Physical And Chemical Differentiation Of West Malaysian Limestone Formations

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Abstract: A large and representative collection of specimens from each of the Malayan limestone formations was analysed chemically, mineralogically and petrographically in an attempt to find a criterion for differentiation independent of palaeontology. Limited success has been obtained only where the limestones are sufficiently far removed from granitic intrusions—in Perlis and Kedah, where significant lithological differences occur between limestones of different formations. Unfortunately the results cannot be extended to other areas and no generally applicable criterion for differentiation has been found.

Extensive studies of the thermoluminescence of limestones show that all collected specimens have been recrystallised during the revolutionary phase of the Thai-Malayan orogeny so that thermoluminescence cannot be used in Malaya to differentiate limestones stratigraphically.

It must therefore be concluded that the differentiation of Malayan limestones must continue to be based on fossils.

"Bright Phosphor, fresher for the night."—Tennyson.

INTRODUCTION

Limestones outcrop in many scattered localities in the northern half of West Malaysia (fig. 1). They range in age from Ordovician to Triassic. Generally the limestone is a white, pale grey or slightly yellowish recrystallised rock. In some places it is dark grey to almost black because of carbonaceous or argillaceous impurity. More rarely it is red because of hematite inclusions, or by iron staining. Generally, however, Malayan limestones are remarkably pure. At numerous localities, grey or black bands, due to minute carbonaceous inclusions, occur in white limestone and in many cases these bands, which are often stylolitic (Hutchison, 1963), represent the original bedding. Three main outcrop areas which have been chosen for this study are indicated by arrows on figure 1. In addition, several specimens were collected from most of the limestone areas throughout Malaya (fig. 1).
In the Perlis-Kedah area, the limestones are differentiated into the Setul Limestone (Ordovician-Silurian) and the Chuping Limestone (Permian) (Jones, 1961). The stratigraphy of this area is reasonably reliable and the limestone ages are firmly based on fossil evidence (Jones, in manuscript) (fig. 2).

In the Kinta Valley, it is known from fossil evidence that limestones of Ordovician-Silurian, Middle Devonian, Late Carboniferous, and Lower Permian ages occur (D. J. Gobbett, personal communication; Suntharalingam, 1968) (fig. 3A).
Fig. 2. Sketch geological and limestone specimen locality map of Perlis and Kedah, northwestern Malaya.
Fig. 3. Sketch geological and limestone specimen locality map of (A) the Kinta Valley, Perak, and (B) the Kuala Lumpur area, Selangor. The positions of these areas in Malaya are shown in figure 1.
In the Kuala Lumpur area, the limestones appear to be confined to the Silurian (Gobbett, 1964) (fig. 3B).

The stratigraphic thicknesses of the limestones of Lower Palaeozoic age have been estimated as 7,500 feet in the Langkawi islands (Jones, 1961) and 6,000 feet in the Kuala Lumpur area (Gobbett, 1964). An accurate estimate of the thickness of the Upper Palaeozoic limestones is not available.

The limestone formations have presented a problem to the Malayan stratigrapher in that they were largely found to be unfossiliferous. Up till 1956, "the practice has been to assume that anything calcareous is Carboniferous" (Scrivenor, 1931), and all limestones were ascribed to the "Calcareous Series", which was formerly known as the "calcareous formation" (Richardson, 1950). Wherever calcareous rocks were mapped in Malaya, they were ascribed to this "Calcareous Series" purely on the basis of their being "lithologically similar to fossiliferous strata... elsewhere" (Roe, 1951).

Since 1956, discoveries of Ordovician-Silurian graptolites in shales inter-bedded with limestones, and of Lower Palaeozoic brachiopods in the limestones themselves, in the Langkawi Islands (Jones 1959), in Perlis (Jones, in manuscript), in the Kinta Valley (Ingham and Bradford, 1960) and in the Kuala Lumpur area (Gobbett, 1964), have made the term "Calcareous Series" obsolete. Limestone areas which were formerly held to be Permocarboniferous, through lack of fossil evidence, have recently been re-classified; some as Ordovician-Silurian (Jones, 1961), some as Silurian (Gobbett, 1964), some as Devonian (Alexander & Müller, 1963), and others as Triassic (Ishii & Nagami, 1966).

The classification of one large discrete area of limestone as being of a single age is usually based on a fossil discovery in a small part of the area. As an example, the 1 : 63,000 provisional (pre-publication) geological map of the Kuala Lumpur area (Yin, 1962) shows the main limestone mapped as the "Hawthornden Formation" as being of middle Silurian age. Yet fossil localities are very restricted in this area. Subsequent discoveries of fossils in different parts of an area, which may initially have been thought to belong to one formation, often have shown that the initial classification and stratigraphy require modification. A good illustration of this point is offered by the recent discovery of Triassic conodonts (Ishii & Nagami, 1966) in the limestone of Bukit Kalong and Kechil, Kodiang, Kedah; which limestone had been taken by Jones (in manuscript) to be Permian of the Chuping Formation.

Limestone stratigraphy in Malaya has been and is dependent almost entirely on palaeontology and fossil localities are sporadic, with the result that a new fossil discovery may mean the necessity for a complete re-appraisal of the stratigraphy. Furthermore, correlation on lithological grounds has led to serious mistakes of stratigraphic interpretation.

For these reasons, I felt that any attempt to date or to differentiate limestones in West Malaysia by a method independent of palaeontology would be worth making. Attempts were made on the petrology and chemical composition and on the non-soluble residues. Although these are applicable for differentiation of the limestones to a limited extent in the Perlis area, no universally valid criterion was discovered which had any wider geographic or stratigraphic significance.
A method of radiometric dating based on the thermoluminescence of calcite was investigated in detail. The results have shown that the Mesozoic orogeny has so obliterated the thermoluminescence of Malayan limestones which is related to their stratigraphic age with the result that only the tectonic ages, and not stratigraphic ages, can be ascertained. I therefore conclude that for limestone stratigraphy in Malaya, there is apparently no substitute for palaeontology.

CHEMICAL COMPOSITION

184 limestone specimens were analysed for calcium, magnesium and insoluble residue using basically the method of Archer, Flint and Jordan (1958), an undated manual by E. Merck, and Bisque (1961), but incorporating modifications after Morris (1961) and Lewis, Nardozzi and Melnick (1961). The detailed procedure is fully described in Hutchison (1966).

The results of the chemical analyses are shown in histogram form in figure 4. The specimens were collected in as random a manner as possible, therefore the frequency distribution of figure 4 should give a rough indication of the relative proportions of limestone and dolomite in Malaya. The distribution indicates that 81% of the collected samples are limestone, and 19% dolomite.

Fig. 4. The frequency distribution of calcium ions as a percentage of the total calcium + magnesium in 184 analysed limestones.
The limestones have a distinct compositional mode of around Ca 99 cationic percent, with a computed arithmetic mean of 97.0 Ca cationic percent. The dolomites have a mode of around Ca 54% with a computed arithmetic mean of 56.2 Ca cationic percent.

Several attempts to correlate the dolomite localities with age of the formation, or with geography, have failed. The occurrence of dolomite throughout Malaya is apparently very patchy and sporadic with no easily discernable pattern. Perhaps the most obvious relationship might be expected with the granite outcrop pattern, but no definite correlation could be made. Several dolomite localities abut the granite, but others are several miles from the nearest outcropping granite. One can conclude that dolomitization is a secondary process, not obviously related to the age of the formation or to geographic position, but perhaps related to the emplacement of granite during the revolutionary phase of the Thai-Malayan orogeny.

Localities where dolomite has been determined are marked in figures 2 and 3. These occurrences are in all formations with the single exception of the Setul Limestone, which alone appears to be free from dolomitization.

Twelve specimens of secondary calcite veining and of stalactitic limestone were analysed, giving an arithmetic mean value of 99.2 Ca cationic percent with a range from 100 to 95. Secondary veining is therefore remarkably pure and free from Mg compared with the main limestone in which it occurs.

PETROLOGY

In hand specimen the limestones vary from fine-grained black argillaceous-carbonaceous limestone (1723*, locality 9), to white coarsely crystalline marble (1690, locality 66), and also include buff coloured fine grained marble (1682, locality 30), fine grained pale grey saccharoidal marble (1705, locality 72), pale reddish fine grained marble with hematite inclusions (1744, locality 30), light grey crystalline limestone (1741, locality 29), and pink coarsely crystalline marble (1800, locality 49); but the most common variety is the white or pale grey coarse to fine grained marble, while black limestones are locally important, especially in the Setul Formation. All limestone specimens have been found to be recrystallised: many occur as recrystallised breccia. The generalisation may be made that Malayan limestones are in fact marbles. Sedimentary structures have been almost completely obliterated by recrystallisation, except perhaps for some relict bedding which is accentuated by bands of darker colour or by stylolitic bands which follow the bedding (Hutchison, 1963).

Thin sections always show the typical crystalline texture of marble. The purity is remarkable and generally thin sections show nothing but pure calcite with the occasional exceptions of localised dolomites (1703, locality 72; 1773, locality 34). The identification of dolomite in thin section was greatly aided by using the modified staining technique of Dickson (1965).

* Numbers of specimens housed in the Geology Department of the University of Malaya.
The exceptional purity of Malayan limestones is illustrated by the frequency distribution of insoluble residues of the 182 analysed samples (fig. 5). There is a pronounced mode at about 2.5% insoluble residue. The computed arithmetic mean is 4.4% insoluble residue for all analysed specimens.

Fig. 5. The frequency distribution of insoluble residues, in weight percentage of the whole specimen, of 182 analysed limestone samples.

In the Perlis-Kedah area (fig. 2) the Setul Limestone can be differentiated from the Chuping Limestone in that the Setul is black in hand specimen. Thin sections show a disseminated opaque argillaceous or carbonaceous material. The insoluble residue of these black limestones is a black carbonaceous clay-grade material which is retained on the filter paper simply as a black stain. The Chuping Limestone normally leaves no insoluble residue, but occasionally there are numerous silicified remains of fossils, especially crinoids. Local silification often leads to complete replacement of the fossils and sometimes of the whole rock (1743, locality 29). Such limestones are characterised in outcrop by sharp jagged edges and irregular weathered surfaces. The distinct differences in purity between the Setul and Chuping limestones is well displayed in figure 6, which shows the frequency distribution of insoluble residues of 20 Setul specimens and 23 Chuping specimens. The Setul specimens give a rather variable distribution from 1 to 16% insoluble residues with an arithmetic mean of 7.4%, whereas...
the Chuping specimens gives a distinct mode in the range 0 to 3% with an arithmetic mean of 2.3%. The difference of purity between the two formations is well reflected in their contrasting geomorphology in the Perlis area: the Setul Limestone is more resistant to erosion and forms a continuous range of hills, whereas the Chuping is more easily eroded and forms a line of isolated limestone hills.

The petrographic distinction between the two limestones which occur in Perlis-Kedah is not applicable elsewhere. The Lower Palaeozoic limestones of the Ipoh and Kuala Lumpur areas are white in colour and pure in composition, more resembling the Chuping than the Setul in appearance. Colour or any petrographic character is therefore of no general guide to the differentiation of limestones, except in the limited area of Perlis-Kedah, and to some extent in the Langkawi islands.

Variable mineralogy as a result of metamorphism is common in the areas of Malaya close to the granite intrusions. Regional metamorphism has generally given a pure marble because of the chemical purity of the limestone. However, thermal metamorphism associated with metasomatism at the granite contact has given localised garnet and vesuvianite skarns in Langkawi and the production of phlogopite and chlorite marbles in the Kuala Lumpur area. In the Ampang area near Kuala Lumpur (fig. 3), periclase and brucite are common minerals in the metamorphosed dolomite. Excellent euhedral cubes of pyrite are widely found in Malayan marbles.
STUDIES IN MALAYSIAN GEOLOGY

THERMOLUMINESCENCE

Seventy carefully selected specimens were measured for thermoluminescence, using the method of Hutchison (1965), in an attempt to determine whether or not the method can be used in Malaya to determine the stratigraphic age and hence to differentiate the limestone formations.

The $R_a$ value, or thermoluminescence index, is directly proportional to the stratigraphic age of the specimen, or to the time that has elapsed since the calcite or dolomite crystals in the limestone have crystallised or recrystallised. $R_a=$the relative equivalent radiation dosage as determined from a calibration curve, and $\alpha =$the alpha activity of the limestone specimen, in counts per hour, as measured in a scintillation counter (Hutchison, 1965).

The results of alpha counting

Each of the 70 specimens which were measured for thermoluminescence was also measured for alpha activity over a period of 2 days in a scintillation counter (Hutchison, 1965, 1966). The alpha activity level of the limestones of the Perlis-Kedah area (fig. 2) gives an example of how this kind of data can be employed in addition to its necessity for calculating the thermoluminescence indices. Fourteen measured specimens of the Setul Limestone gave a mean alpha activity of 6.5 counts per hour, but with individuals ranging from 4.5 to 9.3. Twelve measured specimens of the Chuping Limestone gave a mean alpha activity of 5.2, with individuals ranging from 1.9 to 14.4. In the Perlis-Kedah area, therefore, it can be seen that the Setul Limestone generally has a high radioactivity level compared with the Chuping. However, individual values deviate more from the mean in the Chuping than in the Setul. The higher radioactivity level of the Setul reflects the fact that Setul limestones contain an argillaceous impurity.

The results of thermoluminescence indices

The determined thermoluminescence indices of the Chuping Limestone (Permian) are not smaller than those of the Setul Limestone (Ordovician-Silurian). Therefore, the thermoluminescence indices in the Kedah-Perlis area (fig. 2) are not related to stratigraphy! Folding and recrystallisation during the revolutionary phase of the Thai-Malayan orogeny has drained from the Setul Limestone any 'pre-Chuping' thermoluminescence. Results from the Kinta Valley and from Lower Palaeozoic limestones from the Kuala Lumpur area (fig. 3) give even younger thermoluminescence indices than for the Chuping Limestone. It is obvious, therefore, that the thermoluminescence indices are related not to stratigraphic age but to orogenic phenomena, including folding, recrystallisation and the heating effects of orogenic granites. Thermoluminescence methods cannot therefore be used in Malaya to stratigraphically differentiate the limestone formations!

The results of the thermoluminescence studies have given considerable information regarding the revolutionary phase of the Thai-Malayan orogeny. These results, together with the tabulated thermoluminescence indices, alpha activity counts, and detailed method, have been accepted for publication in two separate parts (Hutchison, 1968, a & b).
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REFERENCES


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**DISCUSSION:** J.H. Leow asked if colour changes caused by the rising temperature of the sample affected the results in thermoluminescence studies. The speaker replied that this did not affect the results as they were all relative to a standard model.