Chemical composition and varieties of sulfosalts from gold mineralization in the Gaching ore occurrence (Maletoyvayam ore field)

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Abstract. The Gaching ore occurrence within the Maletoyvayam ore field is located in the southwestern part of Kamchatka volcanic belt. The ore occurrence belongs to high-sulfidation epithermal type. The sulfosalts of the fahlore-group (tennantite, tetrahedrites and goldfieldite) and enargite-groups (enargite, luzonite, famatinite) found in the gold mineralization of the Gaching ore occurrence. The presence of Cu-rich and Se-bearing fahlores as well as enargite-group sulfosalts in ores indicate a high oxygen volatility at temperature less than 250º during their formation.

1. Introduction
Fahlore belong to the class of sulfosalts. The general chemical formula can be given as: $M(1)\{Cu^{1+}, Fe^{2+}, Zn^{2+}, Mn^{2+}, Hg^{2+}, Cd^{2+}\}^4X^{12}Y^{13}Z^1$ where $M(1) = Cu^{1+}, Fe^{2+}, Zn^{2+}, Mn^{2+}, Hg^{2+}, Cd^{2+}; M(2) = Cu^{1+}, Ag^{1+}; X = Sb^{3+}, As^{3+}, Bi^{3+}, Te^{4+}; Y = S^{2-}$ and $Z = S^{2-}$. Fahlores show complete solid solution between the Sb and As end-members. Due to the widespread isomorphism, the minerals of the fahlore-group are indicators of ore genesis. The characteristics of their composition, the data on mineral associations and the sequence of their crystallization, are of great importance for establishing regularities in the evolution of the chemical composition of ore-generating solutions [2,3,4].

The minerals of the fahlore-group are widely developed in various types of hydrothermal deposits, including epithermal type. The epithermal type deposits with high Au and Ag contents have been discovered in the last 50 years. In Russia, this type includes Au-Ag deposits in the folded regions of Chukotka and Kamchatka [5]. In this paper we presented data on fahlores and other sulfosalts from the Au-Se-Te mineralization in the Gaching high-sulfidation epithermal ore occurrence (Maletoyvayam ore field) (figure 1). Gold mineralization with unique Au-Te-Se-S minerals of this ore occurrence has been investigated recently [6,7].
2. Analytical methods

First samples of oxidized alunite-quartz rocks were crushed and concentrated to a heavy fraction obtained by hydroseparation followed by concentration in the heavy liquid bromoform, (CHBr₃). The specimens (with heavy fraction) polished of the Gaching ore occurrence were investigated by polarized light microscopy and by electron microscope. Au-bearing minerals together with minerals of fahlore-group from the concentrate were analyzed. The chemical compositions of the minerals and texture of the mineral aggregates and separate grains were examined in the Analytical Center for multi-elemental and isotope research of the Sobolev Institute of Geology and mineralogy SB RAS in Novosibirsk using a LEO-413VP scanning electron microscope (SEM) with INCA Energy 350 microanalysis system (Oxford Instruments Ltd) equipped with EDS (analysts Dr. N. Karmanov, M. Khlestov, V. Danilovskaya). To cover the whole range of possible elements in the sulphides and sulphosalts, 16 elements (S, Cu, Fe, Zn, Hg, Ni, Pb, Au, Ag, As, Sb, Bi, Se, Sn, Te, O) was analysed. The obtained analytic conditions were 20 kV accelerating voltage and 0.4 A beam current. The counting times were 50 s and ~1 μm beam diameter.

Figure 1. Volcanic belts [8] and Au-Ag deposits [9] of the Kamchatka Peninsula.
3. Geological setting and ore geology

The Gaching ore occurrence within the Maletoyvayam ore field is located in the southwestern part of the Koryak Highland (figure 1), Kamchatka [10, 11]. The Maletoyvayam ore field together with Vetrovayam ore field belong to the Vetrovayam volcanic zone of the northeastern Central Kamchatka volcanic belt. The Maletoyvayam ore field consist of sediments of the Vetrovayam suite: andesites, tuffs and tuffaceous sandstones and metasomatic quartzites. The metasomatic quartzites are replaced successively by alunite-, sericite-kaolin-quartz and kaolin-quartz quartzites. The outer parts of the Maletoyvayam ore field are composed of argillizes and propylites [11].

A number of gold-silver epithermal deposits (Ozernovskoe, Asachinskoe, Aginskoe and others) have been identified within the Central Kamchatka volcanic belt (figure 1) [12,13]. The Vilyuchinskoe gold-silver-polymetallic ore occurrence in the south of Kamchatka have been studied also in detail earlier [9]. Three ore occurrences (Gaching, Jubilee, South-West areas) were found within the Maletoyvayam ore field at which geological and exploration works started in 2010. The Gaching represents a quartz stockwork hosted by the secondary quartzites and intensely oxidized quartz-alunite rocks [11] The ore occurrence belongs to high-sulfidation (HS) epithermal type [7].

4. Results

The subject of our research are sulfosalts. Sulfosalts from the Gaching ore occurrence were found in association with Au(±Ag)-telluride and sulfoselentelluride, Au oxides, native gold and unnamed Au₄Te₆(Se,S)₄ [6,7]. Sulfosalts include minerals of fahlore-group (tennantite-tetrahedrite solid solution and goldfieldite), enargite-group (enargite, luzonite-famatinite solid solution). The general chemical formula of minerals of the fahlore-group is \( \text{Cu}_{12}(\text{Sb,As,Te})_4\text{S}_{13} \) and enargite-group \( \text{Cu}_3(\text{As,Te})_4\text{S}_4 \). The calculation of chemical formula of fahlore based on \( \sum \text{apfu} = 29 \) was suggested by Trudu and Knittel [16] and Moëlo [14]. The calculations of chemical formula of the enargite-group series should be based on \( \sum \text{apfu} = 8 \). The sulfosalts contain Fe, Zn, Au, Ag, Bi, Se as trace elements in different quantities. The atoms of Fe, Zn, Au, Ag may substitute atoms of Cu. Bi atoms substitute for cation \( X^{3+} \) or \( X^{4+} \) (Sb,As,Te) and Se atoms substitute for S atoms.

**Fahlore-group.** According to [14] fahlores belong to Cu(Ag)-rich sulfosalts and represented by species of tennantite (\( \text{Cu}_{12}\text{S}_{13} \))-, tetrahedrite (\( \text{Cu}_{12}\text{Sb}_{4}\text{S}_{13} \)) and goldfieldite \( \text{Cu}_{12}\text{Te}_{4}\text{S}_{13} \). The minerals occur as distinct grains (figure 2 a,b,e) or intergrown (figure 2 c-d) with native Au, native Te, Te-Se solid solution, unnamed \( \text{Au}_4\text{Te}_6(\text{Se,S})_{14} \) (\( \text{Sb,Bi,Te,As,Fe})_2\text{O}_5 \) calaverite \( \text{AuTe}_2 \) and krennerite (\( \text{Au,Ag}(\text{Te,Se})_2 \)). The distinct grains sizes do not exceed 90 μm. A several grains contain inclusions native Te, Te-Se solid solution (figure 2 a-b). Also there are grains of fahlore with inclusions of others sulfosalts (figure 2 a,e) The compositions of these minerals are listed in the table 1. Tetrahedrite contains up to 3.30 wt.% Ag. Fe-bearing and Zn-bearing tetrahedrite and tennantite (0.35-1.99 wt.% Fe, 0.72-5.06 wt.% Zn) was detected. Goldfieldite includes Te and Sb (28.41-8.93 wt.%, 1.86-11.36 wt.%, relatively). With the increasing of Te content the decrease of Sb is due to solutions between of goldfieldite and tetrahedrite. Zn is not observed in goldfieldite. The minerals fahlore-group are Se-rich, containing 0.77-11.27 wt.% Se (table 1). Such high selenium contents in fahlores is rare.

**Enargite (luzonite-famatinite)-group.** Luzonite \( \text{Cu}_3(\text{As,Sb})_4\text{S}_4 \) and famatinite \( \text{Cu}_3\text{Sb}_4\text{S}_4 \) are the end-members of the isomorphic series luzonite-famatinite. Enargite with a chemical formulas \( \text{Cu}_3\text{AsS}_4 \) [15] have a rhombic crystallography and its polymorphic modification is luzonite (tetragonal crystallography).

Enargite, luzonite and famatinite occur as intergrowths (figure 2 g,i) with gold, calaverite \( \text{Au}-(\text{Te,Se})_2 \) and unnamed phases \( \text{Au}_4\text{Te}_6(\text{Se,S})_{14} \) and \( \text{Sb,Te,As,Fe})_2\text{O}_5 \) or as inclusions in gold (figure 2 f). The grain of intergrowth of famatinite and luzonite (figure 2 h) has been found also. These minerals have a grain size of 60-70 μm, whereas the inclusions sizes do not exceed 20 μm. Luzonite-famatinite solid solutions contain up to 0.72 wt.% Fe, while the other minor elements substitute for Cu not found (table 1). The compositions of sulfosalts of this group include Se (up to 6.2 wt.%) similar to fahlores.
Figure 2. SEM images of sulfosalts from the Gaching mineralization. Gf – goldfieldite, Td – tetrahedrite, Fam – famatinite, Luz – luzonite, Eng – enargite.

5. Discussion and conclusion
The Se-rich tennantite-tetrahedrite solid solution similar to Gaching ore occurrence have been described only in Mutnovsk, Ozernovsk deposits and Vilyuchinsk ore occurrence on the Kamchatka region [9,16,17]. The Se-rich goldfieldite found in Vilyuchinsk ore occurrence [9]. This suggests the high of the $f_{O_2}$ in gold mineralization of Vilyuchinsk and Gaching ore occurrences [7].

The Gaching high-sulfidation epithermal deposit in the Maletoyvayam ore field contain the Cu-excess (>10 $apfu$) tetrahedrite-tennantite (figure 3). According to table 1, the most Cu-rich tetrahedrites (11.507 $apfu$ Cu) and the most Cu-rich tennantite (11.269 $apfu$ Cu) are comparable with tetrahedrites (11.039 $apfu$ Cu) and tennantite (11.784 $apfu$ Cu) from the epithermal St. Demetrios and
Table 1. Representative electron microprobe analyses of fahlores from the Gaching ore occurrence: 1-4 goldfieldite, 5-8 tetrahedrite, 9-11 luzonite (enargite), 12-15 famatinite. b.d. – below detection limit

| wt. % | 1   | 2   | 3   | 4   | 5   | 6   | 7   | 8   | 9   | 10  | 11  | 12  | 13  | 14  | 15  |
|-------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| Cu    | 41.86 | 40.87 | 0.00 | 43.54 | 42.49 | 41.58 | 36.34 | 39.23 | 46.51 | 44.80 | 43.52 | 40.19 | 43.96 | 43.69 | 42.71 |
| Fe    | b.d. | b.d. | 41.94 | b.d. | 0.77 | 1.30 | 1.69 | 1.99 | 0.00 | 0.30 | 0.39 | 0.34 | 0.28 | 0.44 | b.d. |
| Zn    | b.d. | b.d. | b.d. | b.d. | 2.15 | 5.06 | 3.35 | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. |
| Au    | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. |
| Ag    | 0.47 | 0.46 | b.d. | b.d. | 0.46 | 0.41 | 3.30 | 1.09 | 0.48 | b.d. | 0.44 | b.d. | b.d. | b.d. | b.d. |
| Bi    | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. | b.d. |
| Sb    | 9.69 | 6.52 | 10.25 | 11.36 | 13.36 | 17.41 | 22.87 | 23.02 | 0.54 | 10.02 | 7.84 | 21.97 | 17.80 | 14.32 | 14.11 |
| Te    | 17.15 | 19.60 | 16.56 | 12.83 | 10.18 | 6.22 | b.d. | b.d. | 2.86 | b.d. | 2.82 | b.d. | b.d. | b.d. | b.d. | 1.68 |
| As    | 1.51 | 1.51 | b.d. | 2.78 | 3.68 | 4.18 | 3.76 | 3.07 | 17.15 | 11.10 | 12.88 | 3.29 | 6.53 | 8.46 | 8.17 |
| Se    | 11.27 | 9.78 | 9.60 | 4.74 | 4.88 | 2.77 | b.d. | 0.77 | b.d. | 3.63 | 0.00 | 5.65 | 1.72 | 4.72 | 6.46 |
| S     | 19.35 | 19.62 | 18.65 | 22.15 | 22.38 | 24.25 | 25.07 | 24.87 | 31.58 | 28.79 | 33.28 | 26.13 | 29.34 | 28.02 | 25.82 |
| Total | 101.30 | 98.36 | 97.00 | 97.40 | 98.20 | 100.27 | 98.08 | 97.38 | 99.12 | 98.65 | 101.17 | 98.42 | 99.64 | 99.65 | 98.95 |

*apfu*  

| wt. % | 1   | 2   | 3   | 4   | 5   | 6   | 7   | 8   | 9   | 10  | 11  | 12  | 13  | 14  | 15  |
|-------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| Cu    | 11.62 | 11.58 | 12.13 | 11.92 | 11.50 | 10.82 | 9.586 | 10.350 | 2.962 | 2.993 | 2.750 | 2.892 | 2.964 | 2.958 | 2.992 |
| Fe    | -    | -    | -    | -    | 0.237 | 0.385 | 0.507 | 0.597 | -    | 0.023 | 0.028 | 0.028 | 0.021 | 0.034 | -    |
| Zn    | -    | -    | -    | -    | 0.544 | 1.297 | 0.859 | -    | -    | -    | -    | -    | -    | -    | -    |
| Au    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    |
| Ag    | 0.077 | 0.077 | -    | 0.073 | 0.063 | 0.513 | 0.169 | 0.018 | -    | 0.016 | -    | -    | -    | -    | -    |
| Bi    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    |
| Sb    | 1.404 | 0.964 | 1.548 | 1.623 | 1.889 | 2.365 | 3.149 | 3.170 | 0.018 | 0.349 | 0.259 | 0.825 | 0.627 | 0.506 | 0.516 |
| Te    | 2.372 | 2.766 | 2.386 | 1.749 | 1.373 | 0.806 | -    | -    | 0.091 | -    | 0.089 | -    | -    | -    | 0.059 |
| As    | 0.356 | 0.363 | -    | 0.646 | 0.845 | 0.923 | 0.841 | 0.687 | 0.926 | 0.690 | 0.201 | 0.373 | 0.486 | 0.485 |
| Se    | 2.519 | 2.230 | 2.236 | 1.044 | 1.064 | 0.580 | -    | 0.164 | -    | 0.195 | -    | 0.327 | 0.093 | 0.257 | 0.364 |
| S     | 10.649 | 11.018 | 10.695 | 12.018 | 12.012 | 12.510 | 13.106 | 13.004 | 3.985 | 3.811 | 4.168 | 3.727 | 3.921 | 3.759 | 3.584 |
Petrology of magmatic and metamorphic complexes

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1. \((\text{Cu}_{11.62}\text{Ag}_{0.08})_{11.76}(\text{Te}_{2.37}\text{Sb}_{1.40}\text{As}_{0.36})_{4.13}(\text{S}_{10.65}\text{Se}_{2.52})_{13.17}\)
2. \((\text{Cu}_{11.59}\text{Ag}_{0.08})_{11.65}(\text{Te}_{2.77}\text{Sb}_{0.98}\text{As}_{0.36})_{4.09}(\text{S}_{11.02}\text{Se}_{2.23})_{13.25}\)
3. \((\text{Cu}_{12.15}(\text{Te}_{2.39}\text{Sb}_{1.53})_{3.94}(\text{S}_{10.70}\text{Se}_{2.23})_{12.93}\)
4. \((\text{Cu}_{11.93}(\text{Te}_{1.75}\text{Sb}_{1.62}\text{As}_{0.65})_{4.02}(\text{S}_{12.02}\text{Se}_{1.04})_{13.06}\)
5. \((\text{Cu}_{11.51}\text{Fe}_{0.32}\text{As}_{0.07})_{11.83}(\text{Sb}_{1.89}\text{Te}_{1.37}\text{As}_{0.85})_{4.11}(\text{S}_{12.01}\text{Se}_{1.06})_{13.07}\)
6. \((\text{Cu}_{10.82}\text{Zn}_{0.54}\text{Fe}_{0.38}\text{As}_{0.06})_{11.81}(\text{Sb}_{2.37}\text{Te}_{0.81})_{4.10}(\text{S}_{12.51}\text{Se}_{0.58})_{13.09}\)
7. \((\text{Cu}_{9.55}\text{Zn}_{1.38}\text{Fe}_{0.51}\text{As}_{0.84})_{3.92}(\text{S}_{13.10}\)
8. \((\text{Cu}_{10.37}\text{Zn}_{0.86}\text{Fe}_{0.60}\text{As}_{0.47})_{11.94}(\text{S}_{12.17}\text{As}_{0.69})_{3.86}(\text{S}_{13.00}\text{Se}_{0.16})_{13.16}\)
9. \((\text{Cu}_{2.97}(\text{Sb}_{0.52}\text{As}_{0.37}\text{Te}_{0.01})_{1.00}(\text{S}_{13.45}\text{Sb}_{0.02})_{1.00}\text{Se}_{0.20})_{4.01}\)
10. \((\text{Cu}_{2.99}(\text{Sb}_{0.53}\text{As}_{0.37})_{0.98}(\text{S}_{13.32}\text{Sb}_{0.02})_{1.00}\text{Se}_{0.20})_{4.01}\)
11. \((\text{Cu}_{2.97}(\text{Sb}_{0.53}\text{As}_{0.37})_{0.98}(\text{S}_{13.32}\text{Sb}_{0.02})_{1.00}\text{Se}_{0.20})_{4.01}\)
12. \((\text{Cu}_{2.97}(\text{Sb}_{0.53}\text{As}_{0.37})_{0.98}(\text{S}_{13.32}\text{Sb}_{0.02})_{1.00}\text{Se}_{0.20})_{4.01}\)
13. \((\text{Cu}_{2.97}(\text{Sb}_{0.53}\text{As}_{0.37})_{0.98}(\text{S}_{13.32}\text{Sb}_{0.02})_{1.00}\text{Se}_{0.20})_{4.01}\)
14. \((\text{Cu}_{2.97}(\text{Sb}_{0.53}\text{As}_{0.37})_{0.98}(\text{S}_{13.32}\text{Sb}_{0.02})_{1.00}\text{Se}_{0.20})_{4.01}\)
15. \((\text{Cu}_{2.97}(\text{Sb}_{0.53}\text{As}_{0.37})_{0.98}(\text{S}_{13.32}\text{Sb}_{0.02})_{1.00}\text{Se}_{0.20})_{4.01}\)

Pefka deposits in Greece [20]. Thus, it may believe that the epithermal deposits of Greece and Kamchatka display the most Cu-rich tennantite and tetrahedrites of worldwide.

![Figure 3](image-url)

Figure 3. Chemical variation of tetrahedrite-tennantite solid solution series as a function of (a) Cu-Sb, (b) Cu-As.

Considering metalls contents in the fahlores, the nomenclature of [16], is less suitable while the ternary diagram [17] taking into account the valences of the metalls (figure 4 a) can be used by us. The studied fahlores in this diagram show a broad range of compositions and can be classified as tetrahedrite, tennantite and goldfieldite. A complete solid solutions between tetrahedrite and tennantite exists including the Sb- and As- end members from Laki, Madzharovo [18,19], and Pagoni Rachi epithermal porphyry deposit [20]. However, the end members of tetrahedrites (Cu12Sb4S13) and tennantite (Cu12As4S13) were not found in the studied composition of fahlores. In addition, all the compounds of Sb-tennantite also contain Te (figure 4 a-b). Solid solutions between goldfieldite and tetrahedrite with a variable amount of As are also characteristic of Gatching ore occurrence.

The chemical formula of the fahlores (table 1) not close to ideal chemical formula with S+Se = 13 apfu and ∑metals = 12 apfu. For example, S+Se is in range 12.89-13.45 apfu and ∑metal − 11.47-12.24 apfu in goldfieldite. S+Se is in range 12.84-13.32 apfu and ∑metal − 11.82-12.20 apfu) in tetrahedrite.
This is due to replacement of the main ore mineralization during the oxidation process, in which the calaverite and krennerite transform into secondary mustard gold, often in association with complex Au-oxides containing Sb, Te, As, Fe, Cu – the main elements of fahlores [7]. Most likely, sulfosalts also underwent to transformation, as a result of which such variations occurred in their compositions.

Enargite/luzonite and famatinite in the ore mineralization of the Gaching ore occurrence are the earlier minerals than the fahlores, because the inclusions of enargite/luzonite and famatinite were found in the fahlore (figure 2 a). The presence of pentavalent As$^{5+}$ and Sb$^{5+}$ (enargite Cu$_3$AsS$_4$ and famatinite Cu$_3$SbS$_4$) in ores indicate a high oxygen volatility during their formation [21]. This is consistent with our data on the conditions for the formation of mineral parageneses of the Gaching ore occurrence.

According to [22] fahlores may replace the chalcopyrite while the Fe-excess is separated as pyrite. This process takes place at ~180-300ºC. The conditions of $f_{S_2}$ of tennantite and tetrahedrite is higher than of chalcopyrite formations, but is lower than of enargite and famatinite formations. The goldfieldite is indicator of high $f_{S_2}$. With increase of $f_{O_2}$, Cu-rich as well Zn-bearing fahlores instead of Fe-bearing fahlores are formed. At high $f_{O_2}$, Se-rich sulfosalts appear - hakite Cu$_6$[Cu$_4$Hg$_2$]Sb$_3$Se$_{13}$ and giraudite Cu$_4$[Cu$_4$(Fe,Zn)$_2$]As$_4$Se$_{13}$ [22]. These minerals are not found in the Gaching occurrence, but Se is the ubiquitous element in composition of all minerals of ore genesis, including sulfosalts.

We showed that the sulfosalts as the other ore minerals from the Gaching high-sulfidation epithermal occurrence were formed during the main gold stage at temperature less than 250º [7]. This is agree with the temperatures ranging in 170-260ºC established for the other epithermal gold–silver deposits in the Southwestern Hokkaido, Japan [23]. Se-rich goldfieldite as well as Te-rich tennantite and Te-rich tetrahedrite are indicators of significant differentiation of ore-bearing solutions and, accordingly, of large-scale gold mineralization [24].

**Conclusions**

Two groups of sulfosalts were found in the mineral association of the Gaching ore occurrence: the fahlore-group Cu$_{12}$(As,Sb,Te)$_4$S$_{13}$ and enargite-group Cu$_3$(As,Sb)S$_4$. The fahlore-group include tennantite, tetrahedrites and goldfieldite mineral species. The tennantite and tetrahedrites found in the Gaching ore occurrence are similar to the sulfosalts from epithermal deposits of Greece and they are the most Cu-rich known in the world. The fahlores of the Gaching ore occurrence enriched with
selenium. This feature is characteristic of many Au-Ag epithermal deposits of the Kamchatka volcanic belt.

The presence of Cu- and Se-rich fahlores and enargite-group sulfosalts in ores indicate a high oxygen volatility at temperature less than 250º during their formation. This conclusion is consistent with the previously obtained data based on the Au-Te-Se-S mineral assemblage of the main ore stage.

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