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CORRELATIONS BETWEEN ELEMENTS IN ORES FROM THE GOLD-COPPER DEPOSIT CHELOPECH, BULGARIA

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ABSTRACT: The correlations between major and minor chemical elements in the ores from Chelopech gold-copper deposit are established by statistical processing of 19,200 multi-element ICP analysis of samples, analyzed for content of 36 elements (Ag, Al, As, Au, Ba, Be, Bi, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, La, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, S, Sb, Sc, Se, Sn, Sr, Te, Ti, V, W, Y and Zn). The studied elements are analyzed by computing of the correlation matrix, which marks some well developed relationships between pairs of elements, such as Fe-S, Cu-As, Cu-Bi, Cu-Cd, Cd-As, K-Na and other. An inverse correlation of the elements, involved in composition of main ore minerals in the deposit (Fe, Cu, Au, As, S), and highly mobile elements (K, P, Y, La, Rb), which are extracted from the rocks in the hydrothermal processes is observed. The correlation scatterplots of some typical elements with strong, moderate and weak relationships are prepared. Particular attention in the interpretation of results is paid to тhe elements with high positive correlation coefficients and participating in composition of ore minerals and mineral associations found in the deposit. Trends of correlation of typical trace elements in primary and secondary ore minerals with main mineral-forming elements are established (Cd-Cu, Cd-As, Co-Fe, Co-Au, etc.). The increased content of Cu in the sphalerite from the central sectors of ore bodies in the deposit is the result of so called "chalcopyrite disease", which is the most likely reason for the high correlation between the Cu and Cd. Frequent impurities of Co, Te, and Se in pyrite are the reason for the average high correlation of Fe with these elements. The results of present study indicate that the correlation analysis is a powerful tool for establishing interdependences between the chemical elements, involved in the composition of the ore and host rock minerals in Chelopech deposit.

Key words: correlation, chemical elements, ore minerals

КОРЕЛАЦИОННИ ВРЪЗКИ МЕЖДУ ЕЛЕМЕНТИТЕ В РУДИТЕ ОТ ЗЛАТНО-МЕДНО НАХОДИЩЕ ЧЕЛОПЕЧ, БЪЛГАРИЯ

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РЕЗЮМЕ. Корелационните връзки между главни и съпътстващи химични елементи в рудите от златно-медно находище Челопеч са установени чрез статистическа обработка на 19200 броя мултиелементни ICP анализи на проби, изследвани за съдържание на 36 елемента (Ag, Al, As, Au, Ba, Be, Bi, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, La, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, S, Sb, Sc, Se, Sn, Sr, Te, Ti, V, W, Y и Zn). Изследваните елементи са анализирани чрез изготвяне на корелационна матрица, която маркира някои изявени връзки между двойки елементи като Fe-S, Cu-As, Cu-Bi, Cu-Cd, Cd-As, K-Na и други. Наблюдава се обратна корелационна зависимост между елементите, участващи в състава на главни рудни минерали в находището (Fe, Cu, Au, As, S), и силно мобилните елементи (K, P, Y, La, Rb), които се извличат от скалите при хидротермалните процеси. Изготвени са корелационни диаграми на някои типични елементи с изявена силна, средна и слаба взаимовръзка. Особено внимание при интерпретацията на резултатите е обърнато на елементи с висок положителен коефициент на корелация помежду си, и участващи в състава на рудните минерали и минерални асоциации открити в находището. Установени са тенденции на взаимовръзка на типични елементи-примеси в главни и второстепенни рудни минерали с основни минералообразуващи елементи (Cd-Cu, Cd-As, Co-Fe, Co-Au и други). Повишените съдържания на Cu в сфалерита от централните участъци на рудните тела в находището са резултат от т. нар. „халкопиритова болест", което е най-вероятната причина за високата корелация между Cu и Cd. Чести примеси от Co, Te и Se в пирит са причина за средно високата взаимовръзка на Fe с тези елементи. Резултатите от настоящото изследване показват, че корелационният анализ е мощен инструмент за установяване на взаимозависимости между химични елементи, участващи в състава на рудните минерали и вместващите скали в находище Челопеч.

Ключови думи: корелация, химични елементи, рудни минерали

Introduction

The Chelopech deposit is one of the largest gold and copper deposits in Europe, contributing to the advancement of the Republic of Bulgaria to 2nd place in the gold mining industry in the European Union and the 42nd in the World in 2015th year (Reichl et al., 2015).

The main task in present study is clarification of correlations between major and minor chemical elements in the ores of the gold-copper deposit Chelopech. The received results helps to confirm the known geochemical zonal models and the relationship between ore-forming processes and hydrothermal alterations, took place in the deposit.

Materials and methods

The correlations between major and minor chemical elements in the ores from the gold-copper deposit Chelopech are established by statistical processing of 19,200 multi-element ICP analysis of samples, analyzed for content of 36 elements (Ag, Al, As, Au, Ba, Be, Bi, Ca, Cd, Co, Cr, Cu, Fe, Hg, K, La, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, S, Sb, Sc, Se, Sn, Sr, Te, Ti, V, W, Y and Zn). The multi-elemental analyses of samples are kindly provided by the Geological Department of "Dundee Precious Metals Chelopech" EAD. The statistical analyses are carried out using the software STATISTICA of the company STATSOFT.

The correlation between the studied chemical elements is analyzed by making the correlation matrix and determining the levels of significance of the correlation coefficient. The correlation coefficient (r) takes any value from the range -1 ÷ +1. The sign plus or minus in front of correlation coefficient indicates the direction of this relationship, i.e. whether the elements are positively or negatively correlated. The absolute value of the coefficient shows the size of the correlation. The received correlations provisory are divided into four categories, depending on the absolute value of the correlation coefficient, namely: independent elements ($r < \pm 0.3$), elements with a weak ($\pm 0.3 \leq r < \pm 0.5$), a moderate ($\pm 0.5 \leq r < \pm 0.7$) and a strong ($r \geq \pm 0.7$) correlations.

Results and discussion

Pre-processing of data has the character of independent study that is relatively the most complex and of the utmost importance for the correct implementation of subsequent statistical procedures. The initial "acquaintance" with multi-elemental analysis showed that nearly 2,400 consecutive samples does not analyzed for the content of Pb and Zn, and some of the elements (Hg, Ti, Sn, Be and Ag) are below detection limits in most samples. These consecutive samples that are missing data for content of Pb and Zn are excluded from the total volume of analyzed data.

The statistical processing, analysis and interpretation is carried out on a group of data, obtained after the removal of possibly erroneous chemical analyzes and samples with anomalously high contents of some of the elements, as well as samples that have not analyzed for all chemical elements in the surveyed population. The elimination of anomalously high values of some of the elements increases relationship between some pairs of elements and weakening others. After processing of data, a modest increase in the coefficient of correlation of Au with the elements S, Te and Ag is established. The comparative analysis of the results shows, that the removal of samples with anomalously high contents in the studied group strengthens existing correlations, even at lower contents of the relevant element in the samples. This data processing is useful, regarding establishment of relationships between elements like Rb, Sr, Y, K, P, Mn and others related to hydrothermal alterations of host rocks, with those (Cu, Au, Fe, S, As, Sb , Bi, Ag), participating in the composition of main and secondary ore minerals in the deposit.

The correlation matrix marks some well developed relationships between pairs of elements, such as Fe-S, Cu-As, Cu-Bi, Cu-Cd, Cd-As, K-Na and other (Table 1). Elements such as Hg, W, Mo, Cr and P showed a lack of correlation with all other elements of the group. A typical correlation scatterplots of elements are presented below (Fig. 1). Particular attention in the interpretation of results is given to elements with a high positive correlation coefficient between themselves, and participating in the composition of the ore minerals and mineral associations in the deposit.

The scatterplot of Fe and S shows a relatively homogeneous data, distributed near the correlation line. The group of samples with anomalously high contents of Fe in the amount of 15%, which is the upper detection limit of the element, is established (Fig. 1 a.). Probably these samples are erroneous results from the chemical analysis, but the visual interpretation of figure 1 shows, that they practically do not affect the regression and the correlation between Fe and S. A homogeneity of data, confirming the accuracy of the obtained high correlation coefficient between the elements Cu and As ($r=0.93$), is distinctly observed on the presented figure 1 b. On the scatterplot of the pair of Pb-Zn ($r=0.73$) is seen a less marked homogeneity of the data (Fig. 1 c.). The presence of high correlation ($r=0.83$) between Cu and Cd is very interesting, as typical carrier of impurities from Cd is sphalerite and it is expected a stronger correlation between Cd and Zn. Mineralogical and geochemical analyses, conducted on polished sections from the Chelopech deposit, shows higher content of Cu in sphalerite from the central parts of the ore bodies in the deposit, which is a result of so-called "chalcopyrite disease", typical for ore sectors with a high degree of hydrothermal alteration in such type of epithermal deposits. The widespread "contamination" of sphalerite with chalcopyrite is the most likely reason for the high straight correlation between Cu and Cd (Fig. 1 d.).

A small number of samples have anomalously high content of some elements (P, Mg, Mo, Sn, K), marked with the upper detection limits on the correlation scatterplots of these elements, which deviate the regression line in one or another direction and affects the accuracy of the estimated correlation coefficient. The accuracy of the correlation coefficient is affecting by anomalously high contents, found as well for the typical elements associated with the ore minerals in the deposit as Cu, Au, Fe, Ag, Bi, Sb and Cd.

The absence of a real correlation between pairs of elements with negative correlations, due to the heterogeneity of the data, is established on the analyzed correlation scaterplots. Almost all observed scaterplots shows two sets of data, in which in increasing the content of one element of the pair in the samples, the value of the other does not change. This fact is observed most clearly, when analyzing the correlation scaterplots of Fe and S with the other studied elements, having negative correlation coefficient with them (K, La) (Fig. 1 e., f.). The visual interpretation of the correlation scatterplots shows, that it is more likely not about high negative correlations, but for the inhomogeneity of the two sets of samples, in which the elements are independent of each other.

The overall correlation characteristic of Au shows, a lack of strong correlation of the element with the rest of the group (Table 1). The presence of moderate to high correlation coefficient between the elements Au, Te and S is a result of the existing gold-containing sulfide minerals and sulphosalts as well as gold tellurides such kostovite, sylvanite and krennerite.

Table 1.
The correlation coefficient of elements in the ores from the gold-copper deposit Chelopech.

	Ag	Al	As	Au	Ba	Be	Bi	Ca	Cd	Co	Cr	Cu	Fe	Hg	K	La	Mg	Mn	Mo	Na	Ni	P	Pb	Rb	S	Sb	Sc	Se	Sn	Sr	Te	Ti	V	W	Y
Al	-0.06	1																																	
As	0.38	-0.02	1																																
Au	0.34	-0.06	0.44	1																															
Ba	-0.11	0.14	-0.15	-0.12	1																														
Be	-0.20	0.03	-0.29	-0.25	0.29	1																													
Bi	0.43	-0.02	***0.64***	0.41	-0.11	-0.23	1																												
Ca	-0.20	0.04	-0.22	-0.19	0.28	***0.57***	-0.18	1																											
Cd	0.41	-0.05	**0.81**	0.38	-0.15	-0.24	***0.57***	-0.24	1																										
Co	0.30	-0.04	0.35	0.48	-0.18	-0.29	0.31	-0.26	0.32	1																									
Cr	0.05	0.00	0.06	0.09	-0.04	-0.22	0.07	-0.15	0.06	0.11	1																								
Cu	0.43	-0.02	**0.93**	0.49	-0.17	-0.34	***0.70***	-0.27	**0.83**	0.39	0.08	1																							
Fe	0.38	-0.01	***0.53***	0.47	-0.30	***-0.54***	0.41	-0.46	0.48	***0.61***	0.16	***0.61***	1																						
Hg	0.01	-0.03	0.01	0.00	0.08	-0.01	0.01	0.00	0.01	0.01	-0.01	0.01	-0.02	1																					
K	-0.21	0.05	-0.35	-0.28	0.32	**0.78**	-0.27	***0.52***	-0.30	-0.33	-0.20	-0.41	***-0.61***	-0.02	1																				
La	-0.22	0.08	-0.26	-0.21	0.39	***0.62***	-0.21	***0.60***	-0.26	-0.26	-0.14	-0.31	***-0.51***	0.00	***0.67***	1																			
Mg	-0.16	0.17	-0.16	-0.14	0.27	0.48	-0.14	**0.74**	-0.19	-0.21	-0.11	-0.20	-0.36	0.00	0.38	0.45	1																		
Mn	-0.15	0.01	-0.16	-0.14	0.18	0.46	-0.13	***0.66***	-0.17	-0.20	-0.12	-0.19	-0.34	0.00	0.40	0.45	**0.71**	1																	
Mo	0.05	0.00	0.06	0.07	-0.02	-0.19	0.09	-0.15	0.06	0.06	0.17	0.07	0.10	0.05	-0.19	-0.16	-0.11	-0.11	1																
Na	-0.22	0.00	-0.35	-0.28	0.32	**0.74**	-0.27	***0.55***	-0.30	-0.34	-0.20	-0.41	***-0.61***	-0.02	**0.81**	***0.70***	0.44	0.36	-0.20	1															
Ni	0.18	-0.05	0.23	0.25	-0.07	-0.21	0.20	-0.16	0.22	0.46	0.13	0.25	0.36	-0.02	-0.22	-0.16	-0.13	-0.12	0.07	-0.23	1														
P	-0.25	-0.02	-0.33	-0.27	0.29	**0.76**	-0.26	***0.64***	-0.30	-0.32	-0.18	-0.39	***-0.61***	-0.03	**0.77**	**0.81**	0.47	0.48	-0.18	**0.78**	-0.22	1													
Pb	0.35	-0.06	0.05	0.08	-0.06	0.03	0.06	-0.12	0.35	0.07	-0.01	0.06	0.10	0.00	0.00	-0.12	-0.14	-0.09	0.00	-0.02	0.04	-0.07	1												
Rb	-0.16	0.05	-0.24	-0.19	0.23	***0.59***	-0.18	0.38	-0.20	-0.23	-0.16	-0.27	-0.43	-0.01	***0.68***	0.46	0.31	0.31	-0.13	***0.54***	-0.16	***0.53***	-0.01	1											
S	0.43	-0.07	***0.60***	***0.53***	-0.33	***-0.52***	0.48	-0.44	***0.55***	***0.63***	0.12	***0.69***	**0.92**	0.00	***-0.59***	***-0.50***	-0.36	-0.33	0.09	***-0.59***	0.36	***-0.59***	0.12	-0.41	1										
Sb	0.37	-0.07	***0.62***	0.36	-0.08	-0.27	0.42	-0.22	***0.58***	0.27	0.08	***0.65***	0.39	0.08	-0.27	-0.25	-0.16	-0.16	0.07	-0.29	0.17	-0.31	0.08	-0.19	0.42	1									
Sc	-0.15	0.40	-0.18	-0.17	0.35	0.42	-0.13	***0.58***	-0.18	-0.18	-0.05	-0.22	-0.38	-0.02	0.44	***0.55***	***0.61***	0.45	-0.11	0.47	-0.14	***0.50***	-0.09	0.30	-0.43	-0.18	1								
Se	0.33	-0.02	0.47	0.41	-0.15	-0.32	0.42	-0.28	0.44	0.45	0.08	***0.52***	***0.51***	0.01	-0.36	-0.30	-0.21	-0.20	0.10	-0.36	0.26	-0.36	0.08	-0.27	***0.57***	0.38	-0.22	1							
Sn	0.18	-0.01	***0.50***	0.29	-0.07	-0.22	0.39	-0.17	0.45	0.21	0.11	0.50	0.29	0.04	-0.25	-0.18	-0.13	-0.11	0.12	-0.25	0.16	-0.23	0.01	-0.17	0.32	0.42	-0.12	0.32	1						
Sr	-0.07	0.21	-0.14	-0.10	0.34	0.22	-0.09	0.38	-0.12	-0.12	-0.02	-0.16	-0.26	-0.02	0.25	0.35	0.22	0.16	-0.07	0.33	-0.08	0.32	-0.01	0.12	-0.27	-0.12	0.46	-0.13	-0.09	1					
Te	0.36	-0.04	***0.62***	***0.54***	-0.15	-0.30	***0.57***	-0.24	***0.58***	0.43	0.09	***0.67***	***0.51***	0.00	-0.34	-0.26	-0.18	-0.17	0.09	-0.34	0.27	-0.32	0.06	-0.24	***0.58***	0.49	-0.19	0.49	***0.50***	-0.13	1				
Ti	-0.04	0.23	-0.03	-0.03	0.12	0.02	-0.03	0.08	-0.04	0.00	0.05	-0.04	-0.07	-0.01	0.04	0.16	0.16	0.02	-0.03	0.14	-0.02	0.13	-0.05	0.01	-0.12	-0.03	0.39	-0.05	-0.03	0.17	-0.04	1			
V	0.05	***0.57***	0.24	0.12	0.17	-0.05	0.19	0.11	0.17	0.05	0.05	0.26	0.13	-0.01	-0.07	0.11	0.25	0.08	0.00	-0.07	-0.01	-0.01	-0.08	-0.03	0.09	0.17	***0.53***	0.12	0.15	0.21	0.15	0.44	1		
W	0.02	0.00	0.03	0.04	0.06	-0.10	0.11	-0.08	0.02	0.05	0.08	0.03	0.07	0.03	-0.10	-0.08	-0.07	-0.06	0.12	-0.10	0.08	-0.11	-0.01	-0.05	0.06	0.06	-0.07	0.10	0.11	-0.03	0.06	-0.01	0.02	1	
Y	-0.22	0.12	-0.26	-0.22	0.34	***0.59***	-0.21	**0.76**	-0.26	-0.28	-0.12	-0.31	***-0.53***	-0.03	***0.62***	**0.77**	***0.69***	***0.62***	-0.15	***0.66***	-0.18	**0.75**	-0.12	0.41	***-0.53***	-0.25	**0.77**	-0.30	-0.18	0.45	-0.27	0.19	0.21	-0.09	1
Zn	0.21	-0.07	0.00	0.02	-0.02	0.12	0.01	-0.05	0.37	-0.01	-0.01	0.00	-0.01	0.01	0.10	-0.02	-0.09	-0.03	0.00	0.09	-0.01	0.04	**0.73**	0.06	0.01	0.03	-0.03	0.01	-0.02	0.03	-0.01	-0.03	-0.08	-0.02	-0.03

The strong correlations are bolded, moderate - bold and italic.

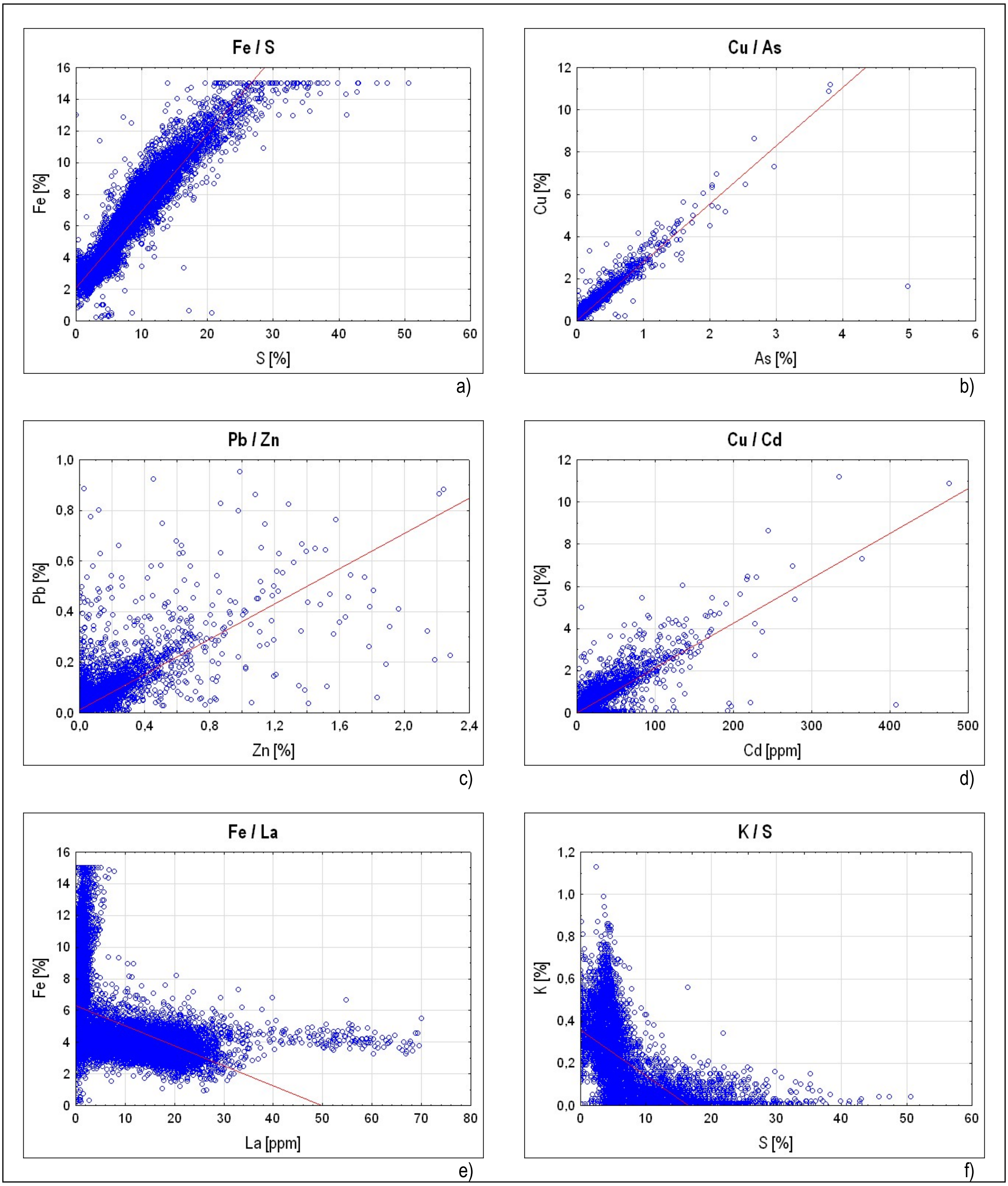

Fig. 1. Scatterplots of some typical correlations, obtained from analysis of elements.

The cobalt is a typical element-impurity in pyrite and its weak, but distinguished correlation with Au, is due to the common mineral association of pyrite with natural gold, gold-containing minerals, as well as a mineral inclusions of natural gold in pyrite. The absence of correlation (independence) of gold with the elemental association Pb-Zn indicates the presence of geochemical and spatial zonation of gold and galena-sphalerite mineral association within the deposit.

The constant presence of the As-containing copper sulphosalts as enargite, luzonite and tenantite in studied samples of the ore bodies is a reason for the very strong correlation of Cu with As and its stable relationship with sulfur. This strong relationship confirms the intermediate Cu-As-S ore-forming stage in the gold-copper deposit “Chelopech”, proposed by Bonev et al. (2002). According to the same authors, this main hydrothermal stage is auriferous and led to formation of the economical mineralization in the deposit with mineral association, represented by enargite, luzonite, covellite, goldfildite, chalcopyrite, tenantite, bornite and natural gold.

The moderate correlation between Cu and Sb, is due to the Sb-containing main (tetrahedrite) and secondary (famatinite) ore minerals in the deposit. The Te-bearing tetrahedrite, when Sb> (Te+As) or goldfildite, respectively when Te> (As+Sb) in the ores from the deposit, are likely to be due for the moderate correlation coefficient of Cu with Sb (0.65) and Te (0.67).

The iron is presented in the form of iron sulphides (mainly pyrite), also in the composition of chalcopyrite, bornite and other main and secondary minerals in the “Chelopech” deposit. The strong correlation between Fe and S (r = 0.92) indicate this mineral expression of the element (Table 1). Microprobe analyzes, carried out on polished sections from the deposit (Petrov et al., 2013), indicate the presence of Cu, Co and Ni, and limited amounts of Te and Se, as elements-impurities in the composition of pyrite crystals. This is the reason for the moderate levels of correlation of Fe with the elements Co, Ni, Te and Se (Table 1).

The weak correlation of elemental association Pb-Zn with other elements is established from the results of correlation analysis for all studied elements. A low to moderate correlations is observed only between Zn and Cd (r = 0.37), and Pb and Ag (r = 0.35), which are result of the frequently established impurities of Cd in sphalerite and Ag in galena, within the ore bodies of the deposit.

A group of chemical elements are particularly sensitive to the hydrothermal processes and resulting wall-rock alterations, taken place into the studied type of deposit, according to some authors (Hikov, 2001, 2005; 2013; Georgieva et al., 2002; Georgieva, 2014). Elements such as Zr, Cr, V, Ga, Ti show an inert character, until other group of elements (Rb, Y, Mn, Co, K) are very mobile and are extracted from the rocks during hydrothermal processes (Hikov, 2001). The behavior of strontium is of particular interest because it is extracted from the external and concentrated in the inner advanced argillic zone, which Hikov (2001) associate with the dissolution of apatite from volcanic rocks and the formation of aluminum-phosphate-sulphate (APS) minerals. In this regard, on the correlations of elements, sensitive to the hydrothermal alterations, is made further analysis, which helped to clarify the mutual dependence of the ore-forming elements with these typical for hydrothermal alterations. The analysis include Co, Mg, Mn, Rb, Sr, Ti, V, Y, K, Sc, La, and P, which indicate high (Co, Mg, Mn, Rb, Sr, Y, K, P) or low (Ti, V, Sc, La) mobility during the hydrothermal alterations, in addition to the elements typical for the ore minerals and mineral associations in the deposit as Au, Ag, Cu, Fe and As.

The inverse correlation of Fe with almost all typical for hydrothermal alterations elements is clearly shown in Table 1. It is noteworthy the moderate to high negative correlation of Fe with the elements K, La, P and Y, whose contents are substantially reduced in the advanced argillic zones in the deposit. These results supports the proposed by Terziev (1965) model, in which ore-forming processes started after the beginning of hydrothermal alterations and run simultaneously with them, such as the formation of Fe-bearing minerals is directly related to the degree of alteration of the host rocks, marked with removal of K, La, P and Y, as well as Rb and Sc from the advanced argillic zones.

The element Cu, participating in the composition of the main and secondary ore minerals from the deposit, logically showed no relationship to weak negative correlation with the typical for hydrothermal alterations studied group of elements (Table 1). Especially clear is this opposite relationship with the elements K, P, Y and La, whose concentrations are lowest within the central parts of ore bodies.

A high positive correlation coefficients of Y with La (r=0.77), Sc (r=0.77) and P (r=0.75), and moderate negative correlations of the element with Fe (r= -0.53) and S (r=-0.53) are established (Table 1). Further more detailed analysis should be made on relationships of Y with other elements included in the composition of the ore minerals, because the element is a highly mobile and typical for the hydrothermal alterations in the “Chelopech” deposit.

Conclusions

The analysis of correlation between the studied elements is helped to clarify the relationship between the ore-forming processes and hydrothermal alterations, taken place in the “Chelopech” deposit.

Some well developed relationships between pairs of elements, such as Fe-S, Cu-As, Cu-Bi, Cu-Cd, Cd-As, K-Na and other are established. An inverse correlation of the elements, involved in composition of main ore minerals in the deposit (Fe, Cu, Au, As, S), and highly mobile elements (K, P, Y, La, Rb), which are extracted from the rocks in the hydrothermal processes is observed. Trends of correlation of typical elements-impurities in main and secondary ore minerals with ore mineral-forming elements are established (Cd-Cu, Cd-As, Co-Fe, Co-Au, etc.).

The increased content of Cu in the sphalerite from the central sectors of ore bodies in the deposit is the result of so called "chalcopyrite disease", which is the most likely reason for the high correlation between the Cu and Cd. Frequent impurities of Co, Te, and Se in pyrite are the reason for the average high correlation of Fe with these elements.

The strong correlation between lead and zinc and absence or their weak correlation with other elements clearly emphasizes the existence of mineral zoning in the deposit and the spatial distribution of galena-sphalerite mineral association in marginal sectors of the ore bodies.

The results of present study indicate that the correlation analysis is a powerful tool for establishing interdependences between the chemical elements, involved in the composition of the ore and host rock minerals in Chelopech deposit.

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