Journal of İstanbul Kültür University
2004/1 pp. 39-43

Natural Radioactivity and EDXRF Analysis of Ashes from Coal Power Plants of Turkey*

Tanıl Akyüz[1], Sevim Akyüz[2], Ahmet Varinlioğlu[3] and Abdullah Köse[3]

Abstract

Natural radioactivity and the chemical composition of ten ash samples collected from six coal power plants of Turkey have been analysed. Gamma-isotopic analysis indicated that all the samples contain Ra-226, U-238 and Th-232. But, K-40 and Cs-134 have not been found in the samples in the detection limits. On the other hand, Cs-137 was found only in the ash sample taken from Yatağan coal power plant (603±15 Bq/kg). Mineralogical analysis of the samples showed that they contained significant amounts of clays and glass. Less amounts of quartz, feldspar and magnetite minerals exist in the samples. Major and minor element analyses of the ash samples were done by X-Ray Fluorescence, XRF/EDXRF, technique. The natural radioactivity of an ash sample taken from the electro-filter of the Ambarlı fuel oil power plant was also analysed for comparison and it was found that sample contained the least amount of U-238, Th-232 and Ra-226.

Özet

Türkiye'nin altı termik santralına ait on adet ucucu kül örneği doğal radyoaktivite ve kimyasal içerikleri yönünden incelenmiştir. Gama izotopik analiz sonuçları, örneklerin hepsinin Ra-226, U-238 ve Th-232 isotoplarını içerdiğini göstermiştir. Fakat K-40 ve Cs-134 isotopları, sistemin ölçme sınırları içersinde saptanamamıştır. Diğer taraftan Cs-137 isotopu, sadece Yatağan termik santralının kül örneğinde bulunmuştur (603±15 Bq/kg). Minerolojik analiz, örneklerin kil ve camsı maddeleri çok miktarda, feldispat, magnetit minerallerini ise az miktarda bulundurduklarını göstermiştir. Uçucu kül örneklerinin, major ve minor elementlerinin analizleri XRF/EDXRF teknikleri kullanılarak yapılmıştır. Ambarlı fuel-oil termik santralının elektro-filtresinden alınan bir kül örneği de, karşılaştırma amacı ile analizlenmiş ve Ra-226, U-238 ve Th-232 isotop içeriği, diğerlerine göre en düşük miktarda bulunmuştur.

Key Words: *Radionuclides, Gamma Isotopic Analysis, Coal Power Plants, Coal Ash, Lignite.*

Introduction

Coal is one of the most impure of fuels. Its impurities range from trace quantities of many metals, including uranium and thorium, too much larger quantities of aluminium and iron to still larger quantities of impurities such as sulphur. Products of coal combustion include the oxides of carbon, nitrogen and sulphur, carcinogenic and mutagenic substances; and recoverable minerals of commercial value, including nuclear fuels naturally occurring in coal.

In coal power plants, generally low lignite coal, which has no commercial values, is used. It is known that some low quality lignite coals contain uranium, thorium and some other radionuclides in significant amounts [1-2]. The low-grade lignite, in the coal power plant, is pulverized and during the combustion process, the uranium, bounded to lignite organically, is enriched in the inorganic matrix, i.e. it is accumulated in the ash.

[1] *Department of Mathematics, İstanbul Kültür University 34191 Şirinevler / İstanbul,*
[2] *Physics Department, İstanbul University 34118 Vezneciler / İstanbul,*
[3] *Çekmece Nuclear Research and Training Center, P.K 1, 34831 Havalimanı, İstanbul.*

Therefore, there are uranium, thorium and some other radioactive elements in the ashes, arising from combustion by-products. Radioactive elements released in coal ash and exhaust produced by coal combustion contain fissionable fuels and much larger quantities neutrons, including those generated in the air by bombardment of oxygen, nitrogen and other nuclei with cosmic rays; such fissionable and fertile materials can be recovered from coal ash using known technologies. In order to decrease the environmental pollution due to the volatile ashes the plants usually utilize electro-filters or other filters. Very fine particular ash collected fly ash and the larger sized ash particles, which fall towards the bottom of the chimney is called bottom ash. These ashes are deposited outside the fabrication areas. They form ash hills, by time. The deposited ashes can bee dangerous for environment, since they contain toxic and radioactive materials. On the other hand there are some studies in order to utilize these ashes economically, e.g. using the ash together with cement in concrete or an absorber of organic parts of the waste water [2-4]. For these reasons, it is found to be interesting to investigate natural radioactivity and chemical composition of the ashes obtained from coal power plants of Turkey. In this study, ten ash samples taken from electro-filter and bottom products of boilers or chimneys of Seyit Ömer, Soma, Çayırhan, Yatağan and Afşin-Elbistan coal power plants of Turkey. An ash sample taken from the electro-filter of the Ambarlı fuel oil power plant was also analysed in order to compare the isotopic composition of ash samples of coal power plants with that of [5].

Material And Method

The analysis has been done on ten ash samples. Six of them were collected from the electro-filters; four were collected from the bottom products of the boilers or chimneys of the coal power plants of Turkey. The samples, carried by large bags were divided into small portions by homogenisations, grounded to fine particles (200 mesh) and dried at 105°C for 24 h. The mineralogical investigation was done by an Olympus polarizing light microscope. Qualitative analysis of the ash samples taken from electro-filters of the coal power plants was carried out with X-ray fluorescence methods (XRF and EDXRF) using General Electric SPG-7, XRF and Kevex 0700, EDXRF Spectrometers.

For gamma-isotopic analysis, approximately 70-80 g of the samples were put into plastic cups, then sealed and kept for 40 days for radioactive equilibrium. The gamma isotopic analysis was carried out by using high-purity Germanium detectors connected with standard fast electronics. The resolution (FWHM) was of the spectrometer was done with a solid nuclide mixture of gamma reference source. The peak analysis of all counted samples was made by using spectran AT software at IBM personal computer. At least 150.000-250.000 seconds of counting time was applied to all samples to obtain better statistics, so that the detection limit was about 1 Bq/kg (27 pCi/kg).

Resuls And Dicussion

Mineralogical analysis of the ash samples indicated that they contained considerable amounts of glass, produced as by-product and clays. They also contained less amounts of quartz, feldspar and magnetite minerals. On the other hand, X-ray fluorescence analysis showed that samples had the following elements: V, Mn, Na, Al,

Mg, Si, Ca, Ti, Cr, Fe, Ni, Cu, Sr, Mo, Pb, Zn, Sb, Zr, Ba, Y, La, Ce, Nd and Sm. Chemical analysis results of six samples taken from the electro filters of coal powers plants were given in Table 1. Investigation of the results has shown that the chemical structures of ash samples were quite similar to those of silicate minerals.

The main sources of radiation released from coal combustion include not only uranium and thorium but also daughter products produced by the decay of these isotopes, such as radium, radon, polonium, bismuth and lead. Although not a decay product, naturally occurring radioactive K-40 is also a significant contributor. The results of the gamma-isotopic analysis are tabulated in Table 2. The results indicated that ash samples studied contained Ra-226, U-238, Cs-137 and Th-232 isotopes in different amounts. The results are in good agreement with the previous resuts [5-7]. As seen in Table 2, the highest amount of U-238 (140 mg/kg), Th (42 mg/kg) and Ra-226 (123 Bq/kg) are found in the ash samples taken from Yatağan power plant. In all the samples, K-40 and Cs-134 (Bq/kg) are below the detection limit (DL); DL= 1 Bq/kg (27 pCi/kg). Th-232 and Ra-226 are found to be in the 0.9-47.5 mg/kg and 15-122 Bq/kg levels, respectively. On the other hand, the gamma-isotopic analysis of the ash sample taken from the fuel oil power plant of Ambarlı is also given at the Table 2, as can be seen, it has the lowest amounts of radioactive isotopes among the others.

Conclusion

The radioactive and toxicological of ash samples of Turkey are important for environmental pollution, since the ashes are deposited outside the fabrication areas and widely used together with cement in concrete. Therefore, natural radioactivity and chemical composition of ash samples obtained from five coal and one fuel-oil power plants of Turkey have been investigated. It is found that all the samples have Ra-226, U-238 and Th-232, however the ash sample from the fuel oil plant is found to have the lowest amounts of radioactive isotopes in comparison to those of ashes from coal power plants. The highest amount of radioactive isotope content is found in the ash samples from the Yatağan coal power plant; Ra-226 (123 Bq/kg), U-238 (140 mg/kg), Th-232 (48 mg/kg).

References

[1] Derman D., Gönen N.(1983) *"Uranium Recovery from fly ashes of Some Coal Power Plants of Turkey"*, Technical Report, Mineral Research and Exploration Institute of Turkey, No: 82-70.

[2] Yazıcıoğlu E., (1982), *"Türkiye Uçucu Küllerinin Özellikleri ve Kullanılma Özellikleri"*, Technical Report, Electrical Power Resources Survey and Development Administration of Turkey, Ankara No : 21.

[3] Capp J.P. (1978) *"Power Plant Fly Ash Use in the Eastern USA"* In F.W. Schaller and P.W. Sutton (Eds), Reclamation of Drasticall Disturbed Lands, Am. Chem. Soc. Argon., Madison, N.Y, p. 339-354.

[4] Flues M, Moraes V, Mazzilli BP (2002) *"The influence of a coal-fired power plant opera operation on radionuclide concentration in soil"* Journal Environmental Radioactivity, 63, p.285-294.

[5] Bem E.M., Bem H., Wieczorkowski (1998), *"Studies of Radionuclide Concentrations in Surface Soil in and Around Fly Ash Disposal Sites"* The Science of the Total Environment, 220, p.215-222.

[6] Theis T.L. and Wirth J.L. (1977) *"Sorptive Behavior of Trace Metals on Fly ash in Aqueous Systems"* Environ. Sci. Technol. 11, 1096-1100.

[7] UNSCEAR Report, (1988), *"Sources, Effects and Risks of Ionizing Radiation"* Report to the General assembly, United National Pub. , New York, p.109,

Table 1. XRF/EDXRF analysis of the ashes from electro filters coal power plants (%).

Name/Element	SiO_2	Al_2O_3	TiO_2	ΣFe_2O_3	CaO	MgO	Na_2O	K_2O	MnO
Seyit Omer	56.0	22.5	0.6	8.0	9.0	0.7	0.5	0.5	0.2
Soma	46.5	27.5	1.2	6.2	13.5	1.9	0.6	0.9	0.1
Cayirhan	46.0	23.5	1.2	8.5	13.0	2.5	1.5	1.8	0.2
Yatagan	55.0	24.0	1.5	7.7	9.1	0.6	0.4	0.5	0.2
Afşin-Elbistan	51.0	26.0	0.6	7.0	9.4	1.0	0.4	0.5	0.2
Name/Element	Cr	Ni	Cu	V	Co	Mo	SrO	BaO	
Seyit Ömer	0.1	0.05	0.01	0.02	0.01	0.01	0.05	0.10	
Soma	0.1	0.1	0.02	0.04	0.01	0.01	0.10	0.10	
Çayırhan	0.15	0.05	0.01	0.02	0.01	0.01	0.05	0.10	
Yatagan	0.1	0.1	0.02	0.04	0.01	0.01	0.10	0.20	
Afşin-Elbistan	0.15	0.1	0.02	0.04	0.01	0.01	0.10	0.20	

The uncertainty is found to be less than ±10%, which has been calculated as the standard deviation for ten measurements.

Table 2. The results of Gamma- Isotopic analysis of the ash samples.

	Ra-226 (Bq/kg)	U-238 (mg/kg)	Cs-137 (Bq/kg)	Th-232 (mg/kg)	K-40 (Bq/kg)	Cs-134 (Bq/kg)
Seyit Ömer						
	15 ± 2	8.9 ± 1.1	BDL	17.4 ± 1.8	BDL	BDL
(*)						
Soma						
	33 ± 3	32.5 ± 1.6	BDL	41.9 ± 2.6	BDL	BDL
(*)						
Çayırhan						
	26 ± 2	12.2 ± 1.1	BDL	19.4 ± 2.1	BDL	BDL
(*)	22 ± 2	11.8 ± 1.3		11.8 ± 1.3		
(**)						
Yatağan						
	122 ± 7	139.8 ±	603 ± 50	47.5 ± 5.4	BDL	BDL
(*)	123 ± 8	14	15 ± 2	41.8 ± 6.4		
		133.3±				
(**)		13.3				
Afşin-						
Elbistan	70 ± 7	74.0 ± 2.5	BDL	13.3 ± 1.5	BDL	BDL
	74 ± 7	79.5 ± 3.3		4.8 ± 1.4		
(*)	52 ± 4	46.5 ± 2.0		0.9 ± 0.2		
(**)						
(***)						
Ambarlı						
	7± 1	4.0 ± 0.5	BDL	5.7 ± 1.3	BDL	BDL
(*), (a)						

Samples obtained from electro-filters (*), bottom of boilers (**) and bottom of chimneys (***), BDL= Below detection limit, detection limit, Detection limit = 1 Bq/kg (27 pCi/kg). (a) Ash sample from the fuel oil power plant.