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Concentration of Natural Radionuclides in Coal and Its End Products in Steel Production

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ABSTRACT

Coal industry and natural radioactivity in coal ash was one of the first major beneficiaries of nucleonics control system (NCS) utilization. Coal as most earth materials contains low concentrations of ^{238}U and ^{232}Th and their radioactive progeny, also ^{40}K. Fly ash and slag are end products of coal combustion industries.

Burning of coal and other fossil fuels is one source of radiation exposure to naturally occurring radioactive materials (NORM) wastes, which is released to the environment. About one fourth of the produced coal is used in iron and steel production. In this work high resolution gamma spectrometry were applied to measure the concentration of different radionuclides in Bq kg^{-1} in coal ore, coke, limestone, iron ore, also ash and slag produced as waste products from blast furnaces. Concentration of different radionuclides in both ash and slag, measured in this work, show low contents, compared to the concentrations of ash and slag obtained as (NORM) wastes from coal fired power plant. Slag is used in Egypt for high way construction purposes, as embankment. It has concentrations of 90 Bq kg^{-1} for ^{234}Th, 73 Bq kg^{-1} for ^{226}Ra, 46 Bq kg^{-1} for ^{214}Pb and 41 Bq kg^{-1} for ^{214}Bi in uranium series, and 29 Bq kg^{1} for ^{228}Ac, 33 Bq kg^{-1} for ^{212}Bi and 24 Bq kg^{-1} ^{208}Tl in the thorium series as well as 97 Bq kg^{-1} for ^{40}K, so it will give slight increase of exposure compared to natural radiation.

INTRODUCTION

Coal is chosen a main raw material used in several industries, such as aluminum production, cement production, iron and steel industry and several other refining and extraction industries. Coal like most earth materials contains low concentrations of ^{238}U and ^{232}Th and their radioactive progeny.

Burning of coal and other fossil fuels is one source of radiation exposure from naturally occurring radioactive materials (NORM) wastes. Radioactive elements are released to the environment during fuel extraction and burning.

Although carbon is major element in coal, as many as 72 elements have been found in coal, e.g. Na, K, Ca, Al, Si, Fe, Ti, Li, Rb, Cr, Co, Cu, Ga, Ge, La, Ni, W and Zr(1). Coal is also important as the source of coke used in the steel industry, about one fourth of the coal produced every year is used for this purpose. Coke is burned in blast furnace to reduce iron ores, iron oxides to metallic iron. In blast furnace, basic open hearth, coke is used as a burner for preheating the combustion air(2).

Hematite is the main iron ore. Iron may also be incorporated into one of the clay minerals to form other ore or into a carbonate mineral. Iron usually combines with oxygen to form Fe_2O_3 (hematite), one of the most common iron ore oxides. Laterites contain high concentrations of uranium series elements and are used to increase the grade of the iron-bearing materials through iron –ore beneficiating plants.

Limestone is used as a flux, in steel industry, is added to render the impurities in the fusible raw materials and to combine with the impurities to facilitate their removal from the metal. Limestone as a flux is preferable owing to the presence of silica in iron ores and sulfur in coke (2). Limestone is a sedimentary rock composed largely of the mineral calcite, $CaCO_3$, which has been formed by either organic or inorganic processes. The carbonate rock, limestone, constitutes about 22 per cent of the sedimentary rocks exposed above sea level (1).

An accepted principal of engineering is to design a product for minimum cost, consistent with fulfilling the functional requirements of the product. Such approach neglects some important items to reduce the cost of the product to the user, such as the cost of disposing the waste products. In addition to the cost of a product to the user and producer, there is the cost to society as the consumption of raw materials and energy, also the impact of the product and waste products to the environment. Reuse of components and materials not only conserve raw materials such as the ore, also it conserves the energy required to extract the metal from the ore (3).

In the operation of the blast furnace, a blast of preheated air passes upward through a permeable bed of solid raw materials (figure 1). The furnace is charged with a mixture of liquid pig iron, solid pig iron, steel scrap, limestone (Ca CO3) as a flux, and some iron ore are usually used in the flux (2).

The complicated series of reactions that occur produce stack gases and fly ash at the top of the furnace, molten slag and pig iron at the bottom. In this work coal ore, coke (coal), limestone, iron ore, fly ash and slag were analyzed by high-resolution gamma spectrometry to measure concentrations of different natural radioactive nuclides in the different samples.

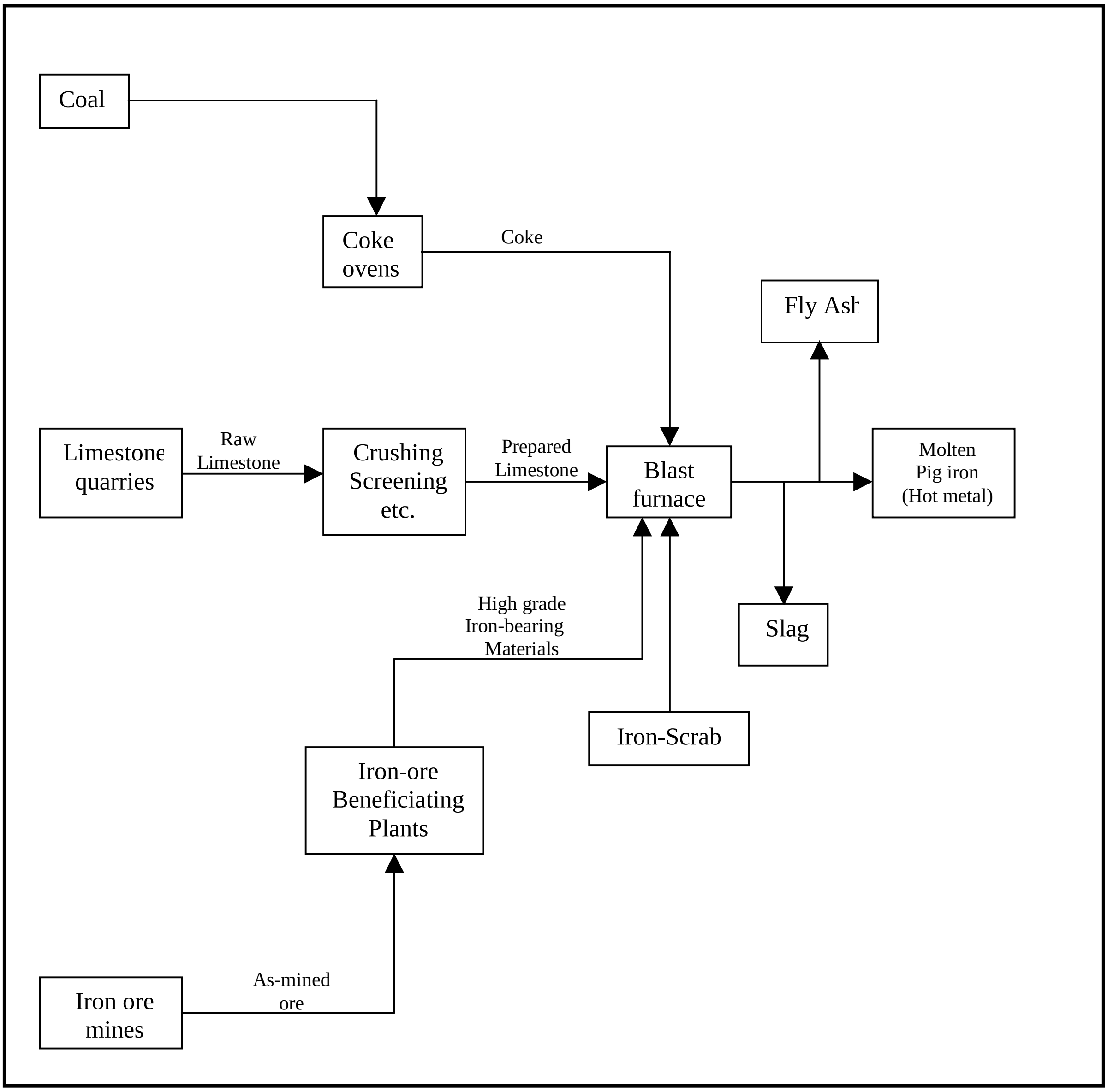

Figure1: Flow diagram of steel making processes [ASM 1978]

EXPERIMENTAL TECHNIQUES

This study was carried out on six samples representing the following materials, hematite, cool ore, coke, limestone, fly ash and slag.

The coal and coke samples were provided by the Egyptian Company for Coke and Batteries in Helwan, Cairo. The iron ore sample were collected from Bahria Oasis at the western desert, the limestone sample was obtained from El-Mokattam quarries Cairo, and the fly ash and waste slag were provided from a blast furnace used for steel production Samples were crashed and homogenized, then transferred to a 750 ml. or 500 ml. Marinelli beakers for gamma measurements. Samples were sealed for 8 weeks to allow the secular equilibrium in both the uranium and thorium series to be reached.

Table I Photon energies and their percentage per disintegration used for the measurements of concentrations for different radionuclides

Nuclide	Energy(keV)	Photon per disintegration %
U-238 series		
Th-232	63.3	3.8
R-226	186.1	3.3
Pb-214	295.1	19.2
	352.1	37.1
Bi-214	609.3	46.1
	768.4	5.1
	934.1	3.2
	1120.0	15.1
Th-232series		
Ac-228	209.4	4.1
	338.4	12.4
	462.1	4.6
	911.2	29.1
	966.6	23.2
Bi-212	727.3	6.7
	785.5	1.1
	1620.0	1.7
Tl-208	583.1	30.9
U-235series		
U-235	143.8	10.5
	163.3	4.7
	185.7	53.1
	205.3	4.7
K-40		
	1460.0	10.7

Environmental Measurements Laboratory, U.S Department of Energy (1990).

A hyper pure germanium gamma spectrometer system based on a HpGe coaxial detector from EG&G Ortec, were used for gamma measurements, the measurements were done for 6 to 10 hours. The HpGe detector has a FWHM of 1.9 keV at the 1.33 MeV ^{60}Co gamma transition, and relative efficiency of about 50%. The HpGe detector was shielded by a lead cylinder with fixed bottom and moving cover, and a concentric copper cylinder, to decrease the gamma background.

Efficiency calibration for the HpGe spectrometer system were explained in a recent publication(4).

Table 1 gives the energies used for the determination of the concentrations of different radionuclides, and their branching ratios. Concentrations were measured in Bg kg^{-1} for, ^{234} Th, ^{226} Ra, ^{214} Pb and ^{214} Bi in the U-238 series, ^{228}Ac, ^{212}Pb and ^{208}Tl in the Th-232 series, also that of ^{40}K.

Table II Concentrations for different radionuclides in Bq kg^{-1}

Material	U-238 series				Th-232 series			K-40	U- 235 series
	Th – 234	Ra – 226	Pb – 214	Bi – 214	Ac – 228	Bi – 212	Tl - 208	K-40	U- 235
Coal Ore	59.2 ± 10.1	25.6 ± 1.3	15.1 ± 0.3	14.8 ± 0.3	8.0 ± 0.4	8.5 ± 0.9	7.9 ± 0.3	30.4 ± 1.2	LLD
Coke	50.1 ± 6.5	25.4 ±1.3	13.7 ± 0.2	11.4 ± 0.3	8.2 ± 0.3	9.6 ± 0.6	8.5 ± 0.3	36.7 ± 1.1	LLD
Iron Ore (Hematite)	460 ± 14	245 ± 10	221 ± 11	212 ± 10	6 ± 4.5	3.2 ± 2.4	LLD	70.8 ± 1.6	10.9 ± 0.5
Limestone	121 ± 10	44.5 ± 3.5	32.5 ± 1.4	30.0 ± 2.2	LLD	LLD	LLD	52.3 ± 8.3	LLD
Fly Ash	41 ± 9.6	23 ± 1.2	17.5 ± 0.25	15.6 ± 0.54	9.5 ± 0.4	11.3 ± 0.84	8.5 ± 0.29	93.5 ± 2.0	LLD
Slag	89.5 ± 10.2	72.7 ± 1.9	46.6 ± 3.7	41 ± 1.4	29 ± 0.58	33.6 ± 1.3	23.9 ± 0.38	97.4 ± 1.75	LLD

RESULTS AND DISCUSSIONS

Table II represents concentrations of ^{234}Th, ^{226}Ra, ^{214}Pb and ^{214} Bi the uranium-238, radium-226 series, concentrations of ^{228}Ac, ^{212} Pb and ^{208}Tl in the thorium-232 series, as well as the ^{40}K concentrations. Concentrations were presented in Bqkg^{-1} for coal ore, coal iron ore, lime stone, fly ash and slag samples .

Concentrations for the individual nuclides in the coal ore and coal, and hematite samples show state of disequilibrium, in both the uranium-radium and thorium series, while Roeck et al (5) and Keller et al (6), considered that the activity concentrations of the different radionuclides in both series are in secular equilibrium. Roeck et al (5) measured the ^{210}Po concentration in coal, then time correction had been done to the obtained value, for time interval between sampling and analysis. ^{210}Po concentration were assumed to be equal to the activity concentrations for the individual nuclides in the uranium radium series

In this work results for concentrations of the ^{234} Th ^{226}Ra, ^{214}Pb, and ^{214}Bi nuclides in uranium- radium series show disequilibrium in both the coke and coal as well as hematite and slag samples.

From table II, coal ore sample, shows low concentrations in all radioactive nuclides. the coal sample gives 23Bq kg^{-1} for ^{226}Ra, which is about the same order for most coal used in different coal power plants, which range between 20 Bq kg^{-1} and 335 Bq kg^{-1} (5,7).

Radionuclide content in ash depends on its concentrations in coal and the ash content in the coal. Concentrations of ^{234}Th, ^{226} Ra, ^{214}Pb and ^{214}Bi are in the same order of that obtained for coal.

The hematite sample shows high concentration in the uranium series which can be explained by adsorption on weathered ferromagnetic minerals. The used hematite in steel production goes through chemical processing also iron scrap were used in high percentage.

The slag in steel production is produced from the combination of coke, iron scrap, iron ore and limestone. It shows lower concentrations for all radioactive nuclides compared to those obtained by Keller et al (6), who measured 186 Bq kg^{-1} for ^{226}Ra, 100 Bg $^{-}$ kg^{-1} for ^{232}Th and 794 Bq kg^{-1} for ^{40}K, for slag obtained as waste product from coal power plant.

In a blast furnace, the temperature is higher than 1200 C, the ash and slag from furnaces burning pulverized coal have a glassy structure, so it has a relatively low emanation rate in comparison with other porous materials, which means the release of radon atoms from solid matter into the pores of the material is very low (6).

Radioactivity content in slag are higher than that obtained in the present work for both the ash and the slag. Also it is lower than the exempt level recommended limits put by council directive 96/92 EURATOM (8).

In Egypt slag and fly ash are used for high way construction purposes as embankment, even it was used in large quantities, it will give a negligible increase of exposure. Also the exposure to workers handling coal ash and slag at the disposal site is slightly increased compared to natural radiation.

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REFERENCES

1. .L.Don Leet and Sheldon Judson, Physical Geology, 3rd edition (1965).
2. ASM, American Society for Metals, Metals Handbook, 9th edition, vol. 1, properties and selections, iron and steels (1978).
3. Charles O. Smith & Bruce E. Broudman, Concepts and Criteria in Materials Engineering, Metals Handbook, 9th edition Vol. 9, P 825 (1978).
4. N.M. Ibrahiem, T.M. Abd. El. Maksoud, B. El.Ezaby, A. Nada, “International Symposium of the Restoration of Environments with Radioactive Residues” Arlington Virginia, USA, 29 Nov-3 Dec. (1999)
5. Douglas R. Roeck, Thomas C. Reavey and James M.Harden, Health Physics vol. 52, No.3, pp 311 - 323 (1987).
6. G. Keller,R.M. Nickels; and K.H. Puch; . The second regional Mediterranean Congress on Radiation, 20th Regional Congress of the Israel Radiation Protection Association Tel-Aviv, Israel, 16-20 Nov (1997).
7. M.S. Baxter, IAEA Bulletin, vol. 35 No.2, pp 33-37 (1993).
8. Anselmo S. Paschoa, Applied Radiation Isot Vol. 49 No.3, pp 189-196 (1998).