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BERYLLIUM, SCANDIUM, YTTRIUM AND YTTERBIUM IN SOME BULGARIAN COALS

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ABSTRACT

The occurrence of Be, Sc, Y and Yb in the coals of different rank (ranging from lignite to anthracite) from fourteen Bulgarian basins and deposits were investigated. The concentrations of all elements in the coal and coal ash were higher than the Clarke values from the Sofia Basins and partly from the Karlovo Basin only. The element content in the coal and coal ash from the Maritza-West, Belibreg, Stanyantzy, Oranovo, Katriste, Suhostrel and Balkan basins were lower than the Clarke. In other coals the concentrations of some elements only were high than the Clarke values. These relations were applied and to the coal shale (with exception of Yb from Samokov, Katriste and Balkan Basin). The correlation coefficients between element concentration and ash content were identical in the coal from Maritza-West, Belibreg, Sofia, Karlovo, Samokov, Oranovo and Pernik (negative coefficients) and Katriste and Suhostrel (positive coefficient). For the other coals the correlation between element and ash content was different. The negative correlation coefficients with ash was highest (from -0.7 to -0.8) for Be (Maritza West and Sofia), Y (Kyustendil) and Yb (Karlovo). Beryllium, Sc, Y, Yb were correlated very often. The associations Sc-Y (Sofia, Maritza West, Belibreg), Y-Yb-Be (Pernik, Svoge, Kyustendil), Be-Sc (Samokov, Suhostrel), Yb-Be (Oranovo, Sofia), Be-Sc-Yb-Y (Karlovo) were established in the investigated coals. Beryllium, Sc, Y, Yb were associated with Ge, Ga, Zr, REE, Al, Si, sometimes with Ca, Mg, S also. The occurrence and concentrations of the investigated elements in Bulgarian coals were depend on some factors: 1) occurrence in the rocks from the peat bog feeding province and 2) the pH values in the ancient peat bogs, which control the fixation of elements in the metal-organic compounds or clay minerals. The degree of fissuring of the coal seams and the presence and composition of mineral waters, which precipitate infiltrational minerals into the fissures of the coal beds were not influenced of the occurrence of the studied elements.

Key words: beryllium, scandium, yttrium, ytterbium, coal, occurrence of elements, factors for accumulation.

INTRODUCTION

The concentration and distribution of beryllium, scandium, yttrium and ytterbium in the coal and coal shale from 14 Bulgarian basins and deposits were investigated. The coal age and rank were different. The lignite from eight Neogene basins and deposits (the Sofia, Beli Breg, Maritza-West, Karlovo, Samokov, Kyustendil basins, Gabrovitza and Katrishte deposits) were studied. The studied sub-bituminous coal was from the Oranovo-Simitli Basin with Miocene age and Pernik Basin with Paleogene age. The bituminous coal was from the Suhostrel deposit with Eocene age and Balkan Basin with Cenomanian age. The studied anthracite was from the Svoge Carboniferous Basin. Data for the concentration and distribution of these elements in other Bulgarian coals were published by Ескенази (1965), Eskenazy (1970, 1978, 1987a,b, 1995).

METHODS

Seven hundred and seventy coal and coal shale samples were studied. All samples were ashed at 800°C and analyzed using Instrumental Neutron Activation Analysis (INAA) and Inductively Coupled Plasma Mass Spectrometry (ICP-MS) analysis. The results were examined statistically and the correlation coefficients between the ash content and the element concentration were determined.

RESULTS AND DISCUSSION

Beryllium. The Be concentration in the coal ash from the Sofia, Karlovo, Kyustendil basins and Gabrovitza deposit is higher (1.4-7.9 times) than the Clarke values after Юдович и др. (1985) (Table 1). The Be content in the coal from the same basins and the Samokov and Svoge basins is higher than the Clarke values and world-wide averages from Swaine (1990) also (Table 1). The concentration of Be in the coals and coal ashes from the other basins is lower of the Clarke values (Table 1). The Be content is very below the Clark and the world-wide averages from Swaine (1990) in the coal from Beli Breg, Oranovo, Balkan and Suhostrel. The Be concentration in the Maritza-West lignite is about the Clarke values. The concentration of the Be in the coal shale ashes is less than its concentration in the coal ashes, but it is higher than the Clark (Table 2). The Be concentration is lower than the Clark only in the Maritza-West, Beli Breg, Stanyantzi and Oranovo coal shale ashes. Probably the Be concentration in the rock formations from the waterside line of the ancient peat bogs is high because the element content in the coal and coal shale from the Karlovo, Gabrovitza and Kyustendil is higher than the Clarke values.

Many authors report data for beryllium affinity (Table 3). Eskenazy (1970) published data for the Be connection with the organic acids. The Be has negative coefficient of correlation with the ash content in the most studied coals. Its concentration decreases, when the ash content increases, with an exemption of the Katrishte and Suhostrel coal (Fig. 1). The correlation coefficient of the element with the ash content in the

Balkan bituminous coal is below the statistical mean value and the concentration of the element varies in short interval (Fig. 1). Beryllium presents mainly into the low-ash coals. It has positive coefficients of correlation with the S (+0.57), Ca and Mg (+0.45) and negative coefficient with the Si and Al in the Sofia lignite (Kortenski and Sotirov, 2002). Probably one part of Be amount is connected with the sulfides and/or sulphates.

Table 1. Average content of the elements in the coal ash

Basins	Number of the samples	Content in the coal ash of:				Content in the coal of:			
		Be, ppm	Sc, ppm	Y, ppm	Yb, ppm	Be, ppm	Sc, ppm	Y, ppm	Yb, ppm
Maritza-West	38	11.1	4.6	10.1	n.d.	2.0	0.8	1.8	n.d.
Beli Breg	38	2.0	1.0	0.9	n.d.	0.4	0.2	0.2	n.d.
Stanyantzi	39	7.7	n.d.	n.d.	n.d.	1.3	n.d.	n.d.	n.d.
Sofia	59	15.0	42.2	80.1	5.0	4.3	19.4	17.4	2.2
Karlovo	23	54.7	8.0	43.5	6.6	11.0	1.6	8.8	1.3
Samokov	31	8.4	26.0	31.0	1.6	2.9	9.1	10.8	0.6
Gabrovitza	40	86.6	4.1	4.1	5.2	32.6	1.5	1.5	2.0
Kyustendil	39	37.9	15.2	21.2	3.2	11.5	4.6	6.4	1.0
Oranovo	61	3.3	n.d.	0.7	1.8	0.3	n.d.	0.1	0.2
Katrishte	22	5.1	n.d.	n.d.	n.d.	1.5	n.d.	n.d.	n.d.
Pernik	35	7.6	18.7	24.1	2.5	1.8	4.5	5.9	0.6
Suhostrel	17	4.1	6.8	n.d.	n.d.	1.2	2.0	n.d.	n.d.
Balkan	58	5.8	4.8	8.3	6.0	1.6	1.2	2.1	1.5
Svoge	90	7.0	22.8	3.0	6.5	2.9	9.3	1.2	2.7
Clark for lignite and sub-bituminous coal[1]		11.0	15.0	37.0	5.0	2.4	2.0	7.0	0.9
Clark for bituminous coal and anthracite[1]		21.0	20.0	47.0	7.0	2.1	3.0	6.0	0.8
World-wide averages[2]						1.5-2	1-10	2-50	0.3-3

1 – after Юдович и др. (1985); 2 - after Swaine (1990); nd – no data.

Table 2. Average content of the element in the coal shale ashes

Basins	Number samples	Be ppm	Sc ppm	Y ppm	Yb ppm
Maritza-West	9	1.6	2.0	6.5	n.d.
Beli Breg	8	1.1	0.4	0.5	n.d.
Stanyantzi	10	2.0	n.d.	n.d.	n.d.
Sofia	19	7.5	33.5	41.5	3.8
Karlovo	9	13.2	4.3	17.3	1.1
Samokov	10	6.2	24.0	23.0	1.5
Gabrovitza	8	71.7	6.0	5.5	5.6
Kyustendil	9	14.6	14.8	9.2	1.3
Oranovo	19	n.d.	n.d.	n.d.	n.d.
Katrishte	10	10.2	n.d.	3.2	7.1
Pernik	9	4.1	15.8	16.9	1.8
Suhostrel	8	10.1	8.6	n.d.	n.d.
Balkan	31	6.0	16.5	6.5	4.0
Svoge	21	5.7	26.8	7.7	4.4
Clark for shales by (Turekian and Wedepohl, 1961)		4.0	13.0	26.0	2.6

n.d.-no data

Probably Be in the coal is not connected with the plants, because it is only 0.1 ppm into them (Bowen, 1966). The acidity of the environment probably had not been important factor for the Be accumulation. Basins with high Be content in the coal had different acidity (Sofia 3.5-7; Karlovo 3.5-6.5; Gabrovitza 5-7.5) (Kortenski, 1992; Kortenski et al., 1997). The fissures and the epigenetic mineral solutions obviously had not influenced on the accumulation and concentration of Be,

because the Be concentration is low in the high-fissured and mineralized Svoge anthracites, Balkan and Suhostrel bituminous coal and the Pernik sub-bituminous coal. Probably, the main reason for the high concentration of the element had been the presence of the Be in the rocks around the basins. For example the relationship between the Be concentration in Sofia lignite and the Cr concentration in the rocks from the waterside line (Vitosha pluton) is established. The Cr content in the andesites from Vitosha pluton is 13.2 ppm (Kortenski, 1986).

Scandium. The concentration of Sc in the most of the studied coals is lower than the Clark by Юдович и др. (1985) (Table 1). The Sc concentration is about 3 times higher than the Clark in the ash of the Sofia lignite and it is little higher than the Clark in the ash from the Samokov, Pernik and Svoge coal. The Sc is approximately to the Clark value in the ash of the Kyustendil lignite (Table 1). The Sc concentration is higher than the Clark by Юдович и др. (1985) in the coal of the Sofia, Svoge, Samokov, Kyustendil and Pernik basins. Only the Sofia lignite contains Sc more than the maximum level of the interval by Swaine (1990) (Table 1). The scandium concentration in the coal shale ashes from the Sofia, Samokov, Kyustendil, Pernik, Balkan and Svoge basins is higher than the Clark (Table 2).

The Sc has positive coefficient of correlation with the ash content in the coals from Gabrovitza, Suhostrel, Balkan and Svoge basins. The element concentration increases with ash content increasing (Fig. 1). The Sc has positive correlation with the aluminum-silicate part of the inorganic matter in the Sofia lignite (Kortenski, Sotirov, 2000). The Sc has negative correlation with the coal ash from the West-Maritza, Beli Breg, Sofia, Pernik and Karlovo basins and Figure 1 shows that with the increasing of the ash content; the Sc concentration is decreasing. The correlation coefficient with the ash is lower than the statistical mean value in the coal from Samokov and Kyustendil. The Sc concentration nearly not changes many with the ash contents change (Fig. 1). Many authors report data for affinity of the Sc (Table 3). Querol et al. (1997a,b) established positive correlation of the element with the sulphur and the aluminum-silicate content of the coal.

Table 3. Reference data of the Be, Sc, Y and Yb affinities

Ele-ment	Reference data of the element affinities		
	Organic	Intermediate	Inorganic
Be	Юровский (1960); Смирнов (1968); Gluskoter et al. (1977); Kuhn et al. (1980); Perrek and Bardhan (1985); Miller and Given (1987); Querol et al. (1992, 1997a); Warwick et al. (1997).	Kojima and Kurusawa (1986); Querol et al. (1996).	Минчев и Ескенази (1972); Beaton et al. (1991); Querol et al. (1997b).
Sc	Минчев и Ескенази (1965); Юдович и Шасткевич (1965); Смирнов (1966); Parrek and Bardhan (1985)	Юровский (1960); Kojima and Kurusawa (1986); Beaton et al. (1991); Warwick et al. (1997); Crowley et al. (1997).	Pippiringos (1966), Минчев и Ескенази (1972), Ескенази и Минчева (1994); Querol et al. (1997a,b)
Y	Gluskoter et al. (1977); Минчев и Ескенази (1965, 1972); Ескенази и Минчева (1983, 1994); Юдович и др. (1985); Miller and Given (1987); Querol et al. (1997a).	Kojima and Kurusawa (1986), Crowley et al. (1997), Warwick et al. (1997).	Юровский (1960); Pippiringos (1966); Spears, Martinez-Tarazona (1993); Querol et al., (1996); Querol et al. (1997b).
Yb	Ершов (1961); Miller and Given (1987); Eskenazy (1995).	Querol et al. (1992); Crowley et al. (1997); Warwick et al. (1997).	Beaton et al. (1991); Ескенази и Минчева (1994); Querol et al. (1997a,b).

The concentration of the Sc in the plants is 0.008 ppm (Bowen, 1966) and it is not important for the accumulation of the element in the coal. But the presence of the element in the rock formations, which build the coal basin waterside, is important. The concentration of the element is higher in the coal and coal ashes from Sofia, Samokov, Pernik and Svoge basins, but it is not so high in the coal from the Maritza-West and Beli Breg basins. It depends from the Sc concentration in the rock formations. Юдович и др. (1985) suggest that if the concentration of the Sc is higher than 50 ppm the element has organic affinity. It has mainly inorganic affinity when the Sc concentration is high, but less than 50 ppm. The environmental acidity of the peat bogs of the Sofia, Samokov, Pernik and Svoge basins had been very different (Kortenski, 1992), but the acidity had been optimal for connection of the Sc with the clay minerals. Юровский (1960) was established that Sc frequently presents in the carbonate minerals. The higher Sc concentration in the Pernik and Svoge coals probably is a result of the high fissuring and presence of the epigenetic carbonate mineralization.

Yttrium. The concentration of the element in the most of the studied coal is low. Only the ash from the Sofia and Karlovo lignite contains Y higher than the Clark value (Table 1). The concentration of Y is below the interval determined by Swaine (1990) in the coal from Maritza-West, Beli Breg, Gabrovitza, Oranovo and Svoge. The concentration of the element in all other coals is into that interval (Table 1). The concentration of the element in the coal shale is much lower than the Clark with an exemption of the Karlovo and Samokov basins (Table 2). It is established that the concentration of the element is high in the coal and the coal shale from the Sofia, Karlovo and Samokov basins, which is probably a result of the presence of Y in the rock formations around the basin.

The reference data for prevailing affinities of Y is shows by Table 3. The Y has positive coefficient of correlation with the ash content only in the coal from Gabrovitza, Svoge and Katrishte, but it has negative coefficient in all other coals. The concentration of Y significantly decreases, when the ash content increases in the coal from Kyustendil, Oranovo, Karlovo and Sofia (Fig. 2). The Y concentration decreases

insignificantly, when the ash content increases in all other coals with negative correlation between Y and ash content. The Y content increases when the ash content increases in the coal from Gabrovitza, Katrishte and Svoge.

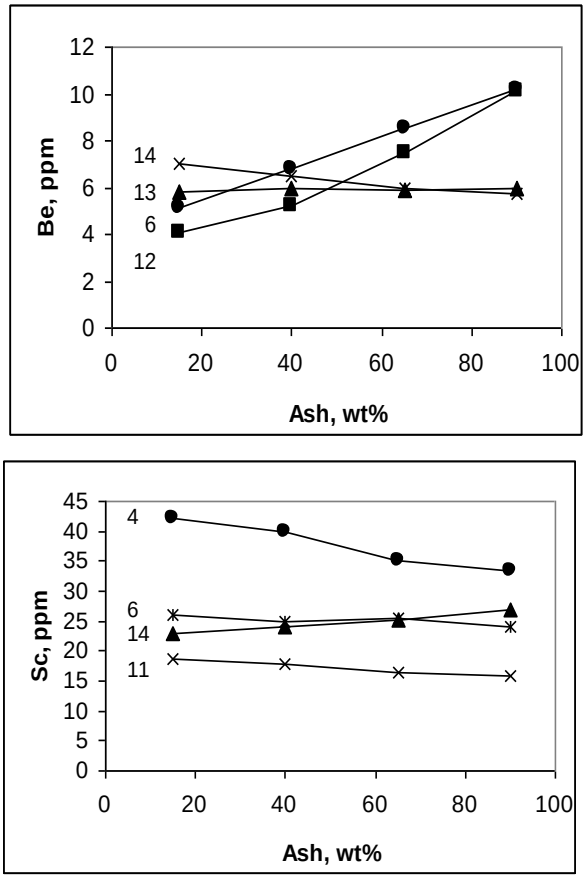

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Figure 1. Distribution of average Be and Sc content versus ash content:
4-Sofia; 6-Samokov; 11-Pernik; 12-Suhostrel; 13-Balkan; 14-Svoge.

The yttrium concentration in the plants is insignificant, about 0.03 ppm (Bowen 1966) and it had not been important factor for the concentration of the element in the coal. The presence of the element in the rock formations, around the basin had been important. The conditions of the peat bogs had not been appropriate for the accumulation of the element. The acidity of the peat bogs for studied basins had been between 3.5 and 7 (Kortenski, 1992). The fissuring and the epigenetic mineralization had not influenced on the Y accumulation also. For an example the coal seams from the Balkan and Svoge basins have high fissuring and epigenetic mineralization, but the concentration of the Y is 8-12 times lower than the Clark (Table 1).

Ytterbium. The concentration of the element is higher than the Clark only in the coal ash from Karlovo. It is about the Clark in the ashes from the Sofia, Gabrovitza, Balkan and Svoge basins (Table 1). The concentration of the element is into the interval of the average rank by Swaine (1990) with an exemption of the Oranovo and Katrishte coal. Some coal shale ashes (from the above-mentioned four basins - Sofia, Balkan, Svoge and Gabrovitza) contain more Yb than the Clark (Table 2). The content of Yb is higher from the Clarke values in the coal shale from the Katrishte deposit, but the element is not established in the coal from that deposit (Table 1, 2).

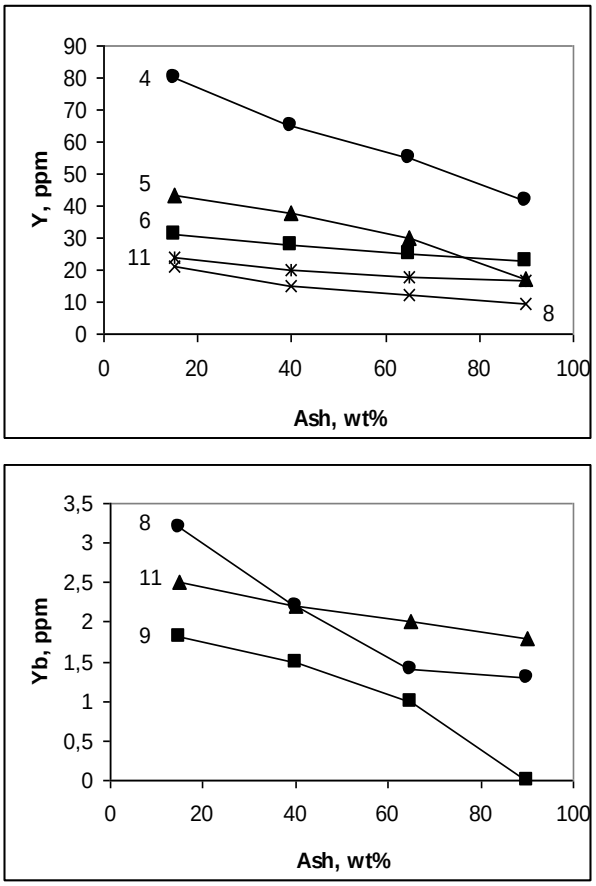

Figure 2. Distribution of average Y and Yb content versus ash content:
4-Sofia; 5-Karlovo; 6-Samokov; 8-Kyustendil; 9-Oranovo; 11-Pernik

The coefficient of correlation of the Yb concentration with the ash content in the Katrishte coal has high positive value. The Yb concentration increases, when the ash content increases. Probably the Yb into that coal has inorganic affinity and it is connected mainly with the clay minerals. The Yb has negative correlation coefficient with the ash content for all studied coals with an exemption of the coal from the Katriste and Gabrovitza. The concentration of the Yb decreases significantly with the increasing of the ash content, especially in the coal with high negative coefficient of correlation (Karlovo, Oranovo and Kyustendil) (Fig. 2). The correlation coefficients between the element and the ash content have values below the statistical mean value and the concentration of the element changes insignificantly with the changing of the ash content in the coal from Samokov and Gabrovitza. The reference data for prevailing affinities of Yb is shows by Table 3.

The concentration of the Yb is low in the studied coals. The major probable reason is the low content of the element in the rock formations from ancient waterside line or inappropriate conditions for Yb accumulation in the ancient peat bogs. For example the rocks from waterside line of Sofia basin contain from 0.7 to 1.5 ppm Yb (Кортенски, 1986).

CONCLUSIONS

Berilium, Sc, Y, Yb concentrations are higher than the Clarke values in the Sofia lignite and coal ash. This is valid and for the Karlovo coal (with exception of Sc). The contents of the four studied elements are below that the Clark and the interval of World-wide averages by Swaine (1990) in the coals from the Maritza-West, Beli Breg, Stanyantzy, Oranovo, Katrishte, Suhostrel and Balkan. The concentrations of the four elements are higher or lower than the Clark in the other coal. The same is valid for the coal shale, with an exemption of Yb concentration in the Samokov, Katrishte and Balkan coal shale.

The elements have similar coefficients of correlation with the ash content in the coal from Maritza-West, Beli Breg, Sofia, Karlovo, Samokov, Oranovo and Pernik (negative) and Katrishte and Suhostrel (positive), but the coefficients are different for all other coals. The negative correlation coefficients are highest (from -0.7 to -0.8) for the Be (Maritza-West and Sofia), for the Y (Kyustendil) and for the Yb (Karlovo). The organic affinity of these elements is high. The elements, which have negative correlation coefficients with the ash content, are with prevailing organic affinity. All studied elements show the inorganic affinity in the coal from Katriste, Suhostrel and Gabrovitza (with exception of Be), Sc has inorganic affinity in the Balkan and Svoge coal and Y – in the Svoge, because its correlation coefficients with the ash content are positive. Be (in Balkan coal), Sc (in Samokov and Kyustendil coal) and Yb (in Samokov and Gabrovitza coal) has the intermediate affinity, because its correlation coefficients with the ash content are below the statistical mean value (positive or negative).

The four elements frequently associate between one with another. The frequently established associations are Sc-Y (Sofia, Maritza-West, Beli Breg), Y-Yb-Be (Pernik, Svoge, Kyustendil), Be-Sc (Samokov, Suhostrel), Yb-Be (Oranovo, Sofia) and Be-Yb-Sc-Y (Karlovo). The elements frequently associate with Ge, Ga, Zn, Al, and Si and sometimes with Ca, Mg and S.

The presence of the elements in the plants is insignificant. Probably the rock formations from the ancient waterside line of the basins had been the main source of the Be, Sc, Y and Y. The clay minerals and sometimes the organic matter had been the main concentrator of the elements, when the conditions had been appropriate. The fissures and the epigenetic mineralization had not been much important for the presence and the accumulation of the studied elements.

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