Analysis of radioactivity and thermal decomposition of fly ash for application in light concrete production

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Abstract. The article deals with the environmental pollution from ash waste, the analysis of radioactivity and thermal decomposition of fly ash at thermal power plants used in the production of light concrete. It shows the research and analysis of fly ash of thermal power plant in Ekibastuz, Republic of Kazakhstan. In these studies, more attention is paid to the comparison of the radioactivity analysis of fly ash of a thermal power plant, for compliance with international standards and requirements for use in light concretes. There is the study of the thermal decomposition of fly ash. The process of thermal decomposition of fly ash of Ekibastuz TPP. The objective of the research is to determine radioactivity and thermal decomposition of fly ash at Ekibastuz TPP and establish the possibility of its potential use in the production of light concrete. The process of decomposition of ash structure began at the temperature of 1100 °C, with heat absorption effects (endothermic) at 240-380° C and heat release effect (exothermic) at 680° C, and a corresponding change in ash mass loss temperature of 1.2%.

1 Introduction

When burning coal, a light part is released from the furnace along with smoke, called fly ash, and the sludge remains in the furnace. There is for fly ash, together with smoke, has toxic effects on air and soil, it is controlled by international and local standards and numerous measures are being taken to reduce its harm. Currently, coal thermal power plants are equipped with filters based on the movement of electric and water flows, capable to hold up to 99% of fly ash. In the United States, 43% of fly ash is stored and processed at power plants. In Europe, more than 40% of fly ash is used in construction. The use of ash in Russia is only 4-5% in 2015, and for Kazakhstan there are no exact data.

The main wastes of thermal power plants in Kazakhstan are ash slags and flue gases. Depending on the type of fuel, they occupy 10-15% when burning brown coal and 30-40% when burning hard coal. The annual amount of ash and slag waste from coal combustion is about 19 million tons in the Republic, and at present the total volume in storage is more than 300 million tons. Ekibastuz TPP in the process of industrial production emits significant amounts of ash and emissions. Currently, ash waste is stored in storage pits and pollutes the environment. Integrated industrial waste reprocessing is an important measure to protect the environment from pollution. A partial solution to this problem is the use of ash in the production of light concrete. Thus, thanks to trade turnover through the reuse of raw materials in the form of ash for production needs, waste-free technology, regional environmental problems and the rational use of local raw materials can be simultaneously solved. Before using the fly
ash of thermal power plants for the production of light concrete, it is necessary to conduct an analysis of their radioactivity and thermal decomposition studies.

2. Main section
When burning coal, most of the uranium, thorium and its by-products are separated from the original coal, distributed between the gas phase and solid combustion products. The amount of transition to the gas phase and the residue of the solid phase depend on the class, structure and mineral and chemical characteristics of the coal [1]. There is about 10% of the volume of the mineral part in the coal, so the radioactive content of ash is about 10 times more than the volume of the original coal. Radon is relatively the most harmful one among the decay products of the uranium group. Since radon is an inert gas, it can penetrate the pores and enter the air inside the building through them. At the same time, only a part of the entire radon gas reaches the surface of the material through air. This ratio is called the radiation of the building material or the radiation coefficient. The radiation effects of building materials on people’s lives are divided into internal and external ones. External radiation is caused by direct action of gamma-radiation, and internal radiation is caused by direct inhalation of products of short decay of radon [2, 4].

The indoor radon concentration allowed by European standards is less than 200 Bq/m$^3$. The average radioactivity of light concrete (Bq/kg) used in the European Union (RP-112, EU, Luxembourg, 1999): radium ($^{226}$Ra) -60, thorium ($^{232}$Th) -40, potassium ($^{40}$K) -430; maximum radioactivity: radium ($^{226}$Ra) -2600, thorium ($^{232}$Th) -190, potassium ($^{40}$K) -1600. The average content of coal fly ash radioactivity (Bq/kg): radium ($^{226}$Ra) -180, thorium ($^{232}$Th) -100, potassium ($^{40}$K) -650, maximum radioactivity: radium ($^{226}$Ra) -1100, thorium ($^{232}$Th) -300, potassium ($^{40}$K) -1500.

3. Research methods
The study of fly ashes radioactivity of Ekibastuz TPP was carried out by Ulan-Bator radiation laboratory using the standards MNS 5072:2018 and MNS 5626:2006 (3), which conform to GOST 30108.

The measurements of the isotopic activity of heavy elements of radium (Ra), thorium (Th), potassium (K), cesium (Cs) in ash were taken with the use of semiconductor detector Germani (Ge) with a highly distinct gamma spectrometer Canberra-GC4018. The radiation power of the instrument is 2.2 keV for the sample analysis. The data obtained with the use of the gamma spectrometer were processed on a computer using the software S-100 and FitzPeak.

The thermal research of fly ash of thermal power plant was executed in the laboratory of materials of Ulan-Bator Corporation of Architecture and Civil Engineering with the use of the derivatographic device of D500 brand manufactured in Hungary, under following conditions: the mass of a sample is 100 g, the speed of heating is 100 °C/min., final temperature is 10000 °C, a stream of air of 350 m/min., sensitivity of TG-500 device of the component DTA-1/10, DTG-1/10.

4. Test and results
The research of the radioactivity of fly ash of Ekibastuz TPP in Kazakhstan was carried out in the Ulan-Bator radiation laboratory with the use the standards GOST 30108, MNS5072:2018. Heavy elements were detected with the use of X-ray fluorescent devices (Logger-HPGe, model-GC4018) and (Ge) semiconductor gamma detectors (Canberra).

Table 1. Content of heavy elements in fly ash of Ekibastuz TPP.

| Element | As | Co | Cr | Cu | Mo | Ni | Pb | Sb | Th | U | V | W | K | Zr |
|---------|----|----|----|----|----|----|----|----|----|----|----|----|----|----|
| Content, gr/ton | 46 | 42 | 78 | 74 | 51 | 37 | -  | 94 | 22 | 25 | 112 | -  | 2  | -  |
Fly ash of Ekibastuz TPP in Kazakhstan contains a large number of heavy elements, such as vanadium, antimony and chromium. The radioactivity of ash depends on the number of radioactive isotopes contained in the elements of the material $^{226}$Ra, $^{232}$Th, $^{40}$K.

Table 2. Comparative parameters of fly ash radioactivity of Ekibastuz TPP.

| Type of fly ash | Individual activity of isotopes, Bq/kg | Radium equivalent, Bq/kg |
|-----------------|--------------------------------------|-------------------------|
| Fly ash of Ekibastuz TPP | $^{226}$Ra: 33±3, $^{232}$Th: 42±4, $^{40}$K: 195±20 | 103.0 |
| Fly ash TPP No.4 | $^{226}$Ra: 663.4±6, $^{232}$Th: 50.6±1, $^{40}$K: 718.7±25 | 789.0±11.4 |
| Fly ash TPP Amgalan | $^{226}$Ra: 386, $^{232}$Th: 108, $^{40}$K: 265 | 545 |

According to the test results presented in Table 2, the individual activity of radium ($^{226}$Ra) isotope of fly ash of Ekibastuz TPP is 10-20 times lower than that of the fly ash of TPP No. 4 and TPP Amgalan Ulanbator, and 5-7 times lower than the radium equivalent.

Figure 1. Derivatogram of fly ash of TPP Ekibastuz in Kazakhstan.

Table 3. Calculation of activation energy.

| No. | Δt, mm | $t$, °C | T, K | 1/T | lnΔt |
|-----|--------|---------|------|-----|------|
| 1   | 2      | 110     | 383  | 0.0026 | 0.69 |
| 2   | 4      | 240     | 556  | 0.0017 | 0.91 |
| 3   | 6      | 380     | 653  | 0.0015 | 1.13 |
| 4   | 8      | 460     | 733  | 0.0013 | 1.35 |
| 5   | 10     | 650     | 923  | 0.0010 | 1.57 |
| 6   | 12     | 680     | 953  | 0.0010 | 1.79 |
| 7   | 14     | 780     | 1033 | 0.0009 | 2.01 |
| 8   | 16     | 860     | 1053 | 0.0009 | 2.23 |
| 9   | 18     | 900     | 1173 | 0.0008 | 2.45 |
According to the derivatogram (figure 1) that shows the process of thermal decomposition of fly ash of Ekibastuz TPP, the process of decomposition of ash structure began at the temperature of 1100 °C, with heat absorption effects (endothermic) at 240-380 °C and heat release effect (exothermic) at 680 °C, and a corresponding change in ash mass loss temperature of 1.2%. Based on the calculation of activation energy given in Table 3, a dependency graph was drawn up.

Activation energy is determined according to the graph in Figure 2:

$$E = R \cdot \frac{b}{a} = 0.0083 \text{kJ/mol} \cdot \text{K}^{-\frac{27}{45}} = 0.005 \text{kJ/mol} \cdot \text{K},$$

where: $R=0.0083 \text{kJ/mol} \cdot \text{K}$ - absolute gas constant; $b, a$ - plotted value in mm (figure 2).

5. Conclusion

According to the results of the study, the natural activity of the contained radionuclides was not limited to the use of radiation factors in the production of all types of building materials. A comparative study of radio emissions showed that the radioactivity of fly ash of Ekibastuz TPP is very insignificant and can be allowed to be used in the production of materials and structures used in the construction of residential and public buildings. The results of the ash thermal decomposition analysis meet the requirements of the standard. Therefore, in accordance with GOST 30108 and MNS5072: 2018 standards, the individual activity of isotopes of heavy elements contained in fly ash of Ekibastuz TPP does not exceed the regulatory requirements and can be used in all sectors of the construction materials industry, including the production of gas concrete.

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