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Roquesite from the Akenobe tin-polymetallic deposits, Southwest Japan

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Abstract: Roquesite grains of the Akenobe mine were analyzed by EPMA. The chemical composition of the roquesite does not show wide range, but changes slightly depending on the mode of occurrence: rods and lamellae of roquesite in sphalerite contain Zn in the range from 2.23 to 3.30 wt% ; roquesite grains at the boundary between sphalerite and chalcopyrite contain Zn (0.00-3.36 wt%) and Fe (0.00-1.62 wt%) ; roquesites as inclusion in Cu-sulfides (bornite, chalcopyrite, etc.) contain Fe (0.29-0.84 wt%). The incorporation of Zn and Fe in roquesite seems to be governed by the following substitution: $Cu^{+} + In^{3+} = Zn^{2+} + Fe^{2+}$. The narrow range of chemical composition and the textural evidences imply that this mineral species exsolves from host minerals.

Introduction

Roquesite, $CuInS_2$, has been described by mineralogists (Picot and Pierrot, 1963 ; Kato and Shinohara, 1965 ; Sutherland and Boorman, 1969 ; Burke and Kieft, 1980 ; Cantinolle *et al.*, 1985 ; Seetharam, 1986 ; Ohta, 1989). This mineral rarely occurs in deposits where cassiterite and Cu-sulfides (especially bornite) coexist. The Akenobe mine is an example of tin-polymetallic veins where the authors found roquesite from the Ginsei and Ryusei veins. Recently syntheses of indium-bearing sulfide system became important (Moh, personal communication) in the development of electronics, and the data on roquesite are necessary still now for this field. Thus the chemical composition of the mineral will be deposited here although Kato and Shinohara (1965) already reported the occurrence of roquesite from the Eisei vein of Akenobe.

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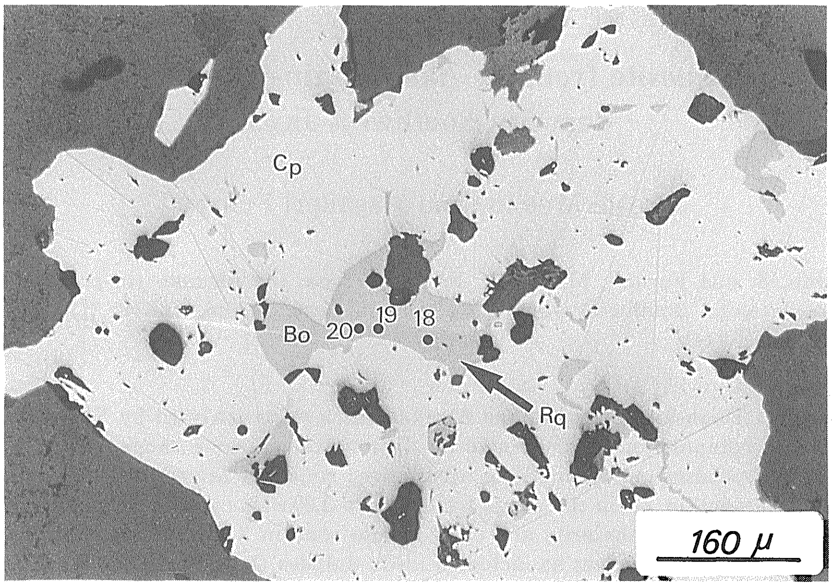
Akenobe mine

The Akenobe mine is a representative tin-polymetallic deposit in Japan. The ore deposit, embedded in the Maizuru structural belt, consists more than 130 veins within the area of 4 × 4 km. The veins show crustification and contain some of the following bands : Pb-Zn, Cu-Zn, Cu-Sn and Sn-W bands. In tensional fractures, the zonal structure of some of these bands is clearly observed, and also cross-cutting relations of veins are often recognized. Due to these facts, structural stages of mineralization, stages I to III, can be established. The stage I ore is, characterized by galena and sphalerite, i.e. Pb-Zn band. Stage II carries Cu-Zn band and chloritization. Stage III contains Cu-Zn[1)], Cu-Sn and Sn-W (cassiterite and

[1)] Stage II Cu-Zn band cuts stage III Cu-Zn band.

Keywords: Akenobe mine, indium, roquesite, tin deposit, polymetallic deposit, EPMA



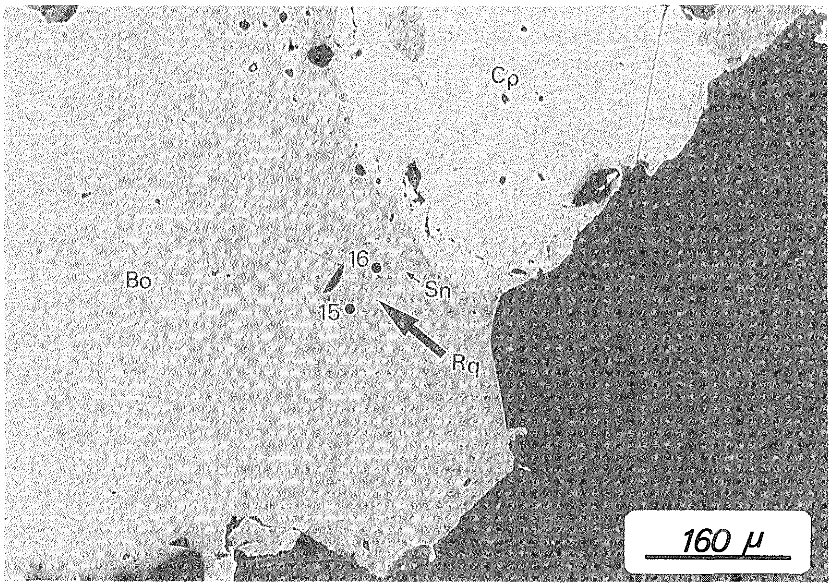


Fig. 1 Microphotographs of roquesite of the Ginsei vein, Akenobe. Rq, roquesite; Bi, bismuth minerals ; Bo, bornite ; Cp, chalcopyrite ; Gn, galena ; Sn, stannite ; Tn, tennantite-tetrahedrite series. Numbers of solid circles correspond to those after GS in Table 1.

wolframite) bands[2] accompanied with silicification, sideritization and dickitization.

The Ginsei vein holds Pb-Zn band on upper levels and Cu-Sn band in the deeper portion. According to Sato and Akiyama (1980), the vein contained 2.93% Cu, 6.52% Zn and 0.63% Sn. The present authors surveyed No.43 stope, −9 L.[3] where stage III mineralization is well developed (Fig. 16 in Murao and Andoh, 1989) and examined the samples.

The Ryusei vein is of Pb-Zn, Cu-Zn, Cu-Sn and Sn-W bands and carried 3.55% Cu, 1.04% Zn and 2.31% Sn in average. The authors studied some stopes on −10 L. where stage III mineralization of Cu-Sn character is distinctive.

Mode of occurrence

In the samples from the Ginsei vein, well-polished tabular crystals of roquesite were found together with bornite and chalcopyrite (Fig. 1) and/or tennantite-tetrahedrite series. The grains are light gray in color and exhibit very weak anisotropism. In the specimen of Ryusei, there are two modes of occurrence : (1) rods and lamellae (ca. 10–20×5 micron) in sphalerite (Fig. 2) ; (2) islands of larger size compared to rods and lamellae (50 × 50 micron) in chalcopyrite (Fig. 3). The type (1) grains are randomely distributed in sphalerite and coexist with rods of chalcopyrite of nearly the same size. In the roquesite-hosting sphalerite, dusts of chalcopyrite which make the sphalerite mostly opaque are observed. The area of roquesite and rod-like chalcopyrite and that of dusty chalcopyrite are clealy separate. This texture implies the formational mechanism of the ore (Murao and Furuno, in preparation).

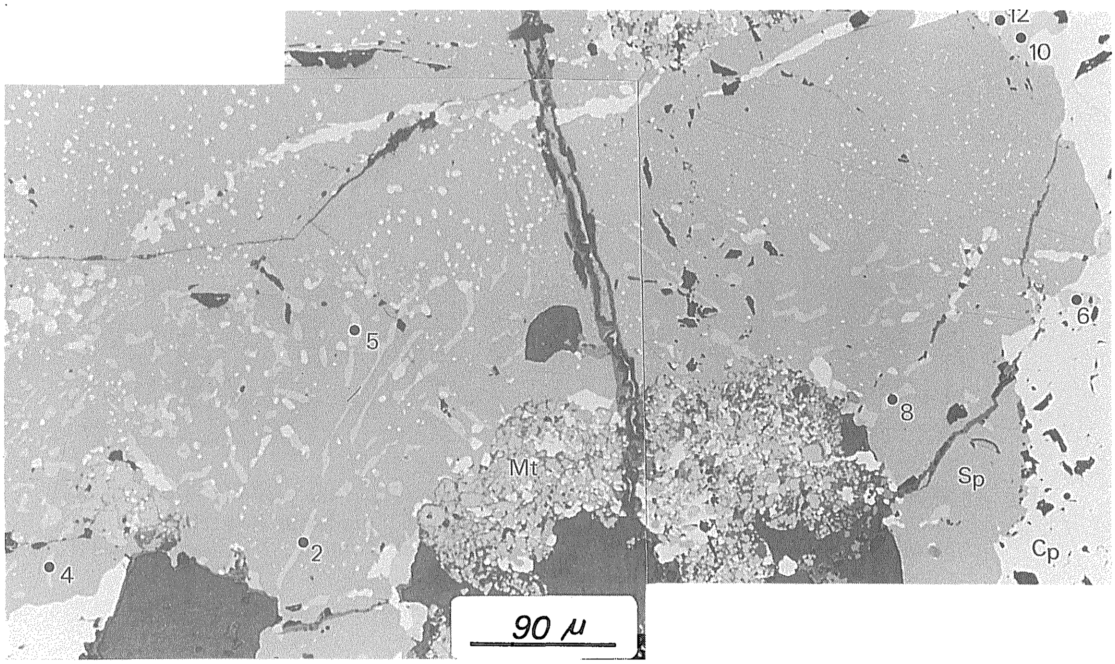


Fig. 2 Microphotograph of lamellae of roquesite in sphalerite. Ryusei vein, Akenobe. Mt, magnetite. Numbers correspond to those after RS in Table 1.

[2] Cu-Sn band grades into Sn-W band.

[3] 0 L. : 380 m above sea level ; level interval : 30m.

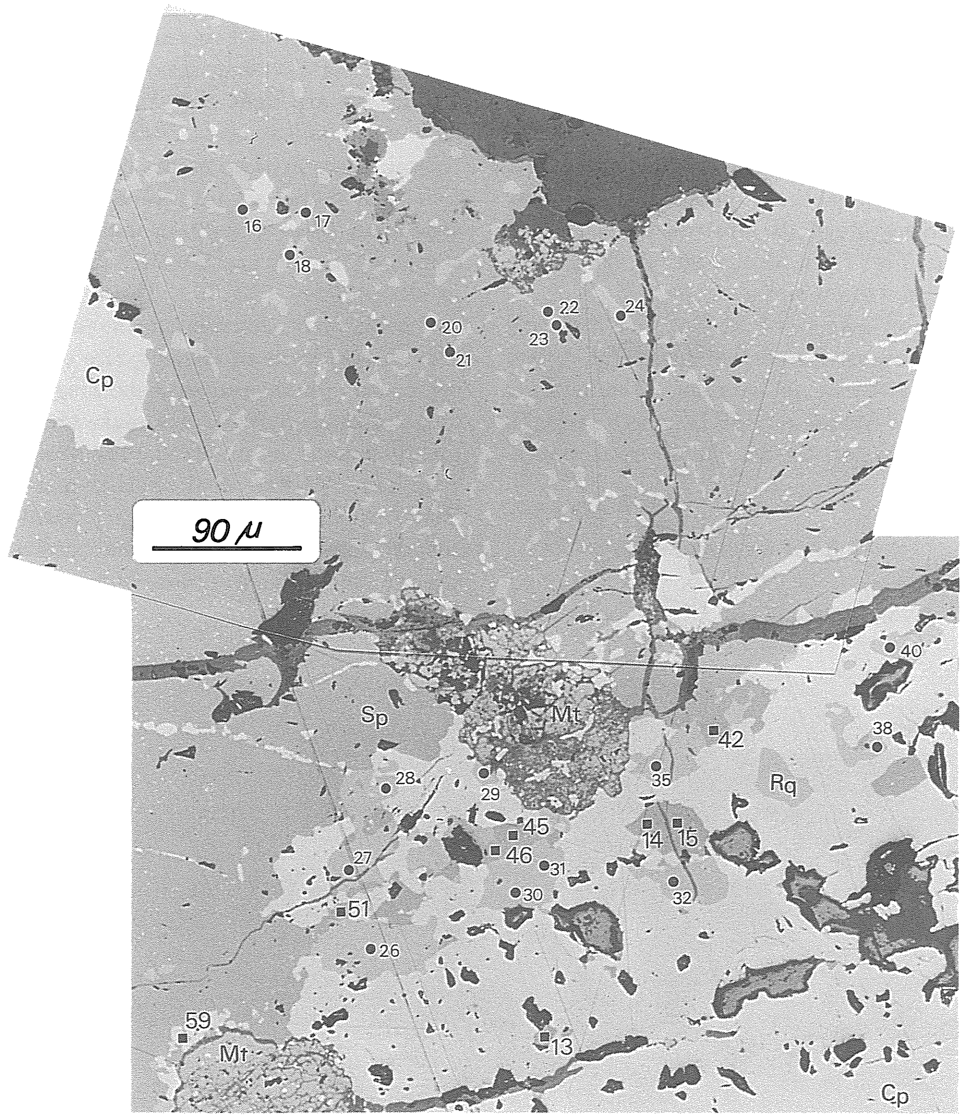


Fig. 3 Microphotograph of lamellae of roquesite (upper side of the picture) in sphalerite and tabular roquesite at the boundary between sphalerite and chalcopyrite. Numbers of solid circles correspond to those of Table 1. Numbers of solid squares, to Table 2.

Chemical composition

The chemical compositions of the three varieties, tabular roquesite with Cu-sulfides, tabular roquesite with chalcopyrite and sphalerite, and rods and lamellae of roquesite in sphalerite, were examined by EPMA. The analytical conditions are as follows : analyzer, TN 5400 of Tracor Northern ; accelation voltage, 20 kV ; probe current, 1.0×10^{-10}A ; take-off angle, 40° ; beam diameter, less than 0.1 micron ; counting time, 10 s. The standards are native $CuFeS_2$, synthetic ZnS, FeS, Ag_2S,

Table 1 Chemical composition of roquesite, Akenobe, Hyogo, Japan.

Sample No.		Cu	In	Fe	Zn	S	Total	Associated Minerals
RS-02	wt%	25.02	45.43	0.21	2.24	25.89	98.79	Sp
	atom	1.92	2.00	0.06	0.07	3.95	8	
RS-04	wt%	24.74	45.50	0.14	2.80	25.45	98.62	Sp
	atom	1.91	1.93	0.05	0.21	3.90	8	
RS-05	wt%	25.00	46.10	0.19	2.85	26.03	100.18	Sp
	atom	1.89	1.93	0.04	0.14	4.00	8	
RS-06	wt%	25.08	46.68	0.64	0.92	25.74	99.06	Cp, Sp, Gn, Bi
	atom	1.93	1.92	0.06	0.12	3.97	8	
RS-08	wt%	24.20	45.79	0.57	2.81	26.18	99.55	Sp
	atom	1.84	1.94	0.00	0.17	4.05	8	
RS-10	wt%	25.63	45.50	0.40	1.93	25.80	99.27	Cp, Sp
	atom	1.95	1.92	0.03	0.25	3.85	8	
RS-12	wt%	26.02	45.94	0.71	1.56	26.22	100.45	Cp, Sp
	atom	1.96	1.95	0.03	0.17	3.81	8	
RS-16	wt%	26.04	45.67	0.00	2.32	25.60	99.63	Sp, Cp
	atom	1.99	1.95	0.04	0.23	3.79	8	
RS-17	wt%	25.15	45.54	0.37	3.36	25.78	100.20	Sp, Cp
	atom	1.90	1.94	0.02	0.25	3.89	8	
RS-18	wt%	25.20	46.15	0.32	2.23	25.91	99.81	Sp
	atom	1.92	1.94	0.09	0.00	4.05	8	
RS-20	wt%	25.00	46.18	0.43	3.08	25.65	100.34	Sp
	atom	1.90	1.95	0.09	0.00	4.06	8	
RS-21	wt%	23.88	45.45	0.20	3.30	25.94	98.77	Sp
	atom	1.83	1.92	0.02	0.14	4.09	8	
RS-22	wt%	25.05	45.81	0.29	2.59	25.77	99.51	Sp
	atom	1.91	1.94	0.02	0.19	3.94	8	
RS-23	wt%	25.25	46.39	0.24	2.28	26.09	100.24	Sp
	atom	1.91	1.95	0.02	0.17	3.95	8	
RS-24	wt%	24.29	45.35	0.24	2.90	26.07	98.85	Sp
	atom	1.85	1.93	0.02	0.22	3.98	8	
RS-26	wt%	26.23	45.91	0.46	0.00	26.54	99.13	Cp, Sp
	atom	1.99	1.94	0.04	0.00	4.03	8	
RS-27	wt%	25.77	45.99	0.43	0.31	26.17	98.67	Cp, Sp
	atom	1.97	1.96	0.04	0.02	4.01	8	
RS-28	wt%	25.62	46.10	1.10	0.71	26.32	99.85	Cp, Sp
	atom	1.94	1.94	0.09	0.05	3.98	8	
RS-29	wt%	26.45	46.26	1.62	0.40	25.91	100.64	Cp, Sp
	atom	1.99	1.94	0.14	0.03	3.90	8	
RS-30	wt%	26.37	46.64	0.38	0.68	26.26	100.33	Cp, Sp
	atom	1.99	1.96	0.03	0.05	3.97	8	
RS-31	wt%	26.23	46.04	0.74	0.36	25.58	98.96	Cp, Sp
	atom	2.01	1.97	0.06	0.03	3.93	8	
RS-32	wt%	26.27	46.02	0.65	0.40	25.90	99.23	Cp, Sp, Gn
	atom	2.00	1.96	0.06	0.03	3.95	8	
RS-35	wt%	26.30	46.54	0.88	0.00	25.86	99.59	Cp, Sp
	atom	2.00	1.97	0.08	0.00	3.95	8	
RS-38	wt%	26.28	46.22	1.00	0.36	25.58	99.44	Cp, Sp
	atom	2.01	1.97	0.09	0.03	3.90	8	
RS-40	wt%	25.69	45.96	1.06	0.00	26.42	99.13	Cp
	atom	1.95	1.94	0.02	0.17	3.82	8	
RS-71	wt%	25.68	45.94	1.06	0.00	26.26	98.95	Sp, Cp
	atom	1.96	1.95	0.01	0.21	3.89	8	

Table 1 Continued

Sample No.		Cu	In	Fe	Zn	S	Total	Associated Minerals
RS-73	wt%	26.21	46.07	0.24	1.91	26.55	100.97	Sp, Cp
	atom	1.96	1.94	0.02	0.21	3.87	8	
RS-75	wt%	24.95	45.32	0.39	2.34	26.03	99.02	Sp, Cp
	atom	1.90	1.92	0.03	0.18	3.97	8	
RS-77	wt%	25.09	46.21	1.11	0.26	26.09	98.76	Sp, Cp
	atom	1.92	1.97	0.10	0.02	3.99	8	
RS-79	wt%	25.90	45.79	1.30	0.00	26.30	99.28	Sp, Cp, Bi
	atom	1.96	1.93	0.11	0.00	4.00	8	
RS-80	wt%	26.12	46.18	1.31	0.00	26.40	100.01	Sp, Cp, Bi
	atom	1.97	1.94	0.11	0.00	3.98	8	
GS-12	wt%	26.19	46.56	0.41	0.80	26.21	100.16	Sp, Cp, Gn, Bi
	atom	1.98	1.96	0.04	0.06	3.96	8	
GS-13	wt%	25.64	46.61	0.20	0.56	25.66	98.67	Sp, Cp, Gn, Bi
	atom	1.98	2.00	0.18	0.04	3.80	8	
GS-15	wt%	27.52	45.95	0.29	0.00	25.26	99.02	Bo, Bi
	atom	2.12	1.97	0.03	0.00	3.88	8	
GS-16	wt%	28.39	45.44	0.68	0.00	25.46	99.97	Bo, Bi
	atom	2.16	1.92	0.06	0.00	3.86	8	
GS-18	wt%	26.92	47.24	0.84	0.00	26.15	101.16	Bo
	atom	2.02	1.98	0.07	0.00	3.93	8	
GS-19	wt%	27.41	46.76	0.67	0.00	25.91	100.75	Bo
	atom	2.07	1.96	0.06	0.00	3.91	8	
GS-20	wt%	26.96	46.66	0.81	0.00	25.86	100.29	Bo
	atom	2.04	1.97	0.07	0.00	3.92	8	
GS-27	wt%	26.79	46.96	0.00	0.88	26.30	100.94	Sp, Cp, Gn, Bi
	atom	2.01	1.96	0.00	0.07	3.96	8	
GS-28	wt%	25.62	46.11	0.37	0.51	25.88	98.49	Sp, Cp, Gn, Bi
	atom	1.97	1.97	0.03	0.04	3.99	8	
GS-35	wt%	26.53	46.33	0.58	0.00	26.22	99.67	Ten, Cp
	atom	2.01	1.96	0.05	0.00	3.98	8	
GS-40	wt%	26.47	46.58	0.43	0.00	26.25	99.74	Ten, Cp
	atom	2.01	1.96	0.04	0.00	3.99	8	
GS-41	wt%	26.81	45.19	0.56	0.00	26.10	98.66	Ten, Cp
	atom	2.05	1.92	0.05	0.00	3.98	8	
GS-41′	wt%	26.77	46.43	0.76	0.00	26.34	100.30	Cp, Ten
	atom	2.02	1.95	0.07	0.00	3.96	8	
GS-43	wt%	26.58	46.90	0.44	0.40	26.09	100.41	Ten, Cp
	atom	2.01	1.97	0.04	0.03	3.95	8	
GS-59	wt%	26.78	45.69	0.67	0.00	26.20	99.34	Bo, Cp
	atom	2.03	1.93	0.06	0.00	3.98	8	
GS-60	wt%	26.60	45.86	0.37	0.00	25.72	98.55	Bo, Cp
	atom	2.04	1.96	0.03	0.00	3.97	8	
GS-65	wt%	25.82	46.48	0.31	0.32	26.35	99.28	Sp, Cp, Gn, Bi
	atom	1.97	1.97	0.03	0.02	4.01	8	

$Au_{80}Ag_{20}$, InAs, Bi_2Se_3, PbTe, CdS, NiS, metallic Sb, Sn and Mn. The data were on-line ZAF corrected.

The results are shown on Tables 1, 2 and Figs 4 and 5. The chemical compositions of roquesite show slight change from $CuInS_2$ depending on the mode of occurrence : roquesites which occur as lamellae of sphalerite contain Zn (up to 3.30 wt%) ; roquesites on the chalcopyrite and sphalerite boundary contain Zn (up to

Table 2 Chemical composition of sphalerite which coexists with roquesite.

Sample No.		Zn	Fe	Cu	Cd	In	S	Total	As. Min.
RS-13	wt. %	61.19	1.30	2.50	0.49	1.48	33.54	100.50	Cp. Rq
	at. %	45.50	1.12	1.88	0.21	0.62	50.65		
RS-14	wt. %	60.74	1.46	1.81	1.00	2.07	33.07	100.17	Cp, Rq
	at. %	45.51	1.28	1.40	0.44	0.88	50.50		
RS-15	wt. %	62.33	1.11	1.41	0.70	1.30	33.15	100.01	Cp, Rq, Bi
	at. %	46.68	0.96	1.08	0.30	0.55	50.43		
RS-42	wt. %	59.92	0.84	2.22	1.09	2.83	32.62	99.51	Cp, Rq
	at. %	45.52	0.74	1.71	0.48	1.22	50.34		
RS-45	wt. %	55.76	1.22	4.31	0.30	5.04	31.75	98.38	Rq, Cp
	at. %	43.20	1.10	3.39	0.13	2.21	49.97		
RS-46	wt. %	57.49	1.06	2.80	0.74	4.79	32.04	98.93	Rq, Cp
	at. %	44.29	0.95	2.19	0.33	2.09	50.15		
RS-51	wt. %	62.69	1.03	0.93	1.00	0.57	32.86	99.09	Cp, Rq
	at. %	47.32	0.90	0.71	0.44	0.24	50.38		
RS-59	wt. %	64.59	0.99	1.05	0.49	0.00	32.43	99.54	Cp
	at. %	48.58	0.87	0.80	0.21	0.00	49.54		
RS-60	wt. %	58.81	0.70	2.92	0.00	4.32	32.36	99.12	Cp, Mt, Rq
	at. %	44.97	0.63	2.27	0.00	1.87	50.26		

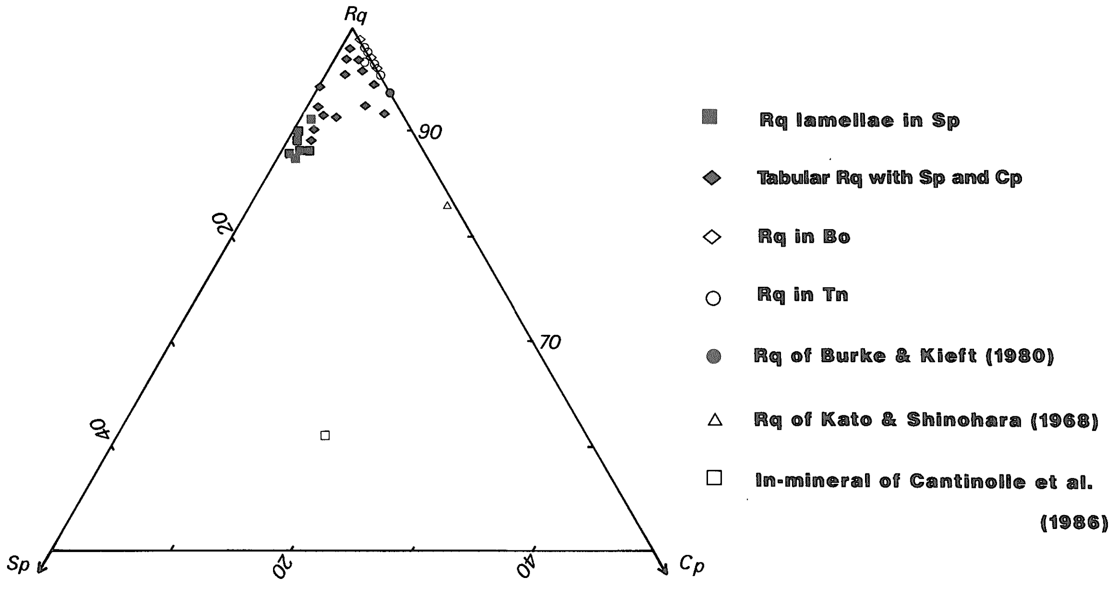


Fig. 4 Data plot in terms of roquesite, sphalerite and chalcopyrite mols.

3.36 wt%) and Fe (up to 1.62 wt%) ; roquesites in bornite and/or tennantite-tetrahedrite series show the narrowest range of the composition. The host sphalerite of roquesite lamellae contains little amount of indium and copper.

Concluding remarks

The chemical compositions of roquesite at Akenobe change slightly with the mode of occurrence. When the data are plotted on the At%$_{(Cu+In)}$ vs. At%$_{(Fe+Zn)}$ diagram, the regres-

Table 3 Texture of roquesite-containing ore at Akenobe and other localities.

Locality	Mode of Occurrence	Mineral Assemblage	Reference
Akenobe, Japan	Lamellae in Sp, when lamellae intersect, their width is thined at the intersection.	Sp-Cp-Rq-Bi minerals -Mgt-Fe-Chl	this study
Akenobe, Japan	Tabular crystals at the boundary between Sp and Cp.	ditto	ditto
Akenobe, Japan	Tabular crystal at the boundary between Cp and Bo.	Cp-Bo-Rq	ditto
Akenobe, Japan	Tabular crystal in Bo near the boundary between Cp and Bo.	Bo-Rq-Sn-Cp	ditto
Akenobe, Japan	Inclusion in Tn.	Tn-Rq-Cas-rutile	ditto
Akenobe, Japan	Lamellae exhibiting graphic texture with Sp.	Cp-Rq-Sp-Wt-Mgt-Cas etc.	Kato and Shinohara (1968)
Toyoha, Japan	Bands or grains intergrown with Ks and In-mineral. Occur near the boundary between Sp and Cp (Fig. 3E, Ohta, 1989)	Rq-In mineral-Sk-Ks-Cp-Sp	Ohta (1989)
Charrier, France	Inclusion in Bo.	Bo-Cp-Rq-Cu-Wt-Fe-Chl etc.	Picot et Pierrot (1963)
Les Clochettes, France	Inclusion in Bo. Rarely accompanies Cp.	Bo-Cp-Rq-Wt-Ak-Po-Sp-Gn-Mw-Cas etc.	Picot (1973)
La Telhaie, France	Inclusion in Gn	Sp-Gn-Rq-Py-Cp etc.	Cantinolle *et al.* (1985)
Vaulry, France	Inclusion in Bo.	Bo-Cp-Rq-Wt-Mw-Std-Tn-Ks etc.	ditto
Långban, Sweden	Small roundish grains (max. 30 μ) in Bo and Cc. Always associated with Cu-In-bearing Sp.	Bo-Cc-Rq-Gn-Sp-Bi	Burke and Kieft (1980)
Gåsborn, Sweden	As mixture of Fe-poor Sp, Cp and Rq replacing In-Sp.	Fe-poor Sp-Cp-Rq	Kieft and Damman (1990)
Tosham, India	Tabular crystal of ca. 16 μ in diameter at the boundary between Cp, Sp and Sn.	Cd and In-free Sp-Cp-Rq-Sn-Bm-Py-Cas etc	Seetharam (1986)
Mount Pleasant, Canada	Subhedral to rounded grains up to 10 μ in size occuring mainly on Cp and Sp grain boundaries, or smaller grains in Cp with calcite and fluorite.	Sp-Cp-Rq	Sutherland and Boorman (1969)

Ak, aikinite; Bm, bismuthinite; Cas, cassiterite; Cc, chalcocite; Chl, chlorite; Cp, chalcopyrite; Ks, kesterite; Mgt, magnetite; Mw, mawsonite; Po, pyrrhotite; Rq, roquesite; Sk, sakuraiite; Sn, stannite; Sp, sphalerite; Std, stannoidite.

sion line can be expressed as (Cu + In) = −0.9 (Fe + Zn) + 50.3 (Fig. 5). Although the data are too partial to discuss the substitution between $CuInS_2$ and $CuFeS_2$ or ZnS, the slope of the regression line is close to −1, and this means that the incorporation of Fe and Zn in roquesite from Akenobe may be governed by the following substitution : $Cu^{+} + In^{3+} = Fe^{2+} + Zn^{2+}$. The fact that the correlation coefficient between Fe and In contents is small (0.03) also indicates that the iron is not trivalent but divalent. This result is in harmony with Ohta's (1989) work. The authors have been studying the indium minerals to clarify their formational mechanism. The narrow range of the chemical compositions and the texture of roquesite (Table 3), together with the chemical composition of coexsisting sphalerite imply that roquesite exsolves from a Cu-Zn-Fe-In-S phase. The detailed analyses and the observa-

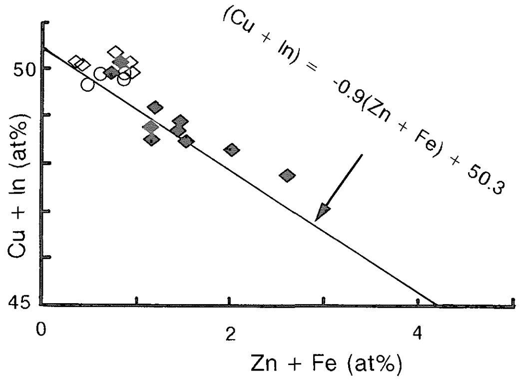


Fig. 5 Correlation of (Cu + In) with (Zn + Fe) in roquesites from the Akenobe tin-polymetallic deposit, Japan. Symbols, see Fig. 4.

tion are desirable to solve the problem.

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明延鉱山産ロケサイトの化学組成

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要　旨

ロケサイトの生成機構解明の一助とするため明延鉱山産鉱石の EPMA 分析を行った．ロケサイトは (1) 閃亜鉛鉱中のラメラ，(2) 閃亜鉛鉱と黄銅鉱の境界部に出現する板状結晶，(3) 黄銅鉱など銅の硫化物中に包有される板状結晶の 3 通りの産状を示すが化学組成は産状に応じてわずかに変動する．すなわち (1) は少量の Zn を，(2) は Fe，Zn を，(3) は少量の Fe を持っている．Fe と Zn は $Cu^{+} + In^{3+} \rightleftarrows Fe^{2+} + Zn^{2+}$ なるメカニズムでロケサイト中に取りこまれると考えられる．鉱石組織や化学組成から本鉱物は親結晶からの離溶によって生成すると想像されるがその証明にはさらなる検討を要する．

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