

Coupled substitutions in goldfieldite–tetrahedrite minerals from the Iriki mine, Japan

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Abstract

Chemical compositions of goldfieldite–tetrahedrite series minerals from the Iriki mine in Japan are reported for the first time. The compositional variations of coexisting goldfieldite [atomic proportion $\text{Te} > (\text{Sb} + \text{As} + \text{Bi})$], tellurian tetrahedrite [$\text{Sb} > (\text{Te} + \text{As} + \text{Bi})$], and Te-free tetrahedrite from this locality are considered alongside data from the literature. These show that the substitution of $(\text{Sb}, \text{As})^{3+}$ for Te^{4+} in natural goldfieldite–tetrahedrite series minerals occurs by the following two mechanisms: (i) tetrahedrite, $\text{Cu}_{10}(\text{Cu}, \text{Fe}, \text{Zn})_2^{2+}(\text{Sb}, \text{As})_4^{3+}\text{S}_{13}$ and goldfieldite, $\text{Cu}_{12}(\text{Sb}, \text{As})_2^{3+}\text{Te}_2^{4+}\text{S}_{13}$ form a continuous solid-solution according to the coupled substitution of $\text{Cu}^+\text{Te}^{4+}$ for $(\text{Cu}, \text{Fe}, \text{Zn})^{2+}(\text{Sb}, \text{As})^{3+}$, and (ii) goldfieldite, $\text{Cu}_{12}(\text{Sb}, \text{As})_2^{3+}\text{Te}^{4+}\text{S}_{13}$ and the ideal end-member, $\text{Cu}_{10}\text{Te}_4^{4+}\text{S}_{13}$ might also form a continuous solid-solution by the coupled substitution of $\square(\text{vacancy})\text{Te}^{4+}$ for $\text{Cu}^+(\text{Sb}, \text{As})^{3+}$. According to this work, the general formulae of tellurian tetrahedrite and goldfieldite are, therefore, respectively, $\text{Cu}_{10+x}^+(\text{Cu}, \text{Fe}, \text{Zn})_{2-x}^{2+}(\text{Sb}, \text{As})_{4-x}^{3+}\text{Te}_x^{4+}\text{S}_{13}$, with $x = 0$ to 2 and $\text{Cu}_{12-y}^+(\text{Sb}, \text{As})_{2+y}^{3+}\text{Te}_y^{4+}\text{S}_{13}$, with $y = 0$ to 2.

KEYWORDS: goldfieldite–tetrahedrite series, coupled substitutions, Iriki mine, Japan.

Introduction

THE tetrahedrite structural formula can be represented in general as: $(\text{Cu}, \text{Ag})_{\Sigma 6}^{\text{TRG}}[\text{Cu}_4(\text{Fe}, \text{Zn}, \text{Cd}, \text{Hg})_2]_{\Sigma 6}^{\text{TET}}(\text{Sb}, \text{As}, \text{Bi}, \text{Te})_{\Sigma 4}^{\text{SM}}(\text{S}, \text{Se})_{\Sigma 13}$, in which TRG refers to trigonal, TET tetrahedral, and SM semi-metal sites, as mentioned, for example, by Sack and Loucks (1985). Goldfieldite was first reported by Ransome (1909) from Goldfield, Nevada, U.S.A., and Thompson (1946) confirmed that this mineral has the tetrahedrite structure on the basis of X-ray powder diffractometry. Although many studies of the chemical composition of tetrahedrite and tellurian tetrahedrite have been made (e.g. Charlat and Lévy, 1974; Ixer and Stanley, 1980; Patrick and Hall, 1983; Johnson *et al.*, 1986), those of goldfieldite are few (e.g. from Goldfield in Lévy, 1967, and Kato and Sakurai, 1970; from Butte, Montana, U.S.A. in Springer, 1969, Criddle and Stanley, 1986; from Kawazu, Japan in Kato and Sakurai, 1970; from Sardinia in Frenzel *et al.*, 1975; from U.S.S.R. in Tsepin *et al.*, 1977;

Novgorodova *et al.*, 1978; Kovalenker *et al.*, 1979, 1987; Loginov *et al.*, 1983; Spiridonov *et al.*, 1984; Sakharova *et al.*, 1984; Igumnova, 1986). Moreover, many of these data deviate from stoichiometry and the charges do not balance, probably because of uncertainty in electron microprobe analyses, impurity, Cu vacancy, and oxidation.

Kase (1986) reports ten chemical compositions of tellurian tennantite from Japanese Besshi-type deposits, and Knittel (1989) shows one analysis of arsenian goldfieldite and three of tellurian tennantite from the Marian gold deposit in Philippines. These data illustrate the coupled substitution $(\text{Cu}, \text{Fe}, \text{Zn})^{2+}(\text{As}, \text{Sb})^{3+} \rightleftharpoons \text{Cu}^+\text{Te}^{4+}$, as experimentally shown by Kalbskopf (1974).

This paper reports chemical compositions of goldfieldite–tetrahedrite series minerals from the Iriki mine, Kagoshima Prefecture, Japan, and discusses the substitution mechanism of (Sb, As) for Te in natural goldfieldite–tetrahedrite series minerals.

Occurrence of goldfieldite-tetrahedrite

The Iriki mine is located in a post-Miocene epithermal Au-Ag mining area in the southern part of Kyushu, Japan. The deposits of the Iriki mine were worked for Au-Ag-Cu in 1917, but later worked for kaolin and closed in the 1950s. The mineralisation was composed of auriferous pyrite-quartz veins and argentiferous pipe-like bodies (Uetani *et al.*, 1966). Kinoshita and Kim (1938) gave a brief description of the geology and ore deposits of the mine; the deposits lie within a brecciated rhyolite dike which cuts hornblende-pyroxene andesite. The latter is overlain mainly by Miocene tuff and mudstone, and all these are covered by Recent volcanic ash and pumice deposits. The rhyolites are strongly silicified and surrounded by kaolinised areas.

Goldfieldite and tetrahedrite occur in two forms in the argentiferous pipe-like bodies. Massive goldfieldite contains tiny inclusions of native tellurium (up to 1.0 wt.% Se), hessite (up to 0.8 wt.% Se), and hemusite of about 10 µm across, and rims strongly silicified rhyolite breccias in the center of the pipe-like bodies. This goldfieldite is surrounded by concentric bands of (i) goldfieldite-native-tellurium-hessite-hemusite, (ii) tetrahedrite, (iii) tetrahedrite-famatinitite, (iv) tetrahedrite-famatinitite-chalcopryrite, and (v) chalcopryrite-pyrite-marcasite-sphalerite-galena, from the centre to the rim, and all these coexist with quartz. The goldfieldite-bearing band can be further subdivided into four goldfieldite-dominant zones ranging from 3 mm to 5 mm in width. Goldfieldite does not contain inclusions near its boundary with tetrahedrite. Here, goldfieldite occurs as euhedral grains of about 100 µm in size.

Another, though rare, occurrence of goldfieldite is as networks coexisting with hemusite, pyrite, marcasite, chalcopryrite, sphalerite, and an unknown mineral in the strongly silicified rhyolite breccias.

Chemical composition

Because goldfieldite often includes minute tellurium phases such as native tellurium and hessite, the polished sections were carefully examined under the microscope at high magnification both in air and oil. The compositions were cross-checked by electron-microprobes of Cambridge Instruments Microscan IX at the British Museum (Natural History) and JEOL 733II at the Geological Institute, Faculty of Science, University of Tokyo, using the conditions as mentioned in Harris *et al.* (1984) and Shimizu *et al.* (1988). Te- $L\alpha$ counts were corrected for partial overlap by Sb- $L\beta$ in the minerals.

Table 1 Chemical composition of Te-free tetrahedrite from the Iriki mine, Japan.

	1	2	3	4	5
Cu	46.00	44.35	45.90	46.10	43.13
Ag	0.07	0.10	0.08	0.13	0.08
Fe	0.30	0.99	0.44	0.28	2.46
Zn	0.29	0.10	0.31	0.41	0.16
Mn	0.56	1.00	0.40	0.41	1.04
Cd	0.01	-	0.02	-	-
Sb	23.85	23.83	23.82	22.65	22.43
As	3.40	3.47	3.54	4.40	5.61
Sn	0.23	-	0.22	-	-
Bi	-	0.17	-	0.46	0.11
S	24.59	24.71	24.48	25.52	25.23
Se	0.46	0.72	0.71	0.24	0.53
total	99.76	99.44	99.92	100.20	100.78
Atomic proportions based on 29 atoms.					
Cu	11.925	11.509	11.901	11.786	11.951
Ag	0.010	0.015	0.012	0.019	0.011
Fe	0.089	0.292	0.130	0.081	0.710
Zn	0.072	0.025	0.077	0.003	0.039
Mn	0.168	0.300	0.120	0.122	0.305
Cd	0.002	-	0.003	-	-
Sb	3.227	3.227	3.221	3.022	2.972
As	0.748	0.763	0.779	0.954	1.202
Sn	0.031	-	0.031	-	-
Bi	-	0.013	-	0.036	0.008
S	12.632	12.706	12.577	12.929	12.694
Se	0.096	0.150	0.148	0.049	0.108
mol. %					
tetr	80.6	80.6	79.9	75.3	71.1
tenn	18.7	19.1	19.3	23.8	28.7

Representative chemical compositions of Te-free tetrahedrite from the Iriki mine are shown in Table 1. The tetrahedrite end-member molecule in the mineral ranges from 62.4 to 89.6 mol.% (no. of analysis, $n = 8$), and the number of copper atoms ranges from 10.95 to 11.85, based on a unit formula of 29 atoms. The empirical formulae on the basis of total atoms = 29 (Table 1) are all close to $\text{Cu}_{12}(\text{Sb,As})_4\text{S}_{13}$.

Representative chemical compositions of tellurian tetrahedrite and goldfieldite are shown in Table 2. The number of copper atoms in tellurian tetrahedrite is also greater than ten, ranging from 10.75 to 11.96 ($n = 10$), and in goldfieldite is from 10.75 to 12.01 ($n = 19$). The tellurium content of the latter is up to 18.5 wt.% corresponding to 59.9% of the goldfieldite end-member molecule (no. 8 in Table 2).

The empirical formulae of tellurian tetrahedrite are close to $\text{Cu}_{12}(\text{Sb,As,Te})_4\text{S}_{13}$ (nos. 1 to 3 in Table 2), and those of goldfieldite are close to $\text{Cu}_{12}(\text{Te,As,Sb})_4\text{S}_{13}$, with $\text{Te} < 2$ (nos. 4 to 6 in Table 2) and $\text{Cu}_{12-x}(\text{Te,Sb,As})_4\text{S}_{13}$, with $\text{Te} = 2$ to 2.74, $x \leq 0.67$ (nos. 7 to 11 in Table 2). Note that $\Sigma(\text{Cu} + \text{Ag} + \text{Fe} + \text{Zn} + \text{Mn})$ atoms is less than twelve in the case of $\text{Te} > 2$.

Substitution of (Sb,As) for Te in natural goldfieldite-tetrahedrite series minerals

The relationship between the proportion of Te and (Sb + As + Bi) in goldfieldite-tetrahedrite

Table 2. Chemical composition of tellurian tetrahedrite and goldfieldite from Iriki (nos. 1 to 8) and Kawazu (9 to 11).

	1	2	3	4	5	6	7	8	9	10	11
Cu	42.68	46.34	45.46	45.80	45.62	47.28	43.37	44.04	44.96	44.97	44.63
Ag	0.03	0.06	0.21	0.11	0.50	0.09	0.26	0.09	0.14	0.13	0.15
Fe	4.42	0.67	0.79	0.21	0.67	0.26	0.67	0.28	0.01	0.12	0.05
Zn	0.64	0.07	0.07	0.01	0.14	-	0.04	-	0.02	0.11	0.02
Mn	0.07	0.24	0.20	0.15	0.17	0.12	-	-	-	-	-
Te	0.32	8.95	10.08	13.32	13.38	15.57	16.55	18.47	20.80	21.18	21.60
As	6.72	4.68	5.79	5.03	7.02	8.26	2.35	1.67	2.82	2.70	2.90
Sb	18.86	12.86	11.40	8.94	6.24	1.72	9.10	9.54	5.22	5.03	4.53
Bi	0.23	-	-	-	0.22	-	1.83	0.32	-	0.24	0.43
S	25.54	25.08	24.93	24.84	25.32	25.49	24.53	24.85	25.12	25.15	24.18
Se	0.36	0.08	0.41	0.64	0.05	0.26	0.69	0.43	0.49	0.52	1.07
total	99.87	99.03	99.34	99.25	99.33	99.05	99.39	99.69	99.58	100.15	99.56
Atomic proportions based on 29 total atoms for nos. 1 to 6, and 4 atoms of (Te, As, Sb and Bi) for nos. 7 to 11											
Cu	10.753	11.958	11.730	11.903	11.692	12.012	11.162	11.229	11.624	11.585	11.364
Ag	0.005	0.010	0.033	0.017	0.075	0.033	0.040	0.013	0.022	0.020	0.023
Fe	1.266	0.197	0.231	0.063	0.195	0.076	0.196	0.081	0.003	0.035	0.014
Zn	0.157	0.018	0.018	0.003	0.034	-	0.010	-	-	0.028	0.005
Mn	0.021	0.072	0.059	0.046	0.050	0.036	-	-	-	-	-
Te	0.040	1.149	1.295	1.724	1.708	1.969	2.121	2.344	2.677	2.717	2.739
As	1.436	1.025	1.267	1.108	1.526	1.781	0.514	0.362	0.618	0.589	0.626
Sb	2.480	1.731	1.535	1.212	0.834	0.228	1.221	1.270	0.705	0.676	0.602
Bi	0.018	-	-	-	0.018	-	0.144	0.024	-	0.018	0.034
S	12.751	12.823	12.747	12.793	12.858	12.832	12.510	12.555	12.869	12.839	12.200
Se	0.074	0.016	0.085	0.134	0.010	0.053	0.142	0.088	0.102	0.108	0.220
mol. %											
goldf	1.0	29.4	31.6	42.6	42.8	49.5	53.0	59.9	66.9	67.9	68.5
tetr	62.4	44.3	37.5	30.0	18.5	5.7	30.8	30.8	17.6	16.9	15.0
tenn	36.1	26.2	30.9	27.4	38.3	44.8	12.8	8.8	15.4	14.7	15.6

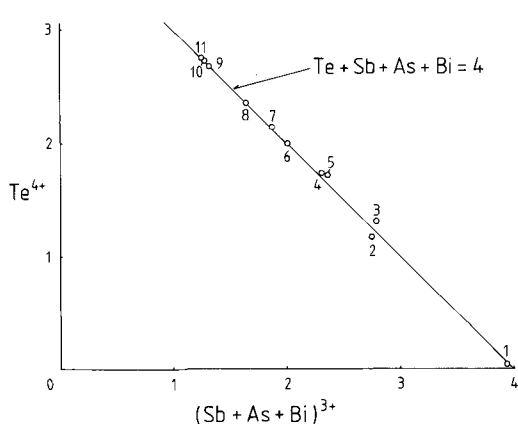


FIG. 1. Relationship between Te and (Sb + As + Bi) in goldfieldite-tetrahedrite from the Iriki and Kawazu mines, Japan. Numbers on the figure correspond to the numbers of the analyses in Table 2.

from the Iriki and Kawazu mines, Japan, verifies the substitution of (Sb + As + Bi) by Te (Fig. 1), through which the charge balance is not attained. Fig. 2 reveals $\Sigma(\text{Cu} + \text{Ag} + \text{Fe} + \text{Zn} + \text{Mn})$ atoms is less than twelve in the case of $\text{Te} > 2$.

Kalbskopf (1974), on the basis of an X-ray study of synthetic goldfieldite-tetrahedrite, pointed out that the substitution of Sb by Te is coupled with a decrease in Cu^{TRG} until the end-member goldfieldite formula

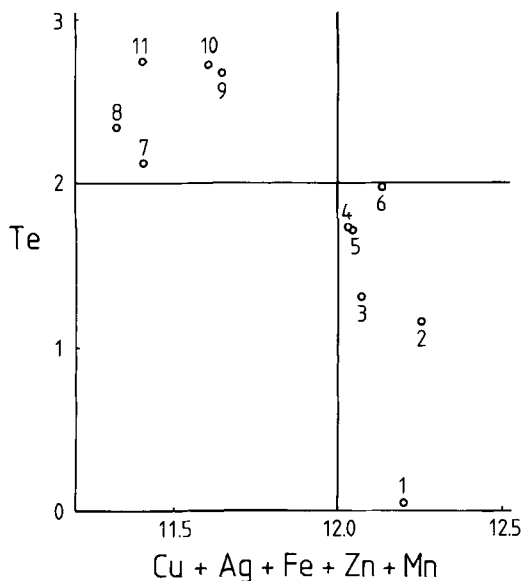


FIG. 2. Relationship between Te and (Cu + Ag + Fe + Zn + Mn) in goldfieldite-tetrahedrite from the Iriki and Kawazu mines, Japan. Numbers on the figure correspond to the numbers of the analyses in Table 2.

$\text{Cu}_6^{\text{TET}}\text{Cu}_{6-x}^{\text{TRG}}\text{Te}_4\text{S}_{13}$, with $x = 2$, is reached. Novgorodova *et al.* (1978), Johnson (1982), Kase (1986), and Knittel (1989) confirmed the coupled substitution of $(\text{Cu}, \text{Fe}, \text{Zn})^{2+}(\text{As}, \text{Sb})^{3+}$ for $\text{Cu}^+\text{Te}^{4+}$ in natural tellurian tennantite, and our data confirm this and also extend it to include tellurian tetrahedrite. Also this paper first confirms the coupled substitution of $\text{Cu}^+(\text{As}, \text{Sb})^{3+}$ for $\square(\text{vacancy})\text{Te}^{4+}$ in natural goldfieldite with $\text{Te} = 2$ to 2.74, based on $\Sigma(\text{Te} + \text{Sb} + \text{As} + \text{Bi}) = 4$. Chemical analyses of Springer (1969), Tsepina *et al.* (1977), Novgorodova *et al.* (1978), Kovalenker *et al.* (1979, 1987), and Knittel (1989) indicate the occurrence of the Cu vacancy as discussed by Kalbskopf (1974) and Johnson (1982), and some of these authors mention that the extra charge caused by substituting Te^{4+} for Sb^{3+} may be compensated by the Cu vacancy. However, their data show the Cu vacancy to occur whether or not the number of Te atoms is greater than two, based on $\Sigma(\text{Te} + \text{Sb} + \text{As} + \text{Bi}) = 4$, and, moreover, do not permit the charge balance to be maintained.

As shown in nos. 7 to 11 in Table 2, the general formulae of natural goldfieldite with $\text{Te} > 2$ from the Iriki and Kawazu mines, Japan are represented by $\text{Cu}_{12-x}(\text{Te}, \text{Sb}, \text{As})_4\text{S}_{13}$, with $x \leq 0.67$. The tabulated data also demonstrate the two-step substitution of (Sb, As) for Te in the goldfieldite-tetrahedrite series, i.e. (i) tetrahedrite, $\text{Cu}_{10}(\text{Cu}, \text{Fe}, \text{Zn})_2^+(\text{Sb}, \text{As})_4^{3+}\text{S}_{13}$ and goldfieldite, $\text{Cu}_{12}^+(\text{Sb}, \text{As})_2^{3+}\text{Te}_2^+\text{S}_{13}$ form a continuous solid-solution according to the coupled substitution of $(\text{Cu}, \text{Fe}, \text{Zn})^{2+}(\text{Sb}, \text{As})^{3+} \rightleftharpoons \text{Cu}^+\text{Te}^{4+}$ as reported by Novgorodova *et al.* (1978), Johnson (1982), and Kase (1986), and (ii) goldfieldite, $\text{Cu}_{12}^+(\text{Sb}, \text{As})_2^{3+}\text{Te}_2^+\text{S}_{13}$ and the ideal end-member, $\text{Cu}_{10}^+\text{Te}_4^+\text{S}_{13}$, only known as a synthetic phase, also form a continuous solid-solution series by the substitution of $\text{Cu}^+(\text{Sb}, \text{As})^{3+} \rightleftharpoons \square(\text{vacancy})\text{Te}^{4+}$.

The general formulae of tellurian tetrahedrite and goldfieldite are respectively therefore, $\text{Cu}_{10+x}^+(\text{Fe}, \text{Zn})_2^{2+}(\text{Sb}, \text{As})_4^{3+}\text{Te}_x^+\text{S}_{13}$, with $x = 0$ to 2 and $\text{Cu}_{12-y}^+\text{Te}_{2-y}^+(\text{Sb}, \text{As})_2^{3+}\text{S}_{13}$, with $y = 0$ to 2.

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