

Studies of pegmatitic cassiterites from the Gunung Jerai (Kedah), Bakri (Johore) and Kathu Valley (Phuket) regions.

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Abstract: Pegmatitic cassiterites are significantly different from cassiterite of other genetic regimes in many of their physical, geochemical and crystallographic characteristics. Such cassiterites from the 3 areas were studied and compared with one another as well as collectively compared with other types of cassiterites present in the Peninsular Malaysia and Thailand Tin Belts. Of particular significance is the discovery that biaxial positive cassiterites are abundant among the pegmatitic cassiterites studied and their distribution appears to be more widespread than previously known within the Tin Belt. Such cassiterites do not appear to be confined solely to the pegmatitic suite though they are more likely to be so. Flattened prismatic rhombic pegmatite cassiterites are also obtained from the lepidolite pegmatites in Phuket at the Ban Guan Tin Mine. Such cassiterites appear to have an orthorhombic rather than a tetragonal symmetry.

Phuket, Gunung Jerai and Bakri are tin mining areas geographically confined to the western tin belt of Peninsular Thailand and Malaysia (Fig. 1).

These 3 areas are main areas where pegmatitic cassiterite are known or are believed (not without reasons) to exist. Pegmatite occurrences characterise the Gunung Jerai, Bakri and Phuket areas but their abundance, nature and relationships with the host rocks differ from area to area.

Simple pegmatites in the Gunung Jerai area are associated genetically and spatially with the 2-mica adamellite of the Jerai pluton of Carbo-Permian age which intrudes into metasediments of the Jerai Formation of Cambro-Ordovician age. In the Bakri area, early Jurassic simple pegmatites intrude into an early Triassic biotite adamellite, though there are no conclusive evidence to suggest that they are comagmatic. These intrusives invade the Jelai Formation of Middle Triassic age. Pegmatites in Kathu, Phuket are the youngest in the 3 areas and are probably comagmatic with early Tertiary 2-mica granites in the area. These granites are probably different from the biotite granite of the Cretaceous "Phuket-Tennasserim Belt Granites". They intrude into the sediments of the Phuket Group which is locally Silurian or younger. Large unzoned massive lepidolite pegmatites are present in the Kathu Valley and such pegmatites are very rare worldwide. The degree of specialization of pegmatites from the 3 areas are indicated by the extensive enrichment of Rb, which resulted in very low K/Rb and high Rb/Sr ratios.

Basically, the mineral assemblage of each of these areas is believed to have a similar character in that they are of pegmatitic origin. Tin mineralization in the 3 areas and also that of columbite-tantalite in the Bakri and Gunung Jerai areas are attributed to the pegmatites. There is no doubt that the deposits in the Kathu Valley are directly related to the pegmatites in the area. However, no *in situ* cassiterite and columbite-

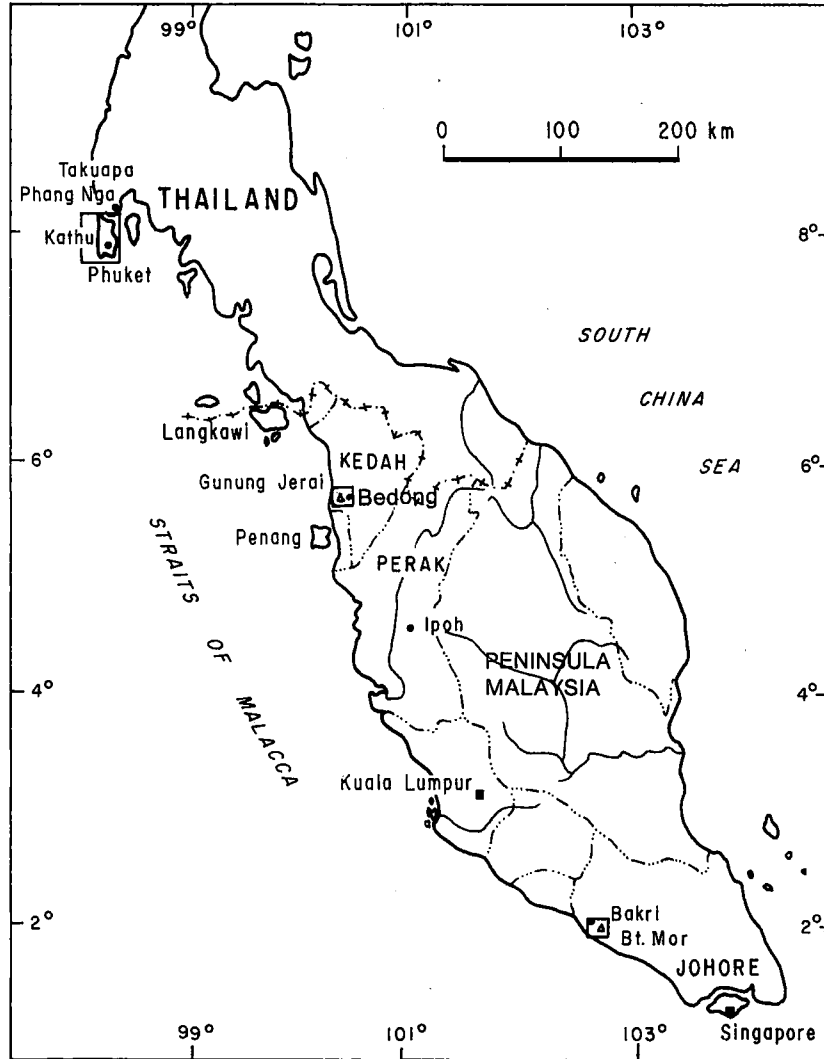


Fig. 1. Location map of the Kathu, Gunung Jerai and Bakri areas.

tantalite have been found in pegmatites of the Bakri and Gunung Jerai area. So the pegmatitic origin is only an assumption based on their close proximity to pegmatite occurrences, the mineral assemblage and general geochemical characteristics of the minerals present.

Cassiterite is the major component common to these ores. In addition, wolframite, which is associated with quartz veins cutting the pegmatites, monazite and some columbite-tantalite constitute the economic assemblage in the Kathu district of Phuket. Wolframite is effectively absent in both the Gunung Jerai and Bakri areas

which have very significant quantities of columbite-tantalite and subordinate monazite and ilmenite. Primary iron ore deposits are also present in the Gunung Jerai area.

Besides these ore minerals, many accessory pegmatitic minerals associated with some of these areas are garnets, molybdenite, mica, struverite, ilmenorutile, euxinite, pyrochlore-microlite, betafite and fergusonite among others (Bradford, 1970; Rishworth, 1960; Garson, *et al.*, 1970).

Though these pegmatitic cassiterites are being mined economically, their contribution is rather insignificant and is relatively unimportant compared with the other types of cassiterites being mined in the Western Tin Belt of the Malay Peninsula which are mainly contact metasomatic or hydrothermal in origin. In Phuket, however, pegmatitic cassiterite forms a major portion of the cassiterites mined.

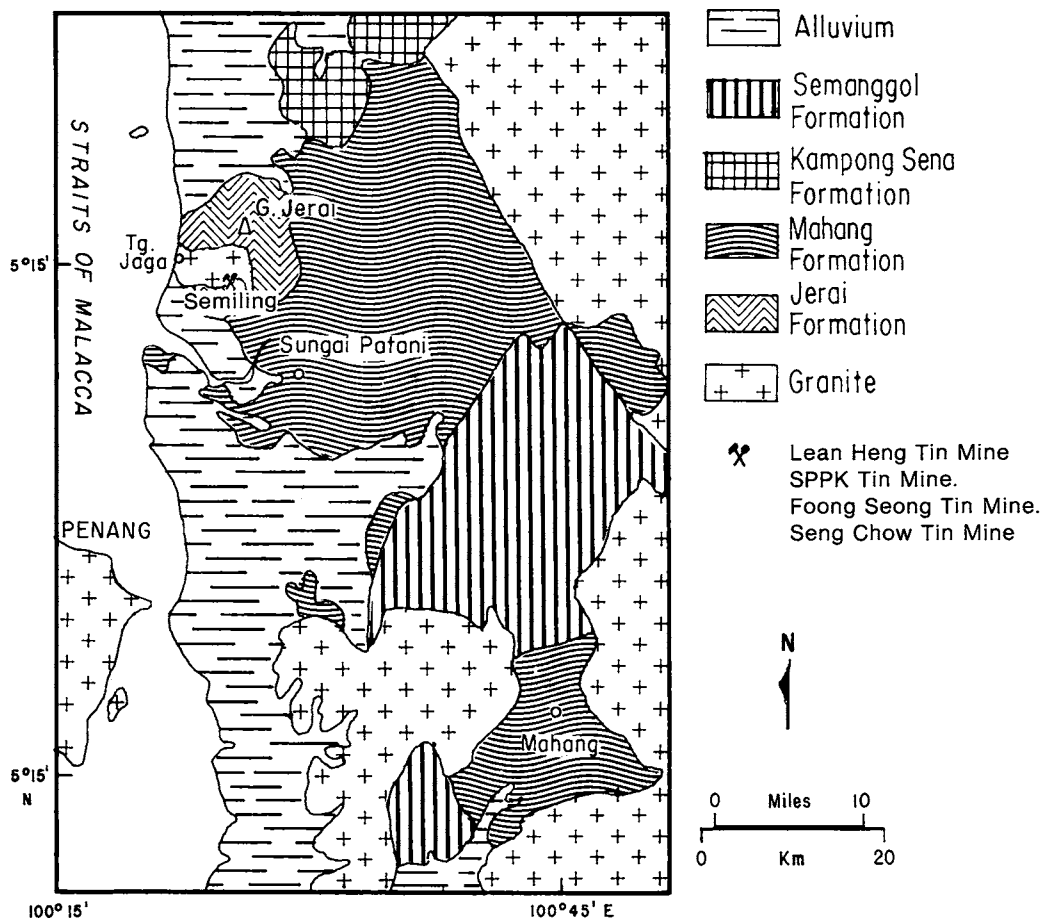


Fig. 2. Geological sketch map of the Gunung Jerai area (after Geological Survey Malaysia's Geological Map of West Malaysia, 1973).

MODE OF OCCURRENCE

No tin sulphides are found associated with cassiterite in these 3 areas. In fact, associated poly-sulphide mineralization and minerals have practically never been found in these areas except for minor and probably genetically unrelated development of such polysulphides in the Gunung Jerai area. Only 2 occurrences were noted, one in the Gurun quarry on the eastern flank of the Jerai massif and the other in a skarn-type limestone on the floor of Lean Heng Tin Mine at Tupah located between Merbok and Semiling (Fig. 2).

Cassiterite occurs *in situ* in the large massive lepidolite pegmatites of the Kathu Valley in Phuket where they are recovered economically directly from these pegmatites (Fig. 3). Probably genetically related to these cassiterite-bearing pegmatites are similar pegmatites in the Phangnga region of Peninsular Thailand (Garson, *et al.*, 1970).

No positively identified *in situ* cassiterite has been found in the pegmatites of the Jerai area. Cassiterite is only recovered from placers. These placers are in fact spatially closely related to the pegmatites and in many cases overlie directly on top of pegmatite occurrences. They vary from alluvial to probably eluvial in origin.

However Bradford (1970) had reported the presence of small amounts of cassiterite in many instances from various rock types in the Jerai area. The author

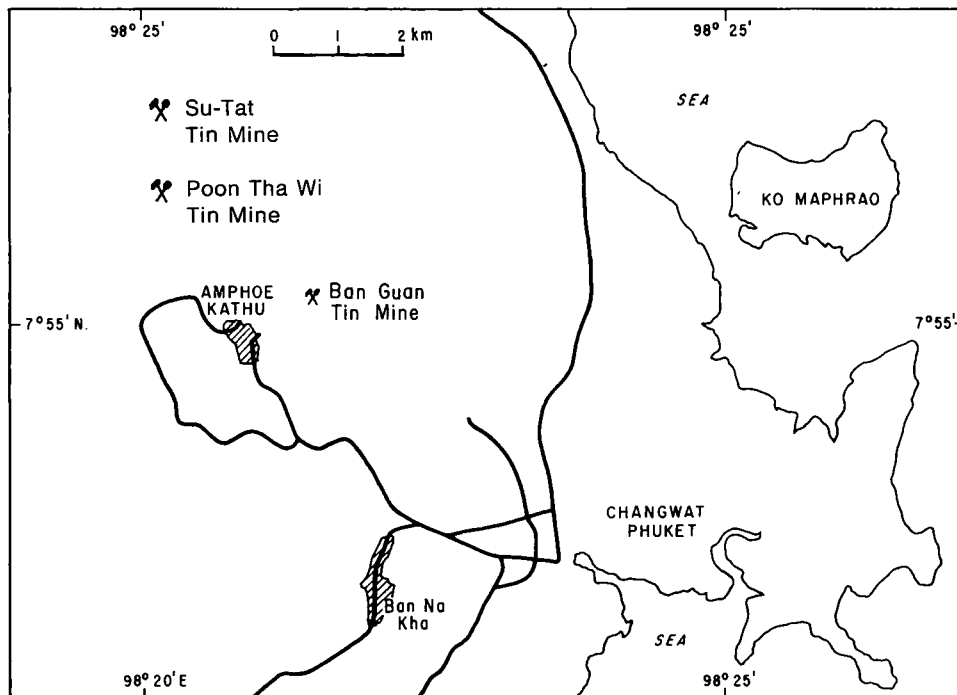


Fig. 3. Location map of the Kathu area, Phuket.

similarly noticed many small minute rhombic shaped crystals in thin sections of many of the country rocks in contact with pegmatites but there is no evidence to show that they are indeed cassiterite. They may very probably be sphene due to reasons given below:

- (i) Rhombic shaped grains of high relief, birefringence and dispersion suggestive of sphene.
- (ii) They are more often observed only in the metasediments especially those in contact with the pegmatites while their presence are not detected within the pegmatites.
- (iii) Chemical analysis of pegmatite and host rock samples both indicate very low

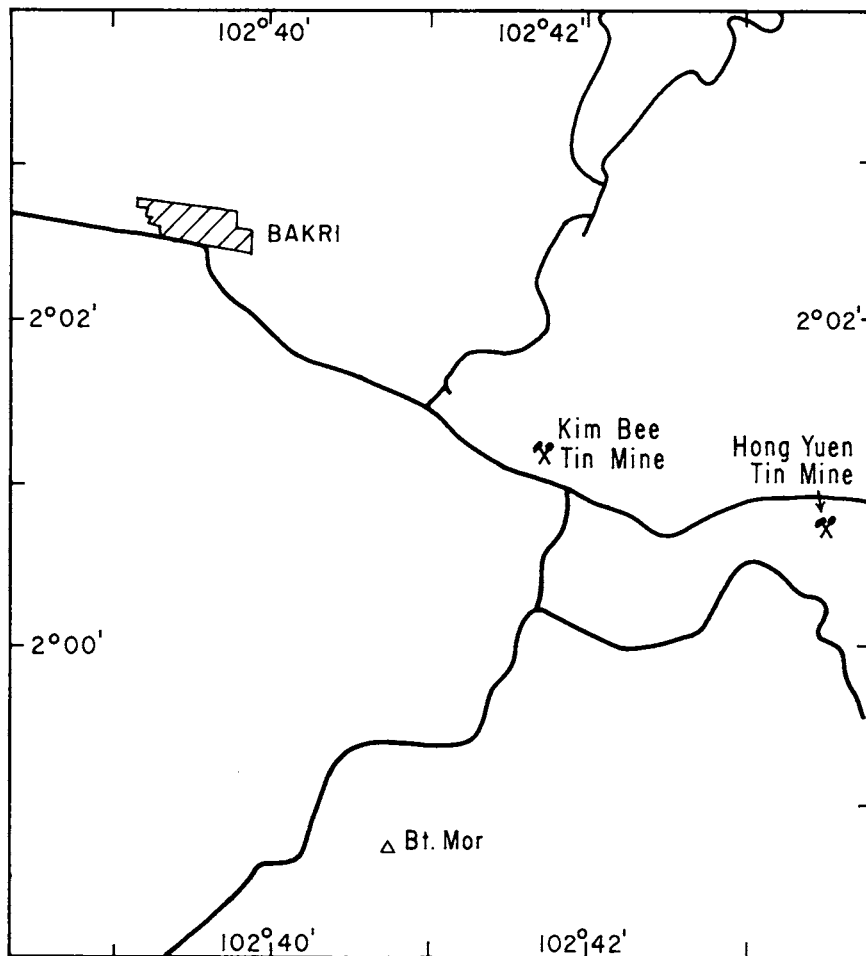


Fig. 4. Location map of the Bakri area, Johore.

concentrations of tin. Such low concentrations probably do not warrant any reason to suspect the presence of relatively large numbers of small discrete cassiterite grains in these rocks.

The placer tin deposits of Bakri are similarly alluvial in origin and are deposited and concentrated in an inland fresh water peat swamp which is of fairly recent origin. It is surrounded by rolling hills of which Bt. Mor is the most conspicuous (Fig. 4).

HANDSPECIMEN FEATURES

Colour and size

Colour of cassiterites from these areas is predominantly greyish to jet black. Various shades of other colours common to cassiterite are also represented, ranging from white, yellow, cream yellow, light to dark red brown. Very rare varieties of colourless to pale brownish transparent vitreous cassiterite with conchoidal fractures are also found associated directly with the jet black cassiterites, though such occurrence is essentially confined to the Syarikat Perlombongan Perbadanan Kedah Tin Mine near Semiling at the SE foothills of the Jerai Massif. It was observed that these transparent parts gradually grade into the opaque black portions. Such transparent varieties are not found in the Bakri and Kathu district. Greyish black to jet black cassiterites are consistently opaque and have a submetallic to adamantine or



Plate 1a. Close up view of cassiterite twinnings and zoning of a pure cassiterite lode. Cyclic twins and zoning are present. This cassiterite lode is hydrothermal in origin. Size of coin is about 2 cm.
Loc: From a mine in Suratani Province, Thailand.



Plate 1b. Mottled nature of the same lode which is characteristic of many hydrothermal lodes. Colours range from pale yellow, yellow brown to dark brown and black. Colour of pegmatitic cassiterite is usually not so variegated and is quite typically black.
 Loc: From a mine in Suratani Province, Thailand.

vitreous lustre with subconchoidal fractures. Paler coloured varieties are often adamantine and translucent with irregular fractures (Plates 1a & 1b).

Cassiterites from the Gunung Jerai area have the greatest spread of colours. The brown coloured varieties often contain subregular zones of black cassiterite, though patchy colour zoning is more typical in the majority of such grains. In contrast, cassiterite from Bakri unfailingly is jet black to greyish black. Intermediate between these two are the cassiterites of the Kathu Valley which contain occasional thin light coloured zones. Separate individual light coloured crystals are absent.

The coarsest cassiterite grains are those occurring in the lepidolite pegmatites of the Kathu district where individual crystals may grow to as much as 3 to 4 cm long, though more usual are those in the 1 to 2 cm range. However, the whole range of sizes below 1 cm are represented. Occasional large blocks of cassiterite weighing up to 15 kilograms are present.

Grains of cassiterite are seldom greater than 1 or 2 cm in the Gunung Jerai area, the mode being in the 2 to 5 mm range.

Representing the finest ore in the 3 areas are the cassiterite grains from Bakri.

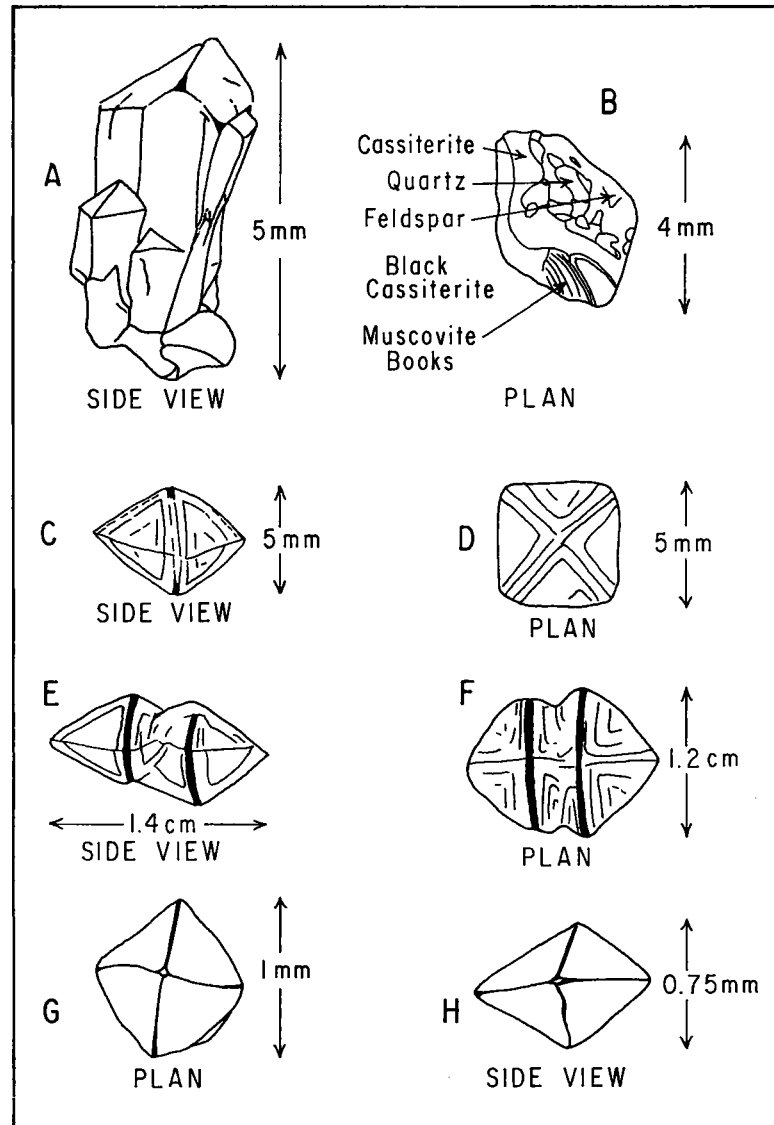


Fig. 5. Common cassiterite forms and habits obtained from the mines in Bakri and Gunung Jerai area.

A: Short to moderately long prismatic cassiterite with low pyramids (Specimen: SKFS, dark brown-black cassiterite, Gunung Jerai).

B: Massive cassiterite with associated minerals (Specimen: SKFS, black cassiterite, Gunung Jerai).

C: Bipyramidal cassiterite with twinned surfaces (Specimen: SKLK, black cassiterite, Gunung Jerai).

D: Plan view of C.

E: Interpenetrating bipyramidal cassiterite twins (Specimen: SKLK, black cassiterite, Gunung Jerai).

F: Plan view of E.

G: Squat, bipyramidal, black, vitreous cassiterite with low pyramidal terminations (Specimen: SJHY, Bakri).

H: Side view of G.

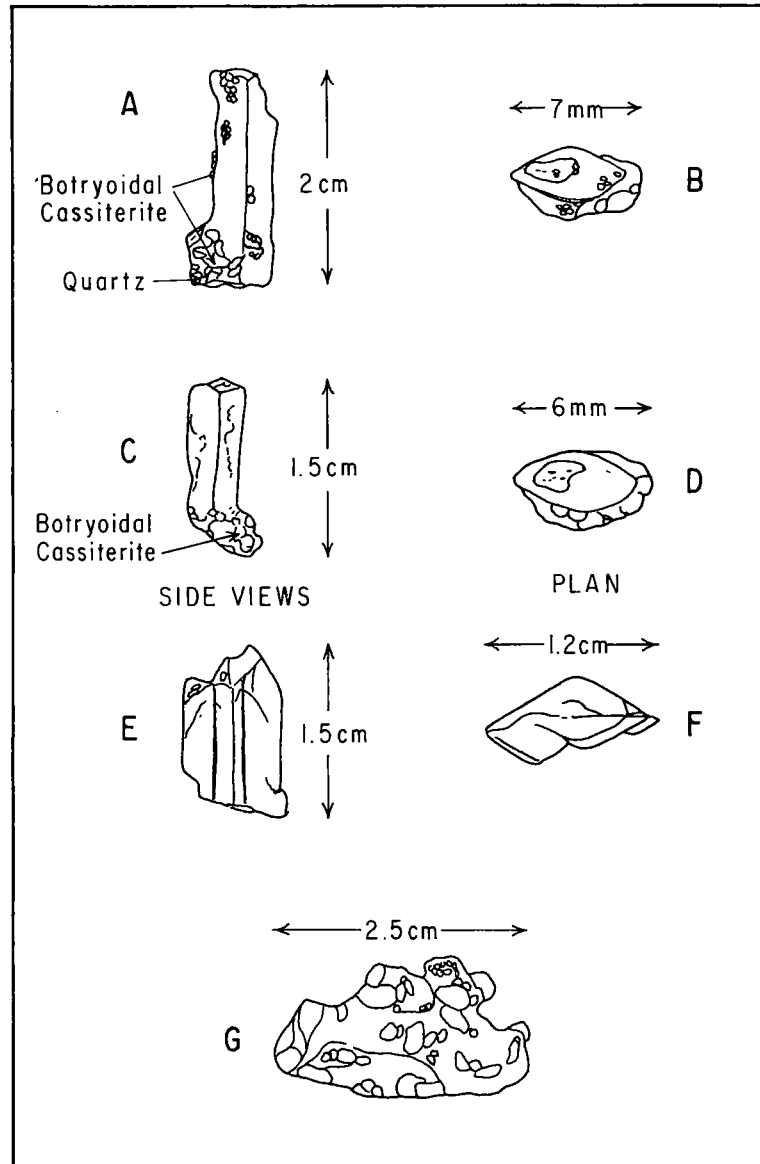


Fig. 6. Common cassiterite forms and habits obtained from lepidolite pegmatite in the Ban Guan Tin Mines, Phuket.

A & C: Extraordinary black cassiterites with deformed sphenic shape associated with botryoidal cassiterite and manganese oxides.

B: Rhombic cross-section of A.

D: Rhombic cross-section of C.

E: Penetration twins of rhombic shaped cassiterite.

F: Rhombic cross-sections of twinned crystals.

G: Black massive botryoidal cassiterite mixed with quartz.

Usual size of grains are the 1 to 2 mm fraction and sizeable amounts are much finer. Grains coarser than 3 mm are rare.

Form and habit

Except for the *in situ* cassiterites of the lepidolite pegmatites, cassiterites of the other 2 areas have been transported and are water-worn to varying extents. Significantly affected are the cassiterites from Bakri where sometimes it is difficult to see the crystal form and thus more often their habits can only be deduced. Though the Gunung Jerai cassiterites are often broken up or abraded, the coarser grain sizes enable some of the crystals forms and habits to be more easily identified (Figs 5 & 6).

Bakri

By a large extent, crystal forms of cassiterites from Bakri are bipyramidal or squat prismatic with rather flattened obtuse terminations. Prismatic forms are practically absent. The squat prismatic forms are often also doubly terminated. These crystals probably occur discretely or in small groups as no massive forms were observed. Another feature of these crystals is that they exist as simple crystals lacking twinning.

Gunung Jerai

A larger variety of crystal forms occur in the Gunung Jerai area where there are a

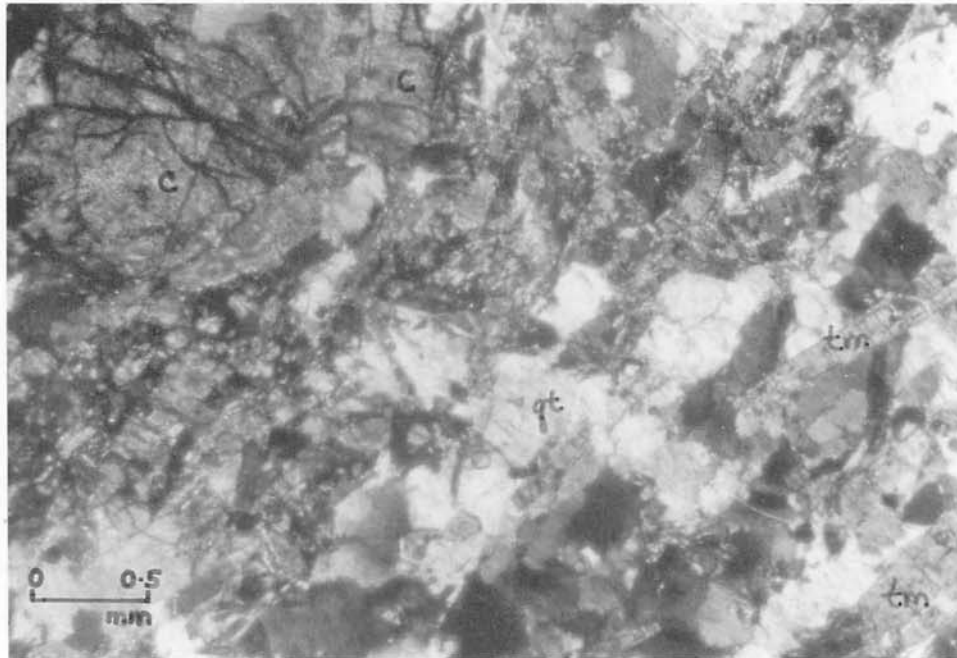


Plate 1c. Photomicrograph showing association of cassiterite (c) with gangue minerals which are mainly quartz (qt) and fine needle tourmaline (tm). The cassiterite grains are subhedral (x polars).
Sample: SKFS 1557816.
Loc: Foong Seong No. 2 Tin Mine, Kedah.

mixture of bipyramidal, squat prismatic and prismatic crystals. Some of the prismatic crystals, in particular, do not seem to be doubly terminated. Crystallographically, these cassiterite crystals are more complex with a large number of twins. Twins range from cyclic, geniculate to interpenetrating and may occur in any of the crystal forms. Massive forms appear to be present and these are usually the light brown variety. Cassiterites of this area appear to develop in crystal aggregates with interpenetrating crystals, although discrete crystals and massive forms are equally significant. Commonly found associated with the cassiterite grains are feldspars, muscovite, tourmaline and quartz (Plate 1c).

Phuket

Cassiterite forms found in the lepidolite pegmatites of Phuket are quite distinct from the other 2 areas. Some of these forms are rarely encountered elsewhere and are unique. They seem to be restricted to these lepidolite pegmatites.

The most interesting are the presence of cassiterite forms which have rhombic cross-sections. They are prismatic but appear to be flattened parallel to their longest dimension or they have been described by Dr. P. Aranyakanon (pers. comm.) as having a 'distorted sphenic shape' (Fig. 6; Plates 2a, 2b). Besides this form, another form of interest is the development of massive botryoidal cassiterite. They are massive and the

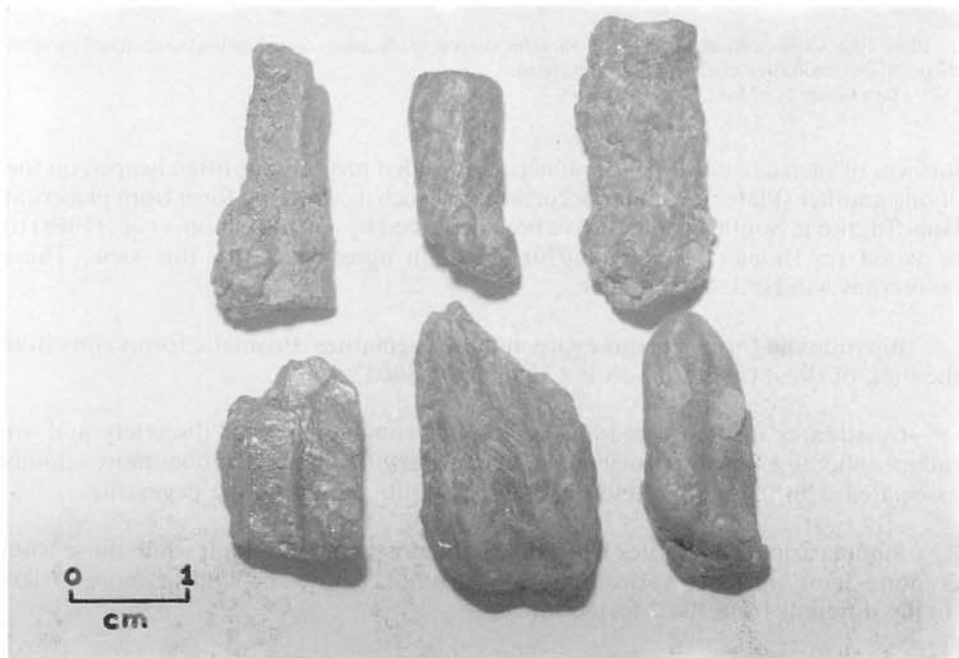


Plate 2a. Specimens of the unique 'rhombic shaped' black cassiterite from the large unzoned lepidolite pegmatites of the Ban Guan Tin Mine. Upper row shows the long flattened single crystals. Lower row shows penetrating twins of such cassiterites. Note the development of botryoidal forms as well as manganese oxides on the walls of such cassiterites.

Loc: Amphone Kathu, Phuket.

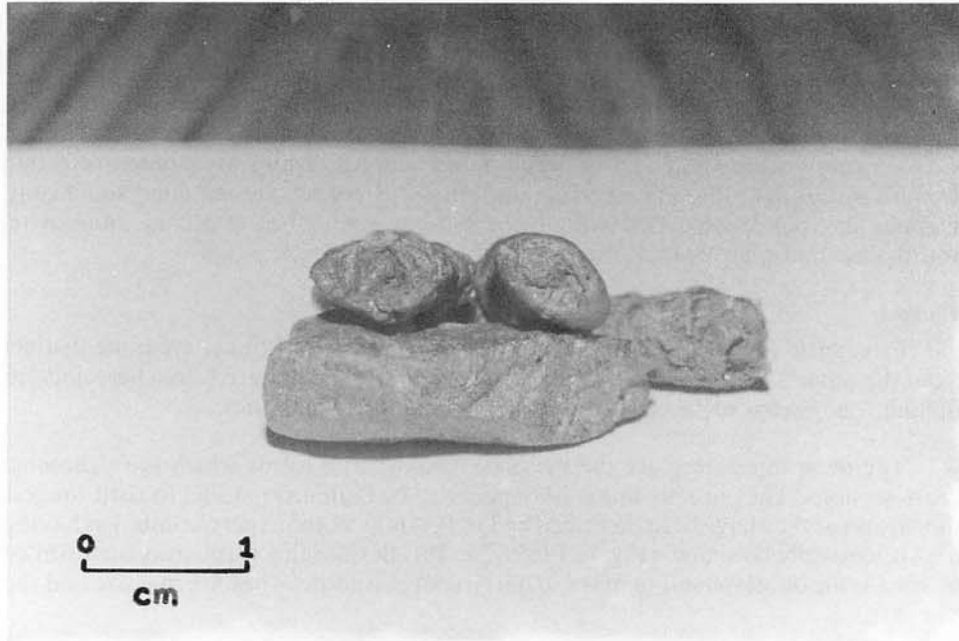


Plate 2b. Cross-sectional view of the 'rhombic shaped' black cassiterites showing the flattened rhombic shape. Such cassiterites often are not terminated.
Loc: Ban Guan Tin Mine, Kathu, Phuket.

surfaces of such cassiterites have numerous rounded projections, often heaped on top of one another (Plate 3). Similar occurrences of such a cassiterite form from placers at Huai Tagrao in South Thailand have been described by Aranyakanon, *et al.*, (1968) to be wood tin though Hosking (1970) is not in agreement with this view. These cassiterites will be described later.

Bipyramidal forms are quite rare in these pegmatites. Prismatic forms constitute the bulk of these cassiterites. Some may be twinned.

Cassiterites in these lepidolite pegmatites commonly occur discretely and are rather inhomogenously distributed. They generally appear to be more closely associated with the quartz, feldspars and lepidolite present in the pegmatite.

Summarizing, cassiterites from Bakri are mostly bipyramidal, while those from Gunung Jerai are more varied. Those from Phuket however, appear more or less totally different from the 2 former areas.

PETROGRAPHICAL PECULARITIES

Introduction

Mineralogical studies of cassiterites using the petrographic microscope have often been neglected in preference to their study by the reflecting ore microscope. As

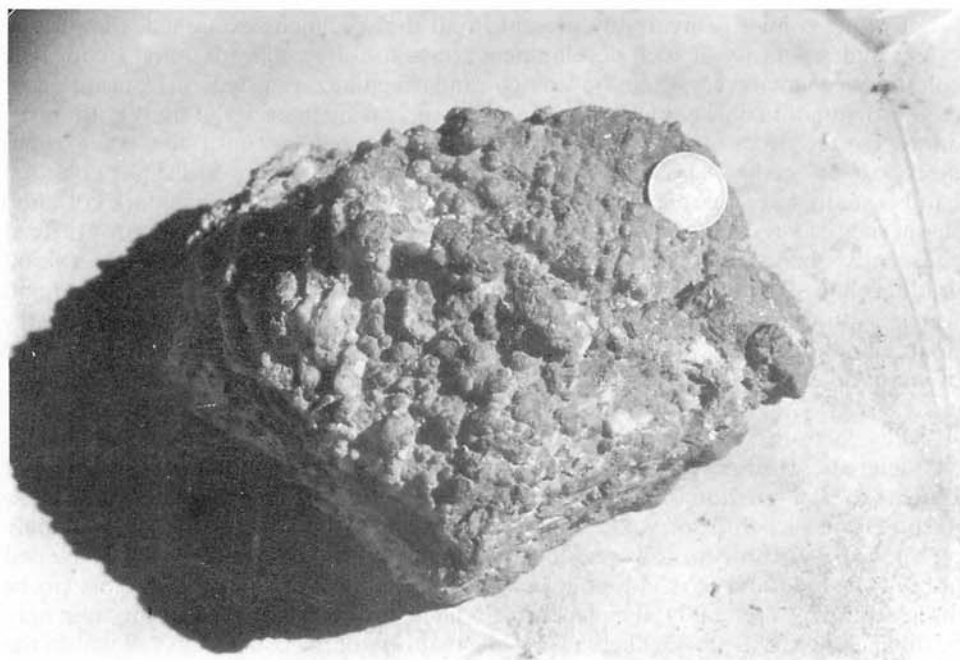


Plate 3. Massive botryoidal black cassiterite associated with some quartz and feldspar. This specimen weights about 12 kgs. and was found within the large unzoned lepidolite pegmatitic in the Ban Guan Tin Mine.

Loc: Amphoe Kathu, Phuket.

cassiterite is an optically transparent mineral it can also be studied fruitfully under a petrographic microscope. It holds many advantages over the ore microscopy studies especially in the study of optical properties, which potentially can help throw light on such perplexing problems as their pleochroism, colour zoning and crystallography.

For these studies, polished thin sections were made of cassiterite samples from the 3 areas. Polished thin sections were preferred over thin sections because these polished thin sections can also be used for ore microscopic as well as electron microprobe studies. In this way the same grain can be studied by the 3 methods.

Microscopic descriptions

Cassiterites from Gunung Jerai

As mentioned earlier, the colour variation of cassiterites from this area is large. This is similarly reflected in the colour of these cassiterites in thin section under plane light. Frequently they show strong body colours with the whole range from very pale to very dark represented. Cassiterites from the Lean Heng and Seng Chow Tin Mines have the strongest body colours while those from the Syarikat Perlombongan Perbadanan Kedah and Foong Seong are paler. Body colour very frequently is directly related to the intensity of pleochroism and the extent of colour zoning.

Colour zoning is invariably present in all the specimens examined, though the extent and regularity of such development seems to follow a gradational trend. Pale coloured specimens very often show patchy and irregular zoning with discontinuous or criss-crossing thin darker blebs or zones. With increasing intensity of body colour, the darker zones necessarily become wider and more regular until the paler zones disappear altogether or occupy only very thin strips. The dark and light coloured bands usually have sharp contacts with each other. Colour banding in dark coloured cassiterites takes another form in that the pale zones may be absent. Instead consecutive bands of generally dark cassiterite are present. These bands exhibit gradual changes in colour intensity until they are sharply terminated by the next band which is quite characteristic of growth zones. Zones are more often planar than wavy or arcuate though these have also been observed. The majority of zones are in parallel or subparallel sets.

Pleochroism is prominently and distinctly expressed with a colour scheme which is characteristic. It shows marked differences in intensity between the various zones and appears to be a function of the body colour. Pale coloured zones normally show weak to moderate pleochroism while darker zones tend to be strongly or very strongly pleochroic. Pleochroism, however, may be completely absent or very weakly developed in both the light and dark colour zones. This probably is due to other factors (to be discussed later). Generally, the pleochroic scheme for the cassiterites of this area may be divided into 3 categories (Table 1.1). The majority of the cassiterites fall within the moderate pleochroism category. Strong pleochroism is shown by a large proportion of the cassiterites from the Lean Heng and Seng Chow Tin mines near Semling. It is noted that the dominant colour in all the 3 categories is brown-red which may occur in any intensity.

Extinction of the cassiterite is straight and fractures are rather uncommon. Faint discontinuous (110) cleavages are present but are usually observable only under high magnifications. Occasionally, 3 sets of cleavages are observed in basal sections, with one set occurring parallel to the extinction position, while the other 2 perpendicular sets make angles of 45° to 46.5° with the former set. These correspond to the (100) and

TABLE 1.1
PLEOCHROISM OF CASSITERITE FROM GUNUNG JERAI AREA
(NB: THE CASSITERITES ARE BIAXIAL)

Pleochroism	Minimum absorption, β to α	Maximum absorption, γ to β
Weak	Usually very pale brown or occasionally colourless	Light brown or light reddish brown or colourless
Moderate	Light brown red or light dark orange brown	Dark brownish red or mallowed honey orange
Strong	Light brown orange (diluted blood red) or dark honey orange	Deep dark blood red or reddish grey or dark orange red

(011) cleavage sets. Weak partings are sometimes present and usually are more continuous and regular than the cleavage traces. Dispersion of the cassiterite is found to be high, but more detail studies reveal that the dispersion is highest for pale or light coloured cassiterite whereas the darker coloured parts exhibit generally weaker dispersion. This difference may be due to masking effects of body colour and is quite marked.

Besides the aforementioned features, twinning is another feature which is quite prominent in these cassiterites. Both simple and polysynthetic twin patterns are present. The simple twins are often cyclically repeated. Twin planes sometimes are parallel to crystal outline but generally make a small angle with the extinction position. The pale coloured parts may develop both the lamella and simple twins whereas the darker coloured parts have the tendency to develop only simple twins. Lamella twins are frequently observed to pinch out when they pass from the pale coloured zones into the dark zones. Generally, twinning is better developed in the paler zones and as much as 4 sets of closely packed lamella twins have been observed (Plate 4).

The interlocking grain boundaries of some of these cassiterites are supported by the observation that many different crystal grains are often present, growing and penetrating into another crystal.

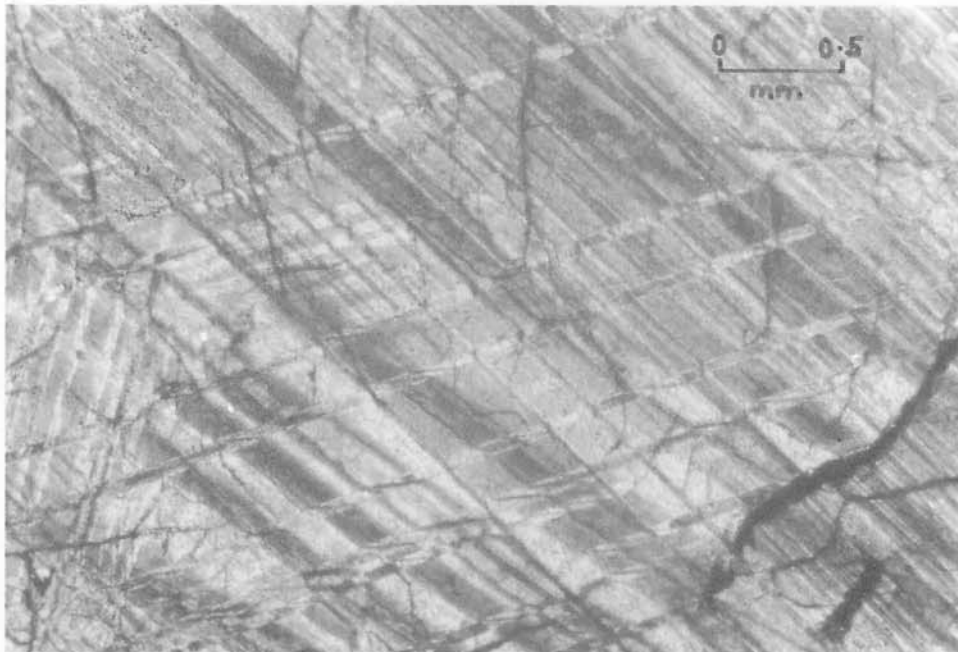


Plate 4. Photomicrograph of intense lamellar twinning in a light yellowish brown cassiterite associated with black cassiterites from the Syarikat Perlombongan Perbadanan Kedah Tin Mine. As much as 4 or more sets of such twins are present. Width of twin zones are variable but are generally thin (X-polars).
Sample: SKLK 74781.
Loc: Semiling, Kedah.

A peculiar and interesting property of many of these cassiterites is that they possess 'anomalous' interference figures. They show distinctive biaxial positive figures with $2V\gamma$ angles ranging from 1 or 2° to as much as 12 or more degrees. True uniaxial positive cassiterite are also present. There appears to be a direct relationship between the biaxiality of these cassiterites and the intensity of body colour and pleochroism. Strongly pleochroic forms with strong body colour are usually the most biaxial of all the cassiterites (to be discussed later).

Inclusions are abundant and these appear to be rather typical of cassiterites from the area. Fluid inclusions are quite plentiful and they often are aligned parallel to the cleavages and sometimes to grain boundaries. The inclusions are mainly elongated or ovoid. In addition, opaque and dust-like particules, some of which may be columbite-tantalite, are invariably present. Rutile, quartz and slightly embayed tourmaline have also been found as inclusions (Plate 5). The light brownish zones of the cassiterites are generally much clearer than the dark coloured zones.

Cassiterites from Bakri

Optically there are little significant differences between the cassiterite of this area from the former. Body colours can be very strong and pleochroism is marked. The

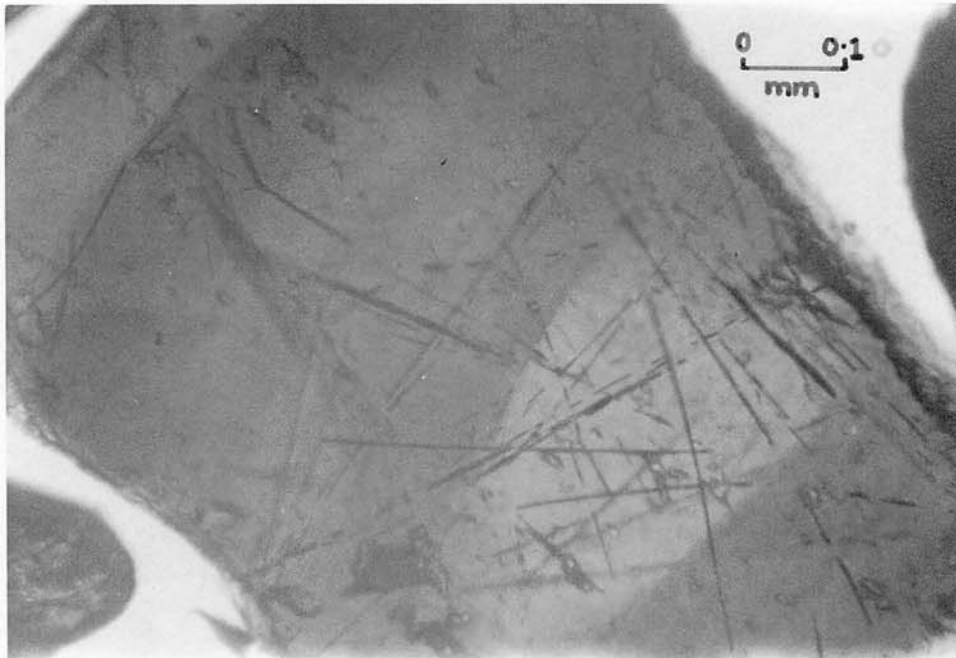


Plate 5. Photomicrograph of a zoned piece of black cassiterite, which has numerous criss-crossing needle shaped rutile crystals. Fluid inclusions are abundant and scattered. Core zone has lighter body colour and is less pleochroic. Some of the inclusions appear to be subparallel to the needle rutile (uncrossed polars).
Sample: SKLK 74785.
Loc: Semiling, Kedah.

TABLE 1.2
PLEOCHROISM OF CASSITERITES FROM BAKRI
(NB: THE CASSITERITES ARE BIAxIAL))

Pleochroism	Minimum absorption. to z	Maximum absorption to y
Weak	Pale light yellowish brown or pale yellow	Light yellowish brown or moderate orangy brown
Moderate	Moderately dark yellowish brown	Mallow honey orangy or blood red
Strong	Blood red	Almost red black

pleochroic scheme is essentially similar to the Gunung Jerai cassiterite and can be represented under the 3 categories (Table 1.2).

Zoning is important and this leads to differences in pleochroism and body colour within the same crystal grain. Twinning is not often encountered but both simple and lamella twins are present and may develop into a complex criss-crossing pattern. However lamella twins are seldom continuous even over short distances and where simple twins are developed they are quite invariably cyclically repeated.

Any number of the possible sets of cleavages and partings may be observed but generally not more than 3 of these sets are observed at any one time. They are weakly developed but cracks and fractures are very frequent. Relief and birefringence are both very high but birefringence is always masked by the extremely strong body colour.

Crystal grains appear to be discrete and no penetrating crystals were observed within another grain in contrast to the cassiterites from Gunung Jerai. Almost without exception, dispersion is markedly more prominent in lighter coloured portions than the darker areas.

Inclusions are mainly fluid inclusions as well as opaque minerals which may probably be columbite-tantalite. In some instances, columbite were observed to be in direct contact with the cassiterite grains (checked under reflected light microscope).

Interference figures are generally slightly biaxial positive with small $2V\gamma$'s. Uniaxial positive grains are also present. The cassiterites from the Kim Bee Tin Mine are much more pleochroic and have stronger body colours than those from the Hong Yuen Tin Mine.

Cassiterites from Phuket

Studies of numerous samples collected from a large number of mines in the Kathu Valley and offshore Phuket indicate that there are substantial degree of variations in the intensity of pleochroism and body colour of the cassiterites found within the area. Strong dichroism is exhibited by cassiterites from the Ban Guan and Poon Tha Wi Tin

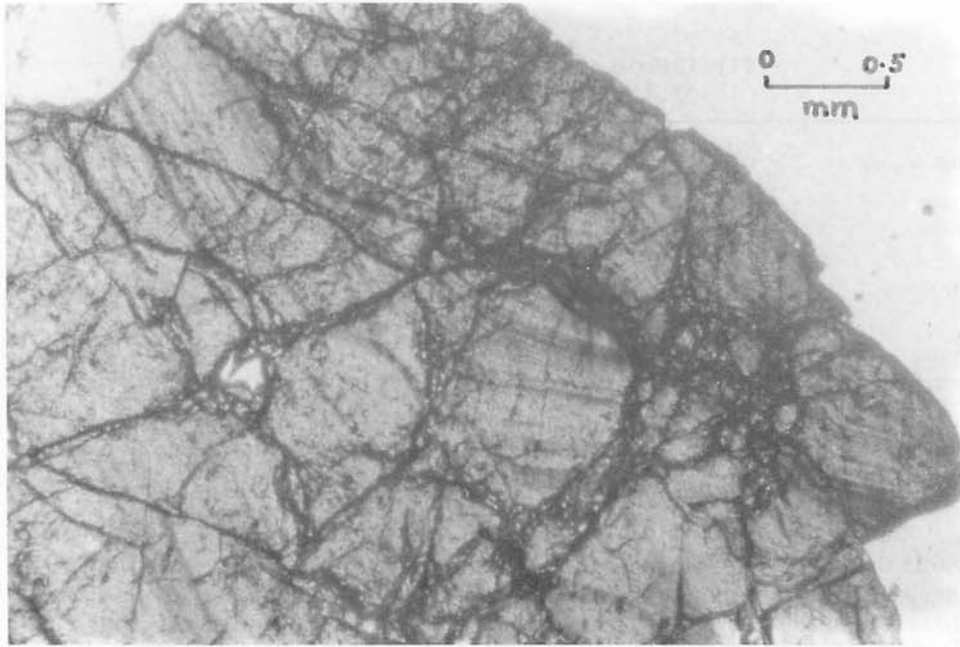


Plate 6a. Photomicrograph of black cassiterite showing numerous zig-zag zones. Irregular patchy zoning is also present and note the blood red colour of the darker zones. The cassiterite contains abundant irregular fractures and the z ray is orientated parallel to N-S polarizer showing minimum absorption colours (uncrossed polars).

Samples: PKPTW 1647818.

Loc: Poon Tha Wi Tin Mine, Kathu, Phuket.

mines in the Kathu Valley (Plates 6a, 6b). This phenomenon is less marked for offshore cassiterites as well as cassiterites from other mines in the vicinity of the 2 mines. Typically the theme colour involved is orangy red and dichroism exhibited ranges from the palest brown or orange to deep blood red. Quite often encountered too are non-pleochroic parts within the grain though these portions may not necessarily be the palest but are quite characteristically brownish with no red tinge.

Dispersion due to internal reflection again shows the inverse relationship; paler regions invariably have higher dispersion. Colour zoning is rather poorly developed and patchy zoning constitutes the most frequent mode of zoning encountered. Simple twinning is more marked than lamella twins which often pinch out in the darker areas. Some particularly well developed repeated twins are broad and in the process reorientate any zones present into a zig-zag pattern. Fractures present are often lined with manganese oxides (Fig. 7).

Of particular interest is the presence of fairly large numbers of cassiterites with 'anomalous' biaxial positive interference figures. Most of the studied cassiterites have $2V\gamma$ ranging from 1° to almost 15° . These $2V\gamma$ values vary from zone to zone with paler zones having the smaller values and the darker zones the larger values.

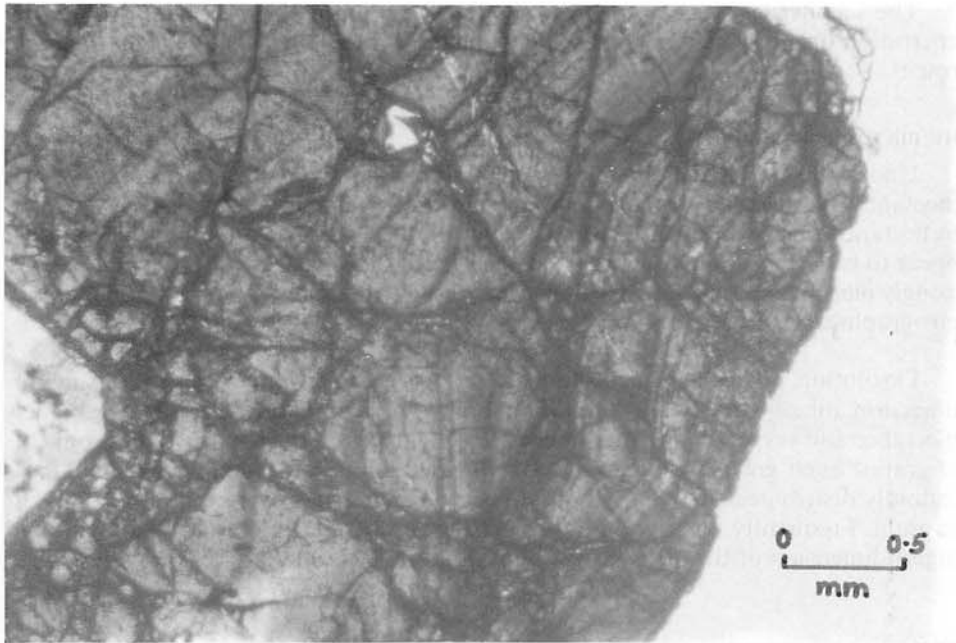


Plate 6b. Photomicrograph of the same cassiterite grain as in plate 6a which exhibits very strong dichroism. The maximum intense blood red colour is shown here when γ ray is parallel to N-S polariser (uncrossed polars).

Sample: PKPTW 1647818.

Loc: Poon Tha Wi Tin Mine, Kathu, Phuket.

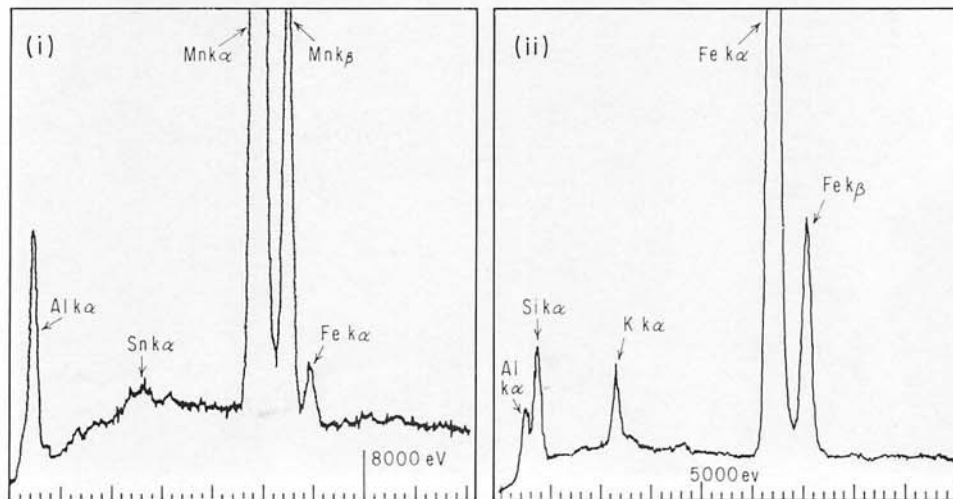


Fig. 7. SEM Energy Dispersion Spectrum of some veins associated with cassiterite
 (i) Predominantly manganese oxides vein in cassiterite from Ban Guan Tin Mine, Kathu.
 (ii) Predominantly iron oxides vein in cassiterite from the Soon Hin-Choy Hin Tin Mine, Kuala Lumpur.

The cassiterites appear to grow discretely or in small aggregates with little penetration or interlocking crystals being apparent. Fluid and opaque inclusions are present.

Ore microscopic studies

Under the reflected light microscope, cassiterites from the 3 areas show low reflectance. Bireflectance is generally weak but some twinned crystals have very strong bireflectance from whitish grey to dark grey. Besides these two features, the grains appear to be homogenous, devoid of any characteristics that may help distinguish the strongly pleochroic and zoned portions from those that are not, as observed under the petrographical microscope.

Exsolution bodies and inclusions are best seen under immersion oil. Using an immersion oil of R.I. 1.515, exsolution bodies of a mineral with slightly higher reflectance and very much weaker bireflectance than cassiterite can be seen in some of the grains. Such greyish white exsolved elongated and ovoid bodies are usually not randomly distributed over the grain but occur in aggregates over certain portions of the grain. Frequently, their orientations are cleavage or partings controlled, with the longest dimension of the blebs being colinear with the cleavage traces (Plates 7a, 7b).

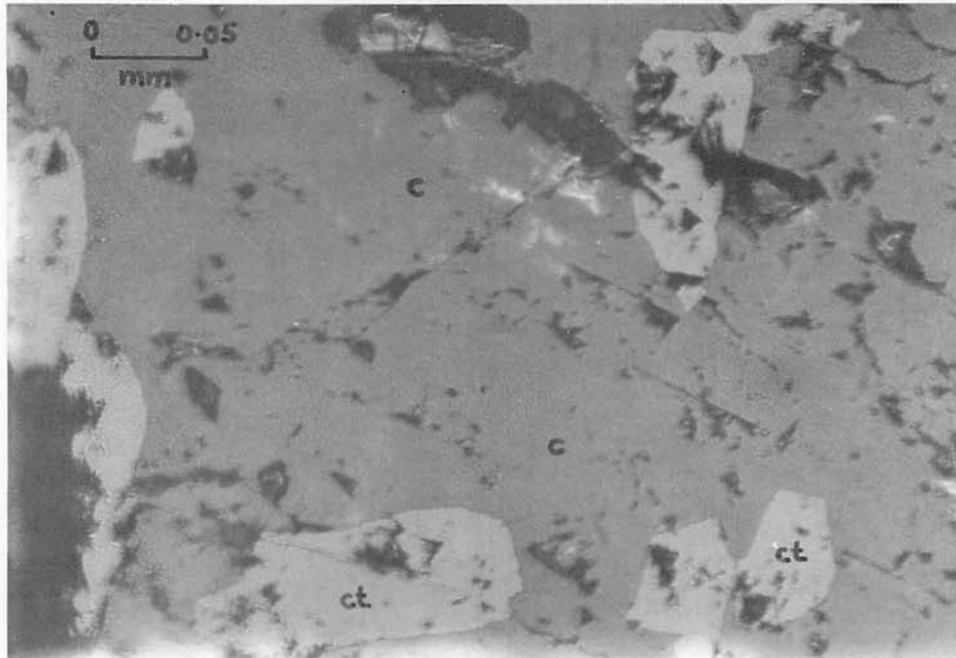


Plate 7a. Photomicrograph of subhedral exsolved bodies of columbite-tantalite (ct) in cassiterite (c). Columbite-tantalite bodies are lighter coloured and appear randomly orientated here.
Sample: PKST 1747824H.
Loc: Su-Tat Tin Mine, Kathu, Phuket.

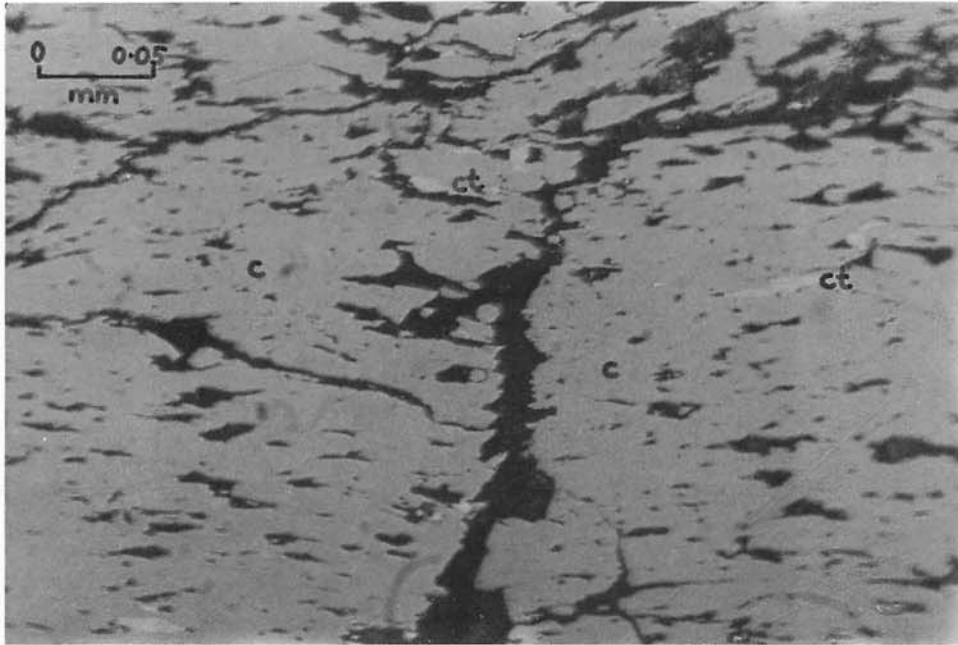


Plate 7b. Photomicrograph of cleavage controlled columbite-tantalite (ct) exsolution bodies in cassiterite (c). The elongated bodies are parallel to the poorly developed cleavages (under immersion oil, reflected light).

Sample: SJHY.

Loc: Hong Yuen Tin Mine, Bakri.

Subparallel, irregular, wavy minute feathery stringers of exsolved bodies are also present at an angle, adjacent to the composition plane of the simple twins, suggesting some degree of twin and zone control of exsolution as well (Plate 8). Such exsolved bodies die out further away from the twin plane indicating that the exsolution occurred in regions of maximum stress within the cassiterite grain. The location of such exsolution bodies present do not necessarily coincide with the strongly pleochroic zones within a cassiterite grain and in fact grains with such exsolved bodies are generally few in number. These exsolution bodies belong probably to the columbite-tantalite or mossaite-tapiolite solid solution series. Electron microprobe studies reveal a predominance of tantalum over niobium, in addition to iron and manganese as well as significant quantities of titanium (Figs. 8 & 9). It is likely that these exsolved bodies are columbo-tantalite (Kuz'mendo's classification, cited in Vlassov, 1964). The titanium may be incorporated within the columbite-tantalite or possibly may form small bodies of struverite or ilmenorutile.

Anisotropism is generally quite subdued for the strongly pleochroic cassiterite though reddish internal reflectance may sometimes be distinguished.

Summary

The marked similarities of cassiterites of the 3 areas, in the author's interpretation

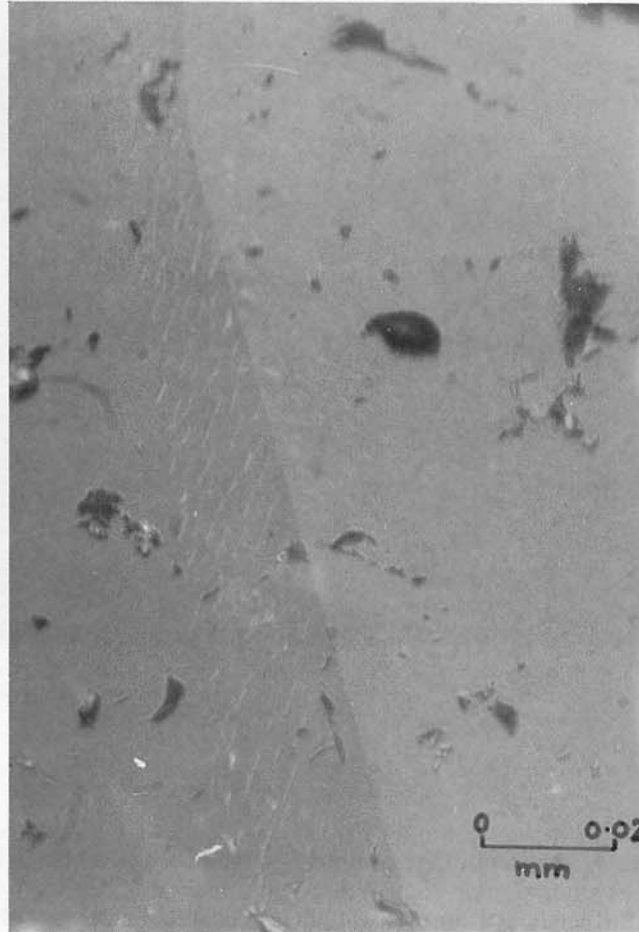


Plate 8. Photomicrograph of a cassiterite with simple twinning. The sharp twin plane can readily be seen. The wavy exsolution stringers of columbite-tantalite are both controlled by twinning and zoning. Note the discontinued nature of the exsolved bodies. (under immersion oil, reflected light).

Sample: SKLH 74785H.

Loc: Syarikat Perlombongan Kedah, Semiling.

are a direct manifestation of a common mode of genetic origin which in this particular case in question, is probably pegmatitic. Columbite and tantalite have been extensively documented to be pegmatite minerals (Turner, 1960; Betekhin, 1964; Zussman, *et al.*, 1962; Rankama & Sahama, 1950) and their close as well as direct relationship with the cassiterites is strong indication of a pegmatitic origin of the cassiterites, especially in the Bakri and Jerai areas. However, though they seem to be of similar genetic origin, there have been no evidence to suggest that they are generated by related events. The subtle variations in the properties as well as age differences of the related pegmatites tend to support the validity of such a contention. These variations may in effect reflect the regional control and modification during ore genesis.

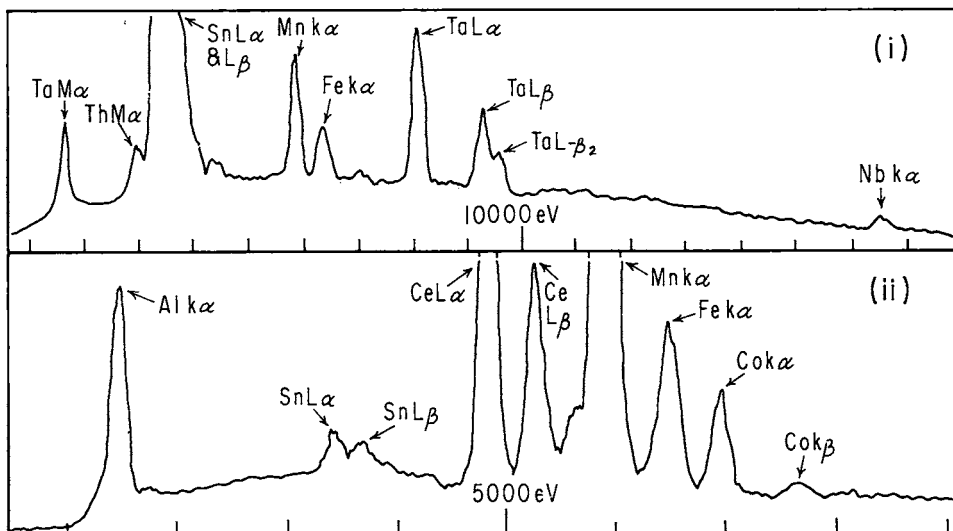


Fig. 8. SEM Energy Dispersion Spectrums of exsolution and inclusion bodies in the cassiterites from the Ban Guan Tin Mine, Kathu, Phuket.
 (i) Incipient exsolution of columbite-tantalite.
 (ii) A rare earth bearing inclusion within the cassiterite.

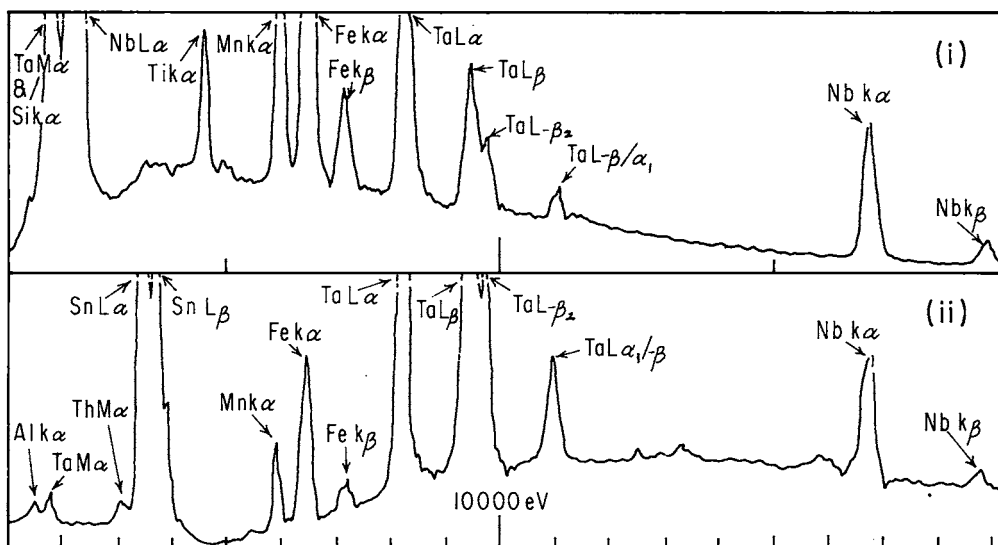


Fig. 9. SEM Energy Dispersion Spectrums of exsolution bodies of columbite-tantalite in cassiterite from the Foong Seong No. 2 Tin Mine, Semiling, Kedah.
 (i) Exsolution body containing abundant Ta, Nb, Ti, Mn and Fe.
 (ii) Exsolution body containing abundant Sn, Ta, some Nb, Fe, Mn, and Th.

RELATION BETWEEN COLOUR AND PLEOCHROISM

The origin of colours in minerals has often been attributed to impurities but many possible explanations have been given for different specific minerals (Nassau, 1978). The black cassiterite in thin section often exhibits orange red body colour and strong pleochroism. This probably is due to impurities which are incorporated within the cassiterite lattice. Chemical analysis and electron microprobe studies have revealed the presence of substantial percentage of impurities (to be given later) such as tantalum, silicon, aluminium, potassium, titanium, iron and manganese. It has been customary to attribute the blood red colour and strong pleochroism of the cassiterites in thin section to be due to the substitution of the tantalum ions for the stannic ions in the cassiterite lattice (Hosking, 1979; Liebenberg, 1945; Hutton, 1950) though Scrivenor (1919) ascribed strong pleochroism to the presence of titanium.

The presence of particular colours in minerals can quite justifiably be attributed to the presence of certain elemental impurities but the pinpointing of the responsible elements understandably is very difficult. For the case in question, it would be necessary to define the impurities present. Tantalum, iron, manganese and titanium appear to be likely candidates as they occur in substantial concentrations and represent the coloured ions present. The intensity of the colour exhibited would probably require impurities beyond the tens or even hundreds of ppm level. Tantalum indeed appears to be responsible for the orange-red colour in thin section and the black colour in hand-specimens, as many cassiterites from many other parts of the country studied which are known to be tantalum-poor but iron rich fail to show this characteristic colour. Instead they are brownish. Pleochroism on the other hand in the author's opinion is basically directly related to the high iron content and further enhanced by the characteristic colours due to tantalum. This contention is supported by the fact that pleochroism in cassiterite is never observed to be confined to tantalum-rich areas exclusively. It is also a well known fact that many mineral species which contain iron are pleochroic while isomorphs lacking in iron do not exhibit this property.

Crystals orientated differently give rise to differences in intensities of pleochroism. Changes in orientation had been observed especially in twinned grains where often different sides of the twins showed different orientations. Zoned crystals only occasionally show this phenomenon.

Collective influence of all impurities especially manganese and titanium in addition to the elements described may possibly be involved in the modification and manifestation of these 2 properties. The role of niobium is doubtful but its concentrations within the cassiterites are often very subordinate to tantalum and thus its effects would be masked by tantalum.

RELATION BETWEEN ZONING AND COMPOSITION

Colour zoning in minerals is frequently shown to be due to compositional zoning. The cassiterites showing intense zoning probably are no exceptions but owing to the relatively low concentrations of the impurities, they sometimes failed to be identified unambiguously, even under the electron microprobe available though Leube and Stumpfl (1963), cited by Hosking (1979) has demonstrated that the intensity of brown

colour in a specimen of brown cassiterite varied with the iron content. There is little doubt however that substantial amounts of iron, tantalum and manganese are present in the darker zones while the paler zones do not seem to contain any significant impurities as shown by electron microprobe traverses of zoned pegmatitic cassiterites (Fig. 18).

BIAXIALITY OF CASSITERITE

Biaxiality in 'dimetric' cassiterites have been noted by Gotman (1938) and Hutton (1950). Literature regarding the properties of such cassiterites is scanty and indeed no mention of such occurrences have been made in many texts on mineralogy. Their presence were cited by Deer, Howie & Zussman (1962). As far as the author is aware, no mention of biaxial cassiterites have been made regarding Malaysian cassiterites. Thus, it is interesting to note that their occurrence have been detected in very significant numbers in all the 3 study areas (Table 2). Studies of a sample from the Choy Hin—Soon Hin Tin Mine located in the Kuala Lumpur tin field similarly shows biaxiality with $2V\gamma$ as large as 20° . This suggests that their existence are not only confined to pegmatitic areas with black cassiterite but probably are much more

TABLE 2

TABLE SHOWING THE POSSIBLE RANGE OF BIAXIALITY OBSERVED IN CASSITERITES FROM VARIOUS LOCALITIES.

Sample Number	Uniaxial/Biaxial	$2V$ range (degrees)	Locality
PKBG1947817a	Biaxial	Probably 10	Kathu, Phuket.
PKBG1947817b	Biaxial	2-10	"
PKBG1947817c	Biaxial	1-6	"
PKPTW1647818a	Biaxial & Uniaxial	probably 0-5	"
PKPTW1647818b	Biaxial	1-3	"
PKSP1547826	Uniaxial	0	"
PKST1747824H	Uniaxial to Biaxial	0-10	"
SKLH74785H	Uniaxial to Biaxial	0-3	Tupah, Kedah.
SKFS1047810	Biaxial	2-10	Semiling, Kedah.
SKLK74782	Uniaxial to Biaxial	0-5	"
SKSC747814H	Biaxial	1-10	"
SJKB	Biaxial	Probably small $2V$	Bakri, Johore.
SKLSHCH	Uniaxial to Biaxial	0-20	Kuala Lumpur.
TS70	Uniaxial to Biaxial	Probably small $2V$	Pelepah Kanan, Johore.
TS4	Uniaxial	0	Kg. Pandan, Kuala Lumpur.
TS71	Uniaxial	0	PCCL, S. Lembing, Pahang.
TS132	Uniaxial to Biaxial	Probably small $2V$	Ayer Hitam, Selangor.
TS74	Uniaxial	0	Sungai Besi, Kuala Lumpur.
TS59	Uniaxial to Biaxial	Probably small $2V$	Perak.

NB: The $2V$ range is determined by visual estimate using Tobi's Method. The series of sample numbers starting with TS are Departmental Slides, from the Department of Geology, University of Malaya.

widespread in their distribution. Gotman (1938) had suggested that such anomalous behaviour in crystals which do not show strain effects is seen in material showing a zonal structure, but Hutton (1950) had noted optic axial angles of approximately 10° in unzoned crystals. Recent studies of biaxiality in 'isometric' and 'dimetric' crystals were made by Foord and Mills (1978) on several different mineral species but they found no systematic correlation between elemental zoning and the $2V$ measured along the same traverses. Similarly, they fail to detect any deviations from previously observed crystallographic symmetries.

From studies made by the author, it seems that biaxiality in cassiterite is not affected solely by whether the crystal is zoned. It occurs in both types. However, it is noted that the $2V$ changes from zone to zone within a zoned crystal whereas in unzoned crystals, they are quite consistent over most of the grain. The dark zones are the more anomalous regions. This implies that compositional control is probably more important than other factors which may include crystallography and induced mechanical strains. Most of the specimens studied do not show strain effects. Anyway it would be difficult to strain a cassiterite crystal to the extent of producing a $2V$ of 20° without any effects of shearing and fracturing of the crystal. The anomalies and relationships between colour, twinning, zoning, pleochroism and interference figures are summarised in a series of sketches of some cassiterites studied in Fig. 10.

From X-ray diffraction data, the unit cell constants of these black cassiterites show slight deviations in the 3rd and 4th decimal places from the pure synthetic tetragonal cassiterites, even though Santokh Singh and Bean (1967) had reported no appreciable effects of tantalum and niobium on the cell parameters of cassiterites. It is not inconceivable that variations do occur when there is ionic substitution by ions of different ionic radius and charge. Stöcher and Collingnes (1957) had documented the increase in cell parameters of cassiterite when Sn is replaced by Zr. Flinter (1960) quoted in Bradford (1970) had also mentioned a larger than normal d-spacing and thus consequently larger unit cell constants of cassiterite from the Kedah Peak area. Unit cell constants calculated for cassiterites from the 3 areas and other non-pegmatite areas show that there is systematic variation in the cell parameters (Tables 4 & 5; Figs. 13 to 17). Black cassiterites occupy a separate field from cassiterites which are brown or yellowish coloured. Shortening of the c-axis of black cassiterite is marked. This probably explains for the shortening of the prismatic cassiterites to squat bipyramidal forms.

It is also of interest that pure as well as analysed cassiterites have cell parameters a and b which are unequal (Table 5). This may form the theoretical and experimental basis to suggest a slightly orthorhombic symmetry of cassiterites to explain the biaxiality observed. Betekhtin (1964) in fact had stated that 'occasional optically biaxial varieties possibly have a crystal structure close to the orthorhombic', as rhombic crystals of pure cassiterite of S.G. 6.70 were obtained artificially by Daubrée (quoted in Betekhtin, 1964) without compositional impurities. The rhombic shaped cassiterites from the lepidolite pegmatites may well be the first rhombic shaped cassiterites found in nature, to correspond with the artificially created ones by Daubrée. The fact that naturally occurring rhombic cassiterite have a higher S.G. (6.85 to 6.90) is not surprising as this is due to impurities present (Tables 3 & 5).

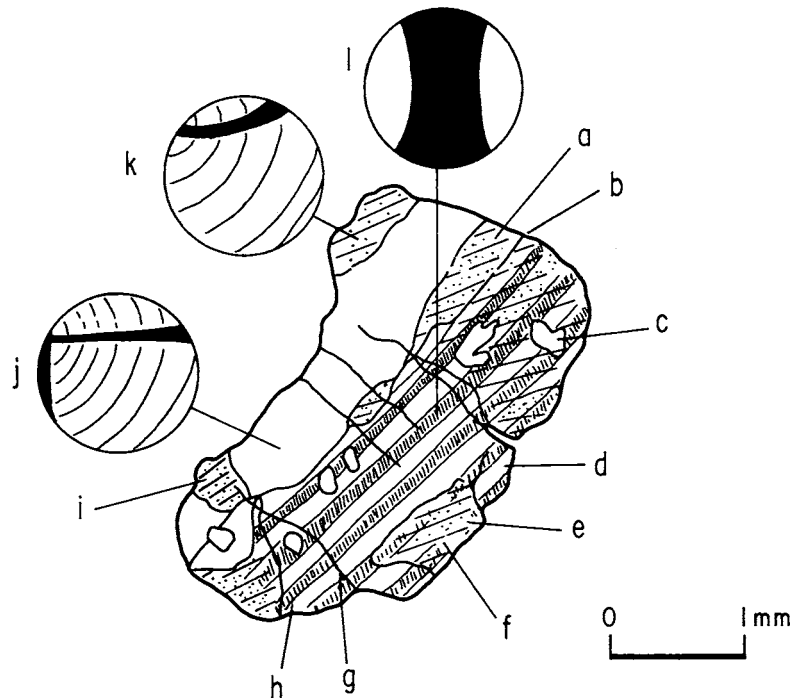
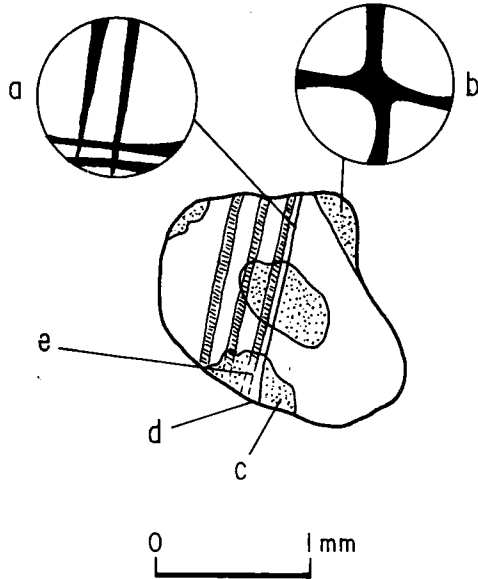
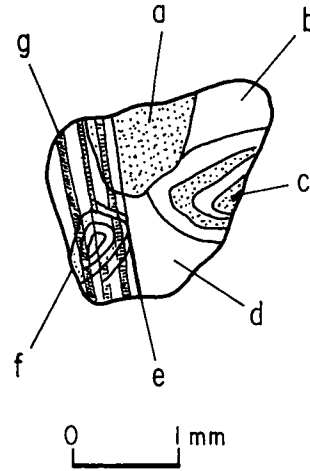
(i) SAMPLE : SKFS 1047810 f

Fig. 10. Sketches of some cassiterite grains showing relationships between crystallographic and optical properties (SKFS, SKLH from Gunung Jerai area, PKST from Kathu Valley, Phuket).

(i) Sample: SKFS 1047810f

The whole crystal is simple twinned, the right hand half shows lamellar twinning while the left hand half does not. Crystal shows patchy zoning and zones may be continuous between the two halves of twinned crystal.

- a. Continuous pleochroic zone with that present on right side half of twinned crystal but not as pleochroic as the right side half. This is probably due to differences in crystallographic orientation between the two sides.
 - b. Simple twin plane.
 - c. Inclusions usually of quartz.
 - d. Light coloured zone. Weak to non-pleochroic.
 - e. More darkish pleochroic zones, blood red pleochroism.
 - f. Dying out of lamella twins in more darkish pleochroic zones.
 - g. Transverse fractures.
 - h. Lamella twins.
 - i. Less pleochroic than similar looking darkish zones on right half of crystal.
 - j. Straight isogyre, uniaxial, off-centered basal.
 - k. Curved isogyre, probably biaxial with small $2V$.
 - l. Diffuse flash figure.
- } Changes in crystal orientation
between the two halves.

(ii) SAMPLE : SKLH 74785 Unsep H.(iii) SAMPLE : PKST 1747824 H

(ii) Sample: SKLH 74785 Unsep. H.

- a. Double straight isogyre observed at simple twin plane indicating changes in crystal orientation.
- b. Slightly biaxial with small $2V$ (at 45° position).
- c. Very pleochroic part (blood red pleochroism).
- d. Simple twin plane.
- e. Pinching out of lamella twins.

(iii) Sample: PKST 1747824H.

- a. Strongly pleochroic.
- b. Weakly pleochroic
- c. Growth zones.
- d. High dispersion, never extinguished.
- e. Simple twin plane.
- f. Concentric zoning.
- g. Lamella twins.

From an analysis of the ionic radii of the common impurities present in the cassiterite, it is obvious that the pentavalent niobium and tantalum of ionic radii 0.70 Å and 0.68 Å respectively (Sahama & Rankama, 1950) and tetravalent titanium of ionic radius 0.68 Å can readily substitute for the tetravalent tin of ionic radius 0.73 Å. It shows the preferential substitution of Ta^{5+} over Nb^{5+} for the Sn^{4+} within the cassiterite lattice. This results in a higher concentration of tantalum over niobium in cassiterite, even though the niobium content in syenitic pegmatites is about 384 times more abundant than tantalum and the average for igneous rocks is 10 to 12 times (Rankama, 1944, 1947). The high concentration of titanium can similarly be explained as Sn^{4+} and Ti^{4+} have similar radii and charge. In fact, Ti^{4+} tends to substitute isomorphously the Sn^{4+} in the cassiterite and vice versa (e.g. malayaite, Sn replaces Ti in sphene).

In the presence of tantalum-niobium, the substitution of Fe^{2+} , Fe^{3+} or Mn^{2+} with ionic radii of 0.76 Å, 0.64 Å, and 0.80 Å respectively are much more subdued according to Goldschmidt's (1937) rules. Among the three, it would be expected that Fe^{3+} will be most favourable for substituting the Sn^{4+} . Observations of chemical analysis results tend to support this theoretical consideration. Though iron is generally much more abundant in pegmatites than both niobium and tantalum, their concentrations within the cassiterites are generally lower than the tantalum. However, the presence of niobium-tantalum within the cassiterite probably also enhances the substitution by iron and manganese, for they are required to balance the electronic charges of cassiterite lattice to maintain neutrality.

Substitution by Al^{3+} and Si^{4+} with ionic radii of 0.51 Å and 0.41 Å respectively are possible but substitution by K^{+} with ionic radius of 1.33 Å is highly difficult and improbable.

It may be summarised that these chemical ionic substitutions are solely or at least partly responsible for most of the variation in optical and structural properties observed in the cassiterites studied. These substitutions may lead to Schottley defects within the crystals which include nonstoichiometric chemistry, inherent crystal defects and differential charges and ionic radii among others. It will also be unrealistic to rule out other possibilities that may have given rise to some of these properties including those probably produced by mechanical deformation and crystal defects caused by temperature or pressure quenches. Temperature and pressure conditions prevailing during the growth of the cassiterite appears to be of significance especially with respect to biaxiality as pure rhombic crystals of cassiterite exhibiting optical biaxiality has been grown artificially by Daubrée.

RELATION BETWEEN COLOUR, PLEOCHROISM AND 2V

Studies were made of the mutual relationships and variations of colour, pleochroism with the 2V of some biaxial cassiterites from Phuket and Gunung Jerai areas. Attempts to correlate these optical properties to chemical composition variations by electron microprobe were not very successful because of relatively low detection levels of the instrument.

Polished thin sections of cassiterite samples made were used for these studies. Tobi's (1956) method of 2V determination was used. Both colour and pleochroism determinations are solely based on qualitative observation and judgement and this was done at the same points along the same traverses where the 2V was determined.

Tobi's method is designed for measuring optic angles from interference figures in which the acute bisectrix is well centered. In the case of cassiterites, this would be the basal section parallel to 001. However, it had been proven that it is equally applicable to acute bisectrix figures which are not very well centered. The error involved would be of the order of 1° or less. Any other errors would be inherited from the approximate nature of Mallard's equation (N_y is taken to be 2.00).

The result of these studies are summarised in Figs. 11 and 12. From the figures, it may be concluded that there is a direct relationship between the 2V γ measured and the

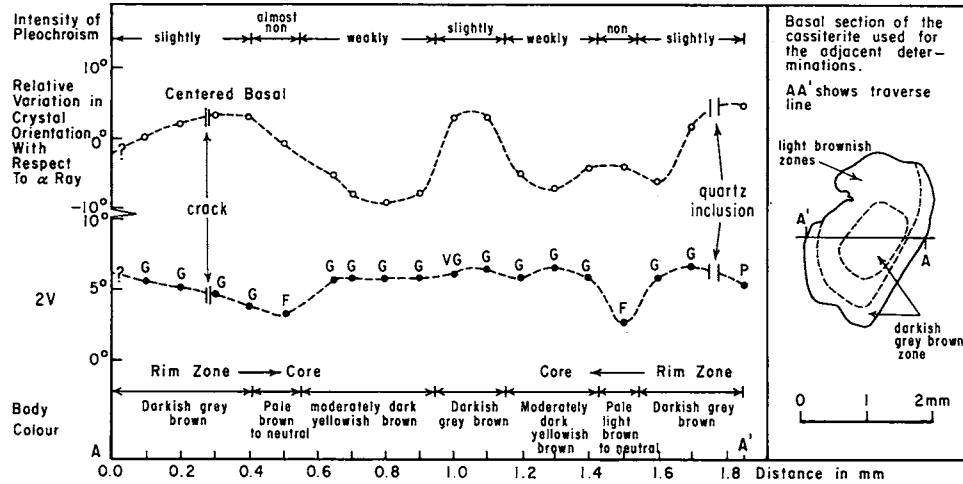


Fig. 11. Relationships between 2V, intensity of pleochroism, body colour and crystal orientation along a traverse of a cassiterite crystal from the Ban Guan Lepidolite Pegmatite. Note especially the direct correlation of the intensity of pleochroism and 2V angle. Points along traverse where determinations were made are plotted. Level of confidence of 2V determinations is indicated as VG-very good, G-good, F-fair, P-poor. (Sample: PKBG 1947317).

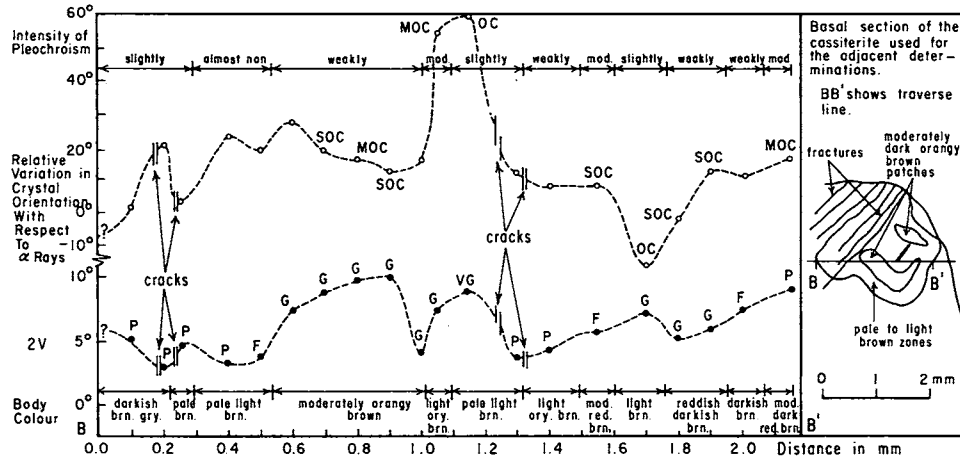


Fig. 12. Relationships between 2V, intensity of pleochroism, body colour and crystal orientation along a traverse of a cassiterite crystal from the Foong Seong Tin Mine. Direct correlation between intensity of pleochroism and 2V angle is evident though it may be modified by crystal orientation. Level of confidence of 2V determination is indicated as in Fig. 11. Degree of deviation of the c-axis orientation is indicated as: OC-off-centered, MOC-moderately off-centered, SOC-slightly off-centered. The rest is centered. (Sample: SKFS 1557816).

degree of pleochroism observed along the traverses. More pleochroic zones are accompanied by generally higher biaxiality which generally vary between 3° to as much as 10° . It also adds further evidence to the belief that the generally darker zones are more pleochroic than the lighter bands and the orange-red body colour is characteristic of the more pleochroic bands.

The fact that the $2V\gamma$, degree of pleochroism and orange-red body colour are mutually dependent imply that they are influenced by factors common to these three properties. As previously discussed, pleochroism and body colour are most probably functions of the extents of elemental substitutions of the Sn ions within the lattice by other elements especially tantalum and iron, thus it is also highly probable that biaxiality is also dependent primarily on this factor.

It is also apparent that the crystallographic orientation of the crystal changes rather randomly and rapidly along the traverse line. This is shown by the different orientations of the α -ray about a fixed arbitrary position. Besides the dislocation of the α -ray, the position of the c-axis also changes slightly, shown by the positions of the interference figure, which shifts from centered basal sections to off-centered basal sections. It has been noted that changes in crystallographic orientation affects the degree of pleochroism observed. Any deviations from the centered basal section would generally increase the degree of pleochroism observed. Manifestation of this effect may be present along the traverse in Fig. 12 whose section varies between centered to off-centered basal.

SPECIFIC GRAVITY (S.G.)

Specific gravity measurements were made of cassiterites from the various areas using the Berman Density balance. Studies were conducted to establish the variability of the specific gravity of cassiterites especially to that relating to their colours. It is the belief of the author that the colour of the cassiterites should show a clear correlation to its specific gravity reflecting the changes in its composition and cell lattice constants. Some cassiterite samples from the Kuala Lumpur tin field were also studied to establish the differences between the non-pegmatitic cassiterites from those which are pegmatitic. The results are summarised in Table 3.

Various works on cassiterites have quoted slightly different S.G. of cassiterites but in general they are expected to range from 6.80 to 7.10. It is confirmed from the studies that there is indeed a whole range of S.G. of cassiterites ranging from 6.75 to 7.12 and there exists a good correlation between the colour and specific gravity. The black opaque and dark brown cassiterites have S.G. ranging from 6.75 to 6.95, the yellowish brown translucent ones having the highest S.G. values, ranging from 7.05 to 7.12. It is noteworthy to state that the clear transparent cassiterites have S.G. in the range of around 7.00, that is, intermediate between the 2 other varieties. This S.G. value of about 7.00 almost coincides with the S.G. of the pure cassiterite given in the JCPDS File. It may be interpreted that the black cassiterites and yellowish brown cassiterites occur in 2 different fields, the former being lighter and the latter heavier. This relationship had in fact been noticed by C.F. Choong (cited by Bradford, 1970), a pioneer miner in the Gunung Jerai area who noticed that bigger standard bags were

TABLE 3
 RELATIONSHIPS BETWEEN THE COLOUR, TRANSPARENCY, GENETIC ORIGIN AS WELL AS
 SPATIAL VARIATION OF CASSITERITES WITH THEIR SPECIFIC GRAVITY AS DETERMINED
 BY USING A BERMAN DENSITY BALANCE (IMMERSION LIQUID USED IS TOLUENE)

Field sample no.	Locality of sample	No. of specimens used in determination	Colour	Transparency, remarks & genetic origin	W ₁ wt in air (mg)	W ₂ wt in toluene (mg)	Temp °C	Specific Gravity (ave)
JCPDS Powder File No. 21-1250	Sample from Johnson, Matthey & Co. Ltd.	-	white	spectro analysis shows no impurities greater than faint trace	-	-	6.995	deduced from XRD
SPSP	Sin Patana Tin Mine, Kathu, Phuket, Thailand	4	yellowish light brown cass. with slight darkish small patches	translucent, coarse grained probably pegmatitic origin	18.34	16.12	25.2	7.112 ₅
SPSP	Sin Patana Tin Mine, Kathu, Phuket, Thailand	4	pale brownish cass.	translucent, coarse grained probably pegmatitic origin	16.32	14.34	25.2	7.095 ₂
SPSP	"	2	pale brown to whitish brown cass.	"	12.98	11.41	25.6	7.113 ₁
SPSP	"	1	brownish black cass.	opaque, from very fine grained up to 3 cm or more, pegmatitic origin	18.94	16.57	25.6	6.876 ₇
SPSP	"	3	jet black cass.	totally opaque, very fine grained up to 3 cm or more, pegmatitic origin	20.02	17.50	25.7	6.834 ₄

TABLE 3 (contd.)

Field sample no.	Locality of sample	No. of specimens used in determination	Colour	Transparency, remarks & genetic origin	W ₁ wt in air (mg)	W ₂ wt in toluene (mg)	Temp °C	Specific Gravity (ave)
SPSP	"	1	"	"	16.59	14.52	25.8	6.894 ₀
SPBG	Ban Guan Tin Mine, Kathu, Phuket, Thailand	3	"	"	22.21	19.43	26.5	6.868 ₅
SPBG	"	2	black cass.	"	24.96	21.87	26.8	6.941 ₄
SPBG	"	1	black cass. but with a slightly metallic bluish tinge	opaque, very coarse grained up to 3 cm or more, pegmatitic origin	22.35	19.53	26.6	6.812 ₂
SPBG	"	1	metallic bluish grey	opaque, very coarse grained pegmatitic origin, have rhombic x-sections	22.40	19.60	22.8	6.904 ₄
SPBG	"	2	"	"	21.73	19.99	22.8	6.845 ₀
SPBG	"	1	metallic bluish black	"	16.96	14.84	23.1	6.902 ₁
SKLK	Syarikat Perlombongan Perbadanan Kedah, Semling, Kedah	1	clear pale brownish or flesh coloured cass.	translucent to transparent, coarse grained prob. pegmatitic origin	19.40	17.02	25.9	7.019

TABLE 3 (contd.)

Field sample no.	Locality of sample	No. of specimens used in determination	Colour	Transparency, remarks & genetic origin	W ₁ wt in air (mg)	W ₂ wt in toluene (mg)	Temp °C	Specific Gravity (ave)
SKLK	"	2	clear with slight pale yellowish & brownish tinge	glassy absolutely transparent, coarse grained prob. pegmatitic origin	18.15	15.93	26.2	7.030 ₂
SKLK	"	2	whitish yellow cass.	translucent, coarse grained prob. pegmatitic origin	21.67	19.05	25.9	7.114 ₀
SKLK	"	5	pale brownish yellow cass.	"	17.15	15.08	25.9	7.126 ₀
SKLK	"	3	"	"	17.99	15.80	26.0	7.065 ₁
SKLK	"	1	jet black	opaque, coarse grained, prob. pegmatitic origin	15.71	13.62	26.1	6.481 ₇
SKLK	"	2	"	"	18.07	15.82	26.0	6.907 ₃
SKLK	"	2	"	"	16.49	14.45	26.2	6.951 ₈
SKFS	Foong Seong Tin Mine No. 1, Semling, Kedah	2	black cass. with slight bluish metallic tinge	opaque, medium to coarse grained, prob. pegmatitic origin	16.60	14.55	26.7	6.925 ₄

TABLE 3 (contd.)

Field sample no.	Locality of sample	No. of specimens used in determination	Colour	Transparency, remarks & genetic origin	W ₁ wt in air (mg)	W ₂ wt in toluene (mg)	Temp °C	Specific Gravity (ave)
SKFS	"	4	light brown cass.	translucent, assoc. with qtz., sample free of gangue	17.18	15.11	26.5	7.134 ₃
SKFS	"	1	black cass. with slight metallic bluish tinge	opaque, coarse grained, prob. pegmatitic origin	24.89	21.83	25.0	7.003 ₄
SKFS	"	2	jet black glassy cass.	very slightly translucent, coarse grained, prob. pegmatitic origin	24.56	21.43	25.0	6.756 ₆
SKFS	"	2	glassy jet black cass. to metallic coloured cass.	slightly translucent to opaque, coarse grained prob. pegmatitic origin	13.93	12.28	25.7	7.263 ₃
SKFS	"	2	black cass.	opaque, coarse grained, prob. pegmatitic origin	15.68	13.74	26.0	6.952 ₅
SJHY	Hong Yuen Tin Mine, Bakri, Johore	3	brownish yellow mixed with some blackish patches	translucent to opaque, zoned crystals, fine to medium grained prob. pegmatitic	4.37	3.84	26.5	7.688 ₇

TABLE 3 (contd.)

Field sample no.	Locality of sample	No. of specimens used in determination	Colour	Transparency, remarks & genetic origin	W ₁ wt in air (mg)	W ₂ wt in toluene (mg)	Temp °C	Specific Gravity (ave)
SJHY	"	2	jet black cass.	opaque, medium grained, very prob. pegmatitic	12.20	10.69	26.8	6.943 ₇
SJHY	"	8	"	opaque, fine grained, very prob. pegmatitic	15.09	13.19	26.8	6.825 ₇
SJKB	Kim Bee Tin Mine, Bakri, Johore	1	black cass.	opaque, coarse grained	23.38	20.44	26.8	6.834 ₆
SJKB	"	2	"	"	17.60	15.43	26.7	6.970 ₅
SKLSHCH	Soon Hin-Choy Hin Tin Mine, Salak South, Kuala Lumpur	1	deep reddish brown to blackish cass. metallic some parts	lustrous, slightly translucent, lode cass. prob. metasomatic assoc. with some clays	17.80	15.57	26.8	6.859 ₁
SKLSHCH	"	2	deep reddish brown to blackish some parts metallic	lustrous, slightly translucent, SG slightly lowered prob. due to clays, etc. cannot be totally removed	23.29	20.40	26.8	6.925 ₁
SKLPM	Pudu Mine, Kuala Lumpur	2	darkish brown black cass.	opaque, contact metasomatic lode cass., coarse grained	17.41	15.24	24.2	6.914 ₈
SKLPM	"	1	brownish black	"	15.80	13.81	23.0	6.851 ₈

required to contain black magnetic cassiterite of constant weights when compared to other tin bearing areas. Visual estimates by Bradford indicate a 5% increase in the volume of the standard bags.

Statistical analysis of the S.G. comprising of the black cassiterites reveal a mode between 6.85 and 6.95, while the light yellowish brown cassiterites range from 7.05 to 7.15. Assuming a statistical average value of black cassiterites to be 6.86 and yellowish brown cassiterites to be 7.10, this would represent a 3.5% increase in the volume of the standard bags. It is possible that an increase of 5% in volume is in order as the electromagnetic separation of cassiterite and columbite ore is never 100% and as such the columbite which have lower S.G., being inevitably present in the concentrations would increase the volume beyond the 3.5% calculated to 5% or more.

It is noticed that there is not much difference in the S.G. of the rhombic cassiterites from Phuket with respect to the other black cassiterites.

Though ionic substitution of the Sn^{4+} of the cassiterite by other ions are directly related to the specific gravity variations, they are also probably related to slight changes in packing and coordination. This becomes more important, especially in the study areas where some of the cassiterites have been shown to be biaxial and increasingly so with increase in colour intensity. As long as the crystal symmetry of these cassiterites remain unresolved, it is difficult to explain completely the seemingly contradictory lower S.G. values of the black cassiterites. It would be expected that with substitution by tantalum ions of much higher atomic weight (180.9) than tin (118.6), this should result in higher S.G. values. A brief attempt to interpret this is made here. It must be stressed here that tantalum possibly is the only significant element with higher atomic weight than tin that substitutes for the Sn^{4+} in the lattice. If it is the sole participant in the lattice, it would expectedly results in an increase in the S.G. However, the substitution by pentavalent tantalum and niobium often results in substitution by other ions of much lower atomic weights like Si^{4+} , Ti^{4+} , Fe^{2+} , or Mn^{2+} for the Sn^{4+} in order to achieve electronic neutrality. The collective effect would result in a lower S.G. In general, the increase in substitution of Sn^{4+} ions by ions associated with pegmatites results in a lower S.G. of the cassiterites. Higher S.G. of brown and light coloured cassiterites is often reflected by lower levels of substitutions and probably the dominant impurity is iron. Iron substitutes for Sn^{4+} in the ratio of 2:1 or 4:3 and with its atomic weight of (55.9) help maintain more or less the S.G. of pure cassiterites. This may be helped by small amounts of heavier atoms which may be present.

CELL LATTICE PARAMETERS

X-ray diffraction techniques were employed to evaluate the changes in the cell lattice constants and the shifts of the cassiterites peaks in relation to the optical and physical properties like colour and specific gravity. Selected samples, mainly from the 3 study areas, which have been determined for their specific gravities were powdered. Pure silicon powder was used as internal calibration in all the determinations. Twelve samples were determined and the $d(\text{\AA})$ values are tabulated in Table 4.

From the results, it is obvious that there is a significant and consistent decrease in

TABLE 4 (contd.)

Cassiterite Sample And Location	5. Sin Patana SPSP, Phuket, Metallic black cassiterite			6. Sin Patana SPSP, Phuket, whitish yellow brown cass.			7. Ban Guan SPBG, Phuket metallic bluish black cassiterite			8. Ban Guan SPBG, Phuket, Bluish metallic grey, coarse, rhomboh X section		
	dÅ	Lines $\alpha, \alpha_1, \alpha_2$	I/I _o	dÅ	Lines $\alpha, \alpha_1, \alpha_2$	I/I _o	dÅ	Lines $\alpha, \alpha_1, \alpha_2$	I/I _o	dÅ	Lines $\alpha, \alpha_1, \alpha_2$	I/I _o
110	3.334	α	60	3.369	α	80	3.336	α	54	3.341	α	9
101	2.634	"	82	2.652	α_1	56	2.632	"	66	2.633	"	78
200	2.360	"	29	2.365	"	25	2.362	"	30	2.365	"	7
111	2.300	α_1	4	2.309	"	3	2.304	"	4	2.305	α_1	3
210	2.106	"	2	2.119	"	2	2.113	"	2	2.114	"	1
211	1.757	"	100	1.766	"	57	1.763	α_1	100	1.761	"	100
220	1.673	"	18	1.674	"	25	1.625	"	34	1.673	"	3
002	1.590	"	15	1.592	"	6	1.591	"	15	1.588	"	3
310	1.495	"	18	1.500	"	21	1.497	"	19	1.498	"	5
112	1.438	"	24	1.440	"	11	1.438	"	25	1.437	"	6
301	1.414	"	59	1.417	"	100	1.415	"	48	1.413	"	12
202	1.321	"	17	1.324	"	6	1.323	"	16	1.322	"	8
321	1.215	"	18	1.215	"	16	1.215	"	28	nd	"	
400	1.185	"	12	1.185	"	2	1.185	"	22	nd	"	
222	1.155	"	12	1.155	"	5	1.155	"	14	nd	"	
330	1.117	"	5	1.117	"	5	1.118	"	20	nd	"	
312	1.091	"	24	1.092	"	20	1.092	"	16	1.091	"	26
411	1.081	"	43	1.081	"	11	1.081	"	48	1.081	"	16
420	1.059	"	15	1.059	"	6	1.059	"	9	1.058	"	7
103	1.035	"	21	1.036	"	2	1.036	"	4	1.034	"	1
402												
510		lines not determined beyond this			lines not determined beyond this			lines not determined beyond this			lines not determined beyond this	
332												
501												
422												
303												
521												
440												
323												
530												
512												
Other lines present		lines not determined beyond this	compared to cass. (211)		lines not determined beyond this			lines not determined beyond this			lines not determined beyond this	
	2.969	α	10				1.983	α_1	2	3.161	α_1	36
	1.834	α_1	2				1.931	"	4	1.840	"	4
		Both are prob. Columbite- Tantalite lines					1.290	"	1			
							1.111	"	3			
	0.077						0.096					
										0.051		

TABLE 4 (contd.)

Cassiterite Sample And Location	9. Foong Seong SKFS Semiling, Black cassiterite			10. Perlombongan Kedah, SKLK, Semiling, yellowish brown cassiterite			11. Perlombongan SKLK, Semiling, clear transparent cass.			12. Soon Hin-Choy Hin, Salak South, SKLSHCH, dark brown blackish cassiterite		
	dÅ	Lines $\alpha, \alpha_1, \alpha_2$	I/I ₀	dÅ	Lines $\alpha, \alpha_1, \alpha_2$	I/I ₀	dÅ	Lines $\alpha, \alpha_1, \alpha_2$	I/I ₀	dÅ	Lines $\alpha, \alpha_1, \alpha_2$	I/I ₀
110	3.340	α	59	3.358	α	100	3.353	α	100	3.349	α	38
101	2.644	"	52	2.650	α_1	70	2.648	"	48	2.646	"	36
200	2.367	"	36	2.372	"	27	2.372	"	19	2.371	"	16
111	2.312	"	5	2.311	"	3	2.310	"	2	2.304	"	4
210	2.119	"	3	2.115	"	2	2.125	"	4	2.119	α_1	2
211	1.767	α_1	100	1.767	"	84	1.766	"	60	1.767	"	100
220	1.678	"	17	1.690	"	36	1.678	"	33	1.677	"	11
002	1.591	"	9	1.594	"	8	1.594	"	31	1.598	"	11
310	1.499	"	14	1.499	"	30	1.500	"	50	1.498	"	9
112	1.439	"	16	1.440	"	23	1.441	"	10	1.440	"	7
301	1.415	"	17	1.416	"	34	1.417	"	40	1.416	"	11
202	1.322	"	17	1.323	"	13	1.325	"	45	1.322	"	4
321	1.216	"	18	1.216	"	22	nd	"	nd	nd	"	nd
400	1.185	"	3	1.185	"	11	nd	"	nd	nd	"	nd
222	1.154	"	9	1.155	"	13	nd	"	nd	nd	"	nd
330	1.117	"	2	1.117	"	13	nd	"	nd	nd	"	nd
312	1.091	"	15	1.092	"	19	1.092	"	14	1.091	"	9
411	1.081	"	17	1.081	"	17	1.082	"	48	1.081	"	16
420	1.059	"	12	1.059	"	8	1.060	"	6	1.060	"	2
103	1.036	"	4	1.036	"	9	1.036	"	8	1.037	"	7
402												
510												
332												
501												
422												
303												
521												
440												
323												
530												
512												
Other lines present	3.165 1.596	α α_1	22 9	3.391 3.189	α "	21 89	3.400 3.185 2.671	α "	107 38 19	3.194 2.700 2.493 2.448 2.189 1.720 1.565 1.453 1.358	α " " " α_1 " " " "	60 14 9 11 7 9 9 4 11
	0.005			-0.035			-0.035			-0.011		

NB: X-ray Diffractograms internally calibrated using Si standard nd—not determined nn—not present

the d-spacings of the black cassiterites compared with the pure cassiterites. Additional lines besides those of cassiterites given in the JCPDS File were also present. Some of these lines had been determined to be those of columbite-tantalite. These probably existed as exsolution bodies within the cassiterites. The decrease is reflected in many lines or peaks though some peaks do not show any significant shifts. Yellowish brown and colourless transparent varieties generally have slightly higher d-spacings or are almost similar to the d-spacings of the standard pure cassiterites. A pure cassiterite sample synthetically prepared by B.D.H., containing 500 ppm of Pb and 20 ppm of As as impurities were determined and were found to be not significantly different from the pure cassiterite peaks. This shows the cassiterite crystal lattice can probably tolerate substitution on the tens or hundreds of ppm level without substantially changing its cell parameters. Only a very slight increase in the d-spacing was noticed after scrutiny and this probably reflects the effects of the large Pb^{4+} ions (0.84 Å) substituting the Sn^{4+} (0.73 Å). On similar grounds, the black cassiterites with a smaller d-spacing is consistent with the argument that they are caused by the smaller pentavalent tantalum (0.68 Å) and niobium (0.70 Å) or Ti^{4+} (0.68 Å) ions substituting for the tetravalent tin (0.73 Å) ions (Table 4).

To derive the unit cell constants of these cassiterites, the various cassiterite peaks were indexed making use of the lines given in the JCPDS File. This indexing results in the assumption of a tetragonal symmetry for these cassiterites even though optically they are known to be slightly biaxial. The indexing of these lines according to the orthorhombic system was not attempted by the author. The presence of unidentified lines and the significant shifts in the lines measured may be suggestive of other possible crystal systems besides tetragonal, the most probably being orthorhombic.

Peaks measured with high levels of confidence were utilised in the computation, using a modified computer program designed for computing the lattice constants of crystals in the cubic, tetragonal, orthorhombic and hexagonal systems (Chan, 1976; Lye, 1978). The applicability of the method have been shown to be very good (Lye, 1978) especially for the cubic and tetragonal systems while for the orthorhombic system it is quite good.

For the purpose of comparison, the unit cell constants of pure cassiterites were recalculated using this program from the lines given in the JCPDS File. In the author's opinion, this would give a more realistic comparison in the changes of the cell parameters of other cassiterites as the unit cell parameters computed may vary slightly, depending on the algorithm of solution used. Results of the computed cell lattice constants for cassiterites are shown in Table 5.

It is apparent that there is a clear relationship between the unit cell parameters, colour and specific gravity of cassiterites studied (Figs 13 to 17). In all cases there seem to be distinct separations of the field into 2 distinct groups representing the black and yellowish brown or transparent cassiterites. However, it is conceivable and in fact very likely that there exists a continuous variation between the 2 fields representing gradational and corresponding changes in colour, specific gravity, unit cell parameters and probably composition, if all varieties of cassiterites are similarly studied. Thus, it is very probable that the present study is principally concerned with the 2 extreme

TABLE 5

COMPARISON OF THE VARIATION OF COMPUTED CELL LATTICE CONSTANTS AND AXIAL RATIOS OBTAINED USING THE MODIFIED PROGRAM OF CHAN (1976) OF CASSITERITES FROM BAKRI, SEMILING, PHUKET AND KUALA LUMPUR IN RELATIONSHIP TO THEIR PHYSICAL PARAMETERS OF COLOUR AND DENSITY.

Field Sample Label & Locality	JCPDS Powder 21-1250	Data File No. Cassiterite	U.M. Departmental Pure Cassiterite Standard	Hong Yuen, SJHY, Bakri, Johore	Kim Bee, SJKB Bakri, Johore	Sin Patana, SPSP, Phuket Thailand
Colour & Transparency	1a White	1b White	2 Pale yellowish white. Translucent	3 Jet black. Opaque	4 Jet black. Opaque	5 Jet metallic black. Opaque
aÅ		4.737 ₂	4.737 ₃	4.740 ₂	4.741 ₈	4.735 ₆
bÅ		4.739 ₅	4.740 ₅	4.734 ₆	4.737 ₆	4.739 ₅
Average of a & bÅ		4.738 ₉	4.738 ₄	4.737 ₈	4.739 ₇	4.737 ₆
Assume tetragonal symmetry. See Text.						
cÅ	3.188	3.187 ₅	3.187 ₈	3.186 ₅	3.183 ₈	3.184 ₅
A = a/b	1	1.000 ₂	1.000 ₅	1.001 ₂	1.001 ₀	0.999 ₂
C = c/b	0.673	0.673 ₅	0.672 ₄	0.673 ₀	0.672 ₀	0.672 ₈
T = c/a	—	0.673 ₇	0.673 ₇	0.672 ₁	0.671 ₄	0.672 ₄
Average of C and T	—	0.673 ₆	0.673 ₆	0.673 ₅	0.672 ₇	0.672 ₁
Specific Gravity, ρ (density)	6.995 inferred from XRD	6.995 inferred from XRD	not known	6.848 ₃	6.943 ₄	6.878 ₈
Remarks	Pure cassiterite sample from Johnson, Matthey & Co. Ltd. No impurities greater than trace. Data obtained from File.	ρ assumed from JCPDS File. Data computed using all lines given in File. See Text.	Pure cassiterite powder prepared by BDH Ltd. Impurities: 500 ppm Pb 20 ppm As Computed using 25 lines.	Data computed using 20 lines. See Text for impurities.	Data computed using 20 lines. See Text.	Data computed using 20 lines. See Text.

TABLE 5 (contd.)

Field Sample Label & Locality	6	7	8	9	10	11	12
Sin Patana SPS, Phuket, Thailand	Whitish yellow brown. Translucent	Bluish metallic black. Opaque	Metallic bluish black. Opaque	Bluish metallic black. Opaque	Yellowish brown. Translucent	Colourless to pale yellowish brown. Transparent	Dark brown-black. Opaque
Ban Guan, SPBG, Phuket, Thailand							
Foong Seong, SKFS, Semiling, Kedah							
Perlombongan Kedah, SKLK, Semiling, Kedah							
Soon Hin-Choy Hin, SKLSHCH, Kuala Lumpur							
aÅ	4.740 ₈	4.736 ₀	4.737 ₀	4.738 ₇	4.738 ₈	4.742 ₅	4.737 ₀
bÅ	4.736 ₂	4.734 ₈	4.724 ₅	4.742 ₀	4.751 ₀	4.745 ₀	4.746 ₃
Average of a & bÅ	4.738 ₀	4.735 ₀	4.730 ₀	4.740 ₀	4.744 ₄	4.743 ₃	4.742 ₀
Assume tetragonal symmetry. See Text.							
cÅ	3.187 ₈	3.185 ₄	3.182 ₀	3.185 ₅	3.187 ₅	3.188 ₁	3.188 ₀
A = a/b	1.001 ₈	0.998 ₄	1.003 ₈	0.999 ₁	0.997 ₂	0.999 ₂	0.998 ₀
C = c/b	0.673 ₂	0.672 ₅	0.674 ₀	0.672 ₀	0.671 ₇	0.672 ₀	0.672 ₇
T = c/a	0.672 ₃	0.673 ₀	0.672 ₈	0.672 ₂	0.673 ₆	0.672 ₄	0.673 ₀
Average of C and T	0.673 ₀	0.672 ₁	0.673 ₇	0.672 ₀	0.672 ₇	0.672 ₂	0.672 ₃
Specific Gravity, ρ (density)	7.114 ₄	6.864 ₈	6.905 ₀	6.896 ₀	7.123 ₅	7.030 ₂	6.892 ₀
Remarks	Data computed using 20 lines. See Text.	Data computed using 20 lines. See Text.	Data computed using 14 lines. See Text. Rhombic distorted sphene shaped cass.	Data computed using 20 lines. See Text.	Data computed using 20 lines. See Text.	Data computed using 14 lines. See Text.	Data computed using 14 lines. See Text.

NB: Lattice constant values given to 3rd decimal place with the 4th decimal places written as subscript. Density measurements obtained using Berman precision density balance and XR Diffractograms are internally calibrated with pure Si standard.

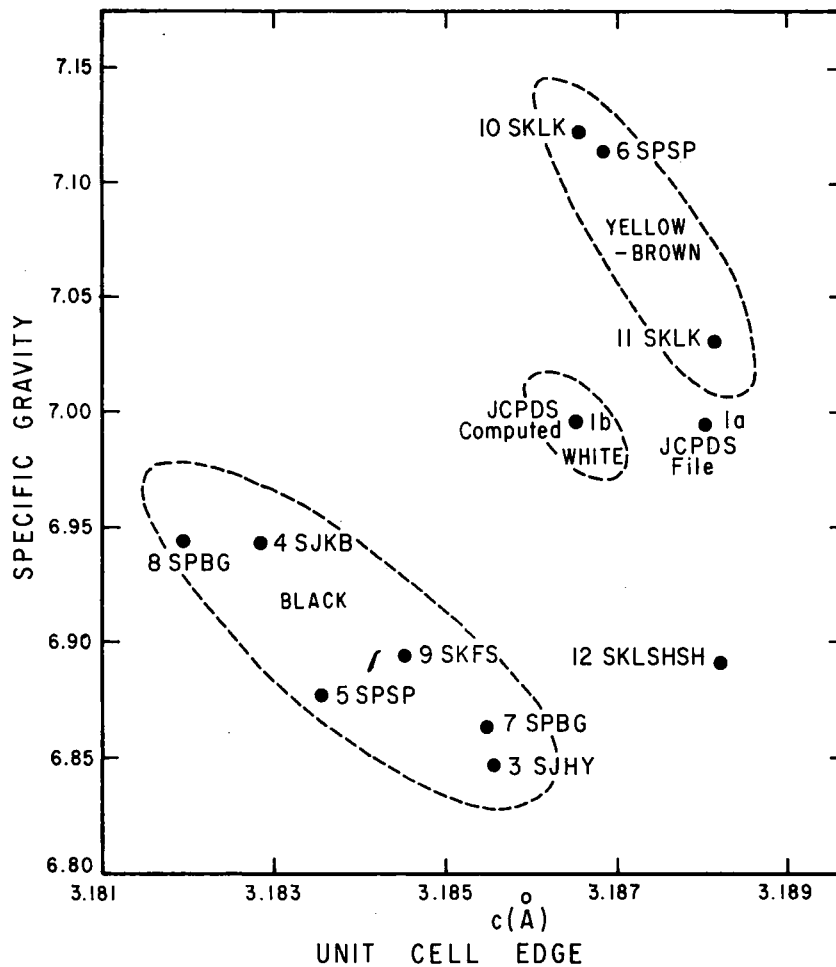


Fig. 13. Relationships between specific gravity and unit cell edge c for cassiterites from Bakri, Kathu and Gunung Jerai areas. The fields delineate the various colours of the cassiterites studied. Samples are numbered according to Tables 4 and 5.

variations of cassiterites properties which may in fact delineate the extreme extents of variation of these properties for most of the naturally occurring cassiterites. It is apparent from most of the figures, that the pure cassiterite always lies more or less intermediate between the 2 fields. The location of the 2 fields do not necessary delineate the region of pegmatitic cassiterites only. It must be viewed that the majority of pegmatite cassiterites probably have the properties which will plot them in and around these 2 fields but cassiterites of other genetic origin may occur over the whole range.

From Figs. 13 and 15, it is obvious that the black cassiterites have shorter c -axis parameters than the paler coloured varieties. This may explain the generally more abundant bipyramidal or short stumpy prismatic cassiterite crystals found in the 3

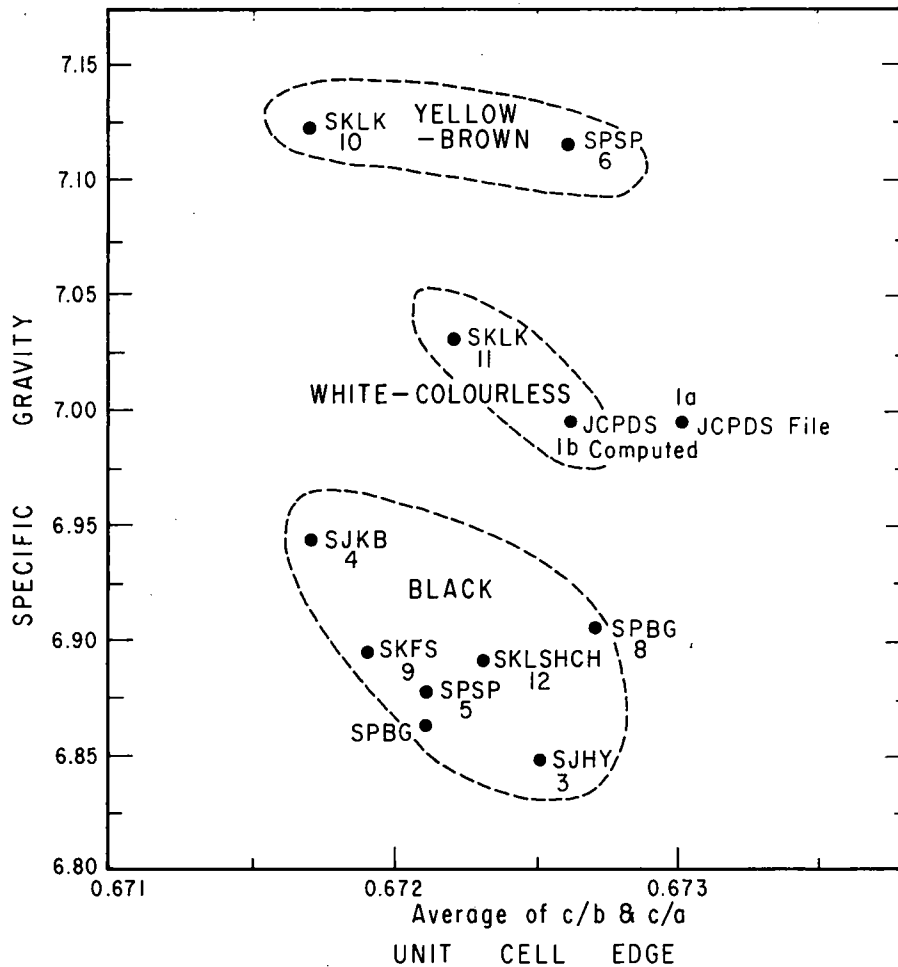


Fig. 14. Relationships between specific gravity and average unit cell edge ratio of c/b and c/a for cassiterites from Bakri, Kathu and Gunung Jerai areas. The fields delineate the various colours of the cassiterites studied. Samples are numbered according to Tables 4 and 5.

pegmatite areas. Fig. 13 also shows that there is a tendency for the c -axis to be longer with increasing density. Similarly from Fig. 14, it is evident that the a or b axis also increases from black to pale coloured cassiterites. However, the axial ratios c/a or c/b appear to be constant, irrespective of colour or origin of the cassiterite. This may mean that the axial parameter ratios are maintained in all cassiterites.

A noteworthy feature observed in the unit cell parameter of cassiterites is that the values of the a and b axis parameters are not equal to each other. More often, the b -axis appears to be slightly larger than the a -axis for pale coloured, yellowish or brownish cassiterites while the converse appears to be the case for black cassiterites (Fig. 17).

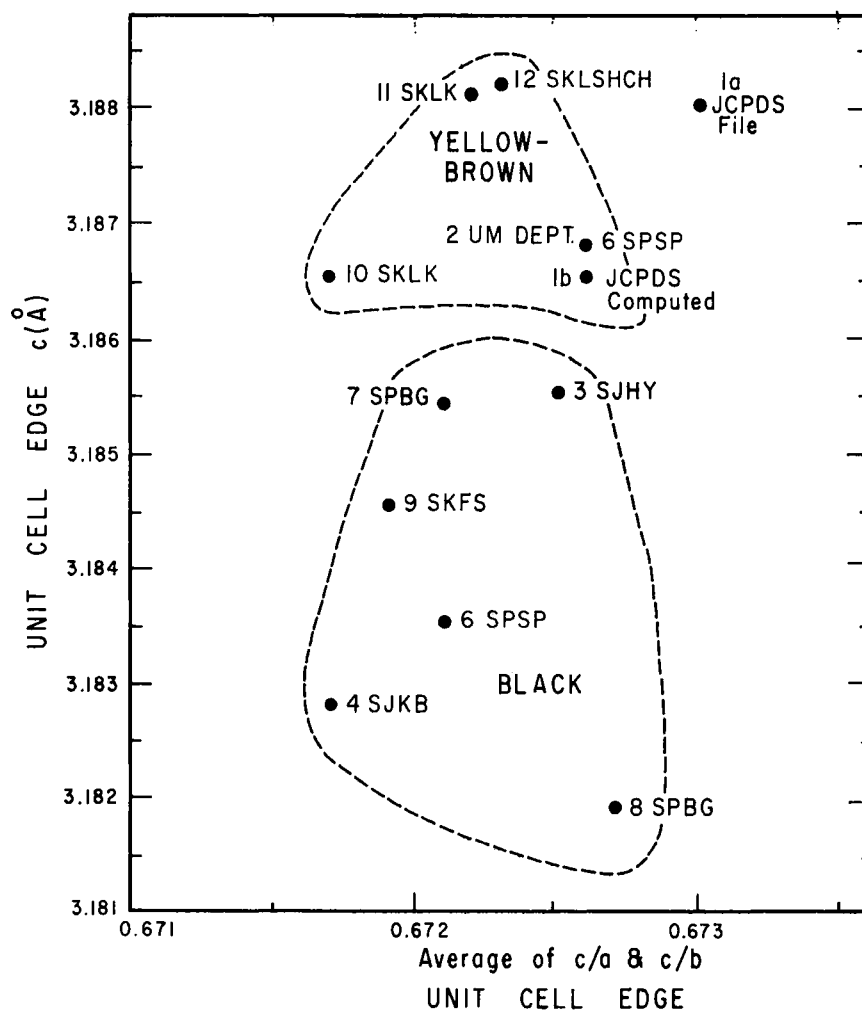


Fig. 15. Relationships of unit cell edge c against the average unit cell edge ratio of c/a and c/b for cassiterites from Bakri, Kathu and Gunung Jerai areas. The fields delineate the various colours of the cassiterites studied. Samples are numbered according to Tables 4 and 5.

However this seems to be not consistent. The greatest variation between these 2 axes belongs to the rhombic shaped cassiterites from the lepidolite pegmatites of Phuket, which shows a 0.014 Å difference between the longer a axis and shorter b axis. This is very suggestive of an orthorhombic crystal symmetry rather than tetragonal symmetry.

The author would like to point out that these findings do not seem to agree well with those of Flinter (1960) (quoted by Bradford, 1970) who obtained greater than normal d -spacing values for dichroic cassiterites from the Gunung Jerai area and that

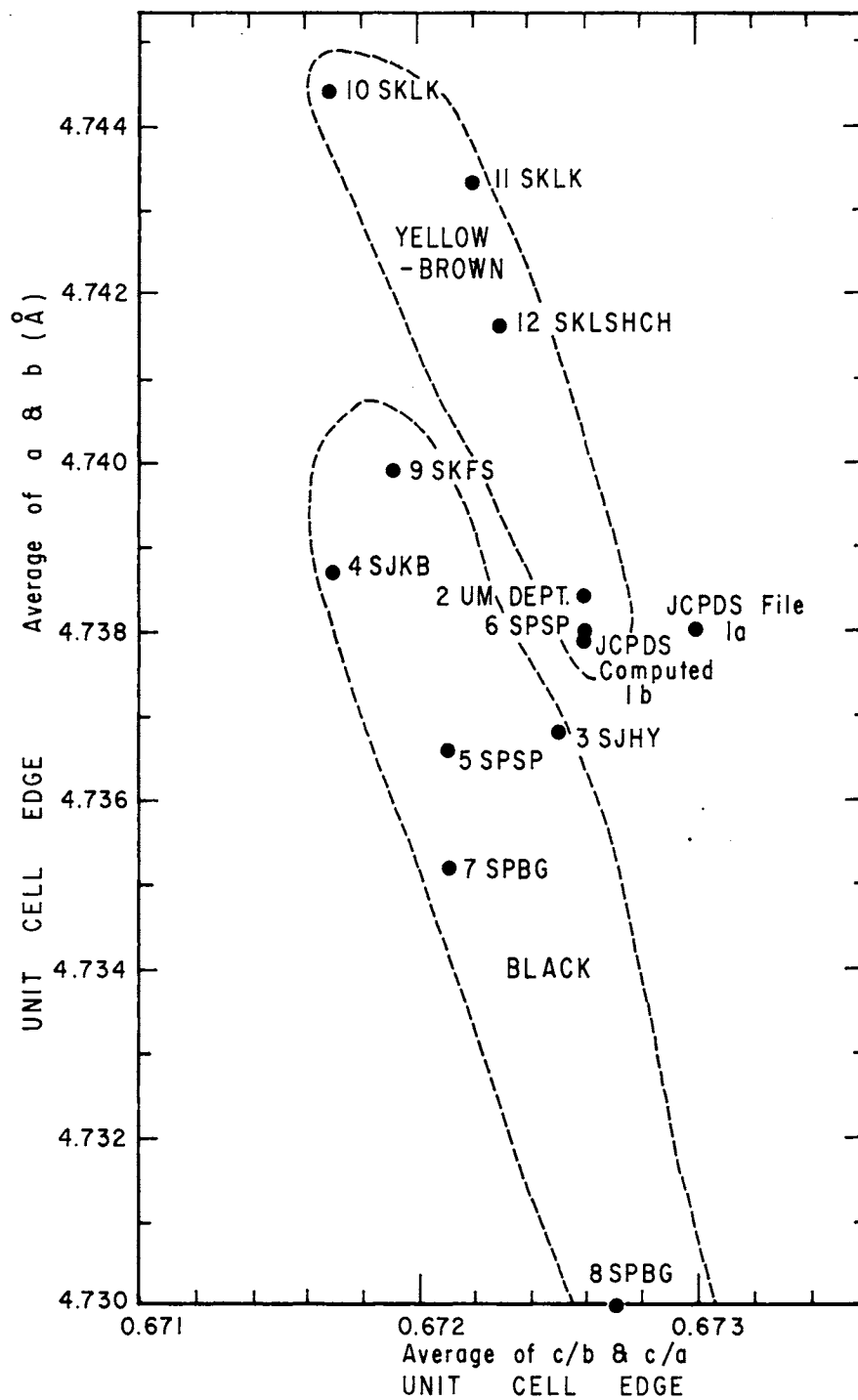


Fig. 16. Relationships of unit cell edge a and b against the average unit cell edge ratio of c/b and c/a for cassiterites from Bakri, Kathu and Gunung Jerai areas. The fields delineate the various colours of the cassiterites studied. Samples are numbered according to Tables 4 and 5.

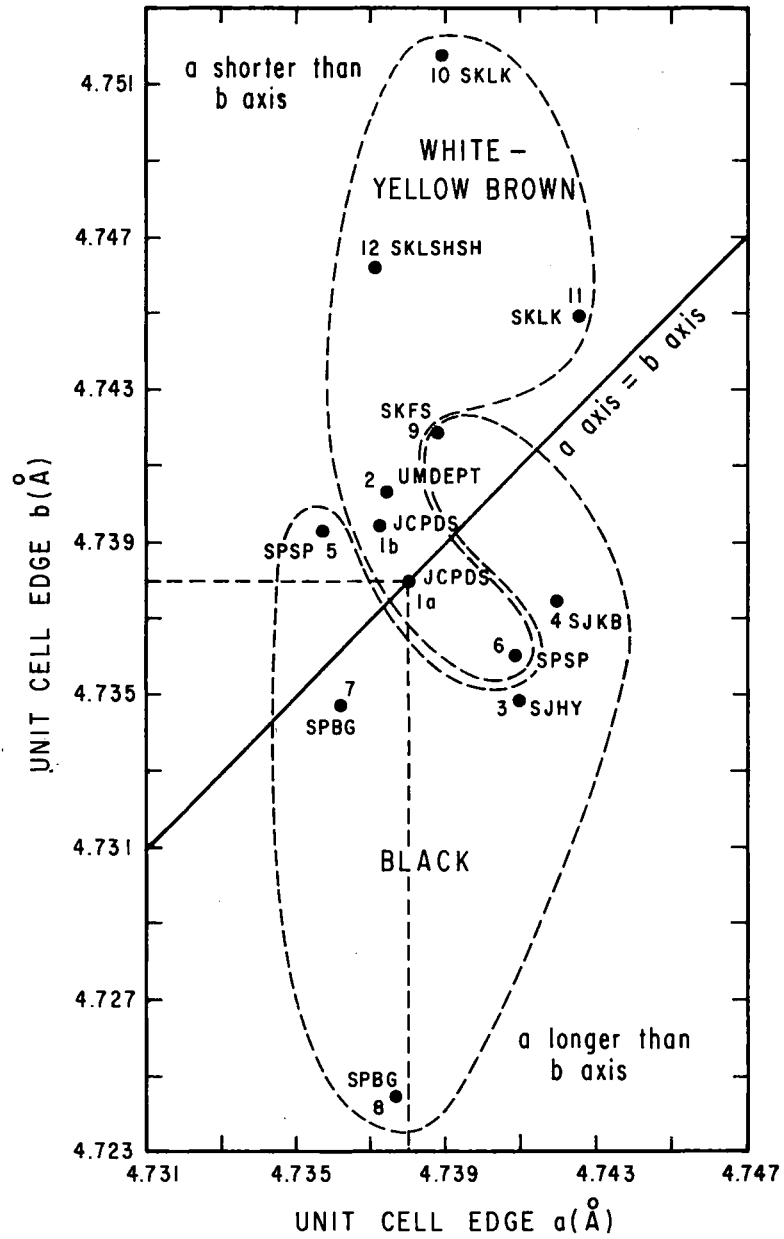


Fig. 17. Relationships between the a and b unit cell edges for cassiterites from Bakri, Kathu and Gunung Jerai areas. Samples are numbered according to Tables 4 and 5.

of Santokh Singh and Bean (1972) who stated that there is no appreciable effect of Ta and Nb on the cell parameters of cassiterite.

COMPOSITION OF CASSITERITES

In order to elucidate the chemistry of these black cassiterites as well as the yellowish brown cassiterites from the 3 areas, chemical analysis by atomic absorption spectrophotometry was carried out. A dark brown sample from the Kuala Lumpur tin field was also analysed. Selected samples were checked for impurities before they are crushed and grounded into a very fine powder for analysis. Concentrations of silicon, aluminium, potassium, tin, tantalum, manganese, iron, cobalt and titanium were determined and the values are shown in Table 6. However niobium concentrations was not determined as a niobium lamp was unavailable.

There appears to be a rather wide range of concentrations for each of the elements determined. Tin concentrations generally are correlatable with the colours of the cassiterites. Pale coloured and colourless varieties are definitely purer in that they contain much higher concentrations of tin and much lower concentrations of other elements compared to the black varieties. The cassiterite from the Kuala Lumpur tin field is shown to be very different chemically from the cassiterites from the 3 pegmatite areas. While titanium and tantalum are found in all the samples from the pegmatite areas, these 2 elements are evidently lacking or absent in the particular cassiterite sample from the Kuala Lumpur area which is of known contact metasomatic origin. It is also noticed that it has a very much higher FeO/MnO ratio which indicates that iron is an element much more closely associated with these cassiterites than those from pegmatite areas. Similarly the high $\text{SnO}_2/\text{Ta}_2\text{O}_5$ ratio reflects the low concentrations of Ta_2O_5 present in the Kuala Lumpur cassiterite.

Cassiterites from Phuket, especially, seem to be very much richer in manganese than iron, shown by the very low FeO/MnO ratios. The anomalously high FeO and MnO values obtained for some samples may be due to fine Fe or Mn oxide veins within the cassiterite grains. This had been proved to be present by the electron microprobe. Similarly, the anomalously high values of tantalum obtained for cassiterites from Bakri may not reflect the true concentration of tantalum incorporated within the cassiterite lattice. The true concentrations are probably slightly lower. The tantalum may be due to exsolved bodies of columbo-tantalite (Figs. 8 and 9) within the cassiterite grains which are often found in the cassiterites from the 3 areas, but it is also likely that they are due to fine discrete columbite-tantalite grains in the cassiterite samples, as complete separation is difficult due to the fine to medium grained nature of the samples and the similarity of the 2 minerals in hand specimens. The below 0.4 ampere fraction of the electromagnetic separated ore using the standard 25° slope and 17° tilt, was used for the analysis. The presence of discrete columbite-tantalite grains may explain also the correspondingly higher values of MnO and FeO values for these samples.

Electron microprobe qualitative analysis (sensitivity ~ 0.5 to 1%) revealed that many of the cassiterites do not contain much impurities besides iron, silicon and sometimes manganese (Fig. 18). Tantalum is detected occasionally in small amounts

TABLE 6
CHEMICAL ANALYSIS OF CASSITERITES FROM BAKRI, G. JERAI, PHUKET AND KUALA LUMPUR AREAS.

Sample & Location	SKLK, Semiling, Kedah	SKFS, Semiling, Kedah	SJKB, Bakri	SJHY, Bakri	PKBG, Phuket	PKBG, Phuket	SKLHCH, Kuala Lumpur	SKLK, Semiling, Kedah	SKLK, Semiling, Kedah
	black	black	black	black	black	black rhombic shaped	dark brown	yellowish brown	colourless transparent
Colour	black	black	black	black	black	black rhombic shaped	dark brown	yellowish brown	colourless transparent
Oxide %									
SiO ₂	0.68	4.55	1.01	0.64	1.85	0.78	0.46	0.46	0.31
Al ₂ O ₃	0.08	0.73	0.38	0.65	1.40	0.26	0.18	0.06	0.01
K ₂ O	0.01	0.01	0.02	0.01	0.05	0.03	0.03	0.01	0.01
SnO ₂	94.03	79.95	69.91	68.73	88.21	87.90	79.10	94.46	96.60
Ta ₂ O ₅	2.45	2.15	10.14	5.43	2.87	5.08	0.15	1.47	1.51
MnO	0.03	2.28	6.53	18.23	2.66	1.61	0.02	0.02	0.02
*FeO	1.33	7.51	9.17	3.38	0.73	2.01	17.50	1.79	0.62
CaO	0.004	0.002	0.003	0.011	0.025	0.012	0.004	0.010	0.011
TiO ₂	0.23	1.62	0.97	1.50	1.02	0.88	trace	0.70	0.16
Total	98.84	98.80	98.13	98.58	98.82	98.50	97.44	98.98	99.25
**Fe ₂ O ₃	1.48	8.35	10.20	3.75	0.81	2.23	19.45	1.99	0.69
SnO ₂	38.38	37.19	6.89	12.66	30.74	17.30	527.33	64.26	63.97
Ta ₂ O ₅									
FeO	44.33	3.29	1.41	0.19	0.27	1.25	875.00	89.50	31.00
MnO									

* Total Fe calculated as FeO

** Total Fe calculated as Fe₂O₃

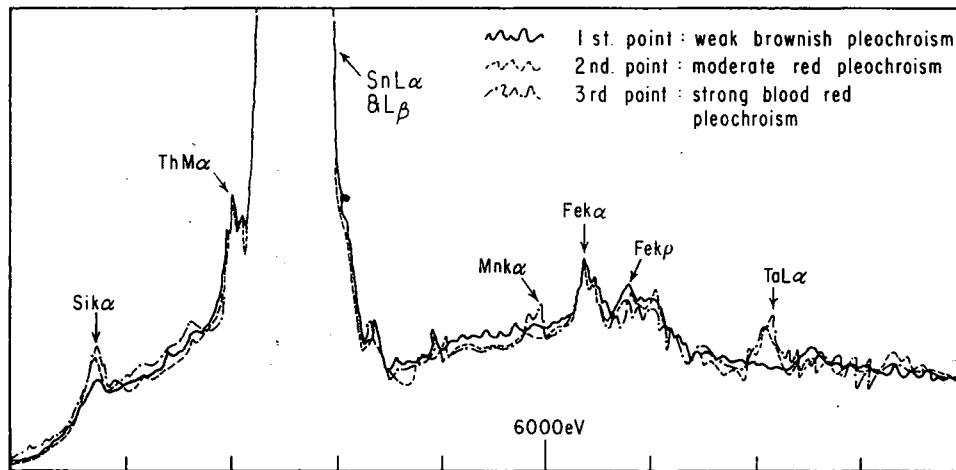


Fig. 18. SEM Energy Dispersion Spectrums of a 3-point traverse of a grain of cassiterite from the Ban Guan Tin Mine, Kathu, Phuket showing variations in the Si, Th, Mn, Fe, and Ta contents. Internal spacing is 1 mm.

which do not explain the rather high Ta_2O_5 concentrations obtained by wet analysis. Thus a proportion of the tantalum may not be incorporated into the cassiterite lattice but rather exists as exsolution bodies. Niobium was never detected, reflecting its low concentrations in cassiterite lattices. The paler cassiterites from the pegmatite area appear to have double the SnO_2/Ta_2O_5 ratio of the black cassiterites from the same areas.

The low concentrations of Al and especially K show the low competitive rates of substitution achieved by these elements compared to Ta, Ti, Mn, Fe and even Si.

ORIGIN OF THE CASSITERITES

Though the provenance of these cassiterites seem elusive, all indications tend to suggest that the cassiterites found in the Bakri and Gunung Jerai area are related to the pegmatite phase or phases immediately subsequent to it like the pneumatolytic and high temperature hydrothermal phases. There is little or no doubt that the cassiterites from the Kathu Valley are of pegmatitic origin. Strong indications include:

- (a) The persistently black cassiterites encountered in these 3 regions. Dr. P. Aranyakanon (pers. comm) noted that there is a 90% level of confidence that all black cassiterites in Thailand are pegmatitic in origin, though the converse does not hold.
- (b) The presence of paramagnetic and ferromagnetic cassiterites from the 3 areas. Paramagnetic cassiterites especially are quite typical of pegmatitic cassiterites.
- (c) The similar mineral assemblage of cassiterites, columbite-tantalite and monazite which have been documented to be a pegmatite suite.

- (d) The substantial substitution of Sn^{4+} by Ta^{5+} and Nb^{5+} also link the cassiterite with the columbite-tantalite found in the same areas as columbite and tantalite are pegmatite minerals.
- (e) The typical squat bipyramidal crystal habit and form of cassiterites believed to be from pegmatites (Varlamoff, 1945; Hosking, 1972) were found in these areas.

In summary, the pegmatite origin of cassiterites from Bakri and Gunung Jerai cannot be proved directly but studies of their physical, chemical and optical properties reveal that cassiterites of these 2 areas have so much in common with the pegmatite cassiterites from the Kathu area. Furthermore, the nature of the cassiterites from Bakri and Gunung Jerai areas are consistent with a pegmatite origin and it is tempting to conclude that they are derived from pegmatite. Whether they are genetically related to the present outcrops of pegmatites observed in the field in the Bakri and Gunung Jerai areas, however is uncertain.

SOME FORMS OF CASSITERITES FROM LEPIDOLITE PEGMATITES, PHUKET

A brief petrographic discussion of the rhombic shaped cassiterite and the botryoidal cassiterite from these pegmatites are made here.

Rhombic shaped cassiterites

In order to understand better the crystallographic orientations of these unique cassiterites, three perpendicular orientated polished thin sections were made of the same grain, each parallel to one of the crystal axis.

The section perpendicular to c-axis when viewed under the petrographical microscope reveal intense blood-red body colour. Pleochroism is strong ranging from blood red to dark mallow red brown. Maximum pleochroism occurs when N-S polariser is parallel to the b-axis which represents the slower of the 2 rays in this section. A series of curved colour zones were observed which are suggestive that the crystal may be twinned with the twin plane more or less parallel to the a-axis which represents the longest dimension of the section. The comparatively lighter zones have moderate pleochroism from light orange brown to honey brown orange. Interference figures obtained for this section consists of a single isogyre which when the stage is rotated some 20° , moves out of the field of view. It appears to be an off-centered optic axis figure. The sign is positive.

The section perpendicular to a-axis reveals a series of parallel colour zones parallel to the c-axis and indicate that the pattern of zoning is parallel to the c-axis. This section shows the most intense pleochroism with maximum pleochroism occurring when c-axis is making an angle of about 51° with N-S polarizer. Interference figure reveals a broad cross which disappears from view when stage is rotated about 8° . This probably represents the optic normal section.

The section perpendicular to the b-axis do not show proper zoning but generally is parallel to c-axis. Pleochroism is the least marked for this section. Maximum

pleochroism is generally about parallel to the c-axis. Interference figure similarly consists of a single isogyre which swings across the field of view. It probably represents an off-centered BXa section.

Interpretation of these observations are complicated by the probable presence of simple twinnings. However, it may be stated that the cassiterite is biaxial positive with a moderate 2V. The cassiterite is probably of the orthorhombic crystal symmetry.

Botryoidal cassiterite

In hand specimens, such cassiterites are black but are massive botryoidal in habit.

Petrographic studies reveal that the botryoidal external form is composed of a series of consecutive rounded zones which parallel the outline of the grain. These zones are not constituted by fine individual needle cassiterite crystals or marked growth layers which are characteristic of acicular and wood tin. Instead, the zones are massive and expressed in the form of consecutive colour zones. The whole grain is optically continuous.

From this study, it is apparent that these cassiterites are not truly wood tin though Aranyakanon (1961) had reported their presence in Thailand. Hosking (1974) came to a same conclusion for similar looking cassiterites from Thailand.

CONCLUSIONS

There is strong evidence that the cassiterites from the Gunung Jerai and Bakri areas appear to be of pegmatite origin even though no *in situ* cassiterite were found in the present outcrops of pegmatites. It also revealed and confirmed the presence of relatively more abundant bipyramidal cassiterite and other related forms which are believed to be of pegmatite origin. If Varlamoff's model (1945) is indeed true, it would suggest a rather high temperature and pressure regime of formation of these cassiterites, probably above 500 to 600°C. This fits rather well with the model of pegmatite intrusion in the Gunung Jerai area and possibly also in the Bakri area.

The absence of cassiterite in the present pegmatites may be due to the localising of cassiterite within certain pegmatites owing to the inhomogenous distribution of minerals in pegmatites and these had been eroded and concentrated to form localised placer deposits. It may also be due to a different phase of pegmatite intrusion other than those exposed presently. Most probable is that the development of cassiterites in pegmatites show vertical zonation. The cassiterites may be concentrated higher up, away from the main pegmatite bodies in pneumatolytic or very high temperature hydrothermal veins which are directly resulted from the pegmatite phase. These ore veins would probably be directly linked to the pegmatites. Present level of exposed pegmatites may be below the cassiterite bearing zone which had already been weathered away.

A rather good correlation was also established here between the various optical, physical, crystallographic and chemical properties of pegmatitic cassiterites and this may even be generalized to include all naturally occurring cassiterites.

Much more studies will have to be done on the rhombic shaped cassiterites from lepidolite pegmatites in Phuket. However it has been established here that their optical and physical parameters are not much more different from the rest of the pegmatite black cassiterites. Only their crystallographic parameters are abnormal or different from the rest of the commonly encountered cassiterites. All evidence suggest that they very likely belong to the orthorhombic crystal system. Chemically, they seem to contain a much higher concentration of tantalum. It is probable that its crystallography is dependent more on pressures and temperatures of formation rather than composition, though quite invariably it is also a factor which can modify the final shape of the grain.

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REFERENCE

- AARONSON, H.I., LORIMER, G.W., CHAMPNESS, P.E. and SPOONER, E.C.T., 1974. On differences between phase transformations (exsolution) in metals and silicates. *Chem. Geol.*, 14, 75-80.
- ARANYAKANON, P., 1961. The cassiterite deposit at Haad Som Pan, Ramong Province, Thailand. *Rep. Invest. R. Dept. Mines Thailand No. 4.*
- ARNANDOV, V. and BOYADZHIEVA, R., 1970. Distribution of niobium and tantalum in the pegmatites from the seven lakes area, Rila Mtn. *Rev. Bulgar Geol. Soc.*, 31, 75-82.
- BETEKHTIN, A., 1964. *A course of Mineralogy*. Moscow, Peace Publishers.
- BIGNELL, J.D. and SNELLING, N.J., 1977. *Geochronology of Malayan granites*. No. 47, Institute of Geological Sciences.
- BRADFORD, E.F., 1958. The occurrence of tin and tungsten in Malaya. *Proc. of the Ninth Pacific Science Congress*, 12, 378-398.
- BRADFORD, E.F., 1970. *Geology and mineral resources of the Gunung Jerai area, Kedah*. Geol. Survey Malaysia, Dist. Mem. 13.
- BURTON, C.K., 1969. The geological environment of tin mineralization in the Malay-Thai Peninsula. *Proc. 2nd Tech. Tin Conf.* Int. Tin Council, Bangkok.
- CHAN, S.H., 1976. A procedure for computing the lattice constants of some crystal system. *Warta Geologi*, 2, 15-26.
- CHOY, K.W., 1977. Tin distribution over granitic rocks in the Kinta Hills area, Perak, Peninsular Malaysia. *Bull. Geol. Soc. of Malaysia*, 9, 233-252.
- DAWSON, K.R., 1974. Niobium (columbium) and tantalum in Canada. *Econ. Geol. Rept., Geol. Surv. Canada*, 29, 157p.
- DEER, W.A., HOWIE, R.A., and ZUSSMAN, J., 1962. *Rock-forming minerals*, vols. 1, 3, 4 & 5. Longmans.
- DERRY, D.R., 1931. The genetic relationships of pegmatites, aplites and tin veins. *Geol. Mag.*, 68, 454-475.
- FOORD, E.E. and MILLS, B.A., 1978. Biaxiality in 'isometric' and 'dimetric' crystals. *Am. Min.*, 63, 316-325.
- GARSON, M.S., BRADSHAW, N. and RATTAWONG, S., 1969. Lepidolite pegmatites in the Phangnga area of Peninsular Thailand. *Rep. 2nd Tech. Tin Conf.* Int. Tin Council, Bangkok.
- GARSON, M.S. and OTHERS, 1975. *The geology of the tin belt in Peninsular Thailand around Phuket, Phangnga and Takuapa*. Overseas Mem. Inst. Geol. Sci. No. 1, 112 p.
- GINZBURG, A.I., 1960. Specific geochemical features of pegmatitic process. *Rep. 21st Sess. Int. Geol. Congr.*, Copenhagen, pt. 17, 111-121.
- GOLDSCHMIDT, V.M., 1954. *Geochemistry*. Oxford, Clarendon Press, 500-505.

- GOITMAN, J.D., 1938. On the properties of cassiterites in connection with conditions of its formation. *Bull. Soc. Nat. Moscow*, 46, 130.
- HESP, W.R., 1974. Geochemical features of Sn-Ta-Nb mineralization associated with granitic rocks in South-Eastern Australia. In Stempok, M. (ed.), *Mineralization associated with acid magmatism*, IGCP Symp., v. 1, 170-180.
- HOSKING, K.F.G., 1975. The search for tin. *Mining Magazine*, v. 113.
- HOSKING, K.F.G., 1973. Primary mineral deposits. In: Gobbett, J.D. & Hutchison, C.S. (eds.), *Geology of Malay Peninsula*. Wiley Interscience, 343-346, 351-374.
- HOSKING, K.F.G., 1974. *The search for tin deposits*. International Tin Council, London.
- HOSKING, K.F.G., 1977. Known relationships between the 'hard rock tin deposits' and the granites of Southeast Asia. *Bull. Geol. Soc. Malaysia*, 9, 141-158.
- HOSKING, K.F.G., 1979. Tin distribution patterns. *Bull. Geol. Soc. Malaysia*, 11, 1-70.
- HUMMEL, C.L. and PHAWANDON, P., 1967. Geology and mineral resources of the Phuket mining district, South Thailand. *Rep. Invest. R. Thai Rep. Miner. Resource.*, no. 5.
- HUTCHISON, C.S., 1975. Granites of the Southeast Asian Tin Province. *Abstract of paper at the ICCP Circum Pacific Plutonism Project 5th Meeting*, Kuala Lumpur, Malaysia.
- JAHNS, R.H., 1933. Distribution of rare elements within granites and granitic pegmatites (abstract). *Bull. Geol. Soc. Amer.*, 64, p.1440.
- JAHNS, R.H., 1955. The genesis of pegmatites. IV. The role of replacement processes. V. Conditions and processes of crystal growth.
- LYE, Y.H., 1978. A note on 'A procedure for computing the lattice constants of some crystal systems by Chan (1976)'. *Warta Geologi*, 4, 149-152.
- LYE, Y.H., 1979. *Petrology, mineralogy and geochemistry studies of pegmatites from the Gunung Jerai, Kedah, Bakri, Johore (Peninsular Malaysia) and Kathu Areas, Phuket (Thailand)*. Unpubl. B.Sc. (Hons) thesis, Univ. of Malaya, Kuala Lumpur.
- MATHEIS, G., 1978. Geochemical exploration around the pegmatitic Sn-Nb-Ta mineralization of SW-Nigeria. *Bull. Geol. Soc. Malaysia*, 11, 333-351.
- NASSAU, K., 1978. The origins of color in minerals. *Am. Min.*, 63, 219-229.
- NUTALAYA, O., CAMPBELL, K.V., MACDONALD, A.S., ARANYAKANON, P. and SUTHAKORN, P., 1979. Review of the geology of Thai Tin fields in Geology of Tin Deposits. *Bull. Geol. Soc. Malaysia*, 11, 137-160.
- PARKER, R.L. and FLEISCHER, M., 1968. *Geochemistry of niobium and tantalum*. Geol. Survey Prof. Paper 612, Washington.
- RANKAMA, K., 1944. On the geochemistry of tantalum. *Finland Geol. Comm. Bull.*, no. 133, 78 p.
- RANKAMA, K., 1947. On the geochemistry of niobium. *Acad. Sci. Fennicae, Annales*, ser. A, III, no. 13, 57 p.
- RAO, A., 1972. *Geology and geochemistry aspects of the southern Gunung Jerai area, Kedah, West Malaysia*. Unpubl. B.Sc. (Hons), thesis, Univ. of Malaya, Kuala Lumpur.
- RAJAH, S.S. 1979. The Kinta Tinfield, Malaysia in Geology of Tin Deposits. *Bull. Geol. Soc. Malaysia*, 11, 111-136.
- SCRIVENOR, J.B., 1928. *The geology of Malayan Ore Deposits*. Macmillan & Co., Ltd., London.
- SINGH, D. SANTOKH and BEAN, J.H., 1967. A technical Conference of tin, vol. 2 pub. International Tin Council, London, 467-478.
- SITNIN, A.A., 1970. On the mobilization of Ta, Nb, Li, Rb, Cs, Be, Sn, W in the process of a high-temperature metasomatism of certain granitoids. In Poubu, Z., & Stempok, M. (eds), *Problems of Hydrothermal Ore Deposits*. Intern. Union Geol. Sci., Ser. A., no. 2, 348-354.
- SMITH, F.G., 1948. Transport and deposition of the non-sulphide vein minerals. III. Phase relations at the pegmatitic stage. *Econ. Geol.*, 43, 535-546.
- STANTON, R.I., 1972. *Ore Petrology*. New York, McGraw-Hill.
- TERPSTRA, P. and TODD, L.W., 1961. *Crystallometry*. Longmans.
- TOBI, A.C., 1956. A chart for measurement of optic axial angle. *Am. Min.*, 41, 516-518.
- VARLAMOFF, N., 1954. Transitions entre les aplites et les pegmatites dan les zones de contact des massifs granitiques des Concessions de Syietain a Kalima (Maniema, Congo Belge). *Soc. Geol. Belgique, Annales*, v. 77, B101-116.
- VARLAMOFF, N., 1972. Central and West African rare metal granitic pegmatites, related aplites, quartz veins and mineral deposits. *Mineral deposita*, 7, 202-216.
- YEE, K.C., 1969. *X-ray fluorescence analysis of tin ores and associated products*. M.Sc. thesis (Unpubl), Univ. of Malaya, Kuala Lumpur.