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Measurement of Natural Radioactivity and Radon Exhalation Rate in Coal Ash Samples from a Thermal Power Plant

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Abstract

Coal is the main energy source for electricity generation in the world. In Morocco, 37% of electricity generation comes from combustion coal in thermal power plants. This combustion process generates large amounts of fly and bottom ashes. In recent years, these ashes became a great topic of interest because of their different uses and especially in construction materials. In this work, we assess radiation risks due to natural radioactivity in samples of fly and bottom ashes collected from JLEC (Jorf Lasfar Energy Company) thermal power plant, and different analyses are performed through two nuclear techniques such as gamma spectrometry and alpha dosimetry based on the use of LR115 films detectors. Our analysis shows that ^{226}Ra activities and ^{232}Th in both ash samples are well above the permissible activity. The values of the external risk index (H_{ex}) and internal one (H_{in}) for these ashes are below unity, with the exception of 1.28 in fly ash for H_{in}. The obtained values for the equivalent radium Ra_{eq} and annual effective doses $\dot{E}$ in fly and bottom ashes are 324 Bq/kg and 210 Bq/kg, and 0.18 mSv/y and 0.11 mSv/y, respectively. The surface radon exhalation rates for the samples of fly and bottom ashes are 276 $mBq \cdot m^{-2} \cdot h^{-1}$ and 381 $mBq \cdot m^{-2} \cdot h^{-1}$, respectively. Based on these results, we have shown that fly ash and bottom one from thermal power plant JLEC didn't have, in any case, a health risk to the public so it can be effectively used in various construction activities.

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Keywords

Natural Radioactivity, Gamma Spectrometry, LR115, Fly Ash, Bottom Ash, Equivalent Radium, Annual Effective Doses, Exhalation of Radon

1. Introduction

The socio-economic development of Morocco and the major projects being initiated in all sectors, particularly the global rural electrification program, contributed strongly to increase the national request of energy. The main source of energy for the electricity production comes from the coal combustion in thermal power plants [1]. This combustion generates large amounts of solid residues such as fly and bottom ashes. The radioactivity content of different mineral coal is very heterogeneous and varies depending on their origin. Generally, coal contains radionuclides with activity ranging from 30 to 100 Bq/kg, 10 to 600 Bq/kg and 10 to 200 kg/Bq, for ^{40}K, ^{238}U and ^{232}Th, respectively [2]. The specific activity of the coal ash can reach three to five times higher than the original one. This is called Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM). While the levels of radioactivity of these ashes remain moderate, a prolonged exposure may become significant when large quantities of these ashes are landfilled or recycled in building materials. The use of coal ashes has economic advantages but can affect the doses received by the human being indoors. Indeed, this use increases the external and internal radiations due to inhalation or ingestion of radon and its descendants [3].

Radon-222 is the most commonly measured because of its relatively long period (3.824 d). It is an odorless natural radioactive gas, and is the main source of radiation exposure for humans. Its contribution is estimated at more than half of the average annual dose after natural radioactivity received by the population [4]. In 1987, the International Agency for Research on Cancer (IARC) of the World Health Organization (WHO) recognized radon as lung human carcinogen [5].

In the present work, we assess the radiation hazards due to natural radioactivity in coal ash from radionuclides of three chains of radioactive decay (^{238}U, ^{235}U and ^{232}Th) and ^{40}K, calculations of external hazard index (H_{ex}) and internal one (H_{in}), the equivalent radium (Ra_{eq}), the absorbed dose rate ($\dot{D}$), the annual effective dose ($\dot{E}$) and the radon exhalation rate (E_x) are performed on samples of fly and bottom ashes delivered by the thermal power plant of Jorf Lasfar Energy Company (JLEC), the first private electricity producer in Morocco.

2. Overview of the Thermal Power Plant of JLEC

The power plant of JLEC is the largest thermal coal one, independent of Middle East and North Africa. It is the main provider of the National Office of Electricity and Water Supply in Morocco with a total annual production capacity about 2056 MW of electricity [6]. It is located in the port Jorf Lasfar, which is 127 km far from the southwest of Casablanca, on the Atlantic Ocean coast. It occupies an area of 60 hectares and is located on a narrow strip of land between a 60 m cliff height on the East and West Atlantic Ocean as shown in **Figure 1**.

In JLEC thermal power plant, coal combustion generates large masses of solid wastes that exceed 640,000 tons per year. Fly ashes are a majority and are around 500,000 tons per year. And for the bottom ones, the production rate exceeds 50,000 tons per year [7].

Today, 80% - 95% of fly ash is valued by some national cement, while the bottom ash continues to be stored in large quarries.

3. Materials and Methods

In the present work, we evaluate radiation hazards due to natural radioactivity in coal ash from radionuclides of three chains of radioactive decay (^{238}U, ^{235}U and ^{232}Th) and ^{40}K. Our analyzes were carried out on samples of fly

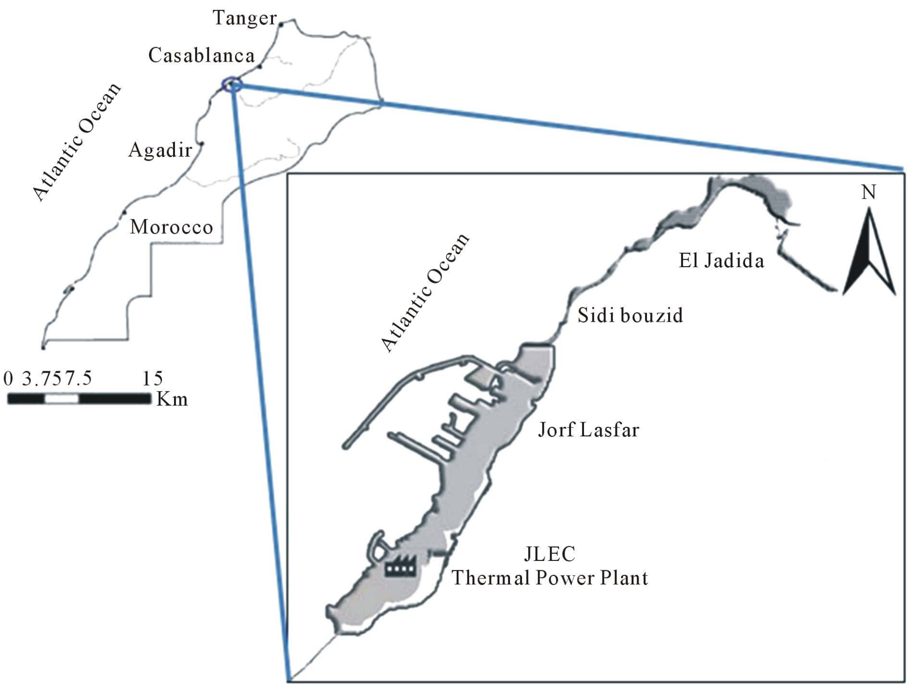

Figure 1. The power plant Jorf Lasfar Energy Company (JLEC).

and bottom ashes delivered by the thermal power plant JLEC using two nuclear techniques: gamma spectrometry for identifying the gamma radio transmitters and quantify their activities, and alpha dosimetry based on quantitative exploitation of Solid-State Nuclear Track Detectors (SSNTD, LR115).

Before any measures and for homogeneous samples, fly and bottom ashes were dried in stove at 40˚C during 24 hours, then crushed and sieved through a sieve of 100 µm. The sieved samples were packaged in sealed radon containers for at least 4 weeks to establish secular equilibrium corresponding to seven half-lives of ^{222}Rn.

3.1. Samples Activities of Coal Ash

To measure the natural radioactivity in coal ash, samples of fly and bottom ashes were studied and analyzed by gamma ray spectrometer with Broad Energy Germanium detector (BEGe) at the Multidisciplinary Institute Hubert Curien in Strasbourg, France. This is a Hyper-Pure Germanium planar type HPGe detector associated with a set of electronic modules for the pulse shaping, amplification and storing of pulses delivered during the passage of gamma radiation through the detector. Its area of energy measurement is 30 to 3000 keV with a resolution of 0.633 keV to 122 keV, and 1.934 keV to 1332 keV [8].

With regard to energy efficiency and calibration of the BEGe detector, a multi-energy certified standard was analyzed under the same conditions and geometry of the studied samples. This standard contains several emitting radionuclides γ such as ^{241}Am (60 keV), ^{109}Cd (88 keV), ^{57}Co (122, 136 keV) ^{139}Ce (165 keV), ^{51}Cr (320 keV) ^{113}Sn (391 keV), ^{85}Sr (514 keV), ^{137}Cs (661 keV), ^{88}Y (898, 1836 keV) and ^{60}Co (1173, 1332 keV). The samples of coal ash have been conditioned by SG50 and geometry set during 263000 seconds counting; a little less than 74 hours. Treatment of amplitude spectra was carried out with automatic counting software (Genie 2000) [8] to give directly the activity concentration of each radioactive element present in the sample.

The radionuclides, in which we are interested, are the radium 226, the thorium 232 and the potassium 40. ^{226}Ra is difficult to differentiate from ^{235}U; as they emit photons with very close energies; 186.1 keV and 185.72 keV, respectively, and the interference can be created. The activity of ^{226}Ra can be performed from the ^{214}Pb (295 keV and 352 keV rays) and ^{214}Bi (609 keV, 1120 keV and 1764 keV rays) after establishment of the secular equilibrium between ^{226}Ra, ^{222}Rn, ^{214}Pb and the ^{214}Bi. ^{232}Th has a ray at 63.81 keV which has a very low emission probability of 0.263%. This ray interferes with that of ^{234}Th at 63.28 keV which has a higher transmission probability equal to 4.1%. The determination of ^{232}Th activity is from the ^{228}Ac (911 keV and 969 keV rays) and ^{212}Pb (239 keV ray). Given the short period (56 s) of ^{220}Rn decay that secular equilibrium is reached quickly for the thorium family. The ^{40}K is determined from the 1461 keV ray with 10.55% emission intensity.

3.2. Surface Radon Exhalation Rate

In this paragraph, we describe the process and experimental set up used to estimate the radon exhalation rate of ashes samples analyzed by gamma spectrometry. Exhalation is the mechanism which a radon atom produces a gas inside the sample reaching its surface. It groups precisely two steps: the emanation and transport. It is commonly expressed as surface flow exhalation radon ($Bq \cdot m^{-2} \cdot s^{-1}$). In the laboratory, for samples measurements we use the term of surface radon exhalation rate ($Bq \cdot m^{-2} \cdot h^{-1}$) [9]-[11].

Each amount of sample fly and bottom ashes (50 g) were placed in the cylindrical "cans" as shown in **Figure 2**. The Solid-State Nuclear Track Detectors (SSNTD) LR115 type II non strippable ($2 \times 2\ cm^2$) was fixed on the top inside of the "can".

These films badges are LR115 type nuclear track ones produced by KODAK and they consist of a 100 µm thick polyester substrate coated with a 12 µm thick layer of red colored cellulose nitrate ($C_6H_8N_2O_9$). It can record energy particles between 1.4 and 4.7 MeV with an incidence angle up to 50˚. Alpha particles traverse the detector leaving various holes with diameters depending to the incident energy. After two months of irradiation, the LR115 was chemically treated in 2.5 N sodium hydroxide solution (NaOH) at 60˚C for time periods of 100 min. An optical microscope was used to read the developed films.

The density of traces per unit area and per unit time D_{LR} in the LR115 and the volumetric activity of radon A_V^{Rn} are related by the following relationship:

$$D_{LR} = \varepsilon_{LR}\left(\theta_c, E_\alpha\right) A_V^{Rn} \quad (1)$$

where $\varepsilon_{LR}\left(\theta_c, E_\alpha\right)$ is the efficiency of detection based on the critical recording angle θ_c, and the energy of the alpha particle E_α calculated numerically [12]. This efficiency is equal to 0.0258 ($traces \cdot cm^{-2} \cdot d^{-1}/Bq \cdot m^{-3}$).

Exhalation rate ^{222}Rn is obtained from the following expression [13] [14]:

$$E_X = \frac{A_V^{Rn} V \lambda_{Rn}}{S_e\left[t + \left(\frac{1}{\lambda_{Rn}}\right)\left(e^{-\lambda_{Rn} t} - 1\right)\right]} \quad (2)$$

where E_X is measured in ($Bq \cdot m^{-2} \cdot h^{-1}$); A_V^{Rn} is the volumetric activity of radon ($Bq \cdot m^{-3} \cdot h$); λ_{Rn} is the decay constant for radon (h^{-1}); S_e is the sample surface (m^2); V is the effective volume of can (m^3) and t is the exposure time (h).

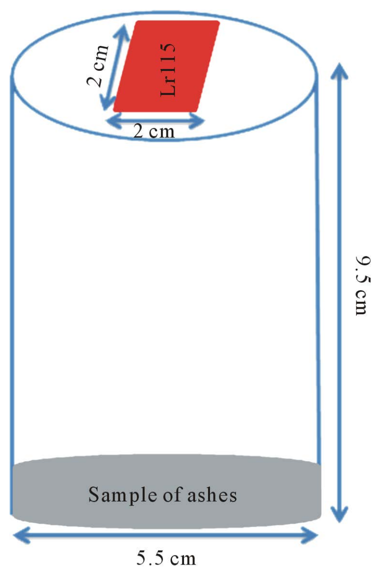

Figure 2. Diagram dosimeter to measure the exhalation of radon in samples of coal ash at thermal power plant JLEC.

3.3. Evaluation of the Radiological Effects and Dose Estimation

In recent years, fly and bottom ashes have become a topic of great interest in the world because of the diversity of their uses in building materials. For this purpose, to evaluate the radiological hazards of fly and bottom ashes from the thermal power plant JLEC due to non-uniform distribution of radionuclides in the samples of coal ash, the UNSCEAR 2000 report [15] offers templates to define some dosimetric quantities such as radium equivalent, the index of internal/external hazard, the flow of the absorbed dose and the annual effective dose. Certain limits not to be exceeded are recommended.

3.3.1. Radium Equivalent Ra_{eq}

To represent the level of activity of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, a common radiological index was introduced. This index is known as the equivalent radium activity symbolized by Ra_{eq} and calculated using the following expression:

$$Ra_{eq} = A_{226Ra} + 1.43A_{232Th} + 0.077A_{40K} \tag{3}$$

This relationship is obtained by considering that the activities 1 Bq/kg of ^{226}Ra, 0.7 Bq/kg of ^{232}Th and 13 Bq/kg of ^{40}K produce the same dose of gamma rays [16] [17].

It should be noted that the maximum value of the activity of radium equivalent in construction materials must be less than 370 Bq/kg.

3.3.2. Internal and External Hazard Indices

The hazard indices are defined by a model taking into account the maximum activity of Ra_{eq} (370 Bq/kg). The H_{ex} external hazard index is given by:

$$H_{ex} = \frac{A_{226Ra}}{370} + \frac{A_{232Th}}{259} + \frac{A_{40K}}{4810}. \tag{4}$$

In addition to the external hazard, the respiratory organs are threatened because of the disintegration of ^{226}Ra and ^{222}Rn and their descendants. The maximum permissible activity for ^{226}Ra has been reduced in half the value of 185 Bq/kg. The internal hazard H_{in} must be quantified:

$$H_{in} = \frac{A_{226Ra}}{185} + \frac{A_{232Th}}{259} + \frac{A_{40K}}{4810}. \tag{5}$$

The maximum value of each index is tolerable with the unit value for the upper limit of Ra_{eq}.

3.3.3. Absorbed Dose Rate and Annual Effective Dose

The absorbed dose rate $\dot{D}$ (nGy/h) from natural radionuclides in the air at 1m height is defined by expression (6):

$$\dot{D}(nGy/h) = 0.462A_{226Ra} + 0.604A_{232Th} + 0.0417A_{40K}. \tag{6}$$

The received dose by the population is called annual effective dose, it calculated by the following equation:

$$\dot{E}(mSv/y) = \dot{D}(nGy/h) \times 8760(h) \times 0.2 \times 0.7(Sv/Gy)10^{-6}. \tag{7}$$

The annual effective dose rates should be obtained to test the health effect of those absorbed dose rates. In order to estimate the annual effective doses, one has to take into account to conversion coefficient from absorbed dose in air to effective and the outdoor occupancy factor. In the UNSCEAR reports [15], a value of 0.7 Sv/Gy was used for the conversion coefficient from absorbed dose in air to effective dose received by adults and 0.2 for the outdoor occupancy factor.

4. Results and Discussion

The specific activities of radionuclides ^{226}Ra, ^{232}Th and ^{40}K were determined by gamma spectrometry in samples of fly and bottom ashes from thermal power plant JLEC and other thermal plants of some European countries are regrouped in **Table 1**.

According to the qualitative analysis of spectra obtained for both samples of fly and bottom ashes, we find practically all radionuclides of natural radioactive families (^{238}U, ^{232}Th, ^{235}U and ^{40}K).

By examining the results in **Table 1**, the first observation is that the specific activities of elements ^{226}Ra, ^{232}Th and ^{40}K in the sample of fly ash are higher compared to those found in the bottom ash with factors of 2.07, 1.77 and 1.32 for ^{40}K, ^{226}Ra and ^{232}Th, respectively.

The activities of ^{226}Ra and ^{232}Th in both ash samples are well above the permissible activity which is in the order of 40 Bq/kg, while the activity of ^{40}K is less than 370 Bq/kg [15] even if the activity of ^{40}K (348 Bq/kg) in the sample of fly ash is very close to the eligible activity.

The levels of ^{226}Ra, ^{232}Th and ^{40}K measured in our samples of coal ash from thermal power plant JLEC are comparable to those found in other thermal power plants in Europe.

Table 2 shows that the values of Ra_{eq}, H_{ex} and H_{in} in fly ash and bottom ash are below the maximum values defined by the UNSCEAR 2000 report [15], except for the value of the H_{in} (1.28) in fly ash that remains a little higher than the unit.

The absorbed gamma dose rates measured in the samples of fly and bottom ashes are of the order of 146 nGy/h and 93 nGy/h, respectively. After conversion of these values, the annual effective dose of these samples of fly and bottom ashes are on the order of 0.18 mSv/y and 0.11 mSv/y, respectively. Those values not exceeding the annual effective dose limit set of 1 mSv/y.

The results presented in **Table 3** show that the values of activity concentrations of radon and the rate of surface exhalation in the bottom ash samples are higher than those of the samples of fly ash. The exhalation of radon levels in coal ash samples are below the world average (57.6 $Bq \cdot m^{-2} \cdot h^{-1}$) [15]. These results agree with those found by other authors (**Table 3**).

Table 1. The specific activities of ^{226}Ra, ^{232}Th and ^{40}K obtained for fly ash (FA) and bottom ash (BA) compared to other works.

Thermal Power Plants	Sample	Activity in Bg/kg		
		^{226}Ra	^{232}Th	^{40}K
JLEC-Morocco (Present Study)	FA	149 ± 26	104 ± 18	348 ± 47
	BA	84 ± 16	79 ± 15	168 ± 30
Serbia (2011) [18]	FA	120	72	360
	BA	65	39	241
Spain (2009) [19]	FA	191 ± 9	74 ± 3	306 ± 13
	BA	149 ± 6	66 ± 3	235 ± 11
Turkey (2008) [20]	FA	149 ± 2	58 ± 4	94 ± 28
	BA	50 ± 1	25 ± 2	376 ± 9
Greece (2004) [21]	FA	904 ± 9	53 ± 5	454 ± 11
	BA	662 ± 9	44 ± 5	405 ± 11

Table 2. Radium equivalent Ra_{eq}, internal hazard index H_{in}, external hazard index H_{ex}, absorbed dose rate $\dot{D}$ and annual effective dose $\dot{E}$ in ash coal.

Sample	Ra_{eq} (Bq/kg)	H_{ex}	H_{in}	$\dot{D}$ (nGy/h)	$\dot{E}$ (mSv/y)
Fly Ash	324 ± 30	0.88	1.28	146 ± 9	0.18 ± 0.01
Bottom Ash	210 ± 28	0.57	0.79	93 ± 6	0.11 ± 0.01

Table 3. Experimental results of the volume activity of radon A_V^{Rn} and the surface exhalation rate E_X in samples of ash coal JLEC.

Thermal Plants	Sample	A_V^{Rn} (Bq/m^3)	E_X ($mBq \cdot m^{-2} \cdot h^{-1}$)
JLEC-Morocco (Our Study)	FA	337 ± 27	276 ± 22
	BA	465 ± 38	381 ± 31
India (2013) [22]	FA	431.70 ± 35.50	155.00 ± 12.8
India (2010) [23]	FA	214 to 590	138 to 381

5. Conclusions

In this study, we assess radiation risks due to natural radioactivity in fly and bottom ashes from thermal power plant of JLEC. For this ending, we measured the natural radioactivity in these samples using gamma spectrometry technique. The activities of ^{226}Ra and ^{232}Th in both ashes samples are well above the permissible activity, which is on the order of 40 Bq/kg, while the ^{40}K activity is less than 370 Bq/kg. Based on the models proposed by the UNSCEAR, our analyses reveal that H_{ex} and H_{in} values for fly and bottom ashes are below the standard value, except for the one of H_{in} (1.28) in the fly ash, which is a little more than unity. The values obtained for the equivalent radium and annual effective doses of the fly and bottom ashes are (324 Bq/kg and 210 Bq/kg) and (0.18 mSv/y and 0.11 mSv/y), respectively. These values do not exceed the recommended limits which are 370 Bq/kg and 1 mSv/y, respectively.

To estimate the Exhalation of radon levels in coal ash, we used the alpha dosimetry based on quantitative exploitation of LR115 detector. Exhalation rates found for samples of fly and bottom ashes are 276 $mBq\cdot m^{-2}\cdot h^{-1}$ and 381 $mBq\cdot m^{-2}\cdot h^{-1}$, respectively; these values remain below the eligible limit (57.6 $Bq\cdot m^{-2}\cdot h^{-1}$).

During our work, we find that the results for our samples of coal ash from thermal power plant JLEC are comparable to those obtained in other countries.

Based on the above results, we can conclude that the fly and bottom ashes from thermal power plant of JLEC can be classified in the category of products exempt from any usage restrictions. Therefore, these coal ashes did not present, in any case, a health risk to the public and can be effectively used in various construction activities.

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