Determination of Uranium Concentrations in Brick Samples from Misan Province-Iraq, Using Kinetic Phosphorescence Analyzer (KPA).

Zahraa A. Ismail AL- Sudani[1],Sawsan Sh. Fleifil[2]
[1]Department of Physics, College of Science, Misan University, Misan, Iraq.
[2] Department of Physics, College of Education, Basrah University, Basrah, Iraq.

Abstract:
Bricks as building materials contain radionuclides. Primordial radionuclides in building materials are one of the sources of the radiation hazards in dwellings made of these materials. It is therefore important to determine the uranium levels in bricks for public health, and radiation protection. The natural uranium concentrations of brick samples, which collected from18 brick factories at 7 various areas of Misan Province ,were measured using Kinetic Phosphorescence Analyzer (KPA-11) ,and an annual effective dose of the brick samples were calculated. The results showed that, the natural uranium concentrations ranged from the 0.692 to 46.481 mg/kg with a mean value of 9.403 mg/kg, and the annual effective dose ranged from the 0.0000052 to 0.00162 mSv/y with a mean value of 0.000391 mSv/y. The obtained results showed that all brick samples contained uranium concentrations with various rates, and the annual effective dose of gamma rays that resulted from the dissolution of uranium isotopes in brick samples were less than the total gamma rays dose which is 0.5 mSv/y, That is part of the total average annual doses of radiation from all natural sources of radiation,2.4 mSv/y (IAEA 2004).

Key words: Uranium, Brick Samples, KPA, Annual Effective Dose, Misan Province.

1. Introduction

An established fact, all construction materials contain trace amount of natural radioactivity. This activity is a major source of external and internal radiation exposure for the occupants of a dwelling. The most commonly radionuclides in the construction material are U-238, Th-232,their decay products, and K-40.Amongst the decay series product, radon is an important source of natural radiation .It founds in the environment due to the radioactive decay of uranium. It is estimated that 50-55% of the average annual dose from the natural background radiation is contributed by Rn-222 alone [1]. All building materials; such as concrete, brick, sand, aggregate, marble, granite, limestone, gypsum…etc.; contain mainly natural radionuclide including U-238, Th-232, their decay products, and K-40 [2, 3].Bricks are used as one of the main building materials, therefore, the disclosure of the basic radiological parameters and radioactive contents in bricks and others construction materials is important. This allows us to calculate the exposure of the population of the radiation from natural source [4].The major aims of this study are to estimate the concentrations and annual effective dose of uranium in brick samples collected from various locations in Misan Province, Iraq, and to provide data which can serve as a basis for the determination of changes in the near future.

2. Experimental Method

2.1 The Study Area

The area covered by this study (Misan Province) is located in the south of Iraq, (Figure 1) [5]. It lies between latitudes of 31° 30′.0″N to 33°0′.00″N and between longitudes of 46° 30′.0″Eto 47° 30′.0″E.It covers an area of approximately 16072 km^2. This area is very important since it contains many brick factories. The residents of Misan Province depend on bricks as a basic building material. The disclosure of the radioactive contents within bricks is important since it allows us to calculate the exposure of the population of the radiation from natural source .The above causes make it necessary to measure the natural radioactivity in building materials which will serve as reference data for future studies.

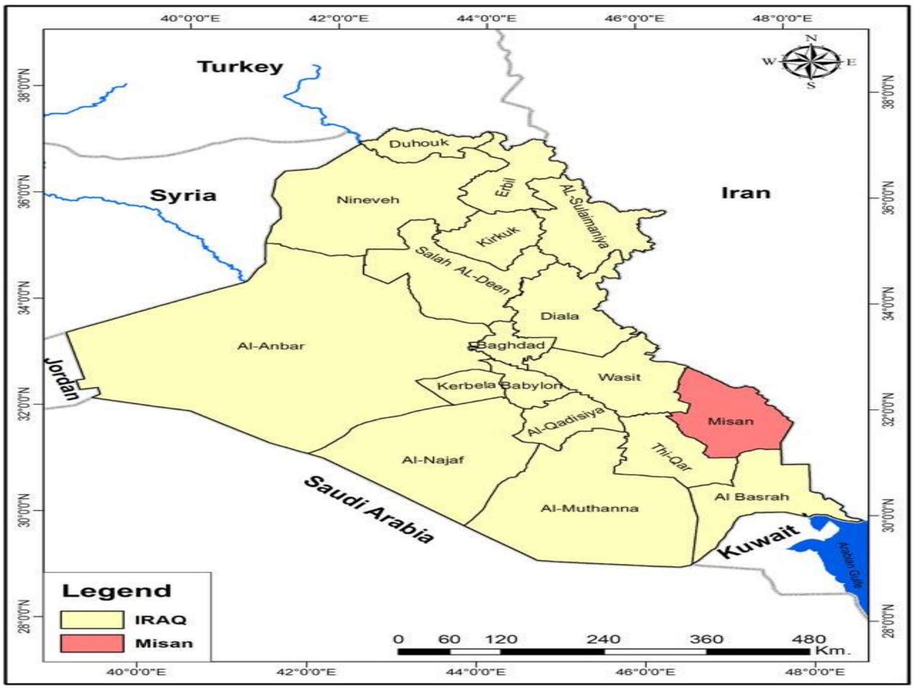

Figure (1): Map of the administrative divisions of Iraq showing the studied area (Misan Province).

2.2 Sample Collection:

In the present work, 18 brick samples were collected from various areas of Misan Province-Iraq, (Almizbania Factory) (S_1), (Almaymuna Factories) (S_2) and (S_3), (Almajar Factories) (S_4) and (S_5), (Altubar Factories) (S_6) (S_7) (S_8) (S_9), (Kasiba Factories) (S_{10}) (S_{11}), (Altayib Factories) (S_{12}) (S_{13}) (S_{14}), and(Albatira Factories) (S_{15}) (S_{16}) (S_{17}) (S_{18}).Each one of collected samples was given a unique code and denoted with its GPS coordinates which was detected by a handheld GPS device, as shown in (Figure 2)[6]. The samples were dried for about two hours by an oven with temperature of 80C°, then crushed to pass through 75 μm mesh sieve to make sure they have homogenous. These crushed brick samples were sealed in plastic containers, and left until analysis[7].These samples were prepared for the analysis of uranium concentrations using a Kinetic Phosphorescence Analyzer (KPA) method[8]**.**In the laboratory, an 0.2gm of the sample is placed in a glass vial, then a 10ml of HNO_3 having concentration of 8M is added along with 0.5 ml of H_2O_2 having concentration of 30%.After wards ,the sample is placed and stirred on a hotplate until it reveals the salt colors that range from yellow to white. Then, the sample is heated to sub-boiling temperature for +2 hours, starting with hotplate set at 200.If sample evaporates to ~2-3 ml, 8 M HNO_3 have to be added in order to return to a volume of ~10 ml. After +2 hours, 8M HNO_3 have to be added again in order to return to a volume of ~10 ml. After that, allow the sample to evaporate to ~2-3 ml and left to cool. Transfer the sample into 50 ml centrifuge tube rinsing vial with 2% HNO_3.Add DI water and 2% HNO_3 in order to return to a volume of 50 ml with a concentration of (0.3-1) M HNO_3.If Sample does not settle, then centrifuge or filter the sample liquid. Then, the solution is analyzed by using the KPA-11 with appropriate further dilution[9].

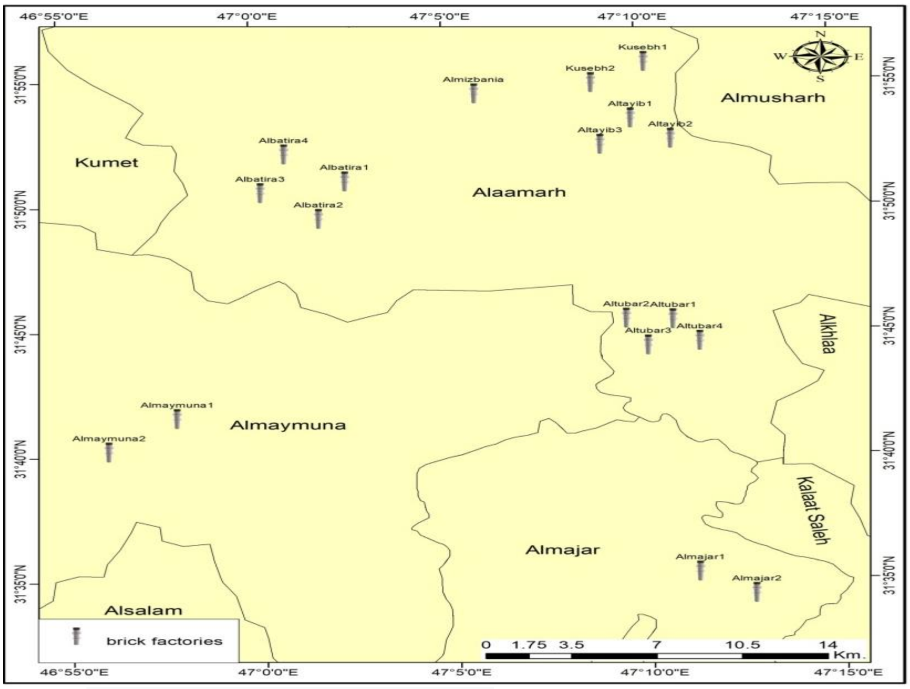

Figure (2):Map of the administrative divisions of the study area (Misan Province) showing locations of brick factories.

2.3 Analysis using KPA technique:

The Kinetic Phosphorescence Analyzer (KPA-11) at Radiation Protection Center (RPC), Ministry of Environment-Baghdad, was used for measuring uranium concentrations in the brick samples.KPA-11 provides a rapid, precise and accurate determination of uranium concentration in aqueous solutions. It is a sensitive and selective analytical technique, with low detection limits (is about 0.01ng·mL^{-1}) [10, 11].The main characteristics of the KPA are showed in Table 1 [8].

Table 1: The main characteristics ofthe KPA-11[8].

Parameter	Laser KPA
Selectivity	Tunable for U, Eu, Sm. Isotopes analysis
Laser source	Nitrogen laser with 337 nm wavelength
Pulse duration	3 nsec
Repetition rate	20 pulses/sec
Emission wavelength	515 nm for U
Pulse power	120 J
Buffer	Uraplex
Sample volume	1 mL
MDL	0.01 ng·mL^{-1} (0.13 mBq·L^{-1})
Precision (RSD)	(1% - 3%) at U > 0.01 ng·mL^{-1}
	(7% - 10%) at U < 0.01 ng·mL^{-1}
Analysis range	0.01 µg·L^{-1} - 5 mg·L^{-1}
Data processing	Computer software

2.4 Uranium Standard Solutions for KPA-11

Uranium standard solutions were prepared using Uranium Octoxide(U_3O_8). Firstly, a stock standard solution of1000 mg·L^{-1} (1000 ppm) was prepared by dissolving117.9 mg of U_3O_8 in 100 mL of 0.82 M nitric acid (HNO_3) in volumetric flask. To construct the calibration curve for kinetic phosphorescence, analysis series of calibration standard were prepared to cover a wide range of uranium concentration which is expected in brick samples. Uranium concentrations in the series of standards were 0.5, 1, 2, 3, 4, 5, 7, 8,and10 µg·L^{-1}.This set of standards was used to construct the calibration curve. In addition, background measurements, as in calibration, were performed using nine calibration standard solutions for each analytical range, ranging in concentration from the detection limit up to 10 µg·L^{-1}. A blank sample of 0.82 M HNO_3

was used to determine the background and reagent uranium concentration. The blanks phosphorescence intensity was subtracted from all KPA measurements [12].

3. Results and Discussion

The result of uranium concentrations, activitiesofuranium-238,uranium-235,and annual effective dose on the public for brick samples of Misan Province are shown in Table 2.

Uranium Concentration (UC) in the sample was calculated using the equation (1) [8]:

$$lnU^{*}t = lnU^{*}O - \left(K_P - K_q\right)t \qquad (1)$$

Where, U_i :population of excited uranium ions at time i = t or 0, k_p is rate constant for phosphorescent decay, and k_q is rate constant for all other relaxation processes. The KPA-11 is controlled by KPA Win software operating under Windows for automatic calculation of uranium concentration [8].

The uranium concentrations in brick samples ranged from 0.692 mg/kg (S_6) (Altubar factory) to 46.481 mg/kg (S_{12}) (Altayib factory)with a mean value of 9.403 mg/kg.

By measuring the uranium concentrations, the activities ofuranium-238,uranium-235can be calculated for these isotopes by knowing the specific activity and the mass fraction of them (presented in Table 3)[13], by using the equation (2) [13]:

$$A\left(\frac{Bq}{kg}\right) = UC\left(\frac{\mathrm{mg}}{\mathrm{kg}}\right) \times I.A.M(\%) \times S.P.A\left(\frac{Bq}{mg}\right) \qquad (2)$$

Where, A: is activity, UC: is Natural Uranium concentration, I.A.M: is the isotopic abundance (%) by mass fraction, S.P.A: is the specific activity.

The activity of uranium-238 in brick samples ranged from 8.545 Bq/kg (S_6)(Altubar factory) to 574.025 Bq/kg (S_{12}) (Altayib factory) with a mean value of 116.126 Bq/kg, and the activity of uranium-235 ranged from 0.398 Bq/kg(S_6) (Altubar factory) to 26.773 Bq/kg (S_{12})(Altayib factory) with a mean value of 5.416 Bq/kg.

The annual effective dose H_E (mSv/y) on the public can be calculate using RESRAD program [14].The annual effective dose on the public in brick samples ranged from 0.0000052 mSv/y (S_6)(Altubar factory) to 0.00162 mSv/y (S_{12}) (Altayib factory)with a mean of 0.00039 mSv/y.

The results (table 2) showed that, for Misan Province, the values of the natural uranium concentrations and activities ofuranium-238,uranium-235 in all brick samples have various rates.

The annual effective dose of gamma rays that resulted from the dissolution of uranium isotopes in brick samples were less than the total gamma rays dose which is 0.5 mSv/y,That is part of the total average annual doses of radiation from all natural sources of radiation,2.4 mSv/y (IAEA 2004) [16].

Table (2): Results of Natural Uranium Concentration, Activities of Uranium -238,Uranium-235,and Annual Effective Dose H_E (mSv/y) in Brick Samples measured within this work.

Code of Brick Samples	Location	Natural Uranium Concentration (mg/kg)	Activity of U-238 (Bq/kg)	Activity of U-235(Bq/kg)	Annual Effective Dose H_E (mSv/y) Unat
S_1	Almizbania	27.718	342.308	15.965	0.00021
S_2	Almaymuna	4.746	58.611	2.733	0.000073
S_3		5.104	63.032	2.939	0.000075
S_4	Almajar	1.870	23.093	1.077	0.000042
S_5		7.652	94.499	4.407	0.000084
S_6	Altubar	0.692	8.545	0.398	0.0000052
S_7		7.284	89.955	4.195	0.0000823
S_8		4.423	54.622	2.547	0.00041
S_9		3.885	47.978	2.237	0.00029
S_{10}	Kasiba	1.827	22.562	1.052	0.000207
S_{11}		5.439	67.169	3.132	0.000611
S_{12}	Altayib	46.481	574.025	26.773	0.00162
S_{13}		4.255	52.547	2.450	0.00063
S_{14}		2.204	27.218	1.269	0.00034
S_{15}	Albatira	2.107	26.020	1.213	0.00031
S_{16}		32.144	396.968	18.514	0.00094
S_{17}		2.668	32.948	1.536	0.00032
S_{18}		8.758	108.158	5.044	0.00079
Mean value		9.403	116.126	5.416	0.000391

Table 3. Radioactive properties of Natural Uranium Isotopes [13].

Isotope	Isotope Half-life (year)	Specific activity (Bq/mg)	Isotopic abundance (%)	
			By mass	By activity
$^{238}_{92}U$	4.51×10^9	12.44	99.2745	48.2
$^{235}_{92}U$	7.1×10^8	80	0.72	2.2
$^{234}_{92}U$	2.47×10^5	230.700	0.0055	49.6

4. Conclusions

The final results obtained indicates that the uranium concentrations in brick samples of the studied area overall have various rates and hence the annual effective dose of gamma rays that resulted from the dissolution of uranium isotopes in brick samples were less than the total gamma rays dose which is 0.5 mSv/y ,That is part of the total average annual doses of radiation from all natural sources of radiation 2.4 mSv/y (IAEA 2004). Therefore, it can be concluded that the brick samples of the area under the study, i.e. Misan Province, are radioactively safe to be used as construction materials and does not impose any serious threat to the health of locals. This study can be used as a reference for more extensive studies within the same subject in future.

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