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H. G. RAGGATT, *Director.*

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BULLETIN No. 10  
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## PIEZO-ELECTRIC QUARTZ

BY

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Issued under the authority of the Honorable W. P. ASHLEY,  
Minister for Supply and Shipping.

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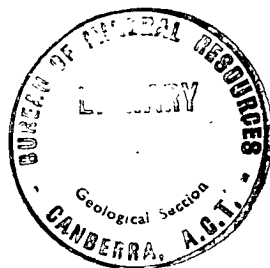
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- Bulletin No. 13—(Geological Series No. 4)—“Notes on the Occurrence of Piezo-Electric Quartz in Australia with Special Reference to the Kingsgate Field”. By M. D. Garretty. Roneoed and issued October, 1944. Printed December, 1947.
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- Bulletin No. 15—(Palaeontological Series No. 5)—“Foraminifera in the Permian Rocks of Australia”. By I. Crespin. Printed and issued September, 1947.

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Plate 1.—Types of Quartz Crystals.

## 1. INTRODUCTION.

In the early days of radio communication, transmitting and receiving sets were not selective, i.e., the transmitter emitted signals whose frequency was only approximately constant. The receiver needed only rough tuning to receive these signals. As the number of broadcast and other stations increased, the available frequency band for each had to be made smaller, and consequently any one transmitter was required to operate more accurately at its allotted frequency to avoid overlapping with adjacent frequencies used by other transmitters. Thus arose the need for an exact frequency control. Early attempts to do this used the vibrations of a tuning fork, but less clumsy and more accurate devices were evolved which made use of the piezo-electric properties of quartz.

Before the recent war the number of broadcast stations and other radio transmitters was relatively small and there was only a limited demand for quartz crystals, but during the war the demand for crystal as a component part of radio equipment, and for other military uses, increased enormously, for, to mention but two examples of its use, the transmitters in all aircraft and tank radios have their frequencies controlled by plates cut from quartz crystals.

Piezo-electric quartz is also used, in a somewhat similar manner but to a lesser extent, in telegraphy and telephony, and in the manufacture of some types of gramophone pick-ups and special radio receivers. The frequency of the current supplied by power houses is often governed by piezo-electric quartz plates.

Broadly speaking, any physical device requiring electrical potential variations of exactly controlled frequency or requiring the precise measurement of small time intervals may use piezo-electric quartz, but the bulk of such quartz is used in radio transmitters.

Brazil and Madagascar are the principal sources of the world supply of quartz crystal. During the early years of the war it appeared that the greatly increased demand for military purposes could not be met from these countries; consequently, it became urgently necessary to look for supplies in Australia and elsewhere. By October, 1943, the position had changed considerably and it appeared possible that Australian requirements of raw crystal could be met from the Brazilian deposits.

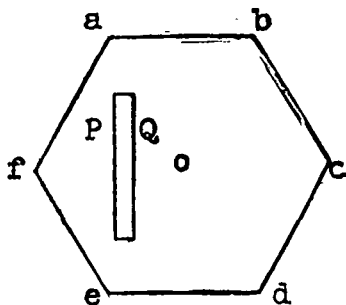
## 2. DEFINITION OF THE TERM "PIEZO-ELECTRIC".

A substance is said to be "piezo-electric" if, on its shape being changed by the application of mechanical pressure, electrical potential differences are created between parts of its surface. Expressed in other words, if a slab of the material is held between metal plates which are connected by a wire, a current will flow in the wire when the "sandwich" is compressed. The current flows only during the change in shape of the slab and ceases when the pressure becomes constant. On release of the pressure, a current flows in the wire in the opposite direction during the return of the slab to its original shape.

The reverse is also true. If a potential difference be created on opposite sides of the slab a small change in its shape results.

The piezo-electric phenomenon is more precisely described by Campbell and Childs (1935) as follows:—

"A section of a quartz crystal cut normal to the optic axis is shown in (the accompanying figure). Lines joining opposite corners, *ad*, *be*, and *cf*, are electric axes; if pressure is applied in these directions the corners become electrically charged, one positively and the other negatively. This property is shared by many other optically active crystals, but quartz alone has the properties required for a stable resonator. A plate cut from the crystal in the manner shown in the accompanying figure would have faces *P* and *Q* charged oppositely by pressure applied in the direction of, or normal to, *cf*. There is no piezo-electric effect parallel to the optic axis.



The inverse effect is also exhibited, namely, if the plate or bar of quartz is supported in an electrostatic field parallel to *cf*, the crystal contracts in this direction and expands in the perpendicular direction, or vice versa. An alternating field thus forces the crystal into vibration, and resonance may be produced between the field frequency and that of one of the modes of vibration of the crystal".

### 3. PIEZO-ELECTRIC SUBSTANCES.

The only common substances which are piezo-electric are crystals of quartz, tourmaline and Rochelle salt (potassium sodium tartrate). Crystals of the latter can be prepared artificially, but as they are neither as elastic nor as robust as quartz they are not suitable for radio use, although they are commonly employed in microphones and gramophone pick-ups. Tourmaline is satisfactory, but suitable crystals are even rarer than those of quartz. The few tourmaline crystals that are available are not made use of, for to do so would necessitate much alteration of the cutting technique which has been evolved for quartz.

### 4. USES OF PIEZO-ELECTRIC QUARTZ CRYSTALS.

Piezo-electric quartz is used primarily in operations which require the precise measurement of time intervals or the precise control of time frequencies.

When a small thin plate of quartz is held between metal plates and a pulsating current applied to the metal plates, the quartz plate will tend to vibrate with the frequency of the applied pulsating current. However, the quartz plate has a natural period of mechanical vibration which is determined by its shape, areal dimensions and thickness. If the dimensions of the plate be so determined that its natural period of mechanical vibration is the same as that of the applied current, the plate will vibrate in sympathy with the current pulsations. However, if the frequency of the current tends to change, the plate, whose rate of vibration is fixed, imposes a potential on the electrical system which acts similarly to the back e.m.f. of a synchronous a.c. motor, and maintains the applied potential variations at their original frequency.

The transmitting valves in a radio transmitter are connected with a quartz crystal plate fitted in a special mount, the mount and crystal being termed a "frequency control". The effect of this control is to maintain a constant rate of oscillation in the transmitting circuit and so to maintain a fixed frequency of the omitted signals.

The rate of vibration of a crystal is altered slightly by changes in temperature and humidity, and to avoid such change some crystals are mounted in enclosed holders which are kept at a constant temperature by a thermostatic control. Such mountings are commonly used only by broadcast stations.

Frequency controls are similarly used in telephony and telegraphy to enable several messages to be transmitted simultaneously by a single line. Each message is transmitted at a different frequency, the control being necessary to avoid overlapping of the various messages.

In microphones and gramophone pick-ups, a piezo-electric substance is subjected to the mechanical movement of the diaphragm or needle respectively. The consequent alteration in shape of the piezo-electric substance causes electrical potential differences to be created which are passed to the electrical system for amplification.

There are many other uses of piezo-electric quartz, such as the establishment of frequency standards, the control of television transmission, the measurement of detonation in aeroplane engines, the determination of depths of water by sonic sounding, the measurement of time intervals in direct reading seismographs and the control of extremely accurate clocks.

## 5. ORIGIN OF QUARTZ CRYSTALS.

During the final phases of cooling of acid igneous magmas, quartz is injected into the country rocks either with other minerals as quartz-pegmatite or by itself as vein-quartz. Sometimes quartz, in association with metallic and other minerals, is injected in the form of a pipe-shaped mass.

Where a vugh or cavity forms in the injected mass, quartz crystals may grow in it, usually more or less normal to the surface of the vugh. The factors controlling this growth are not known with certainty, but it may take place from silica solutions passing through the vugh; the size and perfection of the resultant crystals being affected by the temperature, rate of flow, purity and concentration of the silica solutions.

Quartz crystals are associated with quartz veins and pegmatite pipes which are off-shoots from granitic rock masses, and it follows that crystal deposits are most likely to be found in or near granitic rocks. In Australia such rocks, being old, have been exposed by prolonged erosion and therefore are now exposed at the surface. Consequently quartz veins and pegmatite pipes in this country are usually found either in or within a few miles of granitic rock outcrops.

It may be that quartz crystals are more commonly associated with somewhat glassy types of quartz rather than with the milky "buck" quartz such as is common in large reefs throughout Australia. As insufficient is known at present of Australian occurrences it is not possible to be definite on this point.

Attempts have been made to manufacture suitable artificial quartz crystals but, so far as is known, without success.

## 6. CRYSTALLINE FORM OF QUARTZ.

The great bulk of quartz is composed of an aggregate of small crystalline quartz grains which do not possess crystal faces. The development of separate crystals of quartz is comparatively rare. A normal quartz crystal (Figs. 1 and 2) is composed of a six-sided (hexagonal) prism, terminated at one or both ends by two sets of rhombohedral faces, each set consisting of three faces. Each alternate face belongs to one rhombohedral set, and in most crystals one set is larger, i.e., is more developed, than the other. The six faces are often loosely termed "pyramid" faces. Other small faces may be present near the junction of the prism and rhombohedral faces.

## 7. TWINNING OF QUARTZ CRYSTALS.

The component atoms of a quartz crystal must be arranged in a regular order for it to be of use for piezo-electric purposes. Irregularities in this arrangement are evidenced by several phenomena, the most important of which is "twinning".

Twinning is not easy to describe briefly. A twinned crystal is one in which one or more parts, regularly arranged, are in reverse positions with reference to the other part or parts. Crystals of many minerals may exhibit twinning in a more or less complicated manner. Unfortunately, quartz crystals are subject to several types of twinning, some of which are complex. The more important of these types may be explained as follows:—

(i) All quartz crystals are either "right-handed" or "left-handed", according to the atomic structure of the crystal. It is not possible in this paper to discuss these terms further, but the two types can be distinguished by a polariscope and, for piezo-electric use, it is immaterial whether a crystal is right- or left-handed provided it is one or the other and not a combination of both.

If twinning occurs so that the various sections of the twin are all right-handed, the twinning cannot be detected by optical means but can be detected by etching with hydrofluoric acid for several hours when the twinning can be seen by differences in the texture of the quartz. This type of twinning is known in the trade as "electrical twinning", but the term is somewhat misleading as all twinning affects the piezo-electric properties of quartz.

It is claimed that electrical twinning can be removed from a quartz plate by certain heat treatment, but it is not known that this treatment is used commonly.

(ii) The various parts of a twinned crystal may be alternately right- and left-handed, in which case the twinning can be detected optically. The trade use the term "optical twinning" for this type of twinning. Optical twinning is also shown up by etching and can be differentiated from electrical twinning by differences in the character of the boundaries of the twinned areas.

(iii) Small superficial areas only of a crystal may be twinned. Surface twinning of this type is usually so shallow that, although the piezo-electric properties of the twinned sections are affected, these sections can be avoided during cutting.

The following classification and description of quartz twinning, which is visible to the naked eye, is based on a report by the Post Office Engineering Department, Office of the Engineer-in-Chief (Radio Branch), General Post Office, London, but all the features described have been recognized in crystals of Australian origin:—

(i) *Composite Structure*.—In some crystals reversals of structure are so frequent that the resulting crystal, although of approximately regular shape, is practically a conglomeration of pieces. The presence of such twinning is shown by abrupt changes in level of the prism faces or by changes in texture of the faces (Fig. 3). Such crystals are useless for piezo-electric purposes.

(ii) *Deep Twinning*.—This is evidenced by one face showing two distinct types of characteristic markings, the two types being divided by a fairly well defined line. One or more of the following can usually be seen:—

- (a) An area of well defined striations separated from one of less clearly defined striations (Fig. 4).
- (b) One set of striations discontinuous with an adjacent set (Fig. 4).
- (c) A clear area separated from a dull area (Fig. 5).
- (d) Two separate areas of different roughness or texture (similar to Fig. 5).
- (e) One area characterized by naturally occurring etch-marks separated from an unetched area.

(iii) *Surface Twinning*.—Small areas on the outer part only of the crystal may be twinned. This type of twinning is indicated if on one or more faces (usually alternate faces) small areas have the characteristic features of the adjacent faces. These areas are distinguished from those corresponding to the other types of twinning by their shape, which tends to be regular and triangular in form. These shapes are generally isosceles triangles and may occur on both rhombohedral and prism faces. The apex of each triangle always points towards one end of the crystal (Fig. 6), those on the rhombohedral faces pointing towards the pointed end of the face. This type of twinning generally does not prevent the use of the crystal for piezo-electric purposes.

Certain deposits appear to be characterized by crystals showing one or only a few types of twinning, but the presence of some twinned crystals in a deposit does not indicate that all the crystals in it are twinned. For example, twinned crystals are not uncommon in the Brazilian deposits.

## 8. COMMON FLAWS IN QUARTZ CRYSTALS.

The following flaws, once seen, are easily recognizable in a hand specimen, but the written explanation of the terms is difficult. The flaws are best detected visually by painting the crystal with a liquid having approximately the same refractive index as quartz (e.g., methyl salicylate) and viewing a strong light through the crystal:—

(i) *Bubbles*.—Some holes or “bubbles” containing air, water, salt solutions or other substances are common in quartz crystals. Generally bubbles can be detected visually, and some cutters claim that the



presence of small bubbles which cannot be so detected can sometimes be inferred from the presence of small ridges which interrupt the smooth surface of a conchoidal fracture. It is not known why these ridges indicate the presence of bubbles.

(ii) *Veils*.—These flaws appear as wisp-like clouds in an otherwise clear piece of quartz. They are visible to the unaided eye with difficulty and only when viewed in a strong light. It is possible that the veils are composed of a very large number of minute bubbles.

(iii) *Ghosts*.—Some crystals are formed during more than one period of growth and from different parent solutions. The second or subsequent growths, although conformable with the first, may enclose surface irregularities or impurities of the former crystal, or the material deposited later may be of different composition from that deposited earlier. The form of the earlier crystal is then visible inside the later crystal and is referred to as a "ghost".

(iv) *Needles*.—Fine needle-like shapes are sometimes visible in quartz crystals. These may merely represent cavities but, more usually, are composed of fine crystals of rutile. Such crystals are commonly used for ornamental purposes and are termed "grass stones".

(v) *Inclusions*.—Small inclusions of other minerals, such as bismuth, rutile and hematite, are also found in quartz crystals. The inclusions may be quite irregular in shape or they may have crystal outlines.

(vi) *Irregular Striations*.—The presence of irregular or wavy striations, which are distinct from broken or discontinuous striations mentioned on page 7, Section (ii) (b), usually indicates that the crystal is unsuitable, but if the irregularities are only slight the crystal may be usable.

(vii) *"Tangled" Structure*.—The base of a crystal near its point of attachment to the wall of a vugh generally consists of quartz which is not optically continuous, but is "tangled" rather than twinned. That portion of the end is useless.

## 9. REQUIRED QUALITY AND SIZE OF QUARTZ CRYSTALS.

Quartz crystals suitable for piezo-electric use are subject to very stringent specifications, viz. :—

- (i) The narrowest width of the crystal, measured between two parallel prism faces, must be not less than  $1\frac{3}{4}$  inches.
- (ii) The length along the vertical axis must be not less than 1 inch. (Most crystals are flawed in some places and hence, to satisfy the requirements given in the following section (iii), it is usually found that to be suitable a crystal should be not less than  $2\frac{1}{2}$  inches long.)
- (iii) The crystal must contain a 1-inch cube of unflawed material.
- (iv) The quartz must be perfectly transparent, clear and glass-like and must be free from flaws such as cracks, bubbles, veils, ghosts, needles and foreign particles. Smokiness of the quartz, even if very dark, is not in itself a defect but renders the detection of other flaws more difficult.
- (v) The crystal must not show twinning, other than surface twinning, unless it is large enough for untwinned sections, which are themselves of the required size, to be cut from it.

Broken crystal fragments of water-worn crystals can be used provided they are otherwise satisfactory. Until recently such material could not be used because in the absence of certain crystal faces they could not be oriented. Recent research has devised methods of orienting without using crystal faces, hence material not showing such faces can now be used. Twinning is more difficult to detect visually in water-worn crystals and therefore suitable material cannot be readily recognized.

## 10. MINING METHODS.

Generally the only method of mining quartz crystals is to follow the vein or pipe in which they occur, in search of vughs containing crystals. This procedure usually necessitates the mining of a considerable amount of waste rock to obtain a relatively small amount of quartz crystal, and of the crystals so obtained it is probable that most of them can be rejected by preliminary visual examination at the mine. The uncertainty of finding crystal-bearing vughs during mining and the small amount of usable crystal that can be expected in proportion to the waste rock mined make the winning of crystals both difficult and costly.

Crystals are readily cracked and shattered by the use of explosives in mining, for which reason explosives should be used as little as possible. In some deposits the prospecting workings are better situated adjacent to the quartz body rather than in it. When found the crystals should not be trimmed by "cobbing" or hitting but should be despatched intact to the buyers who are able to cut away unwanted parts without damage to the crystal.

## 11. PREPARATION FOR USE.

Various trade terms are used throughout this report, and especially in this paragraph. To avoid confusion they are explained as follows:—

*Crystal*—The naturally occurring quartz crystal, either fully or partially formed, or even a broken piece.

*Slab*—A piece of quartz about 1 inch thick which is sawn from the crystal.

*Wafer*—A thin slice of quartz sawn from a slab.

*Blank*—A portion of a wafer which is trimmed to its correct areal size, ground or unground, but not lapped to its final thickness.

*Plate*—The blank after it has been lapped to its final thickness.

In cutting the crystal to obtain the final plates (which are made in various sizes according to the use to which they are to be put) the essentials are—

- (i) extreme accuracy of cutting so that the final dimensions of the plate will give it the exact frequency desired;
- (ii) correct orientation of the blank with respect to the crystallographic axes so that the piezo-electric effect will be a maximum and the effects of temperature changes will be a minimum, and
- (iii) repeated testing at successive stages to detect and discard material which is twinned, the twinning being more easily detected as the cutting proceeds and the size of the quartz piece becomes smaller.

There are various methods of cutting, but one common method is described as follows:—

- (i) The crystal is painted with methyl salicylate and examined before a powerful light to detect visible flaws.
- (ii) A slab, about 1 inch thick, is cut from the full length of the crystal so that it is parallel to the vertical and one horizontal crystallographic axis.
- (iii) Provided that twinning is not disclosed when this slab is etched in hydrofluoric acid, thin wafers are cut from it in a direction which bears a fixed angular relationship to the crystallographic axes. This relationship is pre-determined so as to give a maximum piezo-electric effect consistent with a minimum change in frequency consequent on changes in temperature. A wafer having the latter property is said to have a good "temperature-co-efficient".
- (iv) These wafers, which are about 40 thousandths (0.04) of an inch thick, are etched, and portions which do not show twinning and which are large enough for use, are cut off. Recently, portions have been used which show up to 20 per cent. of optical twinning, provided this twinning occurs only around the edges of the portion.  
However the presence of such twinning reduces the quality of the final plate; and hence for plates which have to meet the most stringent requirements no twinning can be tolerated.
- (v) These portions of the wafers are then trimmed to the correct areal shape and size, which vary according to the purpose for which they are to be used, but they are usually rectangular in shape and are from nine-sixteenths of a square inch to 1 square inch in area. They are then ground to within about four-thousandths (0.004) of an inch of the required final thickness and lapped by machine to a thickness which departs from the final thickness desired by an amount which corresponds to a frequency of from 20 to 50 kilocycles.
- (vi) The blank is finally lapped by hand to the exact thickness required, this being determined by matching the frequency of the crystal being cut with that of a frequency-standard crystal plate. Recent research has shown that the frequency of a plate can be reduced slightly by exposure to X-rays, thus enabling the required final frequency to be obtained very accurately.
- (vii) The piezo-electric effect of the completed plate is tested under varying temperatures, ranging from 0° C. to 60° C., and the plate is then mounted. Serious difficulty has been experienced with plates due to "ageing" caused by the action of water vapour on the surface layers of quartz which have been disturbed by grinding, lapping and polishing. This surface material can be removed by etching, the resultant surfaces being stable.

## 12. AUSTRALIAN SOURCES.

Limited inquiries have been made for quartz crystals in Australia as far back as 1926 and since then the Mines Departments of the States have been on the look-out for suitable deposits. However, very few usable crystals were obtained, partly because the minimum acceptable size was much larger then than now. About 1938 the demand began to increase rapidly, but even then, imported crystal was used and it was not until 1941 that more active attempts were made to obtain crystal from domestic sources.

During 1943 and 1944 a comprehensive survey of Australian sources of quartz crystal was undertaken by Mr. M. D. Garretty, Consulting Geologist to North Broken Hill Limited, at the request of the Controller of Minerals Production, Department of Supply and Shipping. The results of this survey are embodied in Bulletin No. 13 of the Bureau of Mineral Resources, Geology and Geophysics, "Notes on the Occurrence of Piezo-Electric Quartz in Australia, with Special Reference to the Kingsgate Field", which should be read in conjunction with Bulletin No. 10.

## 13. AUSTRALIAN DEMAND.

It is difficult to express accurately the annual requirements in Australia of raw quartz crystals, for there is no fixed number of plates which can be cut from a given weight of raw quartz. It has been estimated that of the quartz bought, which appears to be usable on visual inspection, well under 1 per cent. is eventually mounted in holders. It was estimated in 1943 that 150,000 cut quartz plates would be needed annually during the war years, but at the present time 6,000 to 7,000 plates per annum would satisfy requirements. Only a small quantity of raw quartz is being derived from local sources at present.

Ample equipment is installed in Australian factories for cutting the number of quartz plates required in Australia.

## 14. BUYERS AND PRICES.

The principal buyers and cutters are:—Amalgamated Wireless (Australasia) Ltd., Radio Corporation Pty. Ltd. and Quartz Crystal Laboratories.

During the war, the Department of Munitions and the Postmaster-General's Department were also buyers, and the American Army Signal Corps had a mobile cutting unit in Northern Queensland and was prepared to buy suitable crystal direct from producers.

The assessing of the value of quartz crystal is difficult, for often usable crystal cannot be distinguished from unusable crystal without cutting, and valuation for purchase has to be made before cutting. In Australia there is no well defined method of valuation and purchase. Some crystal has been bought on a weight basis provided that it is apparently usable so far as can be seen by preliminary visual inspection. Under these conditions prices have risen from approximately 10s. per lb. weight at the beginning of 1940 to £3 per lb. weight in July, 1943. This price was still operative in February, 1947. However, it is unlikely that cutters would be prepared to buy large quantities of quartz at £3 per lb. on visual examination only. They would probably require to know, from previous experience or by cutting a representative sample, the prevalence or otherwise of twinning in the quartz from the locality.

Another method of purchase is to cut all the crystal supplied and pay for it at an agreed amount per suitable plate obtained.

Both of the above methods can be combined, a low initial payment being made on a weight basis for the apparently usable crystal supplied, plus a later payment based on the number of plates obtained.

Purchase conditions are usually determined by negotiations between the buyer and seller, but it is important that, if prospectors are to be induced to search for quartz crystal deposits, some attractive and fixed price be offered to them and that this price be on a simple basis which the prospector can follow, i.e., he himself must be able to assess roughly the value of any crystal which he may find.

Fig.1.

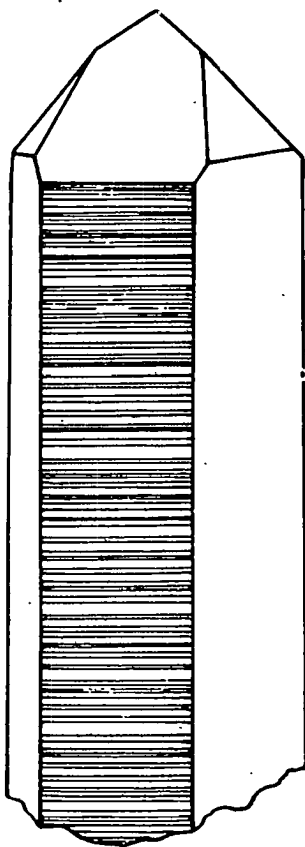


Fig.2

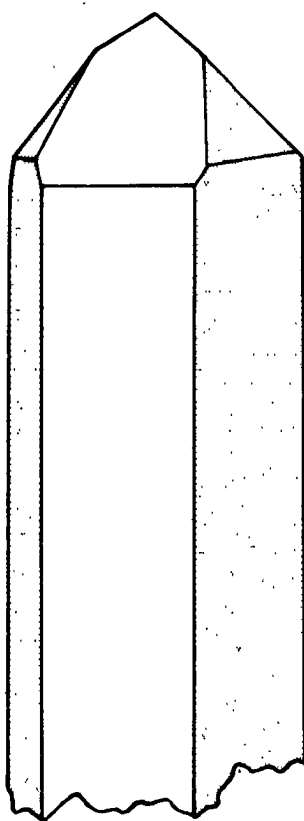


Fig.3

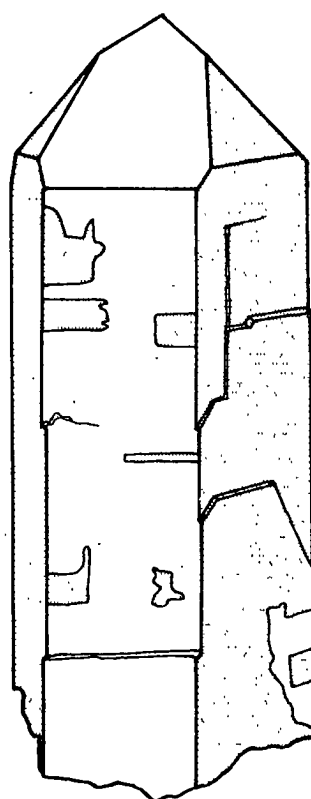


Fig.4

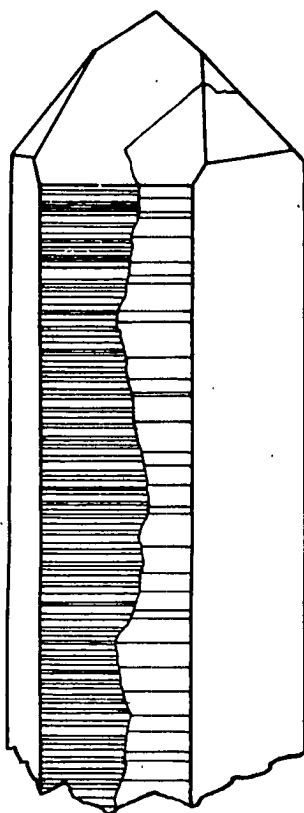


Fig.5

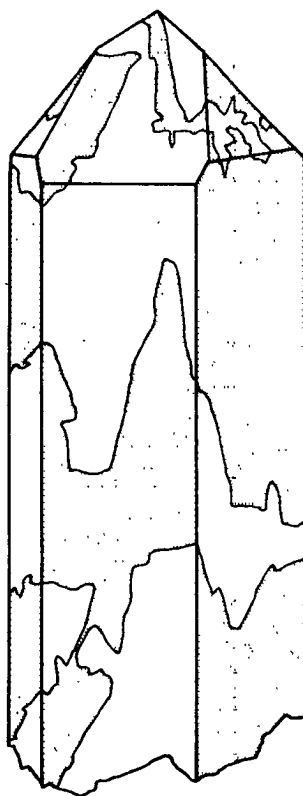
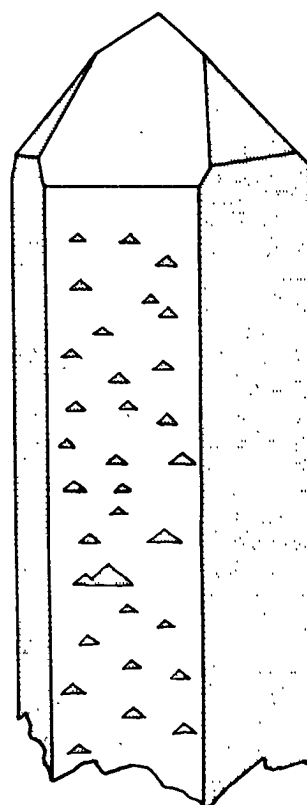


Fig.6



- Fig.1 Untwinned crystal. Striations illustrated on one face only.
- Fig.2 Untwinned crystal showing corresponding textures of alternate faces.
- Fig.3 Unsuitable crystal showing (a) abrupt changes in level of prism faces and (b) small areas having the characteristics of the prism face adjacent to that on which they occur.
- Fig.4 Unsuitable crystal showing broken striations and twinning line.
- Fig.5 Unsuitable crystal showing light and dark areas, or showing changes in texture: indicative of deep twinning.
- Fig.6 Suitable crystal showing triangular shapes: indicative of surface twinning.

# LIST OF SUMMARY REPORTS ON THE MINERAL RESOURCES OF AUSTRALIA.

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No.

- 1 Zirconium.
- 2 Titanium—Rutile and Ilmenite.
- 3 Antimony.\*
- 4 Mica.
- 5 Graphite.
- 6 Fluorite (Fluorspar) and Cryolite.
- 7 Manganese.\*
- 8 Molybdenum.
- 9 Bismuth.
- 10 Chromium.\*
- 11 Magnesium—Magnesite, Dolomite, &c.
- 12 Diatomite.
- 13 Barium—Barite or Barytes.
- 14 Felspar (including Cornish Stone).
- 15 Talc, including Steatite, Pyrophyllite, &c.\*
- 16 Tungsten\*
- 17 Asbestos.
- 18 Beryllium.
- 19 Tantalum and Columbium.
- 20 Mercury.
- 21 Cadmium.
- 22 Arsenic.
- 23 Lead.\*
- 24 Pigment Minerals.
- 25 Lithium.
- 26 Sillimanite, Kyanite, &c.
- 27 Aluminium and Bauxite.
- 28 Uranium and Radium.†
- 29 Phosphates.
- 30 Bentonite and Fuller's Earth.
- 31 Sulphur, including Pyrite and other sulphur-bearing minerals.\*
- 32 Cobalt.
- 33 Zinc.
- 34 Potash Minerals.\*
- 35 Minor Metals.\*

\* To be published shortly.

† Not available for publication at present.