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Ways of Rare Earth Elements Migration and Transportation to the coals of the Shubarkol Deposit

*Blyalova G.G., Kopobayeva A.N., Amangeldykyzy A., Askarova N.S., Ozhigin D.S.

Abylkas Saginov Karaganda Technical University, Karaganda, Kazakhstan

* Corresponding author email: gulim_blyalova@mail.ru

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ABSTRACT
The article presents the results of studying the mineralogical and geochemical features of coals and clay interlayers of the Shubarkol deposit. There were analyzed 71 samples of clay layers and 49 samples of coal using the instrumental neutron activation analysis (INAA), powder X-ray diffractometry, and scanning electron microscopy. A comprehensive analysis of the geochemical and mineralogical characteristics of coals and their enclosing clay interlayers was carried out. A variety of geochemical criteria made it possible to establish the facts of the rare earth elements (REE) migration, and the latest data of mineralogy made it possible to establish the ways of their transportation to the paleobasin in the syn- and epigenetic periods of the Shubarkol deposit formation. The data of the paleoclimate and water composition of the paleobasin of peat formation are presented for the first time. Among other things, the analysis made it possible to identify a number of independent sources and various mechanisms of REE accumulation in the sediments of the Shubarkol deposit. The main patterns of the REE distribution that are expressed in the predominance of light lanthanides over heavy lanthanides, were established, and the role and composition of the rocks surrounding coal deposits in the concentration of REE were considered. Aluminosilicates, sulfides, and sulfates with inclusions of microparticles of rare and rare-earth REE elements were found in coals and clay interlayers.
Keywords: coal, Shubarkol, rare earth elements, mineralogy, geochemistry.

Information about authors:

Blyalova Gulim Galymzhanovna	*doctoral student of Abylkas Saginov Karaganda Technical University, Master of Technical Sciences in Geology and Mineral Deposit Exploration. 100000, Karaganda, Kazakhstan. Email: gulim_blyalova@mail.ru; ORCID ID: https://orcid.org/0000-0001-8801-8683*
Kopobayeva Aiman Nygmetovna	*PhD, Senior Lecturer of the Geology and MD Exploration Department of Abylkas Saginov Karaganda Technical University. 100000, Karaganda, Kazakhstan. Email: aiman_25.87@mail.ru; ORCID ID: https://orcid.org/0000-0002-0601-9365*
Amangeldykyzy Altynai	*PhD, Senior Lecturer of the Geology and MD Exploration Department of Abylkas Saginov Karaganda Technical University. 100000, Karaganda, Kazakhstan. Email: amangeldykyzy@inbox.ru; ORCID ID: https://orcid.org/0000-0002-6665-8804*
Askarova Nazym Srazhadinkyzy	*doctoral student of Abylkas Saginov Karaganda Technical University, Master of Technical Sciences in Geology and MD Exploration, 100000, Karaganda, Kazakhstan. Email: srajadin-nazym@mail.ru; ORCID ID: https://orcid.org/0000-0002-2103-6198*
Ozhigin Dmitri Sergeyevich	*PhD, Senior Lecturer of the Mine Surveying and Geodesy Department of Abylkas Saginov Karaganda Technical University. 100000, Karaganda, Kazakhstan. Email: ozhigin.dima@mail.ru; ORCID ID: https://orcid.org/0000-0002-2443-3068*

Introduction

In recent years, the issue of the nature of impurity element accumulation in coal deposits has been increasingly raised. Understanding the patterns of accumulation of certain elements, and dispersion of others is important for predicting mineralization in coals formed in different blocks of the earth's crust. To these issues, scientists have already formulated theories that explain the anomalous concentrations of various components, the main sources of which are considered to be the rocks of the feeding area of the coal accumulation basin; volcanic-clastogenic inflow of pyroclastic material into the paleopeat bog; hydrothermal introduction of impurity elements into the paleopeat bog or coal seam; hydrological factor, etc [1].

Studying impurity elements, including rare earth elements (REE) in coals and clay interlayers, is of scientific importance due to their static geochemical characteristics, and are studied indicators of the origination of coal [[2], [3], [4], [5]].

But at present, there is not yet sufficient geological information on accumulation and forms of REE occurrence, as well as the mechanisms of their concentration in the coals of Kazakhstan. The Shubarkol deposit is one of the objects, where, alongside a low ash content and a lower sulphur, which makes the coal of the deposit a valuable fuel, there are high contents of impurity elements that often reach industrial significance.

The relevance of the work is due to the need to determine the nature and accumulation of impurity elements in order to obtain comprehensive information necessary for the development of predictive criteria in the future.

In order to elucidate the ways of migration and the mechanism of concentration of impurity elements in the Shubarkol deposit coals, clay interlayers, and coals containing them were tried and explored in detail.

A comprehensive analysis of the geochemical and mineralogical characteristics of coals and their enclosing clay interlayers was carried out. A variety of geochemical criteria made it possible to establish the facts of the rare earth elements (REE) migration, and the latest data of mineralogy made it possible to establish the ways of their transportation to the paleobasin in the syn- and epigenetic periods of the Shubarkol deposit formation. The data of the paleoclimate and water composition of the paleobasin of peat formation is presented for the first time.

Experimental part

The basis for this work is the results of analyzing impurity elements of 49 coal samples and 71 samples of coal-bearing rocks of the Shubarkol deposit. The samples for studying were taken at the Western and Central sections of the Shubarkol deposit, in coal seams 2B, 1B1, 1B2, in sections 1-6 (sections 1 and 2 are located within the Western section, sections 3-6 within the Center section) (Figure 1). The sampling distance in the plots was within 30-35 m. The composite samples weighing 200 g were made using the basic samples. In entire, the group composition of 120 samples was explored.

The samples were studied by the instrumental neutron activation analysis (INAA) to determine the average contents of 28 elements in the nuclear geochemical laboratory of the Geoecology and Geochemistry Department at National Research Tomsk Polytechnic University (TPU) (analyst A.F. Sudyko). To determine the mineral composition of the samples of coal-bearing rocks and coals there was used X-ray powder diffractometry, and scanning electron microscopy.

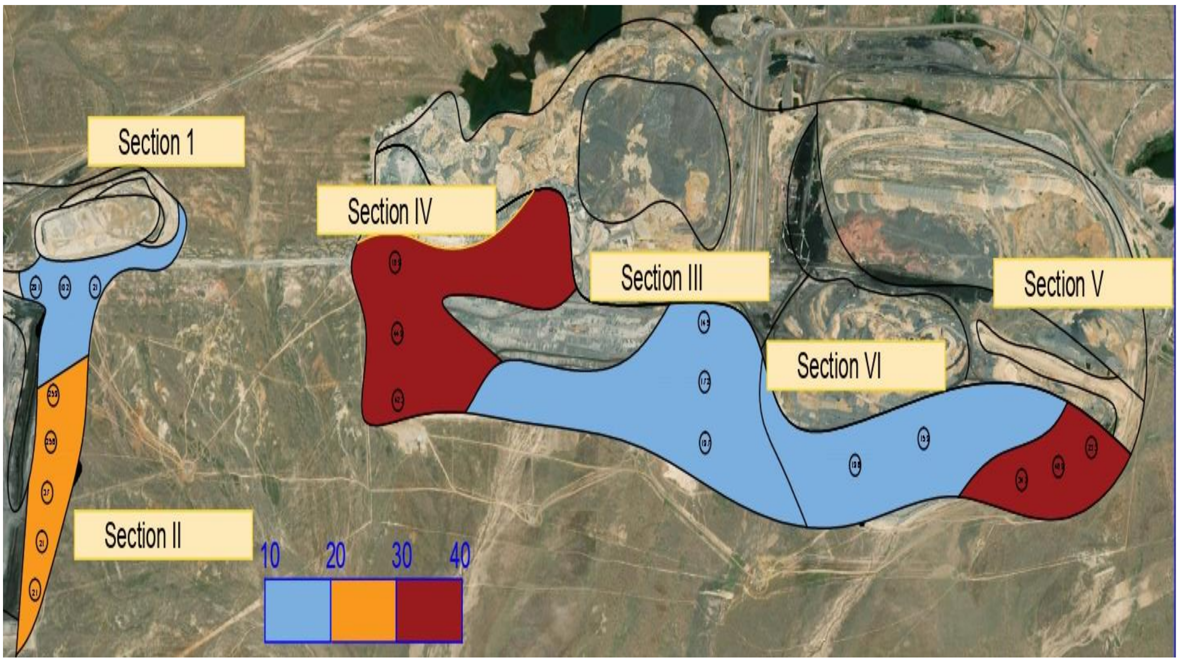

Figure 1 – Total REE distribution (La, Ce, Sm, Eu, Tb, Yb, Lu, Nd) in the Shubarkol deposit coals

Discussion of the results

Compared with the REE content in the UCC (upper part of the continental crust [6]), three types of enrichment are distinguished: L-type ($La_N/Lu_N > 1$), M-type ($La_N/Sm_N < 1$,) and H-type ($La_N/Lu_N < 1$).

In this work, the La_N/ Lu_N ratios of coal can be seen as ranging from 0.44 to 14 (Table 1), which indicates the characteristic predominance of light lanthanides over heavy lanthanides in the coals of the deposit.

The deposit-averaged value of the La/Yb ratio significantly exceeds a unit. REE in coals has low concentrations and is characterized by the distinct L-type enrichment, the formation of which is associated with the REE introduction into the coal accumulation basins, mainly in the composition of clay minerals and LREE-phosphates (Table 1).

Oxidized coal exhibits the H-type distribution of rare earth elements. The normalized La/Yb ratio in the roof coals: 13.8; 15.8; 5.8 - demonstrate a strong relationship between rare earth elements and inorganic matter, since high values of the La/Yb ratio reflect the contribution of the terrigenous (inorganic) component to the light rare earth elements accumulation, as evidenced by significant barium enrichment [7].

The results of the data obtained indicates the existence of a series of unrelated sources and different mechanisms of REE accumulation in the sediments of the Shubarkol deposit.

Table 1 – Geochemical parameters of rare earth elements in the samples of coal and clay interlayers of the Shubarkol deposit

Coal seam	Sample	∑REE	La_n/Lu_n	La_n/Yb_n	δEu	δCe	Enrichment type	Sample	∑REE	La_n/Yb_n	δEu	δCe
	In coals							In clay interlayers				
2B	6	11.14	10.95	13.84	0.8	0.3	L type	7	23.4	5.35	0.82	0.67
	10	9.27	14.05	15.80	0.6	0.3	L type	8	26.1	5.39	0.87	0.80
	41	11.11	1.20	1.34	1.2	0.7	L type	9	14.5	5.83	0.68	0.46
	50	17.01	4.38	5.39	0.9	0.5	L type	42	66.2	1.61	0.74	0.93
	59	19.81	4.68	5.84	0.8	0.5	L type	43	133.9	1.37	0.81	0.92
1B2	68	20.16	4.55	5.01	0.8	0.6	L type	69	40.5	4.18	0.74	1.06
	72	22.88	2.73	3.17	0.8	0.9	L type	70	40.5	4.13	0.76	0.99
	76	13.59	2.42	2.88	1.0	0.5	L type	71	48.7	1.98	0.85	1.17
	85	56.77	1.88	2.04	0.8	0.8	L type	83	48.8	6.42	0.77	0.83
	86	87.06	0.44	0.51	0.8	1.1	H type	88	55.8	5.80	0.80	0.79
	100	43.42	1.47	1.69	0.9	0.9	L type	89	40.0	4.76	0.84	0.91
	104	50.39	1.08	1.14	1.0	1.1	L type	91	38.3	4.83	0.89	0.93
	105	36.29	2.46	2.46	0.8	1.0	L type	92	49.4	4.67	0.73	0.77
	109	32.09	2.26	2.24	0.9	1.0	L type	93	38.2	3.91	0.75	1.01
1B1	115	14.21	4.11	4.75	0.9	0.7	L type	-	-	-	-	-
	116	17.98	6.13	6.88	0.8	0.6	L type	-	-	-	-	-
	117	12.03	8.18	9.87	1.0	0.5	L type	-	-	-	-	-
	118	18.00	4.23	4.68	0.9	0.5	L type	-	-	-	-	-
	119	17.87	5.61	6.08	0.8	0.7	L type	-	-	-	-	-
	120	15.47	5.88	6.45	0.8	0.7	L type	-	-	-	-	-

Note. Alternation of: ∑REY = La + Ce + Nd + Sm + Eu + Tb + Yb + Lu; $\delta Eu = Eu_N/Eu_N^* = Eu_N/[(Sm_N \times 0{,}67)+(Tb_N \times 0{,}33)]$, $\delta Ce = Ce_N/Ce_N^* = Ce_N/[(La_N \times 0{,}67 + Nd_N \times 0{,}33)]$.

This can be explained by the strong enrichment of the peat deposit with terrigenous suspension within the period of transgression (of the reservoir) [8].

Granitoids could be a source of rare-earth. A certain contribution of acidic solutions enriched in rare earth elements is also assumed.

Enrichment of peat with piles of earth could occur during the acidic natural waters infiltration within the epigenetic period, as evidenced by a positive cerium anomaly. It is known that cerium migrates well in acidic waters and precipitates in an alkalized medium [9].

Table 1 also shows the appearance of a negative cerium anomaly in layers 2B and 1B1 (which fluctuate between 0.3-0.8), which is explained by the presence of authigenic minerals [7].

The Eu/Eu* value in clayey shales, silty mudstones, and mudstones is also an indicator of the composition of rock complexes eroded in paleowatersheds. In our case, the samples have moderate negative Eu anomalies (Eu/Eu*, the average value is 0.85-0.9), which indicates the predominance of Archean crystalline rocks or rocks formed by juvenile material that has not undergone significant transformation in the continental crust [10].

The authors revealed the principal regularities and features of the REE concentration in the Shubarkol deposit.

Based on Figure 2 and Table 1, it can be concluded that the elevated concentrations of all the REEs are characteristic of clay interlayers and oxidised coals. On this illustration, the La/Yb reciprocality escalations up the section, indicating a preponderantly clastogenic mechanism of REE supply to coals. This REE distribution is explained by the different sorption capacities of clay minerals.

The analysis of the lateral distribution of lanthanides in coals (Figure 3) showed that the contents clearly decreased from the periphery to the center of the deposit; this fact indicates a certain role of aqueous solutions in the accumulation of REE in coals [11]. A similar pattern of distribution is characteristic of carbonophilic elements.

The content of impurity elements in the coals of seams 2B, 1B2, and 1B1 of the Shubarkol deposit is shown in Table 2. The degree of enrichment with impurity elements of the Jurassic deposits of the Shubarkol deposit is estimated respectively compared with the clarke values for hard coals [12].

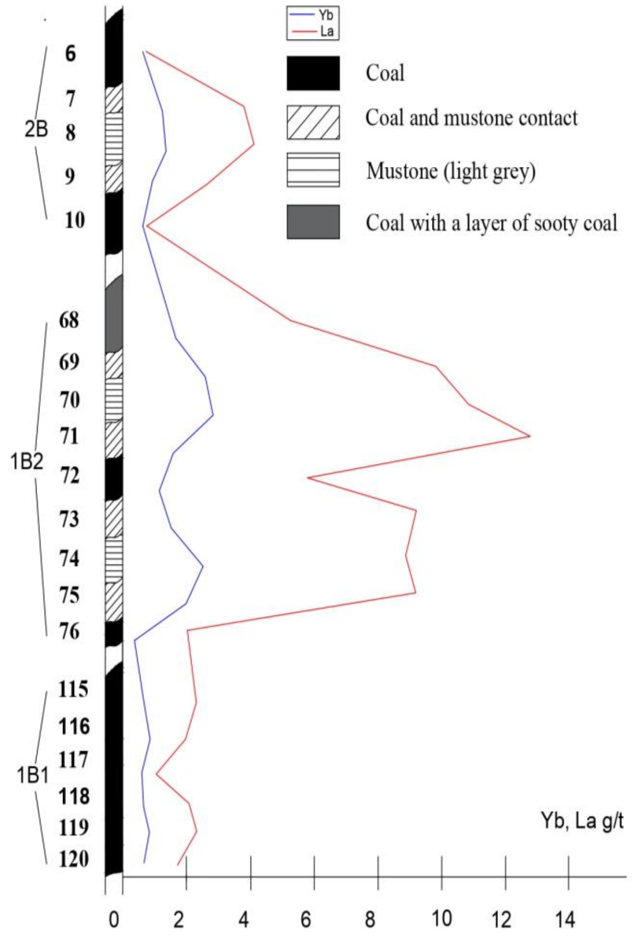

Figure 2 – La and Yb vertical distribution in the section of the Shubarkol deposit coal seams

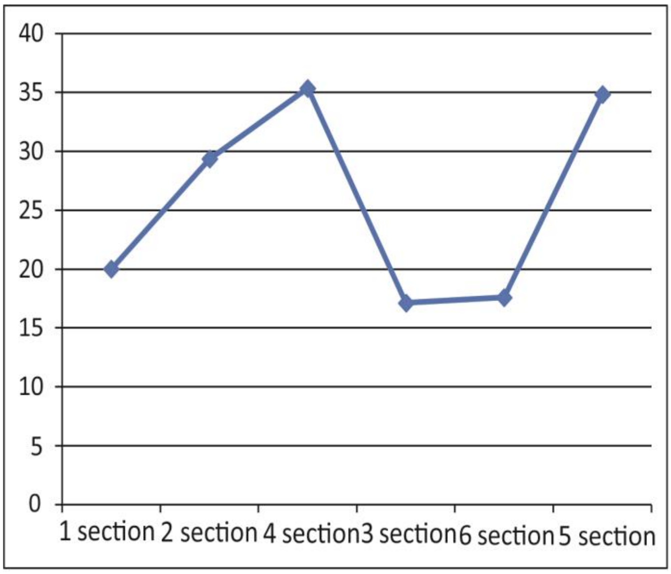

Figure 3 – Graph of lateral variability of rare earth elements from the 1st to the 6th sections of the Shubarkol deposit

The elements with the highest enrichment are Sr and Ba (ionic lithophiles) with the values of 10.1 and 21.7 (10<CC<100); slightly enriched are Th, Cr, As, Br, Rb, Zn, Ta (transit) (CC<0.5). The concentrations of other trace elements are close to the average values for global coals (0.5 < KK < 2) (Figure 4).

Apatite associations that can be obtained from bauxites on a weathered basement in the area of the sediment source have elevated contents of Sr and Ba. In addition, REE enrichment was found to be controlled by both groundwater and acidic hydrothermal fluids based on REE genetic types [13].

In this paper, the indicators of sensitive elements were studied for an attempt to reconstruct the paleoclimate.

The Sr/Ba ratio is one of the most widely used indicators for sedimentary rocks and coals [14], which indicates paleolonetzicity and paleoclimate, respectively.

Sr/Ba values > 1 and < 1 indicate, as a rule, arid and humid climatic conditions, respectively [14]. The values of this ratio in the studied area range from 0.3 to 2.4.

The formation of the Jurassic terrigenous system took place under essentially constant climatic and tectonic conditions, more precisely, the one changing within a very narrow framework. The climate, being generally humid, experienced some shift towards aridization at the very end of the system accumulation. This is evidenced by the Sr/Ba values, as well as the appearance of red-colored rocks and a slight increase in carbonate content [15].

Table 2 – Impurity elements content in the Shubarkol deposit coal samples

No.	Sm	Ce	Lu	U	Th	Cr	Yb	Hf	Ba	Sr	Nd	As	Ag	Br	Cs	Tb	Sc	Rb	Zn	Ta	Co	Eu	La	Sb
2B	1.2	8.7	0.1	0.4	0.6	6.0	0.7	0.2	29.1	39.0	3.9	0.9	0.4	0.7	0.2	0.2	4.1	0.8	7.4	0.0	8.3	0.3	1.9	0.3
1B2	3.4	39.7	0.1	1.0	1.3	5.1	0.7	0.9	3258.3	1010.9	21.2	1.1	0.4	0.8	0.7	0.4	7.1	2.9	8.7	0.0	2.4	0.9	20.6	0.2
1B1	0.8	5.9	0.1	0.2	0.1	1.8	0.7	0.1	7.1	39.0	3.0	0.9	0.4	1.5	0.0	0.3	2.9	3.0	5.7	0.0	10.1	0.2	1.1	0.2
W.C.	2.2	23	0.2	1.9	3.2	17	1	1.2	150.0	100	12	9	0.1	6	1.1	0.31	3.7	18	28	0.3	6.00	0.43	11	1

Note. W.C. – world coal, according to [12].

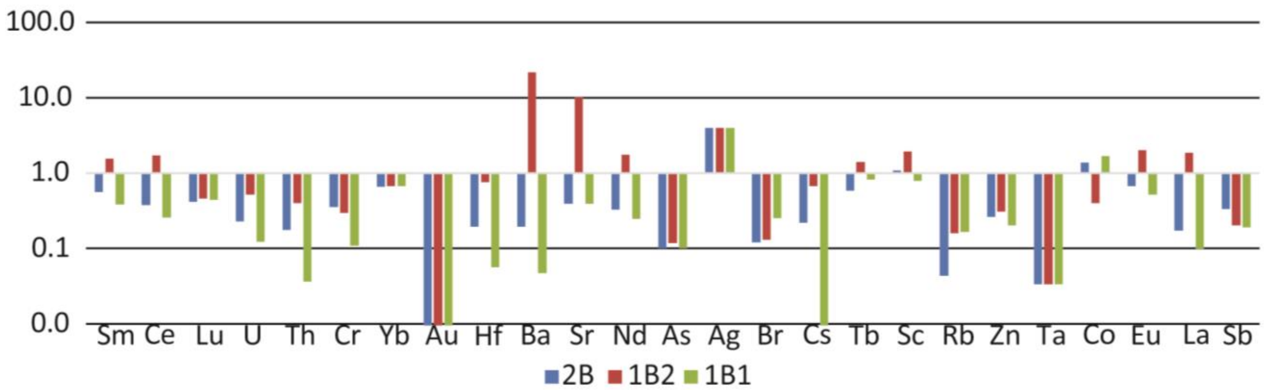

Figure 4 – Element concentration coefficients (CC) in coals that are normalized based on the average concentrations of microelements in hard world coals [12] and based on the classification of microelement enrichment

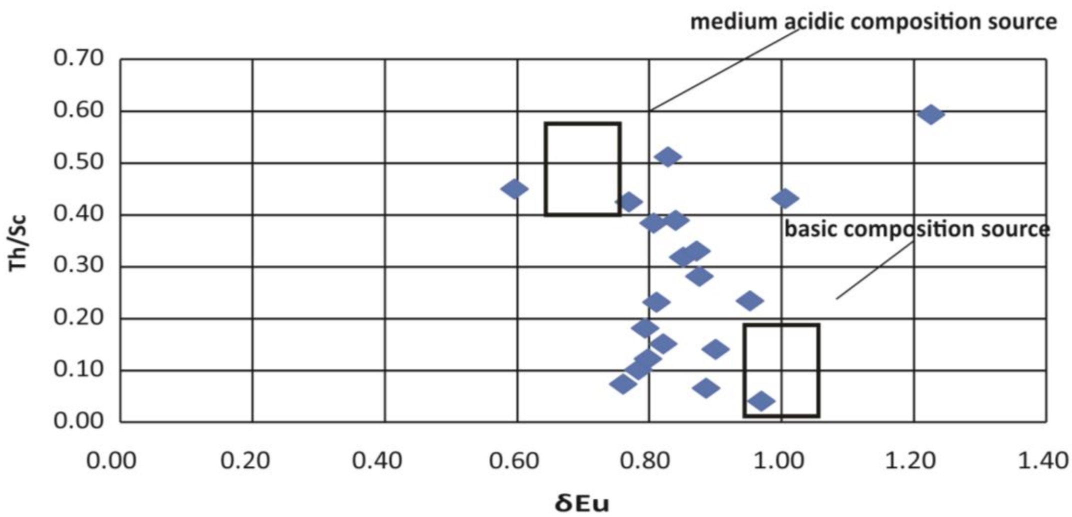

Figure 5 - The position of figurative points of the compositions on the discrimination diagram [17]

Based on the lithochemical features of clay shales, the climate was defined as arid-humid. The systematics of Sr, Ba, and Zr in clayey rocks according to the data of the reference [16] made it possible to establish that the sedimentation basin was freshwater.

The ratios of a number of indicator elements (Sc, Th, Eu) were also used to reconstruct the features of the composition of rocks in the feeding areas and a corresponding graph was built. The sources of rare earth elements for the studied rocks were both sufficiently mature rocks (presence of clay minerals in the initial sediments) and less altered petrogenic material of acidic composition (enrichment of the K-feldspar protolith). Elevated contents of Ba and Rb in the rocks are also indirect evidence of the presence of K-feldspar.

The diagram (Figure 5) used to determine the composition of rocks in the recharge areas shows that the sources of impurity elements for most of the studied samples were sedimentary formations enriched in quartz, as well as igneous rocks of medium acidic and basic composition.

Studying the REE occurrence forms plays an important role in obtaining data on the ways of their migration and concentration in coals and clay interlayers, and they are also significant geochemical indicators that make it possible to assess the nature of REE accumulation in coals, to consider the evolution of matter in the process of coalification and other epigenetic transformations.

According to the studies carried out, it was revealed that the mineral forms of finding rare, rare earth elements in coals and rocks had uneven distribution and differed in composition in the deposit. At the same time, many of the identified minerals were confined to areas of enrichment with organic matter: at the contact of enclosing rocks and coals, or in the coals themselves.

In coals, the bulk of the mineral matter is represented by kaolinite which contributes to the concentration of light REE. With kaolinite, the supply of REE within the peat stage in the composition of water and mineral nutrition should be associated.

Quartz predominates in the composition of petrogenic elements in coal seams in the studied area, which indicates that the source of the material can be mainly terrigenous fragments. Since most of the quartz in coal is terrigenous, it is transported by geological processes such as water or wind to peat bogs and stored in coal seams [18].

Based on the results of microscopic studies of the sample, there were found numerous inclusions of barite ($BaSO_4$) that had angular grains of irregular shape in the form of plates (Figure 6a), inclusions of sphalerite (Figure 6b) of a roundish shape on the microcrack of the sample, well-cut crystals of baddeleyite (Figure 6c) and zircon (Figure 6d).

Fragments of zircon crystals, as well as prismatic crystals, were found in the rocks. Individual zircon grains are irregularly shaped; they look either as rounded intergrown crystals or as neoformations. Baddeleyite is rarer than zircon. The shape of the grains is similar to zircon: prismatic, or grains of irregular shape.

The forms of occurrence of rare earth elements in coals in the form of aggregates of lamellar, foliar, columnar crystals, and fragments of prismatic crystals suggest the authigenic nature of their formation.

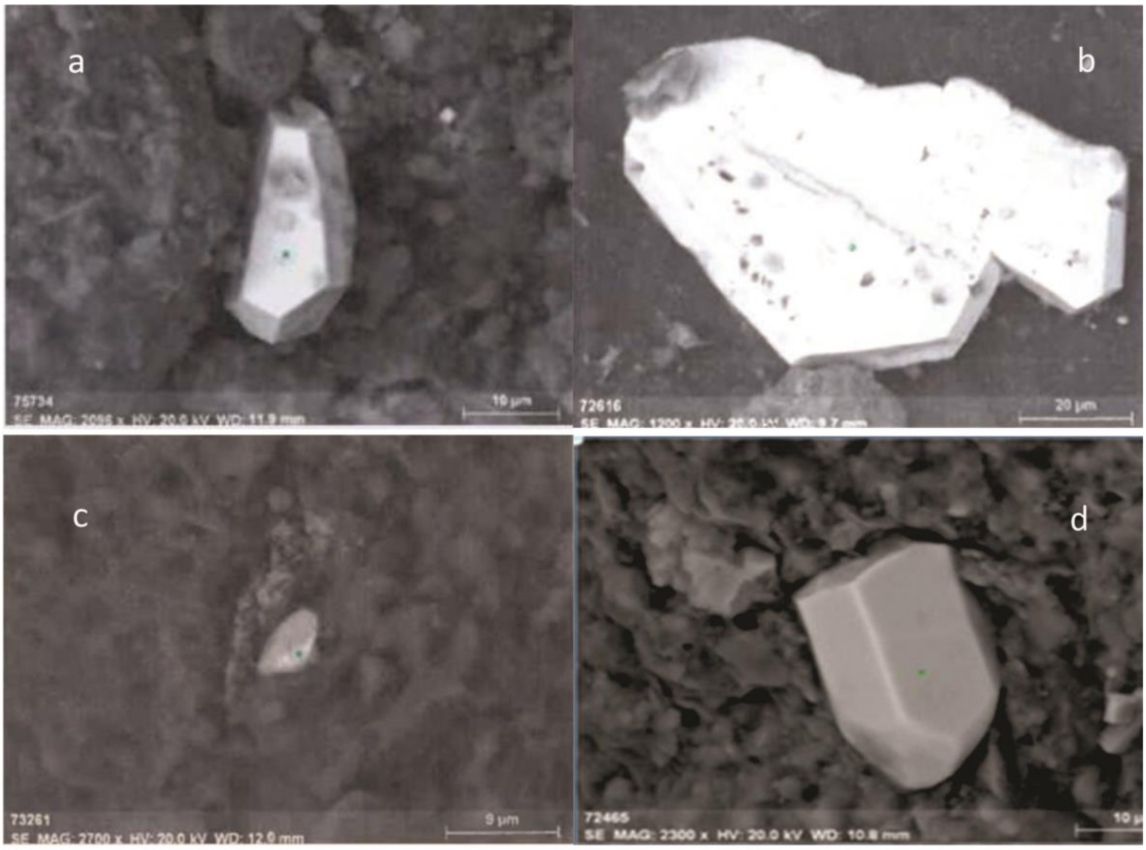

Figure 6 – Barite (a), sphalerite (b), baddeleyite (c), and zircon (d) crystals

Rare earth minerals in rocks are often found in or near organic matter. These minerals are less common in mudstone samples. Oxides form needle-shaped crystals, their aggregates, plates, and grains of irregular shape. This is turn points to various ways of REE migration into coals and coal-bearing rocks of the deposit.

Determining the degree of roundness of detrital grains is important when assessing the porosity and permeability of rocks since increasing the roundness of fragments (in cases of low cement content) contributes to the formation of pores with smooth walls, which in turn facilitates the migration of hydrocarbons. The very fact of roundness indicates that the mineral samples were redeposited, but the range of transfer judging by the poor processing, is small, or such a character indicates that the material was not transported by water flows.

The impurity element migration occurred with capturing denudation plains, stable processes of peat accumulation, and localization of useful components in watercourses adjacent to the plains [19].

Coal swamps/basins are generally low-energy environments not subject to heavy input of clastic materials.

Periodic cycles of geological transgression/regression (cyclic rise/fall of sea level relative to land) can introduce clastic materials into the basin between the coal layers. Finally, diapiric emplacement (upward movement of igneous material through the rock underlying the basin) can introduce REE materials into the region of the basin, where bound hydrothermal fluids can mobilize REE until they find a suitable receptor material such as kaolinite [20].

The weathering of rocks in the conditions of the warm humid Jurassic climate in the territory of Central Kazakhstan also entailed the movement of elements into the dissolved state and their transfer into water solutions. Carbonate rocks in the frame structures led to the organization of alkaline and subalkaline carbonate and hydrocarbonate waters.

Conclusions

The analysis of the obtained complex geochemical and mineralogical data made it possible to identify various ways of REE migration into coals and clay interlayers within the syngenetic and epigenetic periods of the Shubarkol deposit formation. It was established that peat accumulation occurred under essentially constant climatic and tectonic conditions, the climate was generally humid, experienced some shift towards aridization at the very end of formation accumulation, and the sedimentation basin was freshwater.

It was revealed that the impurity element migration occurred on denudation plains and in watercourses associated with plains in stable processes of peat accumulation. Clastogenic transportation took place by alkaline and sub-alkaline carbonate and hydrocarbonate waters from nearby rocks of the framing: sedimentary rocks enriched in quartz, terrigenous rocks, the initial ones that were basic, medium-acid igneous rocks, including granitoid. The addition also occurred with clay minerals.

Conflict of interests

On behalf of all the authors, the correspondent author declares that there is no conflict of interest.

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Шұбаркөл кен орнының көміріне сирек жер элементтерінің миграциясы және тасымалдану жолдары

Блялова Г.Г., Копобаева А.Н., Амангелдіқызы А., Асқарова Н.С., Ожигин Д.С.

Әбілқас Сағынов атындағы Қарағанды техникалық университеті, Қарағанды, Қазақстан

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Сараптамадан өтті: *30 маусым 2022*
Қабылданды: *12 шілде 2022*

ТҮЙІНДЕМЕ

Мақалада Шұбаркөл кен орнының көмір және саз аралық қабаттарының минералогиялық және геохимиялық ерекшеліктерін зерттеу нәтижелері берілген. Сазды қабаттардың 71 үлгісі және көмірдің 49 үлгісі аспаптық нейтронды белсендіру талдауы (INAA), ұнтақ рентгендік дифрактометрия және сканерлеуші электронды микроскопия арқылы талданды. Көмірлердің геохимиялық және минералогиялық сипаттамаларына және оларды қоршайтын саз аралық қабаттарына кешенді талдау жүргізілді. Әртүрлі геохимиялық критерийлер СЖЭ-нің көші-қоны туралы фактілерді анықтауға мүмкіндік берді, ал минералогияның соңғы деректері Шұбаркөл кен орнының қалыптасуының син- және эпигенетикалық кезеңдерінде олардың палеобассейнге тасымалдану жолдарын анықтауға мүмкіндік берді. Торфтың түзілу палеобассейннің палеоклиматы мен су құрамы туралы мәліметтер алғаш рет берілген. Сонымен қатар, талдау Шұбаркөл кен орнының шөгінділерінде сирек жер элементтерінің жинақталуының бірқатар тәуелсіз көздері мен әртүрлі механизмдерін анықтауға мүмкіндік берді. Жеңіл лантанидтердің ауыр лантанидтерден басым болуымен ерекшеленетін сирек жер элементтерінің (СЖЭ) таралуының негізгі заңдылықтары белгіленді, көмір кен орындарын қоршап тұрған тау жыныстарының СЖЭ шоғырлануындағы рөлі мен құрамы қарастырылды. Көмірлер мен саз аралық қабаттарда сирек және сирек жер элементтерінің микробөлшектерінің қосындылары бар алюмосиликаттар, сульфидтер және сульфаттар табылды.

Түйін сөздер: көмір, Шұбаркөл, сирек жер элементтері, минералогия, геохимия.

Авторлар туралы ақпарат:

Блялова Гулим Галымжановна — *докторант, «Геология және пайдалы қазба кен орындарын барлау» мамандығы бойынша техника ғылымдарының магистрі, Әбілқас Сағынов атындағы Қарағанды техникалық университеті, 100000, Қарағанды, Қазақстан. Email: gulim_blyalova@mail.ru; ORCID ID: https://orcid.org/0000-0001-8801-8683*

Копобаева Айман Ныгметовна — *доктор PhD, «Геология және пайдалы қазбалар кен орындарын барлау» кафедрасының аға оқытушысы, Әбілқас Сағынов атындағы Қарағанды техникалық университеті, 100000, Қарағанды, Қазақстан. Email: aiman_25.87@mail.ru; ORCID ID: https://orcid.org/0000-0002-0601-9365*

Амангелдіқызы Алтынай — *доктор PhD, «Геология және пайдалы қазбалар кен орындарын барлау» кафедрасының аға оқытушысы, Әбілқас Сағынов атындағы Қарағанды техникалық университеті, 100000, Қарағанды, Қазақстан. Email: amangeldykyzy@inbox.ru; ORCID ID: https://orcid.org/0000-0002-6665-8804*

Асқарова Назым Сражадинқызы — *докторант, «Геология және пайдалы қазба кен орындарын барлау» мамандығы бойынша техника ғылымдарының магистрі, Әбілқас Сағынов атындағы Қарағанды мемлекеттік техникалық университеті, 100000, Қарағанды, Қазақстан. Email: srajadin-nazym@mail.ru; ORCID ID: https://orcid.org/0000-0002-2103-6198*

Ожигин Дмитрий Сергеевич — *доктор PhD, «Маркшейдерлік іс және геодезия» кафедрасының аға оқытушысы, Әбілқас Сағынов атындағы Қарағанды техникалық университеті, 100000, Қарағанды, Қазақстан. Email: ozhigin.dima@mail.ru; ORCID ID: https://orcid.org/0000-0002-2443-3068.*

Пути миграции и транспортировки РЗЭ в угли месторождения Шубарколь

Блялова Г.Г., Копобаева А.Н., Амангельдыкызы А., Аскарова Н.С., Ожигин Д.С.

Карагандинский технический университет имени Абылкаса Сагинова, Караганда, Казахстан

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АННОТАЦИЯ
В статье представлены результаты исследования минералого-геохимических особенностей углей и глинистых прослоев месторождения Шубарколь. Пронализированы 71 проба глинистых прослоев и 49 проб угля методоми инструментального нейтронно-активационного анализа (ИНАА), порошковой рентгеновской дифрактометрии, а также сканирующей электронной микроскопии. Проведен комплексный анализ геохимических и минералогических характеристик углей и вмещающих их глинистых прослоев. Разнообразные геохимические критерии позволили установить факты о миграции РЗЭ, а приведенные новейшие данные о минералогии позволили установить пути их транспортировки в палеобассейн в син- и эпигенетические периоды формирования месторождения Шубарколь. Впервые приведены данные о палеоклимате и водном составе палеобассейна торфообразования. В том числе проведенный анализ помог определить ряд независимых источников и различных механизмов накопления РЗЭ в отложениях месторождения Шубарколь. Установлены основные закономерности распределения редкоземельных элементов (РЗЭ), которые выражены в преобладании легких лантаноидов над тяжелыми, рассмотрена роль и вывлен состав пород обрамления угольных залежей в концентрации РЗЭ. Обнаружены алюмосиликаты, сульфиды и сульфаты с включениями микрочастиц редких и редкоземельных элементов РЗЭ в углях и глинистых прослоях.
Ключевые слова: уголь, Шубарколь, редкоземельные элементы, минералогия, геохимия.

Информация об авторах:

Блялова Гулим Галымжановна — *докторант Карагандинского технического университета имени Абылкаса Сагинова, магистр технических наук по специальности Геология и разведка МПИ, 100000, г. Караганда, Казахстан. Email: gulim_blyalova@mail.ru; ORCID ID: https://orcid.org/0000-0001-8801-8683*

Копобаева Айман Ныгметовна — *доктор PhD, старший преподаватель кафедры «Геология и разведка МПИ» Карагандинского технического университета имени Абылкаса Сагинова, 100000, г. Караганда, Казахстан. Email: aiman_25.87@mail.ru; ORCID ID: https://orcid.org/0000-0002-0601-9365*

Амангельдыкызы Алтынай — *доктор PhD, старший преподаватель кафедры «Геология и разведка МПИ» Карагандинского технического университета имени Абылкаса Сагинова, 100000, г. Караганда, Казахстан. Email: amangeldykyzy@inbox.ru; ORCID ID: https://orcid.org/0000-0002-6665-8804*

Аскарова Назым Сражадинқызы — *докторант Карагандинского технического университета имени Абылкаса Сагинова, магистр технических наук по специальности Геология и разведка МПИ, 100000, г. Караганда, Казахстан. Email: srajadin-nazym@mail.ru; ORCID ID: https://orcid.org/0000-0002-2103-6198*

Ожигин Дмитрий Сергеевич — *доктор PhD, старший преподаватель кафедры «Маркшейдерское дело и геодезия» Карагандинского технического университета имени Абылкаса Сагинова, 100000, г. Караганда, Казахстан. Email: ozhigin.dima@mail.ru; ORCID ID: https://orcid.org/0000-0002-2443-3068*

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