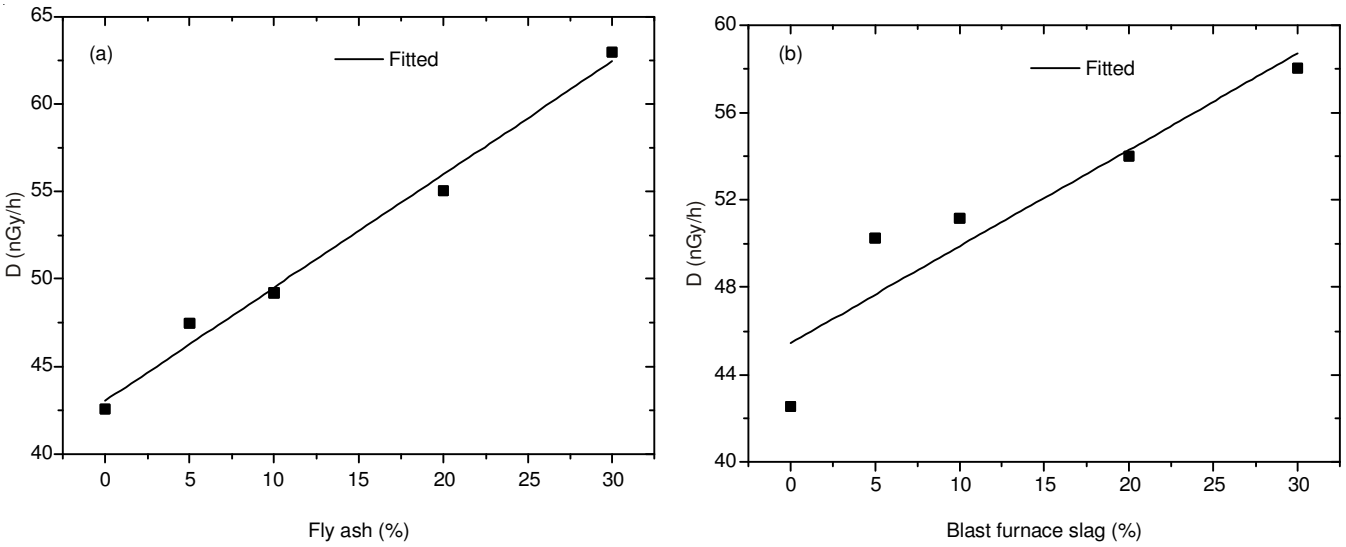

Fig. 3. Dose rate (D) values in concrete mixture samples *versus* per cent of values of fly ash (a) and blast furnace slag (b)

versus per cent of values of fly ash and blast furnace slag, respectively. Fig. 2 demonstrate same noticed feature in previous figure, such as increasing of radium equivalents with mineral additives. Table-4a-e lists the comparison between activity concentrations and radium equivalents (Bq kg^{-1}) in cement, fly ash, blast furnace, aggregate and concrete in different areas of the world, respectively. The comparison shows a reasonable agreement for cement, blast furnace slag, aggre-gate and mixture concretes samples in Table-4a, c, d, e, respectively. As shown in Table-4b, if we compare our results with those of values in different countries for fly ash, it is clear that ^{226}Ra, ^{232}Th and Ra_{eq} is good agreement, while ^{40}K is high in the present work.

The mean values of γ-index (I_γ) calculated from the measured activity concentration of ^{226}Ra, ^{232}Th and ^{40}K are listed in Table-3 (column 2) for all the studied samples. It is observed that the mean γ-index I_γ values do not exceed critical value of unity for all samples. These values of I_γ are < 2, which corresponds to a dose criterion of 0.3 mSv y^{-1}. It is observed that mean I_γ of values for fly ash (1.12) and blast furnace slag (0.90) samples are higher than the values of the rest of the building raw materials and, the concrete mixture samples.

The α-index (I_α) was calculated from eqn. (4) and listed in Table-3 (column 3). As shown in Table-3, the calculated values of I_α for the studied samples are less than unity.

The estimated absorbed dose rate (D) values for the studied samples are also shown in Table-3 (column 4). All the samples measured, except for fly ash (274.01 ± 6.21) and blast furnace slag (234.88 ± 4.75), showed values below the recommended upper level, which is 84 nGy h^{-1}.

The obtained annual effective dose rate results are in Table-3 (column 5) and displayed in Fig. 3. The annual effective dose values for all samples except for fly ash (1.34 mSv y^{-1}) and blast furnace slag (1.15 mSv y^{-1}) are lower than the limit value, which is 0.46 mSv y^{-1} [18].

Conclusion

The specific activity of ^{226}Ra, ^{232}Th and ^{40}K have been measured in ordinary concrete MO, concretes loaded with different proportions of different mineral additives (MF and MB) and building raw materials used in Turkey, using HPGe gamma spectrometry. The radioactivity values of ^{226}Ra, ^{232}Th and ^{40}K in fly ash and those of ^{226}Ra in blast furnace slag are higher than the corresponding world mean specific activities values, while the rest of the samples are less than the critical value of unity. In addition, the mean radium equivalent activity (Ra_{eq}), γ-index (I_γ), α-index (I_α), the indoor absorbed dose rate (D) and annual effective dose rate have been calculated for all samples. The observed these values except for annual effective dose values of fly ash and blast furnace slag are lower than the acceptable level. According to these results, mixture concrete samples examined in this study do not pose any significant source of radiation hazard and thus these materials could be safely used in building constructions.

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