

# Analysis of Radioactive Elements in Coal and Combustion Slag from Kangal Lignite Coal-Burning Thermal Power Plant

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## Abstract

**K**angal thermal power plant in Sivas Province of Turkey utilize about seven million tons per year of low-grade quality lignite coal fuel. During lignite burning, huge amounts of fly ash, bottom ash, and slag are produced as residues or by-products. In this study, the concentrations of uranium (U), thorium (Th), and radioactive potassium (<sup>40</sup>K) in lignite coal and slag samples were analysed using an energy dispersive X-ray fluorescence spectrometer. Enrichment ratio (ER), enrichment factor (EF), and radiogenic heat production (RHP) were also determined. Results revealed average concentrations of U, Th and <sup>40</sup>K for coal of 21.8-, 2.3- and 0.2-mg kg<sup>-1</sup> and slags 45.8-, 10.6- and 0.7- mg kg<sup>-1</sup>, respectively. The estimated values of ER for U, Th and <sup>40</sup>K were found as 2, 8 and 3, respectively. Based on EF results, slag samples contained a very high enrichment of uranium. The average estimated values of RHP for coal and slag samples were 2.6- and 7.0-μW/m<sup>3</sup>, respectively.

**Keywords:** *Coal, Slag, Enrichment Ratio, Enrichment Factor, Radiogenic Heat Production*

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## I. INTRODUCTION

Coal (anthracite, bituminous, sub-bituminous, and lignite) is a solid and carbon-rich organic rock found in the stratified sedimentary beds, which is a widely used as a fossil fuel for generation of electricity. Based on the International Energy Agency (IEA) Coal 2021 report, the global consumption of coal of about 7.91 billion tons in 2021 is projected to reach 8.03 billion tons in 2022 [1]. Turkey is largely dependent on fossil fuel as its main source of energy, accounting for about 83% of the total primary energy supply in the country [2]. Because coal is the primary raw materials, there is a growing interest to develop its coal reserves in the country to reduce dependency on imported coal [2]. Coal, especially lignite, continues to be one of Turkey's most important primary energy sources for electricity generation, although this type of coal can be considered as a low-grade quality due to its low calorific value and high sulphur, dust, and ash contents (16 to 50%) [3]. According to the General Directorate of Mineral Research and Exploration, Turkey has an estimated coal deposits of about 19.14 billion tons of lignite and 1.6 billion tons of hard coal reserves [2]. As of January 2020, the shares of energy generated from incineration of coal accounted for about 22% of the total electricity generation capacity. The installed electricity capacity using domestic coal was about 11,317 megawatts (MW) (10 101 MW lignite, 811 MW hard coal, 405 MW asphaltite), representing about 12.4% of the total installed power. This is higher than the generated power from the imported coal, which is about 8,967 MW.

Major, minor, and trace elements (S, Si, Al, Mg, K, Fe, Mn, Zn, Cd, As, Pb, Hg, Ti, V, Th, U, etc.), which amounts are highly variable depending on coal types, composition, conditions during combustion, and burning efficiency, are released during the incineration process. The combustion of coal to generate electricity in thermal power plants (TPPs) also generates solid wastes (residues or by-products), such as fly ash, bottom ash, slag, acid emissions (SO<sub>x</sub> and NO<sub>x</sub>) into the atmosphere. These coal residues, which may contain radioactive materials (U, Th, and 40K) and toxic elements, (As, Hg, Pb, Cr, Cd, Co, Ni, Zn, etc.) pollute the environmental and pose hazard to human health. Ionizing radiation ( $\alpha$ ,  $\beta$ ,

and  $\gamma$  rays) emitted from these radionuclides in coal and slag may lead to radiological exposure of workers and the local population. Workers and people living near coal beds and slag heaps may be exposed to external irradiation caused by  $\gamma$ - rays. These people may also be exposed to internal exposure caused by  $\alpha$  and  $\beta$  particles due to inhalation of the radioactive noble gas radon ( $^{222}\text{Rn}$ ), which has short-lived decay products [3]. Due to the health hazards posed by the above-mentioned pollutants, it is important to analyse the concentrations of these radionuclides in coal and their residues, which are important data in evaluation of workers and public's exposure to radiation and formulation of guidelines for the use and management of these materials [3,5-14]. This study investigates the elemental concentration of U, Th, and  $^{40}\text{K}$  in lignite coal and slag samples collected from Kangal TPP, which were analysed using an energy dispersive X-ray fluorescence (EDXRF) spectrometer, and then determined the enrichment ratio, enrichment factor, and radiogenic heat production.

## II. MATERIALS AND METHODS

### Study Location

Kangal TPP is situated 25 km southwest of Kangal town of Sivas province of Turkey (Fig. 1). The Kangal TPP has a total installed generating capacity of 457 MW (2x150 MW + 1x157 MW) and has been operating since 1989 [3]. In 2019, about 6.9 million tons of lignite coals were used in Kangal TPP and consequently about 169, 353 tons of slag residues were produced as by-products [13]. These slag residues, which are cooled in a water-filled vessel, are collected in silos and then transported to the storage area by belts in the closed gallery [15].

### Collection and Preparation of Samples

Samples of coal and slag (10 samples each) were collected from the Kangal lignite basin and slag storage area, respectively.

The samples were ground and powdered to make them fit the calibrated powder geometry in the EDXRF spectrometry. A maximum of 5 grams of each sample was taken for elemental analysis.

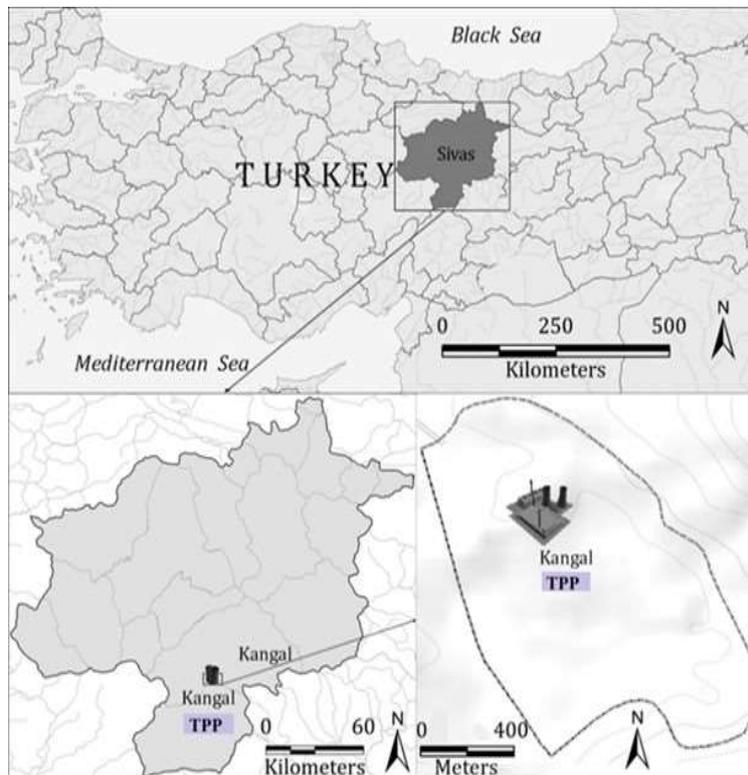


Figure1. Location of the Kangal /Sivas-Turkey TPP [16].

## Enrichment Factor

The enrichment factor (EF) values consist of five classifications as given in table -1 [16].

Table 1. EF Values Consist of Five Classifications

$EF < 2$	Deficiency to minimal enrichment
$2 \leq EF < 5$	Moderate enrichment
$5 \leq EF < 20$	Significant enrichment
$20 \leq EF < 40$	Very high enrichment
$EF \geq 40$	Extremely enrichment

## Elemental Analysis

The analysis of U, Th, and K in the coal and slag samples was carried out using the EDXRF spectrometer (Spectro Xepos, Ametek). Detailed information about the EDXRF spectrometer was given in detail in the study by Turhan et al. [16]. The EDXRF spectrometer was equipped with a thick binary Pd/Co end-window tube (50 W, 60 kV) and a Peltier cooled Si drift detector [16]. The spectrometer utilizes the “standardless” calibration based on the Fundamental Parameters (FP) method. Analyses of the samples placed in the automatic sampler were carried out by counting once for two hours.

## III. RESULTS AND DISCUSSION

### Results

The elemental concentrations of U, Th, K, and  $^{40}\text{K}$  in lignite coal samples are summarized below.

Table 2. Elemental Concentrations of U, Th, K, and  $^{40}\text{K}$  of Lignite Coal Samples

Samples	Elemental concentration (mg/kg)									
	Th			U			K			$^{40}\text{K}$
C1	3.3	±	0.3	25.2	±	0.3	1863	±	11	0.22
C2	0.9	±	0.2	16.7	±	0.3	1619	±	10	0.19
C3	0.7	±	0.2	19.9	±	0.3	1860	±	11	0.22
C4	0.6	±	0.2	14.8	±	0.3	1386	±	9	0.16
C5	1.3	±	0.2	25.7	±	0.3	1963	±	10	0.23
C6	1.0	±	0.2	24.9	±	0.3	2120	±	11	0.25
C7	3.4	±	0.3	18.3	±	0.3	1507	±	11	0.18
C8	4.0	±	0.3	34.5	±	0.4	2018	±	11	0.24
C9	3.0	±	0.3	17.9	±	0.3	1274	±	11	0.15
C10	3.3	±	0.3	19.6	±	0.3	1606	±	10	0.19
Average	2.2			21.8			1722			0.20
Min	0.6			14.8			1274			0.15
Max	4.0			34.5			2120			0.25

### Enrichment Ratio and Enrichment Factor

Enrichment ratio was defined as the ratio of the concentration of the radionuclide in slag to its concentration in coal. The enrichment factor (EF) to Earth’s crustal average was used to eval-

uate the degree of metal or radioactive element pollution and estimated using the following formula [16]:

$$EF = \frac{\left( \frac{C_n}{C_{Ref}} \right)_{\text{Sample}}}{\left( \frac{C_n}{C_{Ref}} \right)_{\text{Background}}}$$

where  $C_n$  is the concentration of element and  $C_{Ref}$  is the concentration of a reference element in the examined sample. Based on reported literature, the enrichment factor is estimated by using one of the elements Al, Ca, Sc, Ti, Mn, Fe, Sr, and Zr as reference elements [14]. In this study, the enrichment factor was estimated with reference to Fe and Yaroshevsky's Earth crust composition [17]. Elemental concentrations of U, Th, K, and  $^{40}\text{K}$  were analyzed in the slag samples and results were summarized below (Table 3).

**Table 3. Elemental Concentrations of U, Th, K, and  $^{40}\text{K}$  a in the Slag Samples**

Samples	Elemental concentration (mg/kg)									
	Th			U			K			$^{40}\text{K}$
S1	10.5	±	0.4	33.9	±	0.5	5832	±	26	0.68
S2	10.5	±	0.4	49.3	±	0.6	5611	±	20	0.66
S3	11.4	±	0.4	52.2	±	0.6	5626	±	21	0.66
S4	10.8	±	0.4	44.0	±	0.6	5621	±	20	0.66
S5	11.6	±	0.4	46.8	±	0.6	6190	±	23	0.72
S6	10.0	±	0.4	45.3	±	0.6	5312	±	20	0.62
S7	9.9	±	0.4	47.0	±	0.5	5052	±	19	0.59
S8	9.5	±	0.4	41.5	±	0.5	4966	±	19	0.58
S9	11.5	±	0.4	50.0	±	0.6	7122	±	27	0.83
S10	10.1	±	0.4	48.0	±	0.6	5263	±	20	0.62
Average	10.6			45.8			5660			0.66
Min	9.5			33.9			4966			0.58
Max	11.6			52.2			7122			0.83

## Radiogenic Heat Production

Radioactive decays of natural radionuclides in the radioactive series of  $^{238}\text{U}$  and  $^{232}\text{Th}$  and  $^{40}\text{K}$  remains as the largest source of internal heat besides the Earth's heat content [18]. Significant proportions of ionizing radiation emitted in these radioactive ele-

ments are completely converted into heat within minerals and rocks during radioactive decay. In general, the total radiogenic heat production of a rock is the sum of the individual contributions of by U, Th, and K. The radiogenic heat production (RHP in terms of  $\mu\text{W}/\text{m}^3$ ) was estimated using the following formula [18]:

$$\text{RHP} = 10^{-5} \cdot \rho \cdot (9.52 \cdot C_U + 2.56 \cdot C_{\text{Th}} + 3.48 \cdot C_K) \quad (2)$$

where  $\rho$  is the density of the samples, in which the average density of lignite coal and slag is about 1200 and 1500  $\text{kg}/\text{m}^3$ , respectively, and  $C_U$ ,  $C_{\text{Th}}$  and  $C_K$  are the concentrations of uranium expressed in  $\text{mg}/\text{kg}$ , thorium  $\text{mg}/\text{kg}$ , and potassium (%), respectively.

Results for the enrichment factor estimated for U, Th, and K in lignite coal and slag samples is summarized below (Table 4). The relative radionuclide elemental abundance in lignite coal and slag samples is found to be in the order of  $U > \text{Th} > {}^{40}\text{K}$ .

**Table 4. Enrichment Factor Estimated for U, Th, and K in Lignite Coal and Slag Samples.**

Material	Radionuclide	EF		
		Average	Min	Max
Lignite	U	29.80	23.00	40.50
	Th	0.60	0.20	1.10
	K	0.24	0.20	0.27
Slag	U	24.40	18.70	27.20
	Th	1.09	1.03	1.14
	K	0.30	0.29	0.35

## Discussion

The concentrations of U in the lignite coal and slag samples range from 14.8 to 34.5  $\text{mg kg}^{-1}$  with an average value of 21.8  $\text{mg kg}^{-1}$  and 33.9- to 52.2  $\text{mg kg}^{-1}$  with an average value of 45.8  $\text{mg kg}^{-1}$ , respectively (Tables 2 and 3). The average concentration of U in the lignite coal samples is approximately 9 times higher than the average U of 2.5  $\text{mg kg}^{-1}$  in the earth's crust whereas

the average concentration of U in the slag samples is approximately 18 times higher than the earth's crust average [17]. The average enrichment ratio (ER) for U was calculated as 8. The U concentration was also highly enriched in the lignite slag than coal. From Table 4, the EF values estimated for U in the lignite coal and slag samples vary from 23 to 41 with an average value of 30 and 19 to 27 with an average value of 24, respectively. The average values of EF denote very high enrichment of U in the lignite coal and slag samples.

The concentrations of Th in the lignite coal and slag samples also vary from 0.6 to 4.0 mg kg<sup>-1</sup> with an average value of 2.2 mg kg<sup>-1</sup>, 9.5 to 11.6 mg kg<sup>-1</sup>, and 10.6 mg kg<sup>-1</sup>, respectively. The average concentration of Th in the lignite coal samples is approximately 6 times lower than the average Th of 13 mg kg<sup>-1</sup> in the earth's crust whereas the average concentration of Th in the slag samples is approximately 1.2 times lower than the earth's crust average [17]. The average ER for Th was calculated as 2. The Th concentration in the lignite was highly enriched compared to coal. The EF values estimated for Th in the lignite coal and slag samples vary from 0.2 to 1.1 with average values of 0.6, 1.03 to 1.14, and 1.09, respectively. The average values of EF denote deficiency to minimal enrichment of Th in the lignite coal and slag samples.

The concentrations of K in the lignite coal and slag samples varied from 1274 to 2120 mg kg<sup>-1</sup> with average values of 1722 mg kg<sup>-1</sup>, 4966- to 7122-mg kg<sup>-1</sup>, and 5660 mg kg<sup>-1</sup>, respectively. The average concentration of K in the lignite coal samples is approximately 14.5 times lower than the average K value of 25000 mg kg<sup>-1</sup> in the earth's crust, whereas the average concentration of K in the slag samples is approximately 4.4 times lower than the earth's crust average [17]. The average ER for K was calculated as 3. The K concentration was enriched in the lignite slag than coal. The <sup>40</sup>K is a natural isotope with an isotopic abundance of 0.0117 % in natural potassium. The concentrations of <sup>40</sup>K in the lignite coal and slag samples varied from 0.15 to 0.25 mg kg<sup>-1</sup> with average values of 0.20 mg kg<sup>-1</sup>, 0.58 to 0.83 mg kg<sup>-1</sup>, and 0.66 mg kg<sup>-1</sup>, respectively. The EF values estimated for <sup>40</sup>K in the lignite coal and slag samples varied from 0.20 to 0.27 with average values of 0.24, 0.29 to

0.35, and 0.3, respectively. The average values of EF denote deficiency to minimal enrichment of K in the lignite coal and slag samples.

The values of RHP estimated for the lignite coal and slag samples varied from 1.7 to 4.1  $\mu\text{W m}^{-3}$  with average values of 2.6  $\mu\text{W/m}^3$ , 5.3 to 7.9  $\mu\text{W m}^{-3}$ , and 7.0  $\mu\text{W/m}^3$ , respectively. The average RHP value of the lignite coal is 3 times higher than the average Earth's crust RHP value of 0.9  $\mu\text{W/m}^3$ , while the average RHP value of the slag is 8 times higher than the average Earth's crust [19]

#### IV. CONCLUSION AND RECOMMENDATION

In this study, the elemental concentrations of uranium, thorium, and potassium in lignite coal and its residue (slag) samples collected from Kangal TPP were analysed in detail by the EDXRF spectrometer. The EDXRF technique, which is a non-destructive method, is fast and fully suitable for simultaneous quantitative radionuclide analysis of coal and slag. Based on values of the enrichment ratios, the concentrations of uranium, thorium, and potassium were higher in slag samples than lignite coal. The enrichment factor of uranium was also very high in coal and slag than the average Earth's crustal, which should raise concerns because of the associated health hazards of these elements.

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