

## RESEARCH ON COALS AND ASH RESIDUES FOR THE PRESENCE OF RARE EARTH METALS

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This article explores the significance of coal mining and processing within the framework of the development strategy of the fuel and energy industry and the transition to a "Green Economy" in the Republic of Kazakhstan. It emphasizes the underutilized potential of rare metals present in coal and its by-products and discusses the importance of developing environmentally friendly technologies for their extraction. The article highlights the issue of ash slag waste from local thermal power plants and proposes ways for its rational utilization, including the extraction of rare metals. The Shubarkol coal deposit in Central Kazakhstan is considered a significant source of rare metals available for extraction during coal combustion. The article also underscores the need for further research and development in this area to maximize the environmental and economic benefits of using coal resources.

**Keywords:** rare metals, coal deposits, ash residues, element extraction, extraction of valuable metals.

## ИССЛЕДОВАНИЯ УГЛЕЙ И ЗОЛОТХОДОВ НА НАЛИЧИЕ РЕДКОЗЕМЕЛЬНЫХ МЕТАЛЛОВ

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Данная статья рассматривает значимость добычи и переработки угля в контексте стратегии развития топливно-энергетической отрасли и перехода к "Зеленой экономике" в Республике Казахстан. Она подчеркивает неиспользованный потенциал редких металлов, содержащихся в угле и его отходах, и обсуждает важность разработки экологически чистых технологий для их извлечения. В статье выделяется проблема золошлаковых отходов от местных тепловых электростанций и предлагаются пути их рационального использования, включая извлечение редких металлов. Шубаркольское угольное месторождение в Центральном Казахстане рассматривается как значительный источник редких металлов, доступных для извлечения в процессе сжигания угля. Статья также подчеркивает необходимость дальнейших исследований и разработок в этой области для максимизации экологических и экономических выгод от использования угольных ресурсов.

**Ключевые слова:** редкие металлы, угольные месторождения, золотходы, экстракция элементов, извлечение ценных металлов.

## ҚЫЗЫҒУШЫ ҚҰРЫЛТАЙЛАР МЕН ҚЫЗЫҒУШЫ АСТАРҒА АРНАЛҒАН ҚАЗЫНАЛАРДЫҢ ТАЛДАУЫ ЖӘНЕ ТҰТАСТАРДЫҢ РЕДКІ МЕТАЛДАРЫНЫҢ БАРЛЫҚТЫҚТАРЫН ТЕКСЕРУ

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Бұл мақалада Қазақстан Республикасының топтық-энергиялық саладағы даму стратегиясы және "Жасыл экономика"ға көшу концепциясы контекстінде көмір екімі мен өңдеу маңыздылығы тексеріледі. Бұл

мақала көмір мен оның жалға материалдарындағы талаптары қолданылмаған қызметті мөлшерлердің бойынша көмір мен оның толқындарындағы күндізгі металлдардың көрсеткіштілігін негіздеуге қарсы ерекше технологияларды дамыту маңыздылығын айқындайды. Мақала орта маңызды термалды энергиялық станциялардың местік қызметінен шығатын мал бұзылу мүлдеміні талқау және бұның рақсетті пайдалануларын көру үрдісіне ықпал етеді, оның ішінде редкі металлдарды шығаруларын қоспаған. Ортағасы Қазақстандағы Шубарқол көмір местінен мәні шамамен көмірдің жауқындысы кезінде шығаруға болатын қызметті металлдардың маңызды құрылымы ретінде сипатталады. Мақала көмір ресурстарын қолдануның экологиялық және экономикалық маңыздылығын көмек көрсету үшін осы жағдайда кейбір басқармаларда үздіктіктер мен зерттеулерді өткізу керектігін түсіндіреді.

**Түйін сөздер:** сирек металдар, көмір кен орындары, күл қалдықтары, элементтерді шығару, бағалы металдарды алу.

**Introduction.** In accordance with the Development Concept of the Fuel and Energy Industry of the Republic of Kazakhstan for the period up to 2030 and the implementation of the Concept for Transition to a "Green Economy," the expansion of coal usage should serve as an incentive for conducting research and development of new, environmentally friendly technologies for its extraction, combustion, and processing [1].

Currently, coal mining and comprehensive processing are gaining increasing importance due to the potential utilization of previously non-commercial deposits. The search and development of rare earth metals, as well as the utilization of waste from black and non-ferrous metallurgy production, play a significant role in the rational use of natural resources. One of the goals of this study is to address this issue. One possible approach to its solution is the utilization of waste as a source of rare earth metals [2,3].

Currently, the rare-metal potential of coals is practically untapped. From coals and their wastes on an industrial scale, only germanium (Ge) and gold (Au) are extracted. Technologies for extracting gallium (Ga), scandium (Sc), rare earth metals, and some other metals have also been developed.

From the perspective of rational land use, coals and their combustion products represent raw materials extracted from the earth's depths, which are transported to other territories and inadequately utilized to satisfy many industrial needs [4].

Ash dumps formed at local thermal power plants pose a serious environmental threat to the region and can have a negative impact on the environment and human health. Due to wind erosion, ash particles can enter the atmosphere and spread over long distances, leading to air pollution. Settled dust, accompanied by chemically active toxic substances, can contaminate soil. Moreover, under the influence of acid rain, toxic

substances from ash dumps are mobilized, which can lead to soil, groundwater, and surface water pollution.

In light of the above, it becomes evident that ash dumps cause significant environmental, economic, and social damage to the region. The problem of ash slag material utilization requires immediate solution [5].

Currently, in global practice, coal deposits are increasingly being considered not only as a source of fuel and energy raw materials but also as a potential source of a range of rare elements and noble metals (including the USA, China, Russia, and other countries). Partial studies of the composition of rare metals in coal organic materials have been conducted in developed economies (USA, Europe, Australia, China), reflected in numerous scientific publications. These studies indicate that coal industry wastes may contain high concentrations of rare elements, which in some cases may be industrially significant.

However, existing methods for extracting rare metals currently have low efficiency, not exceeding 30%. This limits the attractiveness of investments in the development of domestic processing industries [6].

The current global annual volume of gold slag waste (GSW) extraction is approximately 750 million tons, and an increase in this volume is expected in the near future.

**Materials and methods.** Methods of detecting rare earth metals: Mass spectrometry (MS); atomic emission spectrometry (AES); X-ray fluorescence analysis (XRF); laser diffraction method.

Mass spectrometry (MS) is based on the ionization of the sample substance in a magnetic field. The ions are subjected to the Lorentz force, the magnitude of which depends on the mass and charge of the ion. The difference in the trajectories of different ions allows for the determination of the atomic content of substances in the sample. To perform measurements, the sample of the investigated solid substance needs

to be decomposed to eliminate factors that distort the analysis data and converted into a solution, which is labor-intensive and time-consuming [7].

The sensitivity of this method significantly depends on the ionization method and the detectors used. According to [8,9], when using an inductively coupled plasma mass spectrometer (ICP-MS) to determine the concentration of rare earth elements (REEs) in solution, the sensitivity is about 10-12 mg/L. The accuracy of the method depends on the concentration of the determined element (C) in the sample; polyatomic interferences and matrix effects also introduce errors

[10]. Interfering impurities include ions with the same mass-to-charge ratio as the determined atom, as well as certain isotopes; for example, for La, these are LaO<sup>+</sup>, LaOH<sup>+</sup>, and <sup>155</sup>Gd<sup>+</sup> [7]. According to Panteeva's data [11], as the value of C decreases from 1000 g/t to 0.01 g/t, the measurement error increases from 8% to 32%. The reproducibility of the method is considered good [7], but it may also decrease as the value of C decreases. Therefore, the considered method is expedient to use for determining relatively high (tens and hundreds of g/t) concentrations of elements in ash and with relatively small fluctuations over time in the elemental composition of the analyzed ash.

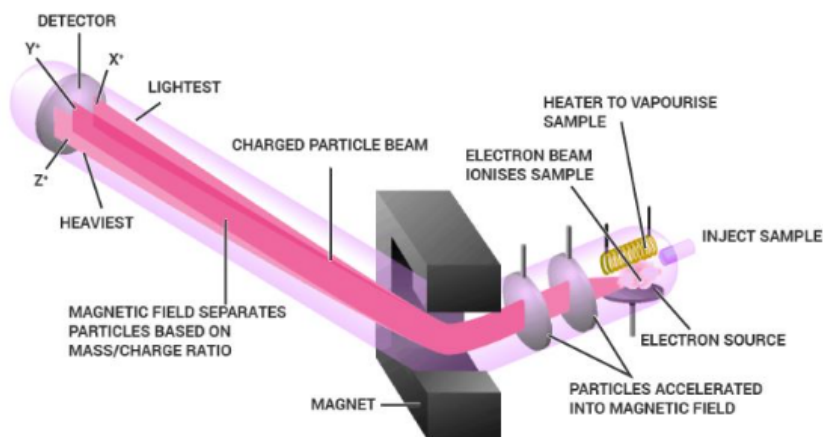


Figure 1 - Operation of Mass Spectrometer: Principle and Schematic Diagram of the Spectrometer

Atomic emission spectrometry (AES) is a method based on the emission of electromagnetic radiation by excited atoms or ions. To achieve this, the sample substance is subjected to high temperatures, resulting in the dissociation of compounds into atoms and an increase in the number of collisions leading to the ionization of atoms. Atoms and ions, being in an excited state, are capable of returning to their ground energy state by transferring thermal or radiative energy and emitting electromagnetic radiation. This allows for the determination of the quantity of atoms, and consequently, the concentrations of elements. The emission spectrum of an element contains more lines than its corresponding absorption spectrum.

For measurements, similar to the MS method, the sample of the solid substance is converted into a solution and heated to evaporate the solvent and excite the atoms of the investigated substance [12]. The emitted radiation from the atoms is decomposed into

a spectrum and recorded. For qualitative analysis, spectral lines are identified, and for quantitative analysis of elements, the intensity of the lines is measured. The concentration of elements is determined using pre-established calibration graphs. The error is caused by the overlap of spectral lines of different elements, as well as the matrix effect. According to data, the relative standard deviation (RSD) during the analysis of geological materials ranges from 5% (Y, Eu) to 10% (Pr). Interfering elements include: Na, K, Ca, Fe. It is also noted that light REEs, such as La, Nd, Ce, Pr, Sm, interfere with the determination of heavy REEs, such as Ho, Er, Tm, Yb, Lu, so separate determination of these groups of REEs is recommended. Zybinski et al. showed that for La, Ce, Eu, Y, Gd, the systematic error of determination does not exceed 5%; for Lu, Yb, Ho, Sc - no more than 15%; for Dy, Er, Tm, Nd - no more than 25%; for Pr, Sm, Tb - tens and hundreds of percent. The reproducibility and

repeatability of AES with an element content above 100 g/t is no more than 5%, less than 100 g/t - no more than 10% [13]. The detection limits of some REEs, in g/t: Y - 2; Zr - 4; La - 2000; Ce - 4000; Pr - 30; Nd - 20; Eu - 1; Dy - 5; Er - 8. From the presented data, it can be seen that the accuracy and sensitivity of this method

for different REEs vary significantly. Therefore, the application of AES is advisable for the determination of elements in ash such as Y, Zr, Dy, Er. At the same time, for a complete analysis of REE content, it is preferable to combine MS and AES methods.

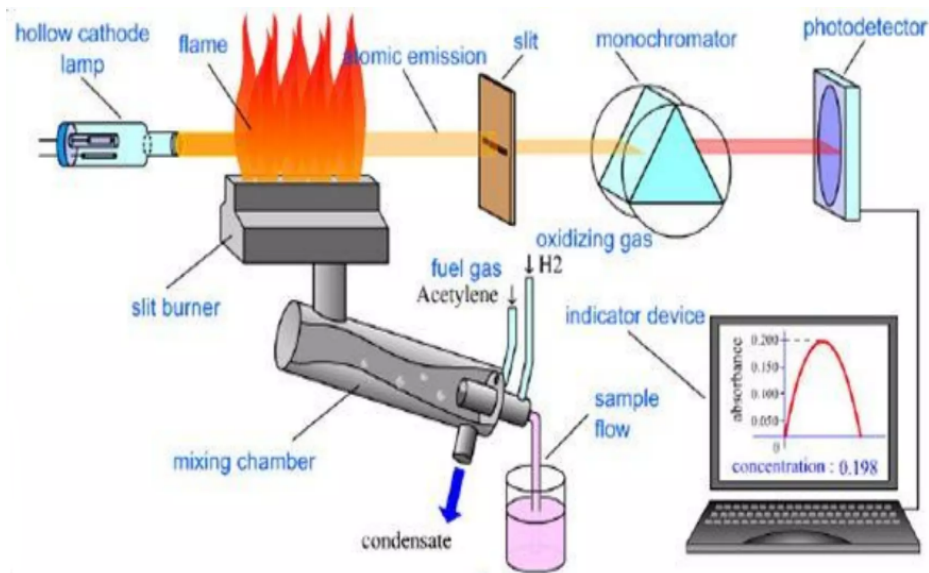


Figure 2 - X-ray Fluorescence Analysis (XRF)

X-ray Fluorescence Analysis (XRF) is based on the analysis of the spectrum generated by using X-ray radiation [7].

When interacting with high-energy photons, atoms of the substance transition to an excited state, resulting in electron transitions from lower orbitals to higher energy levels up to ionization of the atom. In the excited state, the atom remains for a very short time, about one microsecond, after which it returns to its ground state. During this process, electrons from outer shells fill the vacant positions, and the excess energy is either emitted as a photon or transferred to another electron from the outer shells. Each atom emits a quantum with energy of a strictly defined value. The substance structure is judged by the energy and quantity of quanta emitted.

There are several variations of this method:

Wavelength dispersion is used to determine the concentrations of impurity elements; characterized by relatively high sensitivity;

Energy-dispersive method is less sensitive but more suitable for express analysis [10].

The sensitivity of the method in determining the

concentration of REEs in solid materials is at the level of the Clarke content of the element. The method error depends on the nature of the element and its concentration. Thus, according to [14], when determining Y, La, Ce, Pr, and Nd, it ranged from 18 to 28%. Reproducibility is about a few percent. According to [7], the accuracy and reproducibility of the results increase due to pressing the investigated powder material and melting with lithium tetraborate.

Compared to the methods described above, XRF is characterized by less complexity because the material does not need to be decomposed and converted into a solution. The time required for the analysis is relatively short, amounting to several minutes. Disadvantages include relatively low accuracy and the inability to determine the concentration of elements with an atomic mass of less than 40 a.m.u. This method is acceptable for the express assessment of REE content in ash. Other methods of determining the REE content are also mentioned in the literature. In particular, neutron activation analysis, which, according to [7], was effective for determining La, Ce, Nd, Sm, Eu, Tb, Yb, Lu in the 1960s - 1980s but was then replaced

by the more accurate and versatile MS method. The radiometric method, due to its low complexity and speed, has found wide application for the analysis of the

elemental composition of ores, but the low sensitivity (about several hundred g/t) makes it impossible to apply this method for most REEs.

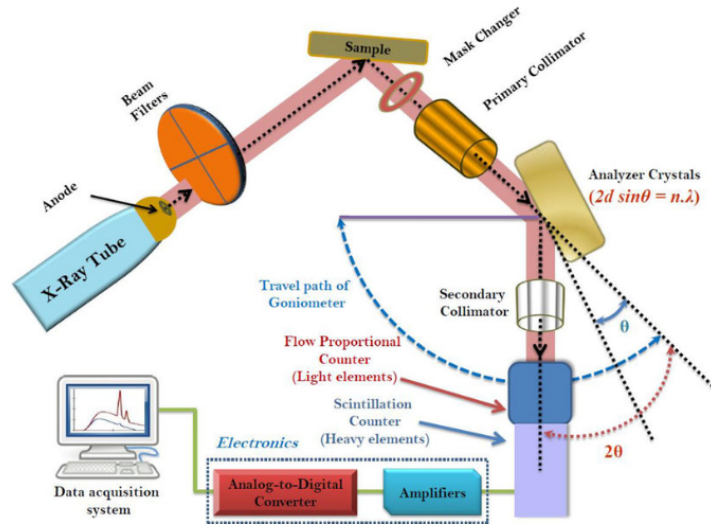


Figure 3 - X-ray Fluorescence Spectrometer

Laser Diffraction Method. Unlike the previous method, it involves the use of laser instead of visible light [12]. This allows measurements of particle diameters in the range from 10 nm to 3500 μm, corresponding to the particle size of ash. The accuracy of the method, according to [15], ranges from 1 to 4%. Advantages include a wide range of measurements, high measurement accuracy, speed, simplicity, the possibility of measurements in a flow-through mode, and good reproducibility. Disadvantages include difficulty in detecting particles

at low concentrations and the need to disperse particles in a liquid [7].

**Results and discussion.** The Shubarkol deposit in Central Kazakhstan has reserves of more than 1 billion tons. These are significant sources of rare earths that can be extracted during coal combustion at thermal power plants (TPPs). Coal deposits contain 64 g/t of scandium, 384 g/t of dysprosium, and 335 g/t of gadolinium (see Figure 4). Thus, these are large reserves of rare earths available for extraction during the coal combustion process at TPPs.

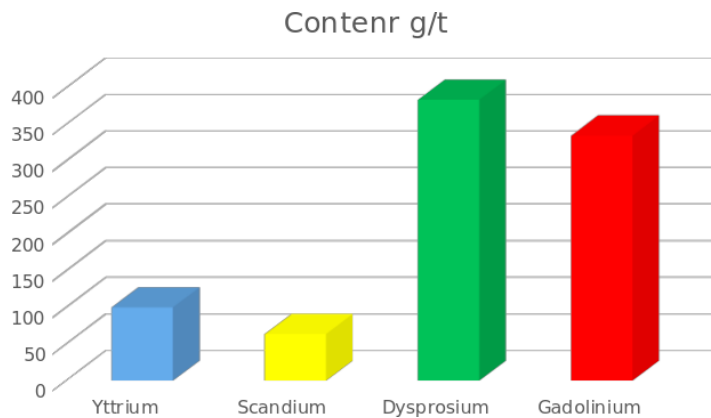


Figure 4 - Rare Earth Element Content in Shubarkol Coal Deposit



The advantages of using waste as resources have both environmental and economic aspects:

It is one of the most rational ways to address environmental issues as it helps reduce anthropogenic pressure on the natural environment.

Waste processing contributes to increasing the economic efficiency of thermal power plants.

As a result of secondary waste processing, companies can reduce expenses on slag and ash storage, and also utilize them as a source of rare and rare earth metals, leading to additional economic benefits.

Surface waste does not require additional extraction expenses, which is a positive factor for geological enterprises. This also contributes to increasing extraction volumes with maximum useful component extraction and reducing the areas alienated for deposit development. In this context, coal ash generated during combustion can be considered as potential ore from which rare earth metals can be extracted in the future. The concentration of these metals in ash can serve as an important indicator for industrial deposit assessment [16].

Table 1 - Rare Earth Content in Coals from Various Geological and Industrial Districts of the Kuzbass Region

Geological-industrial area	Element								
	N	La	Ce	Sm	Eu	Tb	Yb	Lu	Yb
Angers	2	1,8	3,9	0,7	0,1	0,2	0,35	0,15	5,1
Aralichevsky	122	17,6	29,8	3,05	1,31	0,58	1,78	0,38	9,9
Baydaevsky	10	18,3	28,2	2,9	1,03	0,53	2,1	0,41	8,7
Bachatsky	5	24,5	12,7	3,42	1,18	0,15	1,68	0,25	14,6
Bunguro-Chumyshsky	9	13,9	25,6	2,37	0,65	0,37	0,89	0,39	15,6
Kemerovo	116	9,9	23,9	2,33	0,6	0,56	1,06	0,27	9,3
Kondomsky	9	16,8	26,2	3,23	0,93	0,99	2,19	0,63	7,7
Leninist	18	7,9	17,7	1,33	0,33	0,2	1,13	0,33	7
Mrassky	73	24,7	37,9	2,94	0,78	0,58	3,02	2,09	8,2
Osinnikovsky	56	8	15,8	2,05	0,42	1,02	0,8	0,33	10
Prokopyevsky	140	8,4	14,3	1,41	0,31	0,19	1,57	0,26	5,3
Tom-Usinsky	169	12,2	22,6	2,5	0,41	0,75	1,37	0,28	8,9
Uskatsky	15	12,6	38,8	5,1	0,87	-	1,1	-	11,5

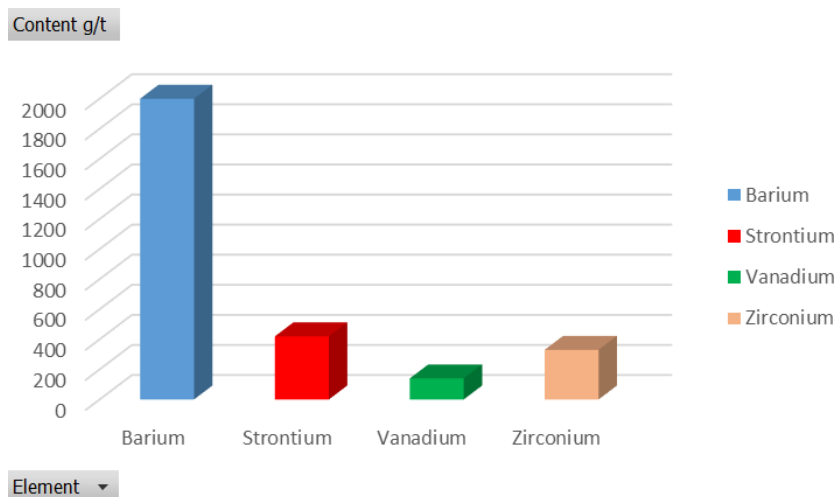


Figure 5 - Results of neutron activation analysis of fly ash

Characterization of Fly Ash from Ekibastuz Coal as Raw Material for Deep Processing includes an analysis of the composition obtained from the electrostatic precipitators of Omsk TPP-4. The main components comprise silicon, aluminum, and iron oxides, reaching

up to 95%. Additionally, the fly ash contains oxides of alkali and alkaline-earth metals, with a total content of 2.3%. Moreover, the presence of 15 trace elements, each exceeding 10-4%, has been identified.

Table 2 - Chemical Composition of Fly Ash from the Ekibastuz Coal Basin

SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	TiO <sub>2</sub>	K <sub>2</sub> O	Na <sub>2</sub> O	P <sub>2</sub> O <sub>5</sub>	MnO <sub>2</sub>	SO <sub>3</sub>
61,5	27,4	5,65	1,17	0,49	1,49	0,42	0,32	0,52	0,17	0,57

Table 3 - Elements of ash slag waste from the Shubarkol coal deposit classified as rare are debated

Sample №	№1	№2	№3	№4	№5	Cp	Min	Max	Price million tenge/gram
Element	Content, g/ton								
1 Boron B	100	200	160	140	100	140	100	200	315
2 Strontium Sr	400	280	220	172	490	312,4	172	490	7,92
3 Selenium Se	0,6	0,6	0,6	0,6	0,6	0,6	0,6	0,6	6,44
4 Tellurium Te	0,06	0,06	0,06	0,06	0,06	0,06	0,06	0,06	3,47
5 Bismuth Bi	60	60	60	60	60	60	60	60	2,25
6 Cadmium Cd	18800	19200	9900	20600	10000	15700	9900	20600	1,39

Table 4 - Rare Earth Elements Present in the Shubarkol Coal Deposit

Sample №	№1	№2	№3	№4	№5	Average	Min	Max	Price million tenge/gram
Element	Content, g/ton								
1 Scandium Sc	20	20	20	20	20	20	20	20	292,5
2 Yttrium Y	6	13	7	33	11	14	6	33	198
3 Germanium Ge	7	7	7	7	7	7	7	7	134,55
4 Lanthanum La	34	93	52	140	48	73,4	34	140	99
5 Beryllium Be	4	4	7	17	10	8,4	4	17	46,58
6 Niobium Nb	10	10	10	10	10	10	10	10	35,69
7 Vanadium V	57	79	110	270	88	120,8	57	270	34,65
8 Indium In	0,09	0,09	0,09	0,09	0,09	0,09	0,09	0,09	18,09
9 Zirconium Zr	30	50	30	70	30	42	30	70	10,89
10 Lithium Li	45	45	45	45	45	45	45	45	9,9
11 Molybdenum Mo	3	4	4	11	3	5	3	11	7,92
12 Titanium Ti	1200	3300	2100	780	2700	2016	780	3300	7,57
13 Thallium Tl	0,2	0,3	0,1	0,3	0,1	0,2	0,1	0,3	7,43
14 Tellurium Te	0,06	0,06	0,06	0,06	0,06	0,06	0,06	0,06	3,47
15 Tungsten W	5	5	6	5	5	5,2	5	6	3,47

Rare metals present the greatest potential as they do not form their own deposits. Among the rare metals contained in this fly ash, the following groups are distinguished: dispersed - Ga; refractory - Ti, Zr, V; rare earth - Y, Yb, Tb, La, Ce, Dy, Sm; radioactive - U, Th.

The phase composition of the ash also plays an

important role in the efficiency of element extraction. X-ray phase analysis, conducted on the Drone-3 setup using the K $\alpha$  line and p-filter, revealed the presence of an amorphous phase in the ash, as well as alpha-quartz and aluminosilicates of the sillimanite type Al<sub>2</sub>O<sub>3</sub>xSiO<sub>2</sub> or mullite 3Al<sub>2</sub>O<sub>3</sub>x2SiO<sub>2</sub>. The fundamental principles of silicon and aluminum

extraction during hydroprocessing were studied, allowing for the complete separation of the amorphous phase [17-18].

During the oxidation of coal seams and coal combustion, a significant portion of rare earth metals transitions into the composition of combustion gas products or settles in the gas cleaning system. At the same time, the concentration of these metals in ash slag waste exceeds their content in the original

coal materials. For example, the gold content in ash slag waste after coal combustion may exceed the gold content in the coal deposits themselves. Similar changes are observed for other rare earth metals, such as platinum.

Research was conducted on the characteristics of ash slag waste generated during the combustion of the Shubarkol coal deposit. The obtained results are presented in the corresponding table.

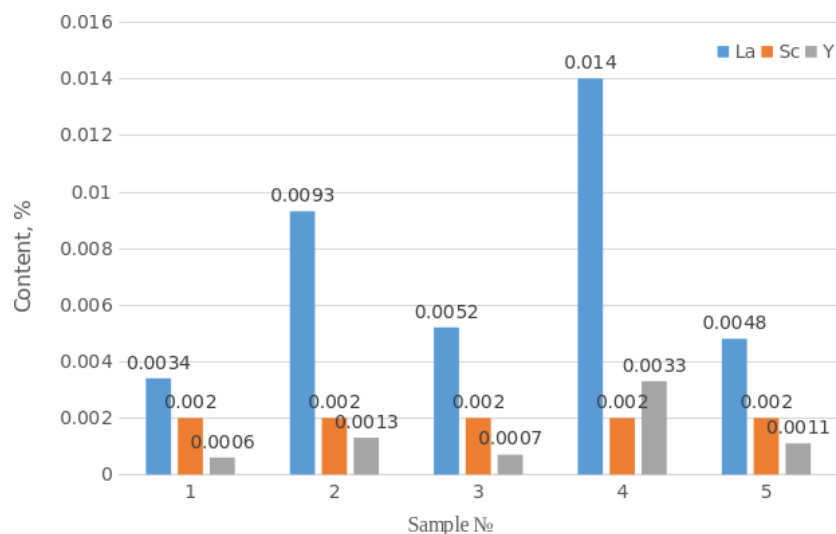


Figure 6 - Rare Earth Metal Content in Coal Ash Slag Waste from the Shubarkol Deposit

The investigation of this fly ash has focused on identifying the content of rare earth metals such as lanthanum, scandium, and yttrium. The obtained results confirm that the extraction of rare metals from secondary sources, including waste processing, represents a promising approach. This helps to reduce the risk of increasing waste volumes and creates a basis for the development of an independent raw material base.

The coal reserves of the Karaganda Coal Basin have not been adequately studied for the content of rare earth metals, and their resources can only be partially assessed. However, in the case of the Shubarkol deposit in Central Kazakhstan, where reserves exceed 1 billion tons, significant amounts of rare earth metals have been found during coal combustion at thermal power plants. The content of yttrium is up to 100 g/t, scandium - 64 g/t, dysprosium - 384 g/t, and gadolinium - 335 g/t. These data indicate a significant potential for the extraction of rare earth metals from coal ash slag at thermal power plants.

**Conclusions.** The article discusses the significance of coal mining and comprehensive processing in the context of Kazakhstan's Development Concept of the Fuel and Energy Industry and the transition to a "Green Economy." It emphasizes the importance of researching and developing environmentally friendly technologies for coal extraction, combustion, and processing.

**Rare Metal Potential:** The study emphasizes the largely untapped potential of rare metals in coal and its by-products. While germanium and gold are currently extracted on an industrial scale, technologies for extracting gallium, scandium, rare earth metals, and other metals have been developed.

**Environmental Concerns:** The article underscores the environmental threats posed by ash dumps from local thermal power plants. These dumps can lead to air and soil pollution, posing risks to human health and ecosystems.

**Economic and Environmental Benefits of Waste**



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Utilization: Utilizing waste from coal combustion not only addresses environmental concerns but also offers economic benefits. Secondary waste processing can reduce expenses on storage and lead to the extraction of rare and rare earth metals, contributing to economic efficiency.

Rare Earth Metals in Coal Deposits: The Shubarkol deposit in Central Kazakhstan is highlighted for its significant reserves of rare earths, including scandium, dysprosium, and gadolinium. These metals can be extracted during coal combustion at thermal power plants.

Research and Development: The article stresses the importance of further research and development in the extraction of rare metals from coal and its by-products. It suggests that existing extraction methods have low efficiency, limiting investment attractiveness.

Overall, the study underscores the potential of coal deposits not only as sources of fuel but also as valuable reservoirs of rare elements. It advocates for the development of technologies to extract these elements efficiently, thereby mitigating environmental impacts and maximizing economic benefits.

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