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On Prehnite and Some Others from the Takase Mine, Okayama Prefecture

By

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With 3 Tables, 3 Text-figures, and 2 Plates

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ABSTRACT: The vein-like lenticular mass composed of prehnite and some others have been found intervening within serpentinite in the Takase mine famous for production of the chromite ores on an industrial scale and studied somewhat genetically with respect to so-called "rodingite". In addition, thermal behaviors of prehnite and garnet have been pursued for inspection of their genesis or interrelations.

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I. INTRODUCTION

The irregularly lenticular masses composed chiefly of garnet, diopside, chlorite and prehnite are found traversing through serpentinite in a close relation to the chromite deposits of the Takase mine situated in Shingō-chō, Atetsu-gun, Okayama Prefecture. It may be that these kinds of occurrence and mineralic assemblages bear an alliance to those of the veins or zones defined previously as *rodingite* enclosed ordinarily in the basic to ultra-basic rocks.

FINLAYSON (1909) regarded the rodingite as a derivative from the basic magma rich in Ca through digestion of limestone, MARSHALL (1911) did so as a primary product resulted from crystallization of magma, GRANGE (1927) as that derived through alteration and metasomatism of gabbro in the case of serpentinitization of lime-bearing pyroxene contained in the ultra-basics, and BLOXAM (1954) as that brought forth from hydrothermal solution predominant in Ca in association with serpentinitization, while WATSON (1953) pointed out the formation of prehnite and diopside through substitution of albite with Ca derived from clinopyroxene-bearing peridotite accompanying serpentinitization but BILGRAMI and HOWIE (1960) demonstrated also that of the related silicates as a result of combination of Ca obtained through serpentinitization of peridotite with

Al supplied from plagioclase through hydrothermal effects.

Concepts of the rodingite, presented in these works, seem to have been offered as a whole simply for an apparently single body composed of several characteristic minerals almost regardless of their respective relations to one another, whereas the related masses recognized in the Takase mine are, though happenly in accordance with those described previously in mineralic constituents, considered to have been produced through successive but not simultaneous mineralogenesis. It appears also of due significance that such minerals as are rich in Ca and Al are found contained as a unique occurrence specifically in serpentinite or its allied rock considered rather poor in the same elements.

On the other hand, the experimental data as to hydrothermal synthesis of prehnite and grossularite (including hydrogrossularite) together with those for their equilibrate relation have recently been yielded by YODER (1950), ROY and ROY (1952) and COOMBO et al. (1959).

There thus remain some rooms for clarifying whether the rodingite is to be distinguished from the allied masses of the Takase mine or not and paragenesis of the minerals in question in more details.

II. OCCURRENCE OF THE CONSTITUENT MINERALS

The chromite ore-bodies of the Takase mine, discontinuously lenticular or irregularly massive in form, are found developed with a general trend of westward plunge in serpentinite and now being incised for mining through five main adits. One of the white-colored vein-like mass under consideration is observed coming into contact with serpentinite on the highest level running into the easternmost part of the ore body and the other is recognized traversing the similar country rock on the lowest level aiming at the westernmost piece of the same body, both pinching out towards both sides in their spindle-shaped sections appeared on the side walls of respective adits and being demarcated irregularly but distinctly with the surroundings without any gradual variation to each other (see Fig. 1). Noteworthy is that these masses are apt to occur relatively close to the bounds of ore formation, although any others have not yet been discovered on the other intermediate levels.

The specimens, named sample A (A1~A4), collected from the uppermost level called "Higashi-ichi-kō" (the eastern No. 1 adit) are abundant in grossularite and diopside associated with a subordinate amount of prehnite, epidote and chlorite. Grossularite is dividable into two species: one (grossularite I) is characterized with pale flesh color and the other (grossularite II), cutting the former as tiny veinlets, with deep reddish brown color. Such an optical anomaly as is apt to be observed commonly in calcareous garnet is not verified in both species accompanying few quantities of inclusions. Diopside is also divided into two

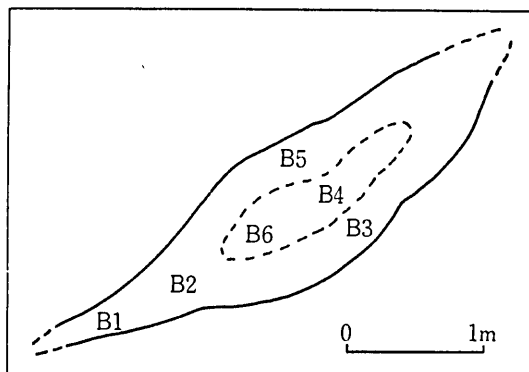


FIG. 1. The lenticular mass observed on the lowermost level (Shinguchi-ko)

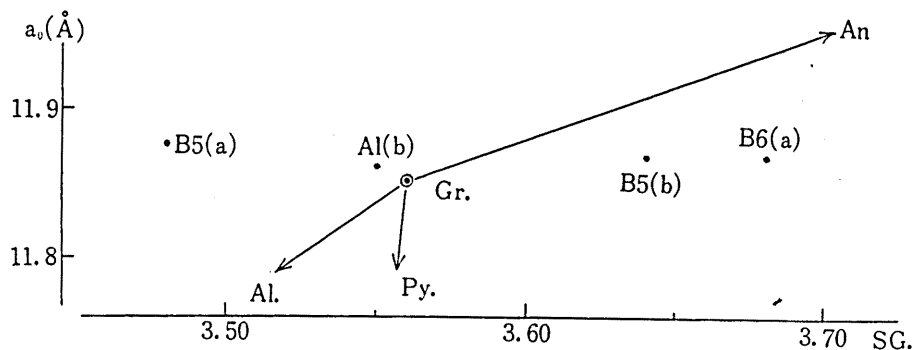
species : those (diopside I) scattering as large-scaled crystals, 3×3 cm² in an average section, in the aggregates of grossularite I are black to greenish black in color and show distinct cleavages, along which opaque minerals produced on alteration are arranged, resulted from folding, while those (diopside II), very small in grain-size, are observed cutting across the former and bear no traces of deformation and alteration. Prehnite replaces grossularite I, diopside I and a part of grossularite II, occurring as veinlets with variable scale or as small masses with scale of more or less than 1 cm in diameter.

The specimens named sample B (B1~B6) are those picked up from the lowermost level called "Shinguchi-kō" (the newly opened adit). Sample B 1 taken out at the end of the elongated mass, rather whitish in color and soft in property on the whole, is constituted of chlorite, diopside, prehnite, grossularite and calcite formed subsequent to the former four. As is evidently exhibited in Pl. 1-3, the part (r), pure white in color, is occupied mainly by prehnite including chlorite and diopside; the part (s), pale greyish green in color, is filled up by diopside indicating the irregularly massive section with diameter of 1~2 cm and a little amount of grossularite; the part (p) colored in the range between (r) and (s) is represented by diopside, chlorite and a slight quantity of prehnite, occurring as veinlets and as irregular masses; and the part (q), deep green to greenish black in color, embraces the chlorite spots with diameter of less than 5 mm. Diopside embedded in (s) is discolored one derived from diopside I, replaced partly by grossularite and demarcated sharply with the surrounding prehnite. Diopside contained in (r), corresponding to diopside II, is only röntgenographically determinable because of its microscopic obscurity in prismatic or radial form, including a little amount of chlorite cut by prehnite. Diopside in (r) is classifiable into diopside I and II and penetrated by prehnite somewhat earlier than that in (r). Diopside I included in the sample B 2 is replaced partly by chlorite only but not so by the later prehnite. The specimens named sample B 3' are those obtained

from the foot-wall situating at the end of the main mass. As is shown in Pl. I-1, the part (m), blackish green in color, is composed of chlorite, forsterite and a slight amount of serpentine, among which the second is surrounded, traversed and replaced by the first and the third. The part (o), blackish grey in color, is represented by diopside I, 4~5 cm long and 2~3 cm wide, characterized with distinct cleavage and trace of deformation, replaced by chlorite and cut by calcite. The part (l) is the veinlet of diopside II, greenish white in color and 1 mm wide, cutting through (m). The part (n), greenish white in general but intensely variable in color, is composed of chlorite, diopside II and a little amount of garnet assumed as the relics from replacement, running along (m) with width of 1~2 cm. The part (p), almost pure white in color, includes prehnite and chlorite, filling up the interstices among the other parts. Moreover, the veinlets of grossularite and calcite are observed traversing all the parts mentioned above. In the sample B 4', as is clear in Pl. I-2, prehnite bearing the inclusions of diopside I and garnet remained from replacement is comprised as irregular veinlets replacing the reddish brown-colored grossularite and accompanied with the later chlorite, colorless to pale green in color, opaque, and fibrous in habit. Prehnite included in the sample B 4 replaces the deep reddish grossularite as irregularly running veinlets. In the sample B 5 taken out near the hanging wall of the main mass, grossularite with color ranging from white to flesh is recognized cutting across the black-colored diopside I, both being cut by another veinlets of the reddish grossularite traversed by the later calcite veinlets. In the sample B 6 obtained from the central part of the main mass, the black- to greenish grey-colored diopside I is surrounded by the deep reddish grossularite replaced partly by prehnite. To be remarked is that in some druses hidden within the reddish grossularite the dodecahedral crystals of transparent, white or colorless grossularite and the other white-colored, slender needle-like ones with length of about 0.5 mm, though still not identified owing to difficulty of sampling, are found grown (see Pl. I-4).

Since each grossularite sampled from the main mass is, as has been alluded to already, apparently too rich in variety of tint and of occurrence to be distinguished from one another, both specific gravity and lattice constant have been determined for reference concerning four specimens. The data are shown in Table I and furthermore plotted on the diagram given by WINCHELL (1958), as is exhibited in Fig. 2.

Inspection of the figure reveals that only B 5 (a) situating considerably distant from the grossularite area is probably capable of being identified with hydro-grossularite if regardless of a_0 value a little smaller than is expected. As for this regard, HUTTON (1943) was of opinion that the garnet derived from replacement of gabbro and basic intrusive is grouped nearly into an end member of garnet-hydrogarnet series, while after HUTTON and YODER (1950) most of the specimen regarded previously as ordinary grossularite are surely classifiable into



Al : Almandite, An : Andradite, Gr : Grossularite, Py : Pyrope

FIG. 2. Relation of a_0 to specific gravity.

Table 1

Sample	Color	Specific gravity	a_0 (Å)
B 6 (a)	deep red	3.68	11.868
B 5 (b)	reddish brown	3.64	11.868
A 1 (b)	pale flesh	3.55	11.860
B 5 (a)	colorless	3.48	11.875
grossularite*		3.560	11.851

Asterisk relating to the data of SRIRAMADAS (1954), SKINNER (1956) and WINCHELL (1958).

hydrogarnet.

In order to confirm whether the results shown in Table 1 may be valid or not, infrared absorption spectra for two specimens have, though limited within the wave-length ranging from 4000 to 700 cm^{-1} , been examined referred to the data offered by LAUNER (1952). The apparatus provided for, and the procedures put into, operation in the experiments have been quite similar to those described formerly by UMEGAKI et al. (1957). Inspection of the principal bands shown in Table 2 evidently points to the identity of two specimens subjected to the experiments with the ordinary grossularite given by Launer (cf. HUNT et al., 1950 and CLARK, 1957).

Table 2

Sample	Principal bands in μ			
Grossularite*	10.4	10.95	11.65	11.92
B 5 (a)	10.43	10.98	11.67	11.94
A 1 (b)	10.48	10.98	11.73	12.00
Andradite*	10.6	11.17	11.90	12.22

Asterisk showing the data given by LAUNER (1950).

Nevertheless, taking account of that any references to the infrared absorption

relating to hydrogrossular have not been presented in the previous literatures and uncertain bands other than the main ones are also recognized in the absorption figures, presence of hydrogrossularite may not be denied based on more elaborate research particularly within the wave-length ranging up to 400 cm.^{-1}

III. CONSIDERATIONS ON THE DATA OBTAINED THROUGH THERMAL EXPERIMENTS

In the light of an intimate relation of prehnite to grossularite in the vein-like masses intercalated within serpentinite of the Takase mine as well as to hydrogrossularite in the other cases (cf. BLOXAM, and BILGRAMI and HOWIE) and of the formal approximation in their chemical formulae, the thermal behaviors of these two minerals have, as a means for pursuit of the necessity bringing on paragenesis, been traced almost after the manner of the precedent made public by UMEGAKI et al. (1963).

(1) Prehnite

The specimens provided for heat treatment are appurtenant to the sample B 4 collected near the central part of the related mass occurring in Shinguchi-kō as irregular replacement veinlets, 2~3 cm wide, penetrating the aggregate of the deep reddish grossularite and include a lick of chlorite and grossularite as artificially unavoidable impurities. X-ray diffraction data obtained for these specimens at the room temperature exhibit the similarity in each spacing to those for prehnite listed in A. S. T. M card excepting the splitting of 3.28Å into 3.30Å and 3.27Å and the interposing of chlorite and grossularite in a slight amount.

Variation in the spacings for the pulverized specimens examined preliminarily has, though within the range of $12^\circ\sim 40^\circ$ in 2θ , been continuously traced during heat treatment for an hour at 100°C up to $1,000^\circ\text{C}$.

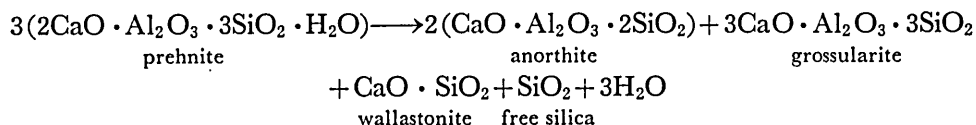
The spacings in Å and the corresponding intensities in cps appearing with increase of each 100°C are classifiable into the strengthened, weakened and invariable ones compared with those revealing at the room temperature, as are illustrated respectively in Figs. 2 (a), (b), and (c). Of three, the first is represented by those developing simply (grossularite) together with those appearing newly and developing gradually (anorthite and wollastonite), the second by those disappearing at ca. 650°C (chlorite) and at ca. 950°C (prehnite), and the third by one of those for prehnite or grossularite.

The spacings for the final product obtained after pulverization of the specimen reversed from heating at $1,000^\circ\text{C}$ to the room temperature are indicated in Table 3 displaying presence of anorthite and wollastonite, absence of prehnite and chlorite, and marked growth of grossularite.

Besides, noticeable is that when the final product inactive for the treatment held merely in water for 3 hours at $70^\circ\sim 90^\circ\text{C}$ is furthermore treated with 1N

HCl in the same conditions all the spacings other than those of grossularite developed far more intensely than expected for those of the impurities mingled, although even with the similar acid treatment the specimen without heat treatment reveals no variation not only in the main spacings but also in those of the impurities.

It thus results in that thermal effects on prehnite might have caused its own dissociation into anorthite, grossularite and wollastonite after the assumed reaction:

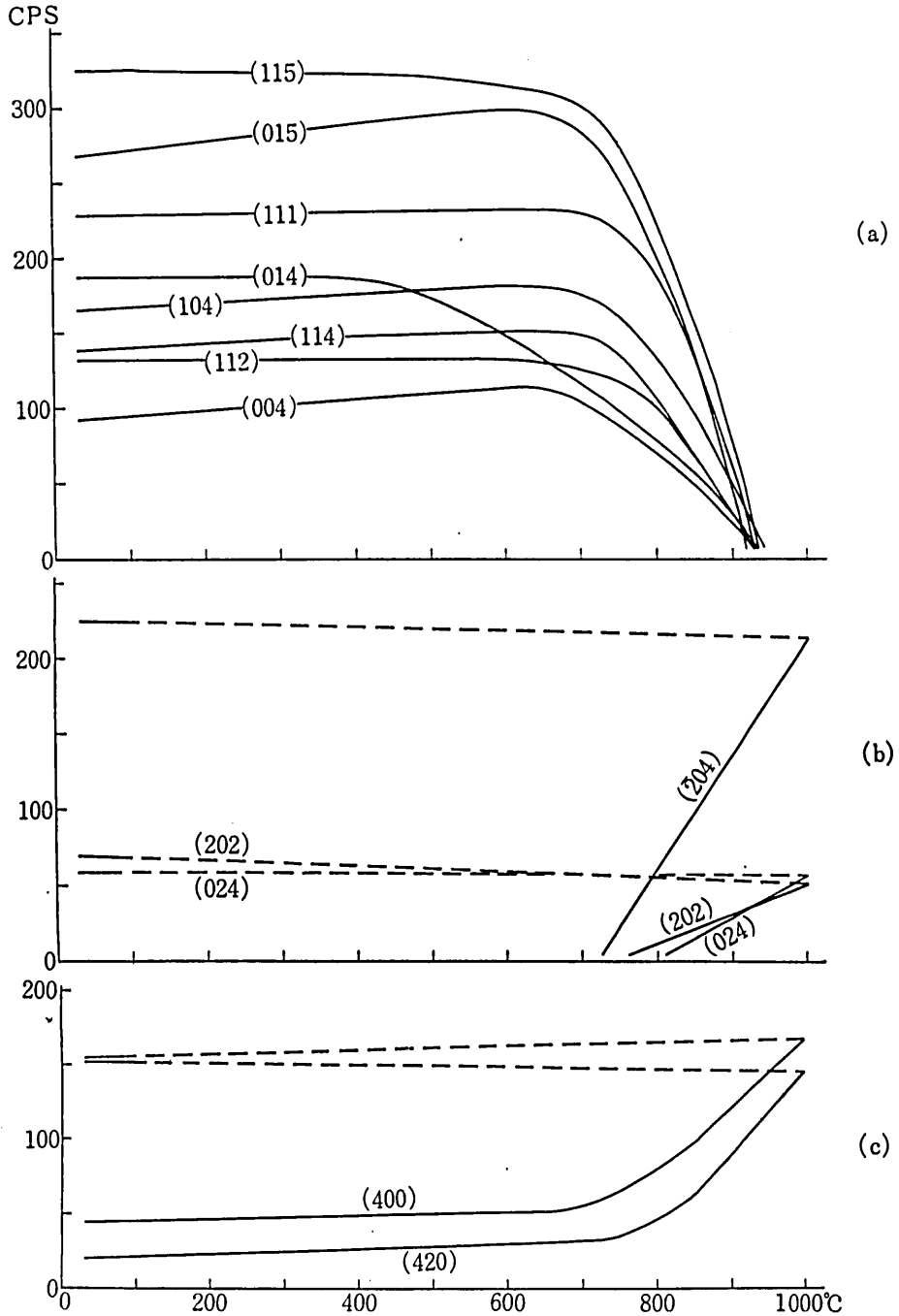


Molecules of each product are difficult to be determined quantitatively but the ratios of diffraction intensities for the respective spacings seem to reflect indirectly the pertinence of the reaction assumed to certain extent. That the spacings representing SiO_2 are absent in the diffraction data is reasonably ascribable to its amorphous state or extremely fine grain-size (cf. RUOTSALA, 1963). As was alluded to already in the other cases by UMEGAKI et al., the temperature suitable for dissociation of prehnite is also in discord with that for formation of each product for several reasons. In addition to this, the reality is that each spacing of prehnite begins to vary at the temperature, different from one another, within the range of $700^\circ \sim 750^\circ\text{C}$ and culminates in completion of its breaking-down at about 930°C , so that its dissociation is not disputable only on the basis of variation appearing in a single spacing. The spacings for anorthite recognized initially at about 750°C bear a tendency of increase in intensity up to $1,000^\circ\text{C}$. It is to be remarked on inspection of the data that $d_{(420)}$ of grossularite mingled originally in a little amount is overlapped on $d_{(002)}$, decreasing already at about 750°C , of prehnite and consequently reveals an apparently gradual growth despite of the really rapid development of newly appeared grossularite. The spacings of chlorite mixed in a small quantity obviously breaks down at about 650°C . Since those of anorthite and wollastonite, though röntgenographically identifiable to certain extent, disappear with acid treatment, their inner structures are, so far as this experiment is concerned, not considered to be perfectly stable at the temperature up to about $1,000^\circ\text{C}$. The data for thermal variation are partly exhibited in Fig. 3.

(2) Grossularite

The spaciments subjected to thermal experiments are selected out from the sample A 1 composed of large crystals of grossularite and diopside, appearing in Higashi-ichi-kō and quite similar to those provided for the manipulation of infrared absorption.

Any variations of their spacings in intensity have not been recognized during



Actual line : heating curve and broken line : cooling curve showing irreversibility.

(a) : Prehnite, (b) : Anorthite, (c) : Grossularite

FIG. 3. Thermal variation in intensity

and after heat treatment in such conditions as were applied to in the case of prehnite. Thermal expansion estimated from variation of a_0 for these specimens, however, amounts to 7.63×10^{-6} in linear coefficient at the temperature from 30° to 900°C , being smaller than 9.21×10^{-6} given by Skinner for synthetic garnet at the temperature up to 850°C .

As for hydrogrossular, endotherm at $650^\circ\sim 690^\circ\text{C}$ together with exotherms at 870°C and 940°C on the DTA curve have been shown by BELYANKIN and PETROV (1941) and the former has been combined with anhydration of hydrous material by SMITH (1952) on the basis of the data concerning decrepitation but with destruction or anhydration of hibschite (hydrous grossular) by Skinner, while any remarkable changes have not been observed in the DTA data given by Frankel (1959) at the temperature lower than $1,100^\circ\text{C}$ suitable for the beginning of decomposition of hydrogrossular. On the other hand, the disappearance of optic anomaly in anhydrous Ca-bearing garnet due to heating at $1,225^\circ\text{C}$ and the fact pointing to no variations other than the mere expansion in the synthetic garnet at the temperature up to 850°C have been brought to light by KÖZU, MACHIBA, TAKEDA and OMORI (1940) and by SKINNER respectively.

Considering all of the thermal data obtained from the present experiments and the previous works mentioned above, it is properly concluded that the presence of hydrogrossular in the specimens concerned is not affirmed and the development of grossularite structure with heating of prehnite cannot help ascribing to the new one derived from dissociation of the latter because of the invariability of the former in its single state.

IV. CONCLUSION

Each vein-like body intercalated within serpentinite in the Takase mine is not regarded as such a simple one as was previously dealt with as a single mass of so-called rodingite, although there still remains a question of whether the former may, just as it is, be correlative with the latter or not.

Noticeable is that two main stages, designated I and II, of mineralogenesis accompanying some other weak ones are found overlapped with one another, causing a confusion resulted from difficulty of discrimination and boundary phenomena within the related mass coming into contact with the country rock, are surely recognizable as if the constituent minerals were seemingly in a sort of zonal arrangement.

The proximity of the mass in question to the chromite deposit seems to display the disposition of the structurally important spaces near or surrounding those suitable for ore formation. There is however no materials proving this directly since any traces of chromite or other ore minerals, for instance, have been discerned nowhere within the mass.

The fact that Ca-rich minerals such as prehnite, grossularite, diopside and so

on occur not as the impregnations but as the aggregates or veinlets in the well-defined vein-like mass demarcated solitarily with Ca-poor serpentinite may be considerably helpful for interpreting the connection of the formers with their country rock and accordingly for ore deposition.

Genesis of grossularite through thermal dissociation of prehnite is believed to justify their intimate coexistence in Nature and to be suggestive for the replaceability of the latter with the former on the spot. This is however not to be tangled with the contrary case replacing the former with the latter formed in the later different mineralogical process, as are revealed commonly in most of the present specimens.

Presence of the primarily hydrous or secondarily hydrated minerals as well as their chemical indifference with the country rock are also considered very significant in relation to genesis of the main mass. Hence, pursuit of the related minerals has been particularly in view, resulting in that hydrogrossularite and another needle-shaped mineral remain yet to be discriminated.

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EXPLANATION FOR PLATE X.

Fig. 1. Sample B 3'

- l: Diopside (diopside II).
- m: Chlorite, forsterite and a little amount of serpentine.
- n: Chlorite, diopside (diopside II) and a small quantity of grossularite.
- o: Diopside (diopside I) and chlorite.
- p: Prehnite and chlorite.
- Ca: Calcite and grossularite other than grossularite I.

Fig. 2. Sample B 4'

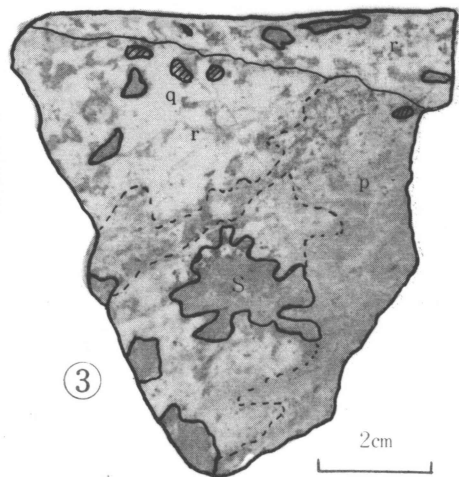
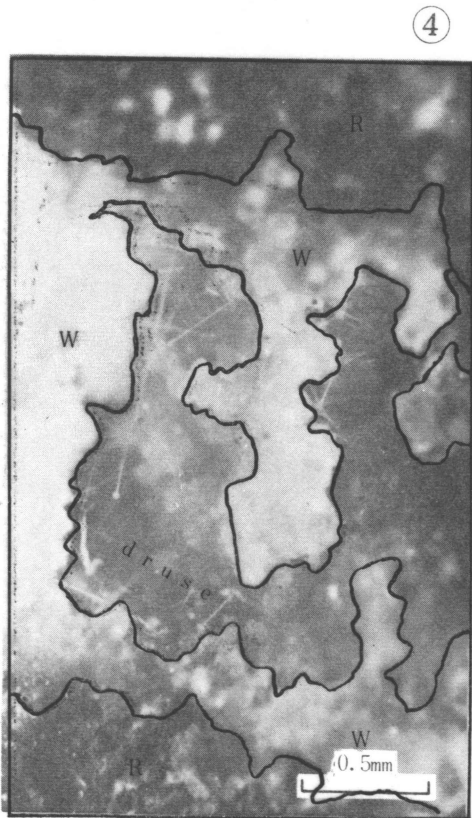
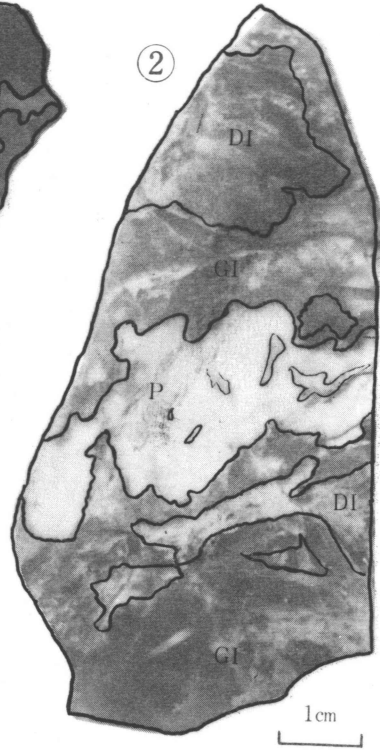
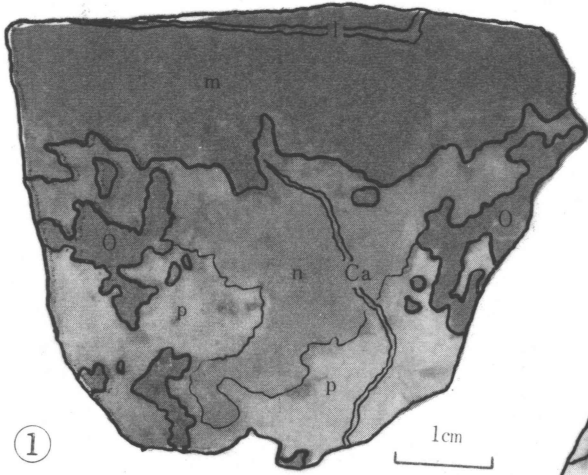
- DI: Diopside (diopside I) and chlorite.
- GI: Grossularite (grossularite I).

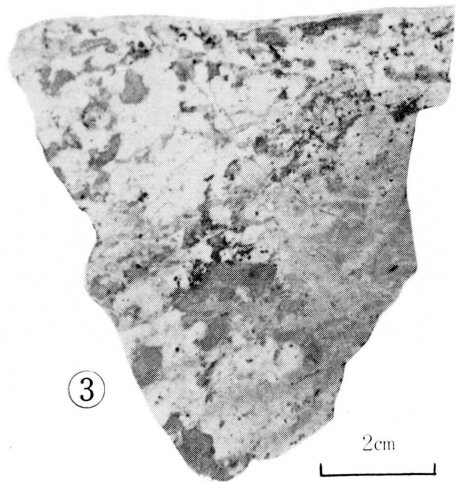
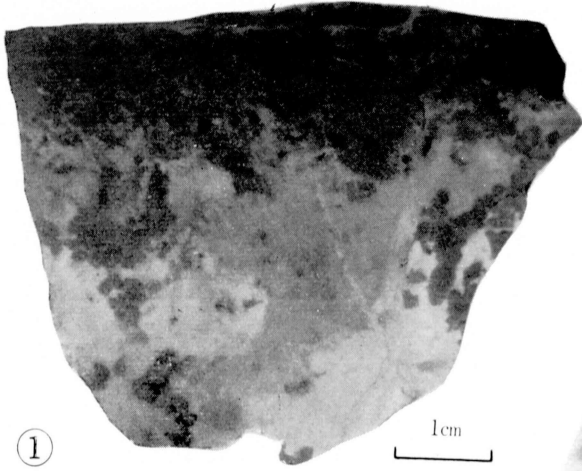
Fig. 3. Sample B 1.

- p: Diopside (diopside I and diopside II ?) and chlorite.
- q: Chlorite.
- r: Prehnite, chlorite and diopside (diopside II).
- s: Diopside (diopside I) and grossularite (grossularite I).

Fig. 4. Sample B 6 and the druse in grossularite.

- R: Deep-reddish grossularite (grossularite I).
 - W: Transparent, colorless (seemingly white-colored) grossularite.
- Acicular crystals are also recognized in the druse.





EXPLANATION FOR PLATE XI.

(All figures x 30)

- Fig. 1. Sample A 1. Flesh-colored grossularite is penetrated by reddish grossularite replaced by the veinlets of prehnite (lower nicol only)
G I: Flesh-colored grossularite (grossularite I)
G II: Red-colored grossularite (grossularite II)
P: Prehnite and chlorite
- Fig. 2. Sample A 2. Diopside is again replaced by grossularite replaced earlier by prehnite (crossed nicols)
D I: Black-colored diopside (diopside I) accompanied with formation of chlorite
G I and P: Same as in the case of Fig. 1.
- Fig. 3. Sample B 3'. Diopside is replaced by forsterite, all being furthermore replaced by chlorite (crossed nicols)
D I: Diopside (diopside I) and chlorite
F: Forsterite
C: Chlorite replacing forsterite
- Fig. 4. Sample B 1. Diopside is traversed by prehnite and chlorite, all being again cut by the veinlet of grossularite-calcite (crossed nicols)
D II: Diopside (diopside II)
P: Prehnite and chlorite
G III: Grossularite (later in stage than grossularite II)
Ca: Calcite

