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Geologic Significance of the Color of Granite Zircon, and the Discovery of the Pre-Cambrian in Japan

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# Geologic Significance of the Color of Granite Zircon, and the Discovery of the Pre-Cambrian in Japan

By

# Tôru TOMITA

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### Introduction

The existence of the Pre-Cambrian gneisses, granites and crystalline schists has long been one of the primest problems in Japanese geology. Since, for reasons of space, it is impossible to make a full historical review of various views on this problem, only some recent views will here be referred to.

Out of many metamorphic provinces of Japan, the Hida Highlands in the Hida Metamorphic Zone, which is supposed to cover also the base of the Noto Peninsula and Dôgo Island (Oki Islands in the Japan Sea), has come into the limelight since the recent successive discoveries of Middle Gotlandian formations in the regions of gneisses and crystalline schists (Kamei, 1949, 1950; Ishioka and Kamei, 1950; Fujimoto et al., 1953). The geologic age of these metamorphites are inferred by Ishioka and Kamei to be Pre-Middle Gotlandian. Minato (1950), however, says that the type of gold deposits at the Amô Mine in the west of the highlands is similar to that of the gold mines in the Korean Pre-Cambrian region and suggests the possibility that the metamorphites are of the Pre-Cambrian. Ishioka (1953, p. 87), on the contrary, holds the view that the gold deposits have no genetic relationship to the generation of the metamorphites. Fujimoto et al. do not think that the age of the Mugishima Gneiss (a member of the Hida Gneiss Complex) is Ordovician or Cambrian, judging from the common non-metamorphic distinctiveness of the Lower Paleozoic formations in the Asiatic Continent. Yet some geologists (Kobayashi and

KAMEI, 1953, and others) hold the saner view that the definite age determination of the Complex is quite beyond them. Such being the case, it seems to me that they trust to luck of discovering radioactive minerals useful for age determination.

In 1949, I devised "Zircon Correlation Method" (for brevity, "Zircon Method") in order to take the place of the well-known heavy mineral (or heavy accessory mineral) correlation method (Groves, 1927a, 1927b, 1930, 1931; Wells, 1931; Reed and Gilluly, 1932; JENKS, 1934; STARK, 1934; MARSDEN, 1935; G.L. TAYLOR, 1935; STARK and BARNES, 1935; BRUCE and JEWITT, 1936; J.C. REED, 1937; J.H. TAYLOR, 1937; TYLER and Marsden, 1937; Dapples, 1940). In my method, a granite of unknown age is correlated with another of known age when no noticeable difference in color displayed by reflected light is recognized between their accessory zircons. Thus their colors are compared between the final concentrates, which are separated by panning method and preserved in small glass-capsules, by placing in juxtaposition on a piece of absorbent cotten, with which a shadow casting by the glass-cupsule can be strictly avoided. Since our present purpose is only to obtain zircon crystals (their relative percentages are left out of consideration), it is not absolutely necessary to use any expensive heavy liquid to concentrate zircon; thus, we are gaining our object only by repeated pannings with one-litre beakers after sizing rock powders with a 50-mesh sieve. In order to dissolve hydrous iron-oxide stains on the crystal surfaces, each concentrate is slowly warmed in hydrochloric acid at a temperature below 100° (at a higher temperature, say above 150°, accessory zircon will begin to discolor) for 20 to 30 minutes. Following this procedure, we can get some thirty specimens of zircon in six days.

By this color matching method, the gneisses and granites making up the Hida Metamorphic Zone were confirmed to be Archean; moreover, unanticipated Archean rocks sandwiched between younger sedimentary formations were discovered in Kumamoto and Wakayama Prefectures, presenting a new problem in Japanese geology.

# I. World-wide Distribution of Purple and Rose-pink Zircons in Archean Rocks

In so far as I know, W. Mackie (1923) was the very first to discover the fact that the accessory zircon of the Lewisian Gneiss (Archean Complex in Scotland) was of distinctive purple or occasionally of rose-colored variety. Early and recent literature leads me to the conclusion—that purple or rose-pink zircon is confined to Archean gneisses or granites. This conclusion was furthermore confirmed by my own examination of the zircons in the Archean granite-gneiss from India, in the Archean gneissic granite from North China, and in the Huronian sandstone-quartzite from Wisconsin (North America), which last should be composed, needless to say, of Archean rocks and minerals.

The distribution of Archean purple and rose-pink zircons hitherto known is as follows:

- (1) Scotland (Mackie, 1923; Boswell, 1927, p. 317; Hutton, 1950, p. 691). Distinctive purple and rose-colored zircons are relatively abundant in the Lewisian Gneiss; commonly rounded or ovoid grains, but sometimes with crystal form; often finely polished as if by wind; sometimes zoned with a darker center, or display colors of varying intensities. According to Boswell, good rose-pink or purple zircon, which differs entirely from the purple-brown variety in the Dartmoor Granite (Armorican or Variscan), are widely distributed in British sedimentary rocks from Pre-Cambrian to Recent, derived from the Pre-Cambrian regionally metamorphosed rocks.
- (2) Canadian Shield (Mackie, 1923; Tyler and Marsden, 1937; Morgan and Auer, 1941; Hutton, 1950, p. 691; Tomita, 1954, this paper). Mackie points out the presence of purple zircon in the Pre-Cambrian gneiss of Canada (Laurentian) exactly similar to that of the Lewisian Gneiss. Tyler and Marsden report the zircon in the Laurentian gneissic granites of the south shore of Lake Superior is of the purple variety, and Morgan and Auer say that the oldest known granites in both the Lake Superior region and the Oxford House area contain light pink to purple zircons. My own examination of a Huronian sandstone-quartzite from the Necedah Mound (Wisconsin) confirms the presence of purple zircon derived from the Pre-Huronian rocks thereabout.
- (3) New Jersey Highlands (Tyler, 1940). In the migmatitic gneisses of the New Jersey Highlands (New Jersey Gneiss) and the Franklin Limestone (tabular lenses conformable to the banding of the gneisses) are found zircons, deep pink or purple through all intensities of shade to almost colorless. The zircon in the limestone was formed, in my opinion, by pneumatolytic transport of material from the igneous source.
- (4) Arkansas River (Boswell, 1927), (5) Tampico, Mexico (Boswell, 1927), (6) Montevideo, Uruguay (Boswell, 1927), (7) Senegambia (15°.0 N. Lat., 15°.0 W. Long.), West Africa (Boswell, 1927). Although Boswell does not describe the geological mode of occurrence of zircon from these localities, it is not unreasonable to infer that it is found in river deposits derived from surrounding Archean shields.
- (8) South Rhodesia, South Africa (BOND, 1948). Purple zircon is found in the Lower Wankie Sandstone of the Karroo Formation. In South Rhodesia, this sandstone is underlain directly by the Archean terrane, from which the mineral in question must have been derived.

- (9) Transvaal, South Africa (Mackie, 1923). The famous Transvaal banket of the Younger Algonkian is another example of the formation that contains purple zircon in South Africa. This zircon is also derived from the underlying Archean rocks.
- (10) Western Australia (HIGGINS and CARROLL, 1940). In the Wandagee Hill sediments (Permian) are found many well-worn crystals of purple zircon. Some are zoned, others clear with a few inclusions. The source of the detrital zircon is, according to the authors, the Pre-Cambrian Complex of granites, gneisses, and schists.
- (11) New Zealand (Hutton, 1950, p. 689). Deep purple and pink to pale pink zircons are reported from Recent deposits. Comparing these with the Lewisian and Laurentian zircons already referred to, Hutton observed that the features of color, zoning, micro-fissuring, and rounding are so quite similar that "distinction between them was impossible."
- (12) New Guinea (Boswell, 1927). The mode of occurrence of purple zircon on this island is not mentioned. The ultimate provenance, however, can be inferred to be the postulated old land that connected this island with the Australian Continent now depressed beneath the Arafura Sea.
- (13) Indian Peninsula (Tomita, 1954, this paper). A single specimen sent from India to place at my disposal is a mesocratic granite-gneiss with a marked banded structure. This gneiss taken from near Bangalore and may belong to the group of "Banded Gneisses of the Bengal type" (Reed, 1921, pp. 260-264). The purple zircon of this gneiss consists roughly of two kinds; one is non-metamict and transparent, and the other is metamict and semitranslucent to opaque, the former being predominant. It is to be noted that the metamict crystals show their good crystal forms in spite of the non-uniform development of metamictization, whereas the non-metamict crystals show fine zonal structure, numerous micro-fissures, and more or less rounded-off edges.
- (14) North China (Tomita, 1954, this paper). During my microscopic examination of the constituent minerals of the "Huangto" (Chinese Loess) in North China, not infrequently were met with purple zircon and malacon, which are believed to be derived from the Older Archean terrane there, because there is no crystalline rock of younger age that contains zircon of a purple color; thus, for example, the zircon in the Younger Archean gneissic microcline-granite from Mt. Taishan (Shantung Province) is rose-pink or rose-lilac, that in the Older Algonkian granodiorite from Mt. Peitai (Wutaishan, Shanhsi Province) is cloudy brownish-purple, and that in the Late Pre-Cambrian porphyrogranite near the Miyün Mine (north of Peking) is

colorless, while that in the Post-Jurassic (probably Lowermost Cretaceous) granite-porphyry on the sea-shore of Tingtao is very pale orange.

- (15) Korea (Boswell, 1927; Geological Survey of Korea, 1941, p. 231). The purple zircon from "Yunan" (Yun-an, or Sun-an), north of Heizyô (Pyongyang), cited by Boswell occurs abundantly in Recent river deposits. It is derived without doubt from the Archean Gray Gneiss extensively developed throughout Northern Korea. It is interesting to refer to two more occurrences of purple zircon in Northern Korea; one is from the limestone layers in the Gray Gneiss near the village of Fûsûdô (Sozan, or Chosan, District), and the other is from the limestone-contact graphite deposits at Shinheidô (Kôkai, or Kanggye, District), both situated in close proximity to the River Ôryokkô (Yalu). It is worthy of more than passing attention that there is no decided difference in color between igneous and pneumatolytic zircons of the same geologic age.
- (16) Formosa (ICHIMURA, 1948). In the Tertiary sandstone and shale is found zircon of a purple or light pink color associated with colorless zircon and monazite. ICHIMURA's view that these minerals have probably been transported from an old land on the west may well be supported, I think, by the existence of the Sankuan System, a remnant of the Pre-Cambrian regionally metamorphosed rocks among the Cretaceous granites in Fukien Province (South China) opposite Formosa.

So far as the above data are concerned, it may safely be concluded that the accessory zircon of the Older Archean (Laurentian) granite-gneiss should be of purple-colored variety, that of the Younger Archean (Algoman) granites or gneissic granites of rose-pink or rose-lilac variety—this is almost universal among all continents, except Fennoscandia, from which, to my great regret, not a single datum or a specimen has yet been available.

## II. Colors of Algonkian and Post-Cambrian Accessory Zircons

Reserving my illuminating remarks for another occasion, some crude notes on the colors of Algonkian and Post-Cambrian accessory zircons will be given for reference in the following:

(1) Algonkian (von Chrustschoff, 1886; Tyler and Marsden, 1937; Morgan and Auer, 1941; Tomita, 1954, this paper). Meager occurrence of the Algonkian granites in the world prevents me from furnishing definite information on this subject. According to von Chrustschoff, the granulite-like and leucocratic gneisses from Utah contain light yellowish, or clear yellow, to brownish and nearly opaque accessory zircons, whereas to Tyler and Marsden the Keweenawan granitic rocks

from the south shore of Lake Superior have colorless to yellow zircons. Morgan and Auer, however, reported the dirty "malacon type" of zircon from the Huronian and Late Pre-Huronian granites, while the clear, colorless, "normal type" from the Keweenawan rocks, of the Lake Superior region. These authors' results are in general accord with among themselves.

On my part, the Peitai Granodiorite (Mt. Wutaishan, North China), which is correlated with the Post-Kalevian or Post-Jatulian, the Older Algonkian of Fennoscandia (Sederholm, 1932; Tomita, 1942, pp. 322, 483), and the Post-Huronian leucogranite from the Necedah Mound, Wisconsin (according to R. Toriyama's oral communication, it is an intrusion in the Huronian sandstone-quartzite already referred to) were examined to find that the zircons of these rocks are of cloudy brownish-purple variety. The Miyün Porphyrogranite (north of Peking) with glomeroporphyritic feldspars of the Rapakivi type as well as the coarse-grained Kung-chuangling Granite of South Manchuria contains colorless zircon, though the rarity of this mineral is certainly remarkable in these Younger Algonkian granites.

- (2) Post-Ordovician and Pre-Devonian: Caledonian (RASTALL and WILCOCKSON, 1915). Of the granitic rocks in the English Lake District (England), the Skiddaw and Shap Granites may here be referred to. Most of the zircons in these granites are of colorless variety, accompanied by some greenish or brownish ones.
- (3) Permo-Carboniferous: Armorican (Brammall and Harwood, 1923; Tomita, 1954, this paper). In the Dartmoor Granite (England), which is unquestionably a mixed rock, there are two strongly contrasted types of zircon; one is the "zoned type," and the other is the "clear type." "The former greatly outnumbers the latter, the relative proportion being of the order 50:1; frequently the ratio is still more in favor of the zoned type" (Brammall and Harwood, 1923, p. 28). Viewed by reflected light, the color of the zoned type is dun-colored to pale mauve, yellowish, or greenish-brown, and that of the clear type is very pale yellowish, mauve, or greenish. This coexistence of variegated zircons may possibly be due to the mixing of materials in this assimilation granite. It may here be remarked incidentally that we must remember Boswell's careful observation that the purple-brown zircon in the Dartmoor Granite differ entirely from the purple variety in the Archean Lewisian Gneisses (see Chapter I, 1).

In Japan, the Higami Granite (Kitakami district) is rich in zircons of a dark reddish-brown to lilac-brown color. To my belief, this granite is Pre-Upper Permian, because the pebbles of this granite—this is also determined by my zircon method—are found in the Usuginu Conglomerate (Kitakami district) and also in a conglomerate bed of the Kuma Formation (South Kyûshû), both being comprised in the Upper Permian. Moreover, the Arisu and Ôno Granites, either of which contains

lilac-brown zircon very similar to the Higami Zircon, are, according to H. Kanô and to T. Senfull (personal communications), intruded into the Lower Permian formation (Sakamotozawa Formation). Accordingly, it may safely be said that the Post-Lower and Pre-Upper Permian zircons should be of the dark reddish-brown to lilac-brown variety.

- (4) Permo-Triassic (Appalachian) and Triassic (von Chrustschoff, 1886; Tomita, 1954, this paper). The color of zircons in the "Variscan" granites and granite-porphyries from various localities in Vosges, Schwarzwald, Sachsen, Schlesien, and near Leipzig is reported to be wine-yellow, clear-yellow, light yellow, or white-yellow to colorless. The Japanese granites of the Post-Permian and Pre-Middle Jurassic (Pre-Torinosu Group)—this is determined from both field evidence and zircon datum on the pebbles of conglomerate—contain the zircons of varied colors as follows:
- (a) Lowermost to Lower Triassic: The Post-Permian zircons begin with a pale yellow variety (Kabano Gneissic Granite, which also contains monazite), passing into colorless (Kiyosaki Granite) and grayish-white, and finally to almost colorless (Miyahohara Granodiorite).
- (b) Early Middle Triassic (probably): Pale grayish-pink or flesh-colored (Ito-shima Granodiorite and Hazu Granodiorite). Since the Hazu Granodiorite (gneissic in parts) in Aichi Prefecture encloses the xenolithic bodies of the Kiyosaki Granite, it is at least certain that there was the long lapse of time involving the complete consolidation of the Kiyosaki granitic magma before the intrusion of the Hazu Granodiorite, even though the latter is assumed to have taken place immediately after the consolidation of the Kiyosaki Granodiorite.
- (c) Early Upper Triassic (probably): Light orange to yellowish-brown (Sumi-kawa Granite). The Sumikawa Granite, which is intruded into the Hazu Granodiorite, shows a banded structure along its margin, indicating that the intrusion of this granite was a distinct event. This implies also that there was an interval of time between the two intrusions of the Hazu Granodiorite and the Sumikawa Granite.
- (5) Jura-Cretaceous and Middle Cretaceous (Tomita, 1954, this paper). For lack of not only the description of the color of granite zircon in American literature but also granitic rocks themselves in Europe, none but the following Japanese data would be useful to develop this line of research.
- (a) Jura-Cretaceous, probably Lowermost Cretaceous: Very pale orange (Yosa Granite, one hundred and twenty million years of age; Takubo and Tachikawa, 1951). It is to noted that the Post-Jurassic Tingtao Granite-porphyry (North China) already referred to belongs to this category.
- (b) Middle Cretaceous: The colors of accessory zircons of this age make a change by easy gradations according to the order of intrusion. Thus, from the older

to the younger, they are: Smoky yellow-brown with a reddish tinge (Kurate Granodiorite), light yellowish-brown (Hirao Granodiorite), smoky light brownish-yellow to light yellowish-brown (Hiroshima Granite and its associated granite-porphyries and nevaditic rocks) to white-yellow (Kôbukuro Granite). By the way, by some geologists, the first two granodiorites are called the Kurate Granodiorites; the rest, the Kaho Granites.

It is interesting to refer to a two-mica granite (Sawara Granite) which was intruded at the close of this period of plutonism. Each heavy fraction concentrated from the rock-specimens that were taken carefully from many different parts of a single granite body is distinguished among our zircon collection (Archean to Pleistocene) by a particularly yellow to brownish-yellow color. This yellow concentrate consists principally of monazite, mingled with a few clear, pale yellowish to colorless zircons. This fact, involving a marked tendency towards simultaneous appearance and disappearance of respective monazite and faintly-colored zircon, suggests that the granite in question is one of the end products of magmatic differentiation undertaken during Middle Cretaceous time. The two-mica granite in Aichi Prefecture (Busetsu Granite) is also particularly worthy of notice in its abundant monazite, its geologic age having not yet been definitely decided, though I am inclined to consider it to be synchronous with the Sawara Granite.

(6) Late Cretaceous and Earliest Tertiary: Laramide (Tomita, 1954, this paper). The granite-pegmatite that yields naegite, a meta-zircon sixty million years old, at Naegi (Gifu Prefecture), is used as the "time-signal" telling us the last day of the Cretaceous in Japan. The concentrate from the Naegi Granite related to the pegmatite just-mentioned is composed of a mixture of transparent colorless zircon and a few opaque gray-brown to dark brown minerals (probably radioactive), their nature having not yet been verified owing to insufficient material. Yet this association of the two kinds of minerals has, in my opinion, much importance to petrogeny, because a very similar association is observed in the Post-Middle Cretaceous granite of the Chûgoku district (Kibe Granodiorite) and also in the Post-Jurassic granite from South China (Hongkong Granite), both of which may probably be correlated with the Naegi Granite, leading me to the conclusion—that this association may well be understood to represent the outstanding characteristics of the granitic magma of this epoch covering a vast area along the eastern border of the Asiatic Continent.

Looking backward, the zircon of the Naegi Granite is preceded by the translucent to opaque yellowish-green to greenish-yellow ones (San-in Granites), probably semi-metamict to metamict, which are inferred to be connected with the Middle Cretaceous zircons of a smoky light brownish-yellow (Hiroshima Granite) and white-yellow (Kôbukuro Granite) color already referred to; looking forward, the Naegi Zircon is without doubt related to a variety of grayish (Hidaka "Late-kinematic

Granite"), white (Tsushima Quartz-porphyry), or colorless (Tsushima Granite) zircon, these igneous rocks being assigned stratigraphically to the Post-Cretaceous.

- (7) Eocene and Miocene (Brauns, 1909, p. 722; Doelter, 1918, p. 134; Kokubu and Ogasawara, 1934; Yien, 1949, p. 19; Yien, 1950, p. 41 and Tables 2, 4; Tomita, 1954, this paper).
- (a) *Eocene* (probably): Dull pink (Ogawa Granite). According to R. Sugiyama (personal communication), the Ogawa Granite is of Pre-Oligocene (Pre-Aikawa Formation), presumably of Eocene. The fact that a very similar zircon is found in an acid volcanic rock (Sobosan Lithoidite) presents a practical as well as a theoretical interest as discussed later.
- (b) Lower Miocene (at its close, probably): Grayish or colorless (Tanzawa Quartz-diorite and Kumano Nevadite).
- (c) Middle to Upper Miocene: Bright red, brownish-red to reddish-brown through all intensities of shade (Kôyama Gabbro-Diorite and Mogami Quartz-diorite). Almost all of the "Tertiary granites" in Northeast Japan and Hokkaidô belong to this category. In the Miocene alkaline syenite of Northern Korea (Taitaku Station near Gôsui in Kankyô-hokudô, or Hamgyong-pukto, District) and in the Miocene olivine basaltic rocks of Formosa (Mabutoku in the Sinchiku District) are found this sort of zircon. The well-known red zircon from the river sands at Espailly near Le Puy in Auvergne (basaltic region of Central France) and that in the basaltic lavas at Niedermendig in the Eifel (Rheinland) may also be classed in this category. The occurrence of hyacinth in the Later Miocene basalts (sometimes as large as 13 mm in diameter as seen among the specimens of Mabutoku) is so quite unusual that their origin, say whether or not they are xenocrysts in the lavas, is well worthy of further study.
- (8) Pliocene to Recent: Cascadian (von Chrustschoff, 1886; Brauns, 1909, p. 723; Doelter, 1918, p. 134; Kunitz, 1936, p. 423; Tomita, 1954, this paper). Though I have least knowledge of Pliocene granite zircon owing to the absence of Pliocene granite in Japan, the striking resemblance in color between volcanic and granite zircons of the Middle Cretaceous and the Middle to Upper Miocene has induced me to pick up knowledge of the volcanic zircons of Pliocene to Recent in order to fill the last chapter of magmatic history of our earth.
- (a) *Pliocene*, probably *Middle Pliocene*: Dull brownish-yellow (Alkaline Trachyte II of Dôgo, Oki Islands).
- (b) *Upper Pliocene*: Clear wine-yellow (Drachenfels Trachyte, Siebengebirge; von Chrustschoff).
  - (c) Plio-Pleistocene: Colorless (Dôgo Comendite).
  - (d) Pleistocene: Colorless (Izu pyroxene-andesite; H. Kuno's personal com-

munication). Colorless zircons of this epoch are also reported from the following localities: the Azores (colorless zircon here is formerly known as "Azorite"; Doelter), and Mt. Vesuvius (colorless with a bluish tinge; Kunitz). The zircon of the last locality may be of Recent, since it occurs in volcanic ejecta.

The foregoing descriptions as to the variation in color of Post-Cambrian accessory zircon may be summed up in Table 1, which is, of course, somewhat tentative awaiting future revision; for the Pre-Cambrian zircon, the reader is referred to Table 3. In Table 1, out of three mental variables of color (Evans, 1948, pp. 117–

Table 1. Geological Classification of the Color of Post-Cambrian

Accessory Zircon

Brightness in Color		More Somb	er ←—→	Bright	ter	
Geologic Age Hue	Caledonian	Appalachian	Trias-Jura	Creta-L	aramide-C	ascadian
Colorless with a bluish tinge						R
Grayish		lmTR		Pa	1M	
Colorless or white	D - G	lmTR		Pa uK**	1M	Ps P-Ps
Yellow-green to green-yellow*	D - G			uK		
White-yellow to yellow	D - G	lmTR uPm		uK mK		uP
Brownish-yellow						1P
Yellowish-brown				mK		
Orange			Early uTR	1K		
Brown		Pm-C				uM
Reddish-brown		Pm-C				mM, uM
Red						mM
Pink			Early mTR			
Rose·pink					E	Í
Purple to violet						
Blue-violet					(Pa or E)	
Blue						

<sup>\*</sup> Semi-metamict or metamict

<sup>\*\*</sup> Mixture of colorless zircon and opaque gray-brown to dark brown minerals

D-G: Devonian-Gotlandian, Pm-C: Permo-Carboniferous, uPm: Upper Permian, lmTR: Lowermost Triassic, mTR: Middle Triassic, uTR: Upper Triassic, 1K: Lower Cretaceous, mK: Middle Cretaceous, uK: Upper Cretaceous, Pa: Faleocene, E: Eocene, 1M: Lower Miocene, mM: Middle Miocene, uM: Upper Miocene, 1P: Lower Pliocene, uP: Upper Pliocene, P-Ps: Plio-Pleistocene, Ps: Pleistocene, R: Recent

137), hue and brightness are presented, neglecting saturation, for which the reader is referred to each description already given.

It goes without saying that, in respect of these three variables (of course, under the same observation condition), the colors of zircons of different ages differ much more distinctively before our eyes than as shown in the Table. However, colorless or white or grayish zircons of different ages cannot be discriminated between each other without knowledge of field evidence of their mother rocks; namely, the knowledge of their ordering in relation to a sedimentary formation or an igneous body, geologic age of which either already known or determined at need by the zircon method, should be required for their discrimination.

## III. Geological Facts about the Color of Accessory Zircon

During our study of accessory zircon, my colleagues and I are always excercising due caution neither to overlook nor to over-refine their characters in discriminating between two kinds of zircon, and yet our inquiry has elucidated the fact that there are some general if not invariable rules with regard to the color of accessory zircon (hyacinth) as follows:

- (A) Within a Single Large Body of Granitic Rock:
- (1) The hue is in general invariant throughout the whole body, except that in some occasions there is more or less notable peculiarity in marginal rock-facies, especially those of smaller bodies (see below, 4).
- (2) The saturation is variant within a single body. In the Higami Granite body, for example, the color is more deeper in the margin than in the center, but it is uncertain whether this variation is gradational or not.
- (3) Neither hue nor saturation has any connection with the features characterizing rock-facies, such as the presence of a certain mineral (say, hornblende in a biotite granite), relative amount of essential minerals, chemical composition of plagio-clases, development of a schistose or a porphyritic structure, and so on.
- (4) The color of our panning concentrate (mass color in our study) separated from a marginal facies is sometimes quite unlike as compared with that from a central facies; that is to say, it does not match any among our zircon collection (Archean to Pleistocene). This is due to either the mixing of two kinds of zircon as the result of the contamination of granite magma by its country rocks along its margin (one is crystallized from the magma, and the other is taken from its country rock) or the unusual character of the zircon (in general, highly radioactive for the accessory zircon of granite). These exceptions, however, do not break the general empirical rule that the color of zircon has no connection with rock-facies within a single granite body when the small magnitude of marginal facies and the rarity of such an occurrence are taken into account.

- (B) In mixed rocks, such as migmatitic gneisses along, and xenolithic inclusions in, a granite body, there are almost always two kinds of zircon as in the case of a marginal facies just-mentioned (YAMAMOTO, 1953).
- (C) Plutonite (Granite or Quartz-diorite) versus Volcanite (Rhyolite or Quartz-andesite): Notwithstanding the difference in their modes of occurrence, the plutonite and its volcanic equivalent, both of which are believed by geologic evidence to belong to the same epoch of magmatic activity, have a common zircon in respect of color in spite of the marked difference in crystal habits (Томіта and Матѕимото, 1935).
- (D) As to Petro-provinces: The difference of petro-provinces is not reflected in the color of zircon. Thus, for example, the alkaline syenite in Northern Korea and the calc-alkaline gabbro to quartz-diorite in the districts along the shores of the Japan Sea, both of Later Miocene age, have a common zircon of red to reddish-brown colors.
- (E) Order of Color Variation with the Lapse of Time: As will be seen in Table 1, the hue of accessory zircon (hyacinth) varies from colorless, through yellow, orange, brown, red, rose-pink, and purple, to bluish-violet with the lapse of time, repeating this order of hue, but changing in brightness from brighter to more somber, throughout the whole geologic time. The blank spaces in each column in the Table may be due partly to the imperfection of my work and partly to the entire absence of corresponding granites on our globe. This fact is clearly recognized by a glance at the standard collection of accessory zircon ("Zircon Scale") preserved in our laboratory and may be of supreme importance in considering the cause of the color of hyacinth as discussed in the next chapter.

#### IV. Theoretical Grounds for the Zircon Method

As will be clear from the foregoing considerations, the zircon method has been initiated with due regard to empirical knowledge. On the other hand, it is most necessary, I think, to find theoretical grounds for the cause of coloration in hyacinth that are consistent with the facts at least hitherto known—facts, not only mineralogical but also geological.

Before setting forth my opinion, the views of various authors on the subject, old and new, hypothetical and theoretical, will be given in formulated form with my short criticisms.

(A) Non-radioactive Pigment Hypotheses: Prior to the discovery of radium (1898) and in those days when the knowledge of radioactivity was not yet universally diffused into the mineralogical science (up to about 1910), every possible effort was exerted only in speculation to find out some element (or its oxide) plausible enough to be the pigment concealed in hyacinth. The postulated pigments were: CuO

(Sandberger, 1845), Fe<sup>3</sup> (Spezia, 1876, 1899),  $Zr_2O_3$  (Weinschenk, 1896),  $CO_2$  (von Kraatz-Koschlau and Wöhler, 1899),  $CO_2$  and  $Fe_2O_3$  (Stevanović, 1903b), and the oxides of Fe, Cr, Mn, or Ti (Herman, 1908).

In view of the already mentioned geological facts on the distribution in time and space of the accessory zircon having the same hue, it is not too much to say that these older views are beneath criticism, and there is no need of referring to various objections in the later days. In short, the non-radioactive pigment hypotheses had been left out of consideration among mineralogists when the radiogenic coloration theories (see below) appeared, though still there remained some authorities who were inclined to regard Fe<sub>2</sub>O<sub>3</sub> and CuO (Eppler, 1927, p. 486) or Fe<sub>2</sub>O<sub>3</sub> (Chudoba and Dreisch, 1936, pp. 78-79) as pigments in certain hyacinth (Mongka Hyacinth, Thailand).

- (B) Radiogenic Coloration Theories: Hyacinth is decolored by heating (Schumacher, 1801; Spezia, 1876; Stevanoviĉ, 1903b; Brauns, 1905; Strutt, 1914; Chudoba, 1935a; Gause, 1936; Lietz, 1937; Tomita and Karakida, 1954) at a temperature between 260° and 900° (K. Simon, 1908; Eppler, 1927, pp. 475-476; Boswell, 1927, p. 313; Chudoba and Dreisch, 1936, p. 70) and rather readily restore its original color by radium irradiation (K. Simon, 1908; Brauns, 1909, 1928; Boswell 1927; Chudoba and Dreisch, 1936). On this interesting character are built up the following radiogenic coloration theories: (1) Impurity-Element Theory, (2) Radio-colloid Theory, (3) Electronic Theory.
- (1) Impurity-Element Theory (Brauns, 1909, pp. 725-726; 1911, p. 139). According to Brauns's belief, natural colorless zircon (Pfitshtal Zircon, almost pure in composition; Kunitz, 1936, p. 423, Table VII) cannot be colored by radium irradiation (10 day irradiation with 10 mg radium-fluoride) making a strong contrast with the behavior of decolored hyacinth. On this belief he erected his theory that no bond of ZrO<sub>2</sub>-SiO<sub>2</sub> (the current view in his days) is broken so as to color zircon, whereas the impurity-elements (foreign elements) within the mineral are displaced by the irradiation to make a new condition favorable enough to produce the color in response to their kind and relative abundance. As to the source of radium radiation in nature, he holds the view that it is of post-magmatic and post-volcanic solutions or gaseous transfer; that is to say, it is of exogenous origin.

This theory was soon disproved by Doelter and Sirk's experiment (1910), which was made on the zircon from the same locality as what dealt with by Brauns but under different irradiation conditions (23 day irradiation with 1/2 g radium-fluoride). Thus they found that even natural colorless zircon could be colored by the radio-irradiation if ample radio-source and sufficient time are allowed. Furthermore, the most recent (so far as I know) experiment along this line of research has proved that artificial zircon become almost completely metamict by 40 month irradiation with 4 mg radium (von Stackelberg and Rottenbach, 1940b).

At present, to the best of my belief, there is very little doubt that every zircon would be colored by radio-irradiation independently of the existence of non-radioactive impurity-elements, provided that the time of irradiation is sufficiently long and/or the irradiation is efficiently strong, though little if any influence of the impurities cannot be utterly neglected.

(2) Radio-colloid Theory (Doelter, 1918, pp. 147-148). Doelter attributes the natural color of hyacinth to the colloid particles of radioactive constituents, such as " $ThO_2$  and  $UO_2$  as isomorphous mixtures" as well as radioactive inclusions, which are dispersed as electrolytic dusts throughout the structure. As to the artificial coloration of pure zircon by radium irradiation, he holds the view that the materials constituting zircon ( $ZrSiO_4$ ) must be dissociated by electrolysis into the color-producing constituents of colloidal dimensions.

Enlightening though it was in his days, Doelter's view will be disputed in many respects. To begin with, we cannot believe in the presence of such radioactive constituents as "ThO2 and UO2 as isomorphous mixtures"; Th and/or U must be present diadochically replacing zirconium in the structure of zircon. Secondly, the color that would rise if colloidal particles were present within zircon must be dependent upon the size, shape, and dispersity of the particles as is in a colloidal solution. On the other hand, in view of the geological facts already mentioned, it is required that there must be a qualitative and quantitative uniformity in the three properties just-mentioned among zircons from say a single granite body. To my opinion, it is not likely that the colloid particles which were contained in any two zircons from different parts of the same granite batholith should not make much difference in the three properties between them. Thirdly, the decoloration by heating, according to DOELTER'S view, is to involve a fairly rapid change in the state of the three properties. However, it is doubtful whether any solid colloid particle could move freely within a rigid crystalline framework, though there is the possibility that some atoms would coagulate by radio-irradiation into colloidal particles, which latter would be dispersed in turn by heating. And lastly, and that this seems to be the most substantial evidence against Doelter's view, no alpha-track images characteristic of radio-colloids (YAGODA, 1949, Figs. 27, 28) are recognized on the autoradiographs of our zircon specimens. Notwithstanding a little weakness, Doelter's excellent view that the source of radio-radiation in nature is of endogenous origin may be highly appreciated.

(3) Electronic Theory (Eppler, 1927, pp. 480-483, 486; Lietz, 1937; Tomita, 1954). As early as in 1927, Eppler suggested the possibility that to the explanation for the cause of the dilute color in zircon Przibram-Belar theory on rock-salt can be applied. Lietz's study in the absorption of the Mongka Hyacinth develops a deeper interest in the subject concerning the cause of the color of hyacinth. He determined the absorption spectra of four laboratory materials: colorless (de-

colored specimen), brown (heating product at  $200^{\circ}$ ), red (heating products at  $110^{\circ}$  and  $120^{\circ}$ ), blue (specimen colored through ultraviolet-ray irradiation). Showing the three absorption curves for the blue, red, and brown materials in a diagram (Fig. 10) and analyzing the features of their absorption spectra, he arrived at the conclusion that there are two distinct and one uncertain absorption-bonds, which closely resemble those for the fluorite from East Turkestan displayed through ultraviolet-ray irradiation at lower temperatures. Of the two distinct absorption-bonds, one ("a-bond," with a maximum at about the middle between 450 and 500 m $\mu$ : 2.62 eV) is interpreted by him to be due to F-centers, and the other ("b-bond," with a maximum at about 650 m $\mu$ : 1.90 eV) to F'-centers, referring to the number of color centers ( $10.4 \times 10^{14}$  of total) as well as the ionization within the structure of the hyacinth (pp. 350–352).

In view of Lietz's opinion, I re-examined W.G. Simon's data (1930) on the absorption of zircons from various localities by constructing a new composite diagram (this diagram will be published in another paper now in preparation), and found that each absorption curve shows discontinuities somewhat similar to those pointed out by Lietz. Thus it is likely that every crystal of hyacinth is ionized and its color is due to color centers. By the way, the physical characters of hyacinth which are interpreted in the light of the electronic theory are listed below:

- (a) Changes of color and decoloration by heating (Schumacher, 1801; Spezia, 1876; Stevanović, 1903b; Brauns, 1905; K. Simon, 1908; Strutt, 1914; Eppler, 1927; Boswell, 1927; Chudoba, 1935a; Chudoba and Dreisch, 1936; Gause, 1936; Lietz, 1937; Tomita and Karakida, 1954).
- (b) Coloration and changes of color by the irradiation of radio-rays (K. Simon, 1908; Brauns, 1909, 1928; Doelter and Sirk, 1910; Doelter, 1918; Boswell, 1927; Chudoba and Dreisch, 1936), of ultraviolet rays (Eppler, 1927; W. G. Simon, 1930; Lietz, 1937), and of infrared rays (Chudoba and Dreisch, 1936).
- (c) Thermoluminescence (Henneberg, 1846; Chandler, 1857; Hahn, 1874; Spezia, 1876; Stevanović, 1903; K. Simon, 1908; Strutt, 1914; Michel and Przibram, 1925; Eppler, 1927; Lietz, 1937). This phenomenon is intimately associated with the decoloration phenomenon (just before the decoloration) independently of the variety of color, and is not observed after the decoloration.
  - (d) Radioluminescence (K. Simon, 1908; Michel and Przibram, 1925).

The problem of color centers is nothing but the problem of lattice defects. Thirty years ago, Lind and Bardwell (1923) discussed the cause of the coloration and thermophosphorescence in transparent minerals (excluding hyacinth) and attributed the coloration to the absorption by the vibration ("with a frequency which may, and frequently does, fall into the visible region") of electrons displaced by irradiation from lattice positions to the space between. In an up-to-date experimental work

of coloring diamond by bombarding the crystal with deutrons and alpha-particles, Hamilton et al. (1952) also put forth the view of electron displacement. According to them, the displaced electrons "possess the property of absorbing energy from visible light and this energy is then re-emitted at a fixed frequency from these electrons," and the difference (or changes) in color is "not due to difference in the frequency of light emitted but rather to an ever-increasing intensity" of the re-emitted light; this intensity may depend upon the number of electrons displaced, which is more and more increasing with the lapse of time during which the mineral has undergone bombardment. Though put forth for the minerals other than hyacinth, these authors' view that the electrons are displaced to abnormal positions may well be accepted also in the case of hyacinth, I think.

Quite recently, three interesting papers concerning the age determination of zircon, metamict and non-metamict, pegmatitic and accessory, have been published successively (Kulp et al., 1952; Larsen et al., 1952; Hurley and Fairbairn, 1953). Of these three, the paper of HURLEY and FAIRBAIRN is a good guide to the study of lattice defects in zircons, though nothing about the color problem is touched upon. With a view to economizing space, essential points concerning lattice defects will be given as follows: (a) Zircon will be excited by atomic collision of alphaparticles radiating from radio-elements concealed in either zircon constituents or enclosed minerals within zircon; (b) these electronic excitations do not appreciably damage the structure, but produce free electrons and excitons; (c) the primary accelerated electrons which possess a considerable range will spread in a considerable volume; (d) more than 99 per cent of the energy of alpha-particle will be dissipated in exciting the electronic system, and the rest (less than one per cent) in atomic collision; (e) this atomic collision causes the disorder of structure characterized by isolated displaced atoms (interstitial atoms) and vacant lattice sites [Frenkel defect]; (f) the displaced atoms remain permanently out of lattice positions; (g) there is the distension of the unit-cell dimension due to disordering effect. In short, the effect of radio-irradiation resolves itself into the following two

In short, the effect of radio-irradiation resolves itself into the following two points: lattice defect and electron displacement. This radio-effect involves the problem of color-centers, the details of which are awaiting a further study. At present, however, I am promoting my geological studies on a working hypothesis which will be discussed in the next chapter.

It may here be remarked incidently that I have some doubt as to whether the "distension" of the unit-cell dimension is due only to disordering effect of atomic collision. In other words, I do not think there is not the least possibility that at least a small portion of the "distension" is due not only to the inherent presence of Th and/or U diadochically replacing Zr, but also to the presence of Pb a posterior, the disintegration product of the radio-elements. Thus, chemical studies are also required in our studies of the structure of hyacinth.

Author

(1923)

# V. Working Hypothesis Involved in the Zircon Method

For both geological and mineralogical reasons mentioned above, I cannot conceive that the variations in hue of hyacinth or transparent accessory zircon are due to the difference in the size and shape of zircon, the amount of enclosed minerals, kind and relative abundance of non-radioactive impurity-elements, or crystallization conditions (the temperature of zircon at its formation as well as the rate of cooling of magma carrying already crystallized-out zircon), but I believe the age (the length of time after its formation) is the most essential factor. So I am inclined to favor the view that the variations in hue is due to the number of color-centers which may probably be proportional to the number of electrons displaced, which last in turn may have been increasing with the lapse of time since the formation of accessory zircon in magma. This view will be discussed a little more fully below.

The experimental data concerning the coloration of zircon by radio-irradiation have been brought together from scattering sources and are given in Table 2. This table brigns out the following important facts:

Table 2. Experimental Data on the Coloration of Zircon

through Radio-irradiation Original Kind of Time of Coloring Radio-Source No. Locality Color Material Irrad. Effect

			1120101101					
1	Laacher See (the Eifel)	Light reddish brown- yellow	Decolored by heating	1 mg Ra*	ca. 1.5d	Faintly brown	CHUDOBA and DREISCH (1936)	
		Brown-red		1 mg Ra**	3d	Brown-red	BRAUNS (1909)	
2	Mongka (Thailand)	Rose-red	do.	1 mg Ra*	4-7d	Brown→ Brownish red → Rose-red	CHUDOBA and DREISCH (1936)	
3	Scotland	Rose-pink Purple	do.	β – and γ – rays	7d	Pale pink Pale purple	Boswell (1927)	
4	Tasmania	Yellow- brown to Brown-red	do.	10 mg Ra*	a few hours	Coloring began	K. SIMON	
4					a few days	Brown	(1908)	
5	Mongka	Colorless	Idiochro- matic	1 mg Ra*	2 mth	Pale brown	BRAUNS (1928)	
6	Artificial	do.	do.	4 mg Ra	4 mth	METAMIC- TIZED	von STACKEL. BERG-ROT- TENBACH (1940b)	
7	Pfitschtal (Italy)	Pfitschtal	do. do.	1 mg Ra**	10d	None	BRAUNS (1909)	
7		ao.		1/2	1/2g Ra**	23d	Gray to violet	DOELTER- SIRK (1910)
8	Artificial	do.	do.	1/2g Ra**	?	Violescent	DOELTER (1918)	
9	Natural	do.	do.	β - and	,	None	LIND and BARDWELL	

<sup>\*</sup> Ra-bromide

<sup>\*\*</sup> Ra-fluoride

- (1) In case of the same radioactivity, the longer the time of irradiation, the more remarkable the coloring effect (Nos. 1a vs 1b; 4a vs 4b).
- (2) By far the richer the radio-source, the lesser time is needed for nearly the same coloration (No. 1a vs 4a).
- (3) Combined action of superior radioactivity and a longer time gives otherwise insensible colorless zircon a violet color or pushes them to the furthest extreme, the metamict state (Nos. 7a vs 7b; 5 vs 6).

Judging from these three facts, it is quite natural to conclude that factors in the coloration of zircon through radio-irradiation should be radioactivity (alphaactivity) and the length of time during which the crystals have undergone bombardment. Generally speaking, there is the third factor: the inherent stability of the crystal structure. Since, however, zircon is one of the most structurally unstable minerals (Goldschmidt, 1924; Machatschki, 1941; Rankama and Sahama, 1950, p. 114), it is not required to take this factor into our account.

By the way, it is of geological interest to induce that the younger zircon of high activity will give the same coloration as the older of low activity and that in case of the equal or nearly equal activity the older zircon will be damaged more intensely than the younger.

- (4) Restoration of color in a decolored zircon seems to take place more rapidly than the first coloration of the originally colorless zircon (Nos. 1, 2 vs 5, 7a).
- (5) No greenish coloration has ever been observed in the experiments so far. Since the "green zircon" (low specific gravity, below 4.2) is metamict or semi-metamict (Damour, 1864; Grattarola, 1879, 1890; Stevanović, 1903a; Koechlin, 1903; Spencer, 1904; Eppler, 1927; Chudoba, 1935b, 1936, 1937a, 1937b; Gause, 1936; Kunitz, 1936; Chudoba and von Stackelberg, 1936a, 1936b; Kostyleva, 1936; von Stackelberg and Chudoba, 1937; Lietz, 1938; Bauer, 1940; von Stackelberg and Rottenbach, 1940a), a prolonged irradiation by low activity with a careful observation or a high-energy bombardment is required to verify the greenish coloration.
- (6) Various colors corresponding to the different intensities of radio-irradiation can be arranged in order of increasing effect as follows: Faintly brown, pale brown, brown, brown-red, rose-red, and violet. This order of changes in color ("color order") bears a close parallel to the natural color order seen in the granite zircon (Table 1), except the greenish one which is without doubt of the metamict state depending upon the relatively high contents of radio-elements concealed (Weigel, 1938).

This striking parallelism is so supremely important that it forms the ground of my zircon method. In short, the first category of the working hypothesis involved in the zircon method is that the color of granite zircon is due to the coloring effect of radioactivity inherent in them; in other words, it is due to the production of color-centers, one of the disordering effects of inherent radioactivity on the structure of zircon.

Next, it is assumed that the radio-elements must have been uniformly distributed in a magma body on the ground that the color of granite zircon does not show any notable difference in a single batholithic body. In connection with this assumption, it may be well to refer to Morgan and Auer's view (1941). They say, "The type of zircon present in a granite may be depedent on the quantity of uranium available at the time of crystallization of the zircon.......The concentration of uranium in granite magmas of one intrusive period may be similar over wide areas, since granite intrusive of similar time relationships can be distinguished by the zircon variety present in them. Such similarity in concentration of uranium may mean a common, deep-seated source for the magma" (p. 310).

Moreover, my method is established on another assumption that the amount of radio-elements in the granite zircon is very small and accordingly the difference in the radio-element contents between any two zircons of different ages is so slight as to be almost negligible. This assumption involves the idea of placing great importance on the length of irradiation-time as the factor to produce the color accordant with geologic age rather than the difference if any in the radioactivity of the accessory zircon. Since recent accurate analytical data covering the radio-elements of the granite zircon are not obtainable, I will refer to the old data furnished by Strutt (1910); namely, in accessory zircon,  $U_3O_8$  is less than 0.13 per cent (but 0.38 per cent for the Vesuvian colorless zircon which is remarkable by its bluish tinge) and ThO<sub>2</sub> does not exceed 0.08 per cent. These data, old as they are, point to the probability that the assumption is not unreasonable. Thus, the number of electrons displaced, to which the number of color-centers is probably proportional, must have been increasing with the lapse of time since the crystallization of granite zircon.

#### VI. The Pre-Cambrian in Japan

From the foregoing considerations, the following two facts will be pointed out: (1) So far as my patient experience dealing with not less than seven hundred specimens is concerned, no zircon undistinguishable from the Archean purple to rose-pink ones has been found in the Post-Archean granite-gneisses and granites, and (2) there is no reason to believe that these zircons can occur as the primary accessory minerals in the Post-Archean rocks. On these empirical and theoretical grounds, I do not feel the least hesitation in suggesting that the purple to rose-pink zircons are the "index minerals" of Archean, serving as "geologic clock."

Among the some seven hundred specimens of Japanese granites and granitic gneisses examined up to the last day of October in 1953, I have found sixteen Archean rocks, which are given in Table 3—so far, this Table is so incomplete as

to be by no means beyond criticism. Some of them came from the Hida Metamorphic Zone, and others from the sandwiched bodies in the Outer Zone of Southwest Japan, though the mode of occurrence of the Ôhira Granite (Fukushima Prefecture) is uncertain.

ie ei	Repre-	Color	G. 1.1.D.	Pre-Cambrian Rocks in Japan			
Geol. Age	senta- tives Zircon		Standard Rocks	Hida Metamorphic Zone	Sandwiched Bodies		
Algonkian	Killarney Granite	Colorless  Tellowish	Miyün Porphyrogranite (North China) Kung-Chuang-lingGranite (South Manchuria)				
	Animikee Granite	Cloudy brownish purple	Necedah Mound Granite (Wisconsin) Peitai Granite (North China)				
Younger Archean	Algoman Granite	Rose-pink  Rose-lilac	Taishan Gneissose Granite (North China)	Unazuki Granite Unazuki Augen- gneiss Unazuki Banded- gneiss Amô Granodiorite Mugishima Granite Dôgo Granite (Oki) Oki Augen-gneiss	Yatsushiro Gneiss (Near Yatsushiro City, Kumamoto Prefecture) Nabae Gneiss (Near Yuasa, Wakayama Prefecture) (?) Ôhira Granite (Fukushima Prefecture)		
Older Archean	Laurentian Granite-Gneiss	Purple  † Dark purple  Purple black	Bangalore Granite-gneiss (Peninsular India) Necedah Mound SS-quartzite (Wisconsin) Shinheidô Ls-contact (Northern Korea)	Oki Granite-gneiss			
	Grenville Leptite Formation	Colorless  Grayish yellow  Brown- yellow	Sankang SS-granulite (Shanhsi, North China)  Colombo Acid Granulite (Ceylon)	Oki Para-gneiss			
	Lei	Orange	Sankang SS-granulite				

Table 3. The Pre-Cambrian in Japan

Now I will make a few passing remarks about the geological and petrological problems connected with the Archean in Japan.

(A) As to Dōgo, Oki Islands (Tomita, 1927; 1936, pp. 80-82). In my previous paper (1936), the Oki Gneiss was correlated with the Hida Gneiss, which was considered to be of Post-Paleozoic and Pre-Jurassic time, the current view in those days. As to the Dôgo Granite (Post-Gneiss and Pre-Miocene), it is pointed out that there was an interval of time between the formation of the Gneiss and the intrusion of

the Granite with the words "a relatively small interval of time." Now I confess that I had my doubts about my own words "relatively small," because I had no positive proof of it. I dare say this view is due only to the fact that some parts of the Gneiss massive are of undoubted lit-par-lit injection-gneiss.

At present, these previous views of my own as to the time relationships ought to be abandoned. The new determination of geologic ages of the gneiss and granite in question has brought me a comprehensive interpretation of the genesis of the gneiss. Thus the Oki Gneisses are composed of three types of gneisses of different ages and origins: the oldest (Tyôshidani, Fuse-minamidani, and between Iibi and Shimoganya) is a para-gneiss, including graphite gneiss, of Older Archean time, the second (Fuse) is a granitic gneiss of the Older Archean, and the youngest (Tyôshidani) is of augengneiss (migmatitic eyed-gneiss) of Younger Archean age. The Dôgo Granite (Fuse-minamidani and Iibi) intruded into all of the gneissess is of the Younger Archean, having no genetic relationships with the Older Archean gneisses, though some polymetamorphic effects of the Granite on the older gneisses cannot be denied.

From my personal experience, I cannot too strongly advise the greatest necessity of the determination of geologic age of plutonic rocks and their associated gneisses for the petrogenic studies of plutonism.

(B) As to the Hida Highlands. So far as my examination of accessory zircon goes, no Older Archean rocks have yet been detected in the Hida Metamorphic Complex. To confess the truth, my examination has not covered all kinds of the Hida Gneiss, and this is the reason why the name of this gneiss finds no mention in the column for the Older Archean in Table 3.

In fact, there is a very fair possibility of the presence of the Older Archean in the Hida Highlands, because I have found abundant purple zircons in the Akaishi Sandstone (upper part of the Tetori Group; Lower Cretaceous), the materials of which must have come from the Highlands.

It may be well to note here that in the Highlands there are also the Lower Permian granitic rocks. They are the Shimonomoto Granodiorite and the Funazu Granite, both situated in the east of the metamorphic province. Gneissose rocks adjoining these granitic rocks have undergone injection metamorphism. This indicates that the so-called Hida Metamorphic Complex is of polymetamorphic rocks generated not only during the Pre-Cambrian Era but also at Lower Permian time, and that the oldest gneiss, if present, must possess every trace of each metamorphism (possibly more than two during the Pre-Cambrian Era).

(C) As to the Sandwiched Bodies in the Outer Zone of South-west Japan. Since Matsumoto and Kanmera (1949, pp. 78-79) have called attention of the existence of the "sandwiched" bodies including gneissic rocks, which are believed to have been "sqeezed out" upward into among younger geologic members by severe

faulting movement, the age of the gneissic rocks has been demanded to be decided. But for my zircon method, however, the demand could not have been met yet.

Up to the present time only two localities of the Archean rock bodies have been recognized as shown in Table 3. Besides these, somewhat similar igneous bodies are found in the middle zone running across Kyûshû and Shikoku. The geologic ages of these igneous rocks will be determined as my study advances. Anyhow, the Archean sandwiched bodies are the representatives of the deep-seated terrane, indicating that the Japanese Paleozoic formations do not "rest on nothing" as formerly said, but on the Archean basement.

(D) Sedimentary Rocks that Contain the Archean Zircon. In order to secure the data for age determination of granitic rocks, many sandstones and pebbles of conglomerate beds have also been examined. In the course of these researches, it has been found: (1) that the Older Archean purple zircon is present in the gneissose pebbles of a conglomerate bed of the Momonoki Formation (Mine Group: Carnic), in the sandstone of the Toyonishi Group (Lower Cretaceous), in the Akaiwa Sandstone (Upper Tetori Group: Lower Cretaceous), and in some Paleocene sandstones in North Kyûshû, and (2) that the Younger Archean rose-pink to rose-lilac zircons are present in the granitic or gneissose pebbles of the Ichinashi Conglomerate-schist (Middle Gotlandian) and of the Murakami Conglomerate (Upper Devonian) as well as in those of the conglomerate beds of the Kuma Formation (Upper Permian), Shiraiwa Formation (Tsunemori Group: Upper Permian), Miharaiyama Group (Skytic), Hinaku Formation (Lower Cretaceous), and the Tomochi Formation (Middle Cretaceous).

From these new data, it is concluded that the Archean zircon is widely distributed in Japanese sedimentary rocks from Middle Gotlandian (the former oldest formation in Japan) to Tertiary, presenting some new, complex problems for solution in paleogeography and structural geology of the Japanese Islands.

# Concluding Remarks

My work presented in this paper is so elementary that no quantitative data have been given, but the fact I found that the color of accessory zircon of granite and gneiss depends principally on the age of the mineral will be a subject eminently worthy of further careful study. I am in hopes that many granite zircons of different ages in foreign countries would be examined and that my method would be revised and developed by any person who is interested in this line of research.

By the way, to recognize a difference in color is one thing, and it is another to describe it. Difficult though it is to describe the colors of zircon, it is a rather easy and rapid practice for the experienced eye to recognize a difference in color between zircon crystals, and for promoting the spread of this method it is desirable to make

"Zircon Scale" (a standard collections of accessory zircons of different ages) so familiar among us as is Mohs scale of hardness in mineralogy.

In addition, it is to be noted that a living discussion is needed on my working hypothesis as to the cause of the color of hyacinth. Setting aside my crude hypothesis, however, the zircon method newly introduced into our geologic science, though some cautions still are required, is practical and effective for every geologist who is obliged to deal with a good many rock-specimens in a limited time, and especially for those research workers who are attached to poorly-equipped laboratories in moderate circumstances.

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