PETROLOGY AND GEOCHEMISTRY OF THE LANGKAWI GRANITES

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KUALA LUMPUR

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Petrology and Geochemistry of the Langkawi Granites, North of Peninsular Malaysia

ABSTRACT

The Langkawi Islands comprises a group of 99 tropical islands lying off the northwest coast of Peninsular Malaysia. The Langkawi granite makes about 40 % of the Langkawi. It can be divided into two main bodies: Gunung Raya and Kuah granites. The Gunung Raya biotite granite shows very coarse- to medium-grained texture, some are strongly porphyritic with K-feldspar phenocrysts reaching 6 cm in length whereas the Kuah granite consists of highly evolved fine- to medium-grained tourmaline granite. Tourmaline occurs in both Gunung Raya and Kuah granite. The tourmalinization is clearly observed in the Kuah granite. Geochemically, the Kuah granite (75.1-77 %SiO_2) has more siliceous and evolved magma than the Gunung Raya granite (68.1-75.3 %SiO_2). The highly evolved magma for the Kuah granites is also supported by high Rb/Sr ratio (~20.55), compared to the Gunung Raya granite which is much less it is differentiated (~7.84). The distinct negative anomalies for Nb, Sr, and Ti are showing typical upper crustal compositions. In addition, the negative anomalies in Ba, Nb, Sr, and Ti that become increasingly marked towards the most felsic and fractionated granitic facies: such trends are typical of collisional peraluminous granitoids. All granites have ASI >1 which is peraluminous. Both the Gunung Raya and Kuah granite show fundamental S-type feature like peraluminous composition, high K_2O / Na_2O ratio (~1.56 %), low total Fe and restricted composition range dominated by high SiO_2, normative corundum > 1%. Strongly negative Eu anomaly is normally subscribed to fractionation of plagioclase due to the similar ionic radius and charge of Eu^{2+} and Ca^{2+}. The rare earth element (REE) pattern of the Langkawi granite constitutes familiar birds-wing REE pattern and strong negative Eu anomaly which is similar to the dominant S-Type granites of the Main Range. Major and trace elements Harker diagram and large ion lithophile (LIL) modeling suggest that the Gunung Raya and Kuah magmas were derived from the different magmatic pulses. Both pluton are controlled by the same mineral assemblages (plagioclase + K-feldspar and biotite) suggest that they may be formed from the same magmatic source. The Langkawi granite has K - Ar and Rb - Sr dates (217 ± 8 Ma and 209 ± 6 respectively). The Late Triassic age suggested that the granites may falls within the syn-collisional granites. The S-type characteristic associated with the tin mineralization clearly indicates that it is related to the Indosinian Orogeny.

Petrologi dan Geokimia Btuan Granit di Langkawi, Utara Semenanjung Malaysia

ABSTRAK

Langkawi terdiri daripada gugusan 99 buah pulau tropika yang terletak di luar pantai barat laut Semenanjung Malaysia. Batuan granit merangkumi 40% daripada Pulau Langkawi yang boleh dibahagikan kepada dua pluton utama iaitu Gunung Raya dan Kuah granit. Biotit granit Gunung Raya menunjukkan butiran yang sangat kasar ke sederhana halus. Terdapat batuan biotit granit Gunung Raya yang menunjukkan tekstur porphyritic dengan K-feldspar sebagai fenokris mencapai 6 cm panjang. Kuah terdiri daripada granit turmalin dan menunjukkan butiran sederhana ke halus. Selain itu, terdapat juga sebilangan biotit digantikan oleh muskovit dalam granit Gunung Raya. Tourmalinization ini jelas diperhatikan dalam granit Kuah. Secara geokimia, granit Kuah (75.1-77 % SiO_2) mempunyai magma lebih bersilika dan berevolusi daripada granit Gunung Raya (68.1-75.3 % SiO_2). Nisbah Rb / Sr tinggi (~ 20.55) yang ditunjukkan oleh granit kuah menunjukkan magma berevolusi dengan pesat manakali Granit Gunung Raya pula menunjukkan differensiasi yang kurang pesat (~ 7.84). Anomali negatif yang jelas untuk Nb, Sr, dan Ti menunjukkan komposisi biasa seperti kerak benua atas. Di samping itu, anomali negatif dalam Ba, Nb, Sr, dan Ti yang menjadi semakin ketara ke arah komposisi yang paling felsik adalah tipikal bagi granit yang terbentuk semasa perlanggaran antara kerak . Semua granit mempunyai nilai ASI> 1 yang menunjukkan sifat peralumina. Sifat granit yang peralumina, nisbah K_2O/Na_2O yang tinggi (~1.56), jumlah Fe yang rendah, normative aluminium oksida > 1 % telah mencirikan kedua-dua granit Kuah dan Gunung raya adalah granit jenis S-jenis. Anomali negatif Eu yang hebat biasanya menunjukkan fraksinasi plagioklas kerana jejari ion Eu_2^+ dan Ca_2^+ yang sama. Corak REE granit Langkawi menunjukkan corak ‘sayap burung’ dan negative anomali Eu yang hebat menunjukkan ciri dominan yang sama dengan granit main-range Jenis-S. Elemen-elemen utama dalam gambarajah Harker dan model ‘Large Ion Lithophile’ menunjukkan bahawa Gunung Raya dan Kuah magma berasal daripada magmatik yang berbeza. Kedua-dua pluton ini dikawal oleh mineral-mineral yang sama (plagioklas+K-feldspar and biotite) menunjukkan punca magma yang sama. Granit Langkawi mempunyai umur K-Ar dan Rb-Sr (217 ± 8 Ma dan 209 ± 6), iaitu Triassic lewat dimana ia terbentuk semasa syn-perlanggaran (syn-collision). Ciri-ciri granit Jenis-S yang berkait rapat dengan mineral bijih timah itu jelas menunjukkan bahawa ia adalah berkaitan dengan Orogen Indosinia.

UNIVERSITI MALAYA

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LIST OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

CHAPTER 1

INTRODUCTION

1 .1 Objectives

The aims of this project are:

(1) to provide a systematic study of the geochemistry, petrology and mineral chemistry of the Langkawi granites,

(2) to model the evolution of the granites : each of the pluton will be modeled separately using the major, trace and REE data,

(3) to classify the granites : the granites will be classified by using the different criteria of granite classification,

(4) to correlate and compare the Langkawi granites with the Main Range granites of Peninsular Malaysia,

(5) to determine the placement of the Langkawi granites into a proper tectonic setting.

1.2 Location and Accessibility

Langkawi comprises a group of 99 tropical islands lying off the northwest coast of Peninsular Malaysia, about 30 km from Kuala Perlis and 51 km from Kuala Kedah on the mainland. The mainland is known as Palau Langkawi. This area is approximately 85 sq. miles (200 sq. km of land surface) bounded by Latitudes N 6° 21.928' and Longitude E 099° 46.661'. The Langkawi Islands can be reached directly by air and car, from Kuala Lumpur to Kuala Kedah and Kuala Perlis. It can also be reached by ferry from Kuala Kedah and Kuala Perlis, taking about one and half hours to reach Kuah Jetty Langkawi. On the main island, road communication is smooth because of the numerous tar roads that run through the area. The inter island communication is by means of fishing and private boats.

Fig. 1.1 : Location map of Langkawi Island.

1.3 Methods of study

1.3.1 Previous work and literature review

J. B. Scrivenor, a geologist visited Langkawi islands in 1919 and 1920. His assistant, E.S. Willbourn re-visited the area at 1922 for additional information. They made many geological investigations in the area and their work was published in a joint paper accompanied by a geological sketch map (Scrivenor and Willbourn, 1923). After their work, there is a hiatus in the Langkawi geology literature until 1963. Hutchison and Leow (1963) described the granites exposed at the coastal area of the Langkawi islands. They also noted the abundance of developing hypothermal dikes cross cutting the granite batholiths.

Although the granites are extensively exposed in central and northwestern part of the Langkawi Islands, little has been published on the petrology and geochemistry of the Langkawi granites. Jones (1981) reported the general geological description of Mesozoic – granite and other igneous rocks in the memoir, which deals with the Geology and Mineral Resources of Perlis, North Kedah and the Langkawi Islands. It is included a one-inch geological map (sheet 150) of Langkawi Islands.

Khoo (1981) made a discussion on the reinterpretation of the earlier version of Hutchison and Leow (1963) regarding the tourmaline greisenization in Langkawi using the available composition model. A general report of the granites prepared by Wan Fuad Wan Hassan (1999) is documented in Warisan Geologi Malaysia (Ibrahim Komoo & Mohd Shafeea Leman, 1999).

A detailed report on the subdivisions of the Langkawi granites on the REE patterns is provided by Wan Fuad Wan Hassan et al., (2001). They divided the Langkawi granites into two groups including the Gunung Raya-Dayang Bunting, Kuah granites and the Pulau Tuba granite. Their report also includes comparison in geochemistry between two groups such as Rb/Sr ratios and degree of partial melting and fraction and a possible source material for the Langkawi granite.

There are a small number of undergraduate theses carried out on the aspects of general geology of the Langkawi Islands (Miskin Fakir Mohd, 1963; Foo, 1963; Fuad Bin Wan Hasan, 1973; Encik Aris Yub, 1981; Haron Abdul Ghani, 1982; Kamarudin Salim, 1982; Soo Meng Fook, 1985; Ku Aziz Bin Ku Sulaiman, 1986; Hairunnizam Mohammed Salleh @ Tajudin, 1996). Kamarudzaman Bin Lokeman (1996) completed his B.Sc thesis on the petrology and geochemistry of granites from the Teluk Ewa area, Langkawi Islands.

There were no further works reported in the field of petrology and geochemistry of the Langkawi granites between 1997 and 2005. In 2006, an undergraduate research work on the geochemistry of the Langkawi granites was carried out by ImanIdrus (2006). He pointed out that the igneous rocks from the Langkawi Islands are included in the granite category as indicated by the geochemical analysis.

1.3.2 Field Work

A topographic map at a scale of 1:63,360 is used as a base map. The fieldwork includes recognizing the types of rock from the representative outcrops, collecting samples from the fresh surfaces, recording necessary data such as the locations where samples were taken. It also involved observing, sketching geological features and characteristics and also the texture of the granitic rocks, recognizing minerals present in the rocks, taking photograph of the area of interest, and studying the general geology of the area. Rock samples were taken from outcrops at roadside, hillside and also from a quarry. Samples collected from 6 locations (30 samples) in the field were wrapped in newspaper and placed in a tough plastic bag, labeled with GPS location(Garmin Oregon 550) and marked the nearest location on the both samples and plastic bag with a black water-resistance felt-tip pen. At the same time, samples were located on the map and their description recorded in the notebook as follows:

L1,L2 (L- location) - N 06° 21 ,636' , E099° 41 , 780' (near Pantai Kok)

Photo No: - DSC 00018 (from GPS built in camera)

Brief Description - Biotite Granite (Feldspar, Mica and Tourmaline). Mica can be divided in to biotite and muscovite and most minerals are visible, as well as tourmaline. Basal part of the outcrops is weathered.

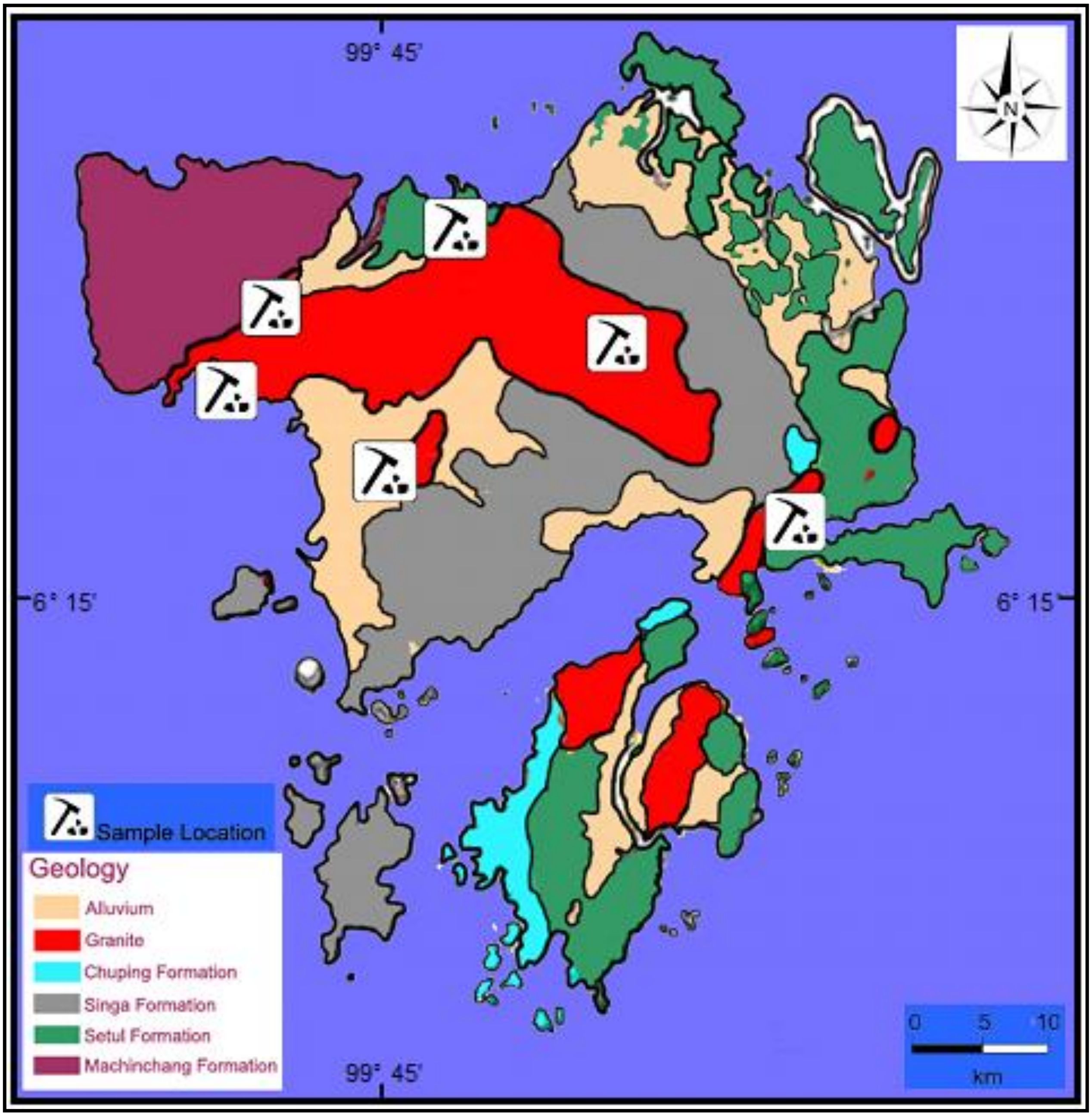


Fig. 1.2 : Sample location map of Langkawi Island.

1.3.3 Laboratory work

The collected granite samples were made into thin section for microscopic study. Thin sections of the samples were prepared to study in detail describing the minerals present in the rock samples. The data analysis for major, trace elements and rare earth elements were carried out by using the following steps and procedures:

(1) Heating

Rock samples of sizes (5-8 cm) and weighing 2-3 kg were put in the oven (temperature 70ºC) overnight for the purpose of quicker disaggregation

(2) Crushing

The samples are then crushed into smaller sizes (up to half a centimeter) using the Jaw Crusher.

After that the samples were milled to 200 mesh and then thoroughly mixed using a tungsten carbide swing mill. The rock pulps of each sample are ready to analyze for major, trace and rare earth elements (REE). Major and trace elements compositions were determined at ACME Labs Vancouver, Canada.

1.4 Physiography

1.4.1 Topography

The islands consist mainly of rugged mountains. This terrain is obvious especially in the areas of limestone bedrock. There are narrow sandy beaches with low ridges present along the south west coast of Langkawi Islands. The highest points are located in the centre of the area and attain heights of at least two thousand feet. A couple of granite batholiths are exposed in the central and eastern parts of the area. The Gunung Raya extends northwards and westwards covering an area of about 30 square miles. The northern portion of granite which forms a 900m high ridge is separated from the central mountain region by a narrow valley through which runs a path. Alluvium covers the valleys between the elevated areas.

The rock-type are granites, shale and sandstone (some of the shale and sandstone are metamorphosed to slate and quartzite) and metamorphosed limestone. The granite forms the western part of the area. They appear hilly with a rounded topography rising to 2890m (as in Gunung Raya the highest point on the island). Smooth granite cliffs up to 100m high give rise to impressive water-falls, especially along the eastern slopes of Gunung Raya. The slate and hornfels produce a more craggy topography. They are well-jointed and also form steep and high water-falls, the highest of which is about 150m.

1.4.2 Climate and Vegetation

The area is under the seasonal influence of the two monsoons. The first season that has an important effect on the vegetation is comprised of three months of dry weather lasting from January to March. This is the period of the north-east monsoon with the average monthly rainfall in January and February hardly exceeding 2 inches. Two wet seasons occur during the change of monsoons when monthly rainfall average is over 9 inches. A dried period runs between these two wet seasons. The average temperature is more uniform with a main daily maximum of about 88° F and a mean daily minimum of 74° F. The temperature fluxes are influenced by the sea. The area is part of big exposures of virgin jungle, partly untouched. Most of the jungle is tropical typically evergreen rainforest. Part of the forest has been cleared to cultivate rubber, coconuts and other crops. The alluvial areas at the middle of Langkawi Islands have been drained to grow rice.

CHAPTER 2

GENERAL GEOLOGY

2.1 Regional geology of the Peninsular Malaysia

Peninsular Malaysia has been divided into two blocks by a suture (Stauffer, 1974; Hamilton, 1979) (Fig. 2.1):

(1) Western Block

(2) Eastern Block

Stauffer (1974) named these blocks the East and West Malaya Blocks and Hamilton (1979) used Western and Eastern Peninsular Blocks. Peninsular Malaysia forms part of the Sunda Shelf. Cenozoic deposits are all superficial and relatively thin except at the costal margin. The origin of rocks in Peninsular Malaysia shows significant continuation in their deposition. The pre-Triassic rocks are characteristically marine meanwhile, Triassic rock are made up of both marine and non-marine deposit. Furthermore, all post-Triassic rocks are non-marine in origin (Gan, 1980).

Almost half of the total surface area of Peninsular Malaysia is covered by granitoids. There are at least four major episodes of granitic emplacements shown by radiometric dating, ranging from Upper Carboniferous to Lower Tertiary (Rajah and Yin 1980). The emplacement peaks are during the Permian and Triassic periods (Bignell, 1972; Hutchison, 1973), and contemporaneous submarine extrusive activities from the Upper Cambrian to the Middle Triassic characterized the geological history of Peninsular Malaysia.

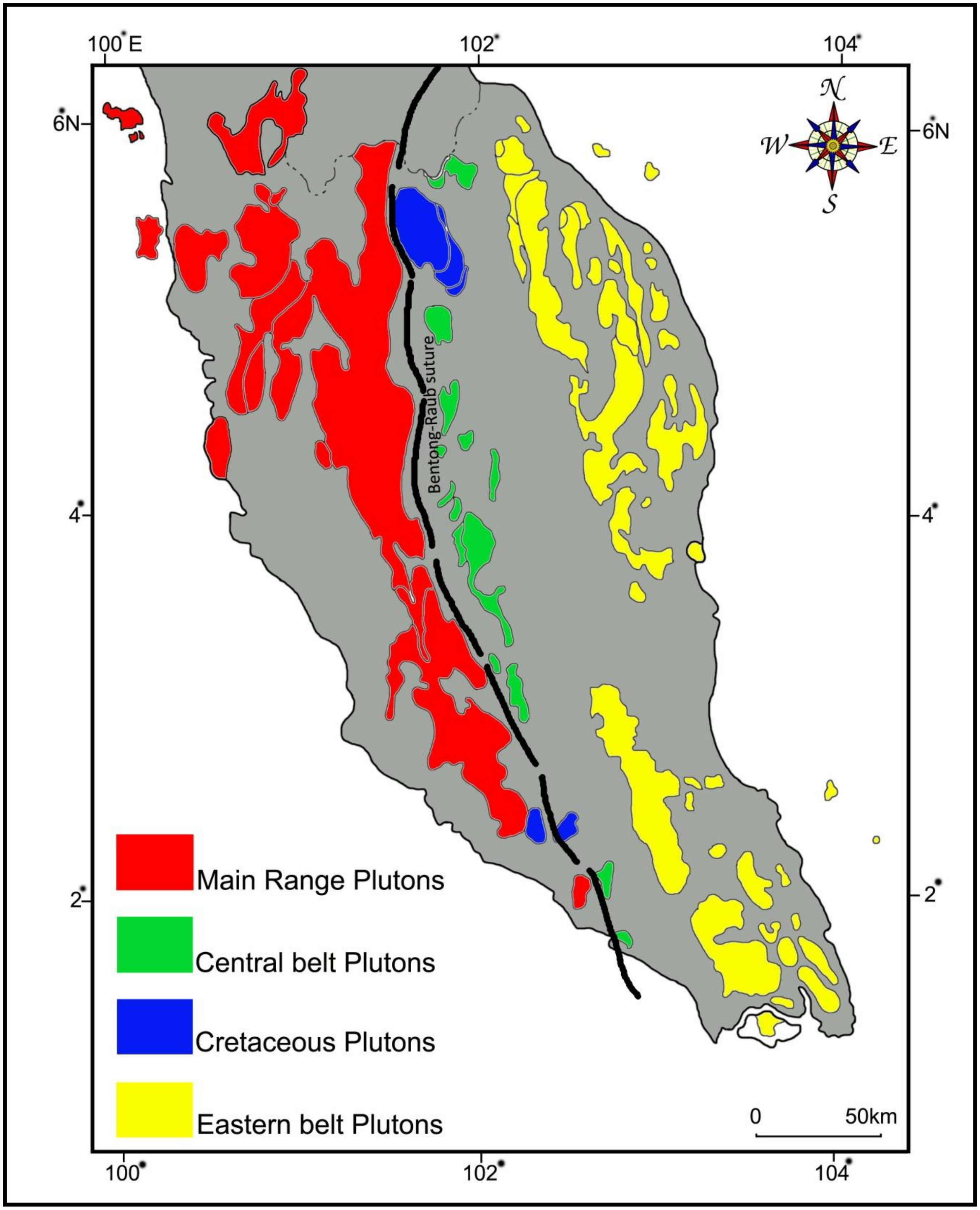


Fig. 2.1: The Granitic provinces map of Peninsular Malaysia.

In the Lower Paleozoic, rhyolitic volcano activity is predominantly concentrated in northwest Peninsular Malaysia. This activity reached its paroxysmal stage in the Permian to Lower Triassic, when activity is centered in axial Peninsular Malaysia. The andesitic rocks are more abundant towards the Triassic. These rocks indicate a period marked by widespread volcanism of mixed rhyolitic and andesitic character. In the upper Triassic to Lower Jurassic, volcanism seems to have waned. But during Late Mesozoic and Cenozoic, reactivated volcanism has changed from sub-marine to sub-aerial (Gan, 1980; Hutchison, 1973).

The geological structure of Peninsular Malaysia is characterized by three sedimentary basins, which are separated by two major granitic batholiths namely the Main Range belt and Eastern Belt. The three basins are the Western Basin, Central Basin, and the Eastern Basin. Because of their distinct mode of mineralization, the three basins are also called the Western Tin Belt, Central Gold Belt and Eastern Tin Belt respectively (Scrivenor, 1928; Rajah and Yin, 1980; Yeap, 1993).

The Straits of Malacca marks the western boundary of the Western Basin. This Basin includes the eastern foothills of the granitic Main Range. Meanwhile, the Bentong Line (Haile, 1973) or Bentong – Raub Ophiolite Line (Hutchison, 1975) marked the eastern boundary. Halmiton (1979) and Hutchison (1973) interpreted this lineament as a Lower Paleozoic Subduction zone. This subduction zone is characterized by phyllitic mĕlange in which blocks of serpentinite, ophiolite and metabasic rocks occur along with radiolarian chert and conglomerate. The Lebir fault zone marked the eastern limit of the Central Basin (Hutchison, 1973). According to Grubb (1968), the Lebir fault zone may have been shifted by left lateral displacement along the Kuala Lumpur – Endau fault zone into the South China Sea immediately east of the Johor shoreline (Gobbet and Tjia, 1973).

In Peninsular Malaysia, the oldest sedimentary sequences are found in the northwest portion of Peninsular Malaysia and indicate a Late Cambrian age. These sequences are characterized by bedded sands mixed with small amount of mud, silts and pebbles indicating a shallow water depositional environment. By the Silurian, the sea continued to transgress southwards. In this period, thick successions of limestone and graptolitic shale were deposited under deeper shelf – like conditions (Rajah and Yin 1980). During upper Silurian-Lower Devonian, a mild orogeny occurred. Post-orogenic sedimentary sequences exhibit uncomformities in the Western Basin, but in the other basins sedimentation was uninterrupted from Devonian until Carboniferous. It is shown by the sequences predominantly of Limestone in the south and non-calcareous material in the north (Gan, 1980, Rajah and Yin, 1980).

An upper Carboniferous-Lower Permian orogeny disrupted the deposition and resulting in granitic emplacement in many parts of Peninsular Malaysia. Sediments are mainly calcareous during Permian. In the Lower Permian, sedimentation was restricted. Meanwhile in the Middle Permain the sea has transgressed to the southern part of Peninsular Malaysia. Tuff and lesser flows of acid to intermediate composition are found in the sedimentary sequences of the Central and Eastern Basins towards Late Permian. During Upper Permian, sedimentation become restricted again and marked the beginning of another orogeny which reaches its peak during Lower Triassic. During Upper Permian-Lower Triassic orogeny, the large scale granitic intrusions and the rise of volcanic island arc also occurred (Rajan and Yin, 1980).

In Middle Triassic, Limestone is common but the strata became more argillaceous and arenaceous. The Middle and Upper Triassic sequences are characterized by flysch type sedimentation. Deposition in the Central and Eastern basins occurred simultaneously with widespread volcanic activity with the eruption of andesite and other intermediate to acid tuff and lavas. The Upper Triassic orogeny which is accompanied by granitic intrusions has changed all sedimentary basins into landmass and consequently accounts for the continental character of Post-Triassic sediments. Sediments are widespread and are described as molasses during Upper Triassic to Lower Jurassic. Sandstones, conglomerates and shale with minor coal layer and volcanic made up these sediments.

Tertiary rocks are typically carbonaceous and crop out as small outlinear trending north-south in the western portion of Peninsular Malaysia. They lie unconformably over the older beds and are non-marine. Meanwhile, the older Quaternary sediments are semi consolidated fluviatile and grade into less consolidated ones (Gan, 1980).

2.2 Granite from Peninsular Malaysia

2.2.1 Introduction

Hutchison & Taylor (1978); Beckinsale (1979); Cobbing *et al.* (1986) documented two granite provinces of Peninsular Malaysia: Western province and Eastern province. The Main Range and Eastern Belt granites of Peninsular Malaysia are made up of different petrological and geochemical varieties. These provinces are separated by the Bentong-Raub suture (Metcalfe, 2000).

2.2.2 Main Range Granite

The Main Range forms the backbone watershed mountain range of the Peninsular. It extends to the north into Peninsular Thailand, and southward to the Indonesian 'tin' islands. The main plutons of the Main Range Granite Province of the Peninsular were emplaced into phyllite and limestone, predominantly of Ordovician to Devonian age. The plutons commonly contain large metasedimentary enclaves. The Main Range Batholith is made up of very large plutons which range from 80×25 km^2 to 10×15 km^2 and comprise predominantly primary textured biotite granite and located west of the Bentong-Raub suture. Although, predominantly of igneous rocks are deep-seated emplacement, there are also some occurrences of rhyolite and microgranite (Ghani, 2009). Smaller intrusive centers occur father to the west, for example at Bukit Mertajam-Kulim, Penang, Dindings, Jerai, in the Langkawi Islands and Satun, Phuket.

U-Pb zircon dates of West Malaya Main Range granite range between 220 and 198 Ma (Liew & McCulloch, 1985; Liew & Page, 1985 and Searle et al., 2012) are in agreement with the result of Cobbing et al., 1992. K-Ar mica age from Penang Island generally range between 205 and 185 Ma (Bignell & Snelling, 1977).

The High initial $^{87}Sr/^{86}Sr$ ratios are also in agreement with the S-type characteristics. Searle et al. 2012 has done the U-Pb zircon dating from Kuala Lumpur granite, ranged between 215±7 Ma and 210 ±7 Ma.

Krächenbuhl (1991) has done the several dating of Main Range granite by using Rb-Sr, and K-Ar methods. He suggested that Upper Carboniferous granite occurs in the Cameron Highlands, Rb-Sr dated 291±1 Ma with an initial Sr isotope ratio of 0.7094±0.0002. He also concluded that no Jurassic granite intrusive exist there. He carried out the apatite and zircon fission track data from the Ipoh and Kuala Lumpur areas. He deduced that the K-Ar and fission track dates have been reset and have resulted in post – magmatic unroofing and slow cooling in the range 1-5 °C Ma^{-1} throughout the Cretaceous and early Tertiary.

2.2.3 Eastern Belt Granite

In general plutonic rocks of the Eastern Belt form smaller batholiths consisting of zoned and unzoned plutons of compositionally expanded rocks (SiO_2: 50 to 78 %). The main rock types range from monzogranite to granodiorite with minor gabbro and diorite. A notable difference of the Eastern Belt granite, compared to the Main Range, is that texturally the rocks are more equigranular and of smaller grain size. The average grain size of K-feldspar, plagioclase and quartz in the Eastern Belt granite is 1-2 cm across. Another contrast with the Main Range is that the K-Feldspar from the Eastern Belt granite contains orthoclase to intermediate microcline. This is taken by Hutchison (1977, 1983) to imply Eastern Belt epizonal emplacement resulting in rapid cooling.

Eastern belt comprise of several batholiths such as the Boundary Range, Lawit, Kapal and Jerong batholiths. The Boundary Range Batholith ranges from felsic (granitic, granodiorite) to basic (diorite/gabbro) and minor ultrabasic rock pyroxenite. The Kapal and Jerong batholiths are composite bodies ranging from diorite to monzogranite, dominated by granodiorite.

On the basis of the summary of the Rb/Sr radiometric dating of the Peninsular Malaysia (Cobbing et al., 1992), the plutons seem to be younger towards the suture ranging from Upper Permian to Lower Triassic (220 – 240 Ma) and around 200 Ma adjacent to the suture. The oldest intrusive age is found along the eastern coastline, which is Early Permian

(255 -270 Ma). There are quite a number of granites in the Eastern Belt that have Permian age, including Kuantan (250-275 Ma), Besar (280 Ma), Jerong (258 Ma) and Perhantian granite (258 Ma). The younger granite age are dated by Darbyshire (1988) using K-Ar method on Benom granite gave an age of 197±16 Ma (Cobbing et al., 1992). The main age for granitic magmatism in the Eastern Belt is Triassic (197-240 Ma) as dated by Darbyshire (1988) using K-Ar and Rb-Sr whole rocks.

2.2.4 Cretaceous Granite

The Cretaceous plutonic rocks of the Peninsular Malaysia are the Stong Complex, Batang Melaka, Gunung Ledang, Gunung Pulai and Pulau Tioman granites. These granitic bodies have been dated between 96 and 60 Ma (Bignell & Snelling, 1977; Darbyshire, 1988).

Whole-rock Rb-Sr isochron ages of the Noring Granite and Kenerong Leucogranite of the Stong Complex gave 90 ± 30 Ma and 79 ± 3 Ma respectively (Darbyshire,1988). The Tioman granite gave a U-Pb zircon age of 80±1Ma (Searle et al., 2012).

2.3 General Geology of the Langkawi Island

A full account of the Palaeozoic stratigraphy is provided by Jones (1961) and Gobbett (1972). Rocks of Langkawi Islands can be easily divided into four sedimentary rock formations and one granite unit (Fig. 2.1). These sedimentary rock formations, in chronological order, are:

(a) Machinchang Formation, mainly of quartzite of Upper Cambrian age,

(b) Setul Formation of Lower Palaeozoic – Devonian limestone and minor detrital bands

(c) Singa Formation mainly of mudstones with age ranging from uppermost Devonian to Lower Permian,

(d) Chuping Formation of Middle to Upper Permian limestone.

Summary of important Palaeozoic geological history of Langkawi Islands are shown in Figure. 2.3. These sedimentary rock formations is generally younger towards the east (Jones, 1981), only to be interrupted by the presence of Kisap Thrust Fault in the eastern part of Langkawi. The westward transport direction of this fault has brought older Setul (and possibly Machinchang) Formation(s) from the east to overlay younger Chuping and Singa Formations in the central axis of Langkawi Islands. The Triassic Gunung Raya granite intruded older sedimentary rock formations and turned them partially into various types of contact metamorphic rocks (Jones, 1981). Since the Late Trisassic time, not much geological records are found in Langkawi. Tropical weathering upon rocks in Langkawi might have taken place since Early Jurassic and continues until present day. The youngest geological unit in Langkawi is the unconsolidated recent sand and clay deposited in narrow valleys and coastal plains.

2.3.1 Machinchang Formation

The Cambro-Ordovician Machinchang Formation is well exposed as strong ridges in the North West of Langkawi Island reaching heights of 750 m, 1350 m thick. The Machinchang strata consist of grey, brown, purple and red, coarse to medium-grained quartzite, arkose and subgreywacke. Subordinate beds of gritty sandstone and conglomerate composed of rounded pebbles of white vein quartz and quartzite in a gritty matrix occur on the north coast of Pulau Langkawi. Brown, grey and black silty and flaggy beds are abundant in the lower half of the sequence and an interval of red and grey shale and mudstone is developed near the top of the formation. The age of the formation as evidenced by the saukiid trilobite, is late Cambrian to Lower Ordovician. It was first described by Jones (1981), and later by Lee (1981, 1983), and the detailed stratigraphy was described by Lee (2006). The granite intruded into the easterly dipping limb of the Machinchang anticline and caused marked structural distension in the surrounding sediments.

2.3.2 Setul Formation

The Cambrian to Lower Ordovician Machinchang Formation is overlain by thick grey shallow-marine shelly limestone of the Ordovician to Silurian Setul Formation (Jones, 1961). The formation is fossiliferous and geological range from the Lower Ordovician to Lower Devonian. The Setul Group is exposed on the eastern seaboard of Langkawi Island. Most of the exposure on the main Island are more metamorphosed and tectonised, due to close to the central granite core of the island.

2.3.3 Singa Formation

Singa Fromation (upper most Devonian to lower Permian) is composed of dark grey argillaceous rocks, which is overlying the Langgun Redbeds and other transitional units above the Setul Group (Jones, 1961). It is well exposed in the Langkawi islands and Pulau Singa Besar. The lighter color rocks of Singa Formation are equivalent to the Kubang Pasu

Formation in the mainland (Jones, 1961, 1981). The Singa Formation comprises thin- to poorly-bedded rapidly alternating black mudstone, silty shale and lithic to quartzitic sandstone with pebbly horizons in parts within the black mudstone. There are six lithofacies carried out from the Singa Formation on Pulau Tepor, Pulau Ular and Pulau Singa Besar in Langkawi Island (Stauffer & Lee, 1986):

1. Laminated clean sandstone facies
2. Rhythmically interbedded sand-mud facies
3. Megaclast-bearing rhythmically interbedded sand-mud facies
4. Sandy mudstone with thin graded sandstone facies
5. Diamictite facies
6. Laminated sandstone-siltstone facies

2.3.4 Chuping Formation

The Chuping Formation (middle to upper Permian) is exposed in central Perlis, north Kedah and Pulau Dayang Bunting, Palau Jong and Pualu Singa Besar in Langkawi. The Chuping Formation is comprised of massive, pale colour, finely crystalline and pure calcitic limestone. The top of the formation is overthrusted by the Lower Palaeozoic limestone of the Setul Group in Langkawi (Lee,1981). The base of Chuping Formation is transitional to the Singa and Kubang Pasu Formations. The most part of Chuping Formation is massive and unfossiliferous, except for the basal part, composed of well-bedded dark grey shelly limestone with chert nodules in layers parallel to the bedding. The basal part of this formation is exposed at the base of Bukit Temiang in Perlis and Pulau Singa Besar and Pulau Singa Kechil in Langkawi Island (Lee, 1981).

2.3.5 The Langkawi Granite

Forty four square miles of the Langkawi Island are underlain by granite. Large and small igneous stocks and bosses are found scattered, particularly in the central part of Langkawi Islands. Out of these, there are two larger igneous stocks namely the Gunung Raya and Bukit Sawar stocks. Other bodies including the Kuah, Tuba and Dayang Bunting stocks are comparatively smaller than these two. The Langkawi igneous bodies have been collectively named as the Gunung Raya Granite, which has been dated by Bignell & Snelling (1977) using both the K/Ar and Rb/ Sr method. These rocks are of Late Triassic (217 ± 8 Ma and 209 ± 6 Ma). The granites of Langkawi intruded several types of host rocks, transformed them into various types of metamorphic rocks, and bringing along with them some minerals such as tourmaline, ilmenite, zircon and garnet.

The largest body on the main island forms the Gunung Raya massif and extends westwards to Telok Borau. Smaller stocks also found on Palau Langkawi near Padang Mat Sirat and around Kuah and Telok Apau. The granite was intruded into the easterly dipping limb of the Machinchang anticline.

Jones (1981) believed that these granite bodies are connected to each other along the sea floor. But based on the REE patterns, Wan Fuad Wan Hassan et al. (2001) divided into two groups:

(1) Gunung Raya – Dayang Bunting granite

(2) Kuah – Pulau Tuba granite

They also reported comparison in geochemistry between two groups such as Rb/Sr ratios and the degree of partial melting and fraction and a possible source material for the Langkawi granite.

In this study, Langkawi granite can be divided into two units based on textural features. These are summarized in Table. 2.1.

Table. 2.1: Summary of textural feature of Langkawi granite

Granite Unit	**Gunung Raya**	**Kuah granite**
Textural feature of granite :	- coarse grain strongly porphyritic texture with K- Feldspar phenocrysts reaching 6 cm in length .	- composed predominantly of coarse-grained quartz and contain veins of tourmaline, muscovite and fine-grained quartz.
Distinguished feature :	-Well-jointed - rich in biotite	- tourmalinization is commonly manifested as segregated round clots of 5 cm in diameter which stands up as prominent resistant projections of black colour against the light coloured granite surface in the background.

The intrusion of granite into the sedimentary formations is responsible for the copper, bismuth and galena mineralization together with associated skarn minerals. This granite body itself is altered in parts into tourmaline greisens. Some minor granite sills are found in the sedimentary rock layers on Pulau Tuba. The granite covered about half the area of Palau Tuba. The rock is mostly coarse - grained, porphyritic and biotite – bearing. Occasional late differentiates occur intruding into both the parent granite and country rock. Ilmenite, topaz and zircon are conspicuous accessories and scheelite occurs along the limestone contact zone on the northeast side of the island (Jones, 1981). The northern part of Pulau Dayang Bunting is covered by the granite about half of the area. The rock at Tanjong

Tinlin is coarse-grained, porphyritic, xenomorphic granular, biotite granite. Quartz dykes and late-phase differentiates of tourmaline greisens are similar to the Kuah granite.

There is an occurrence of Precambrian K/Ar aged trodhjemite megaclast at the Palau Tepor, which has been interpreted as glacial-marine dropstones (Stauffer & Lee, 1986). This evidence is further supported by the presence of fresh feldspars in the matrix and granitic clasts, angular limestone clasts and cold-water fossils, notably brachiopods associated with them (Waterhouse,1982; Mohd Shafeea,1996 and 2003; Mohd Shafeea & Asmaniza, 2002; Shi & Archibold, 1995; Shi et al., 1997).

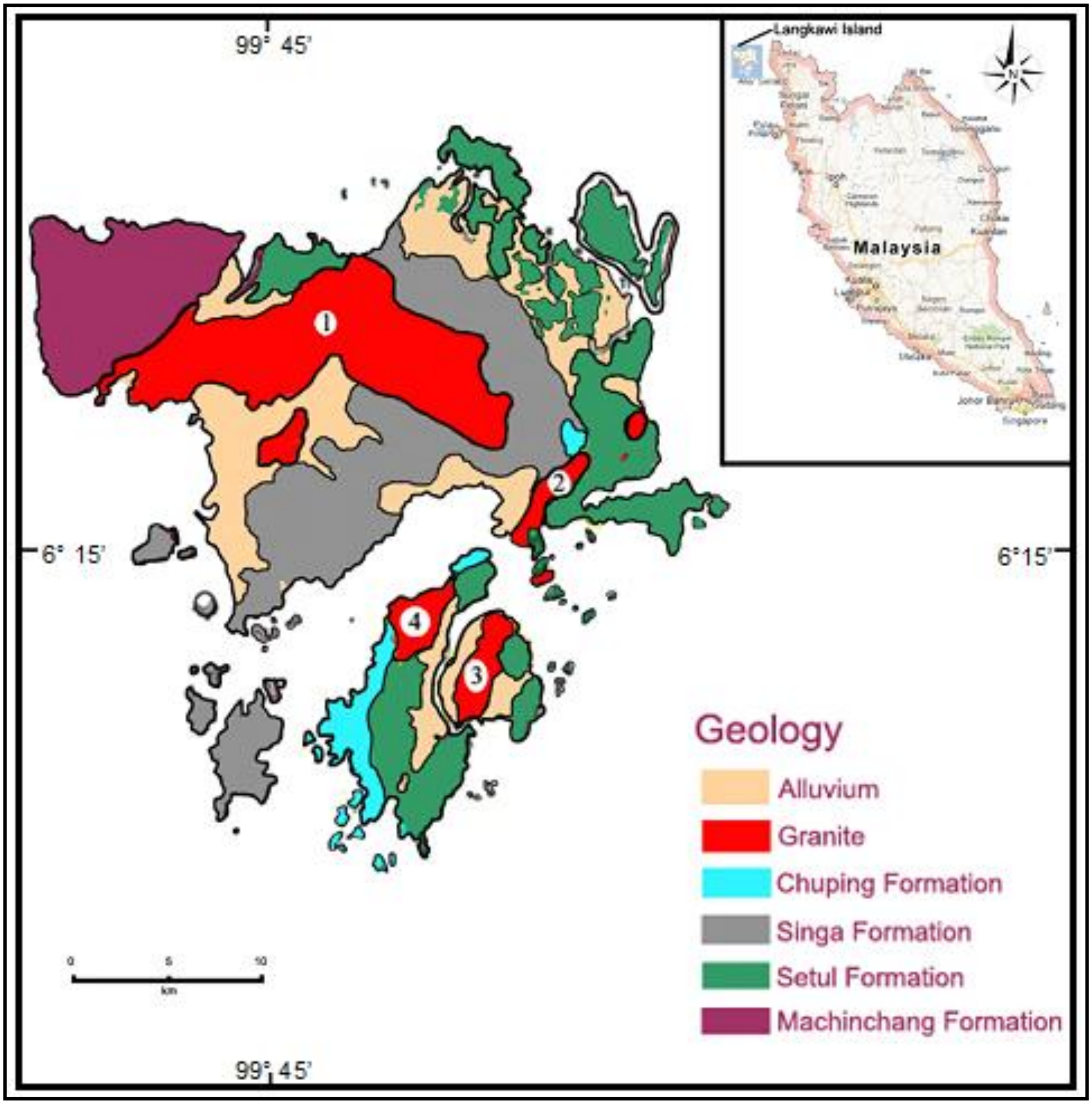


Fig. 2.2: Geological map of the Langkawi Island showing distribution of granites 1- Gunung Raya granite, 2 – Kuah granite, 3 – Tuba granite, 4 – Dayang Bunting granite.

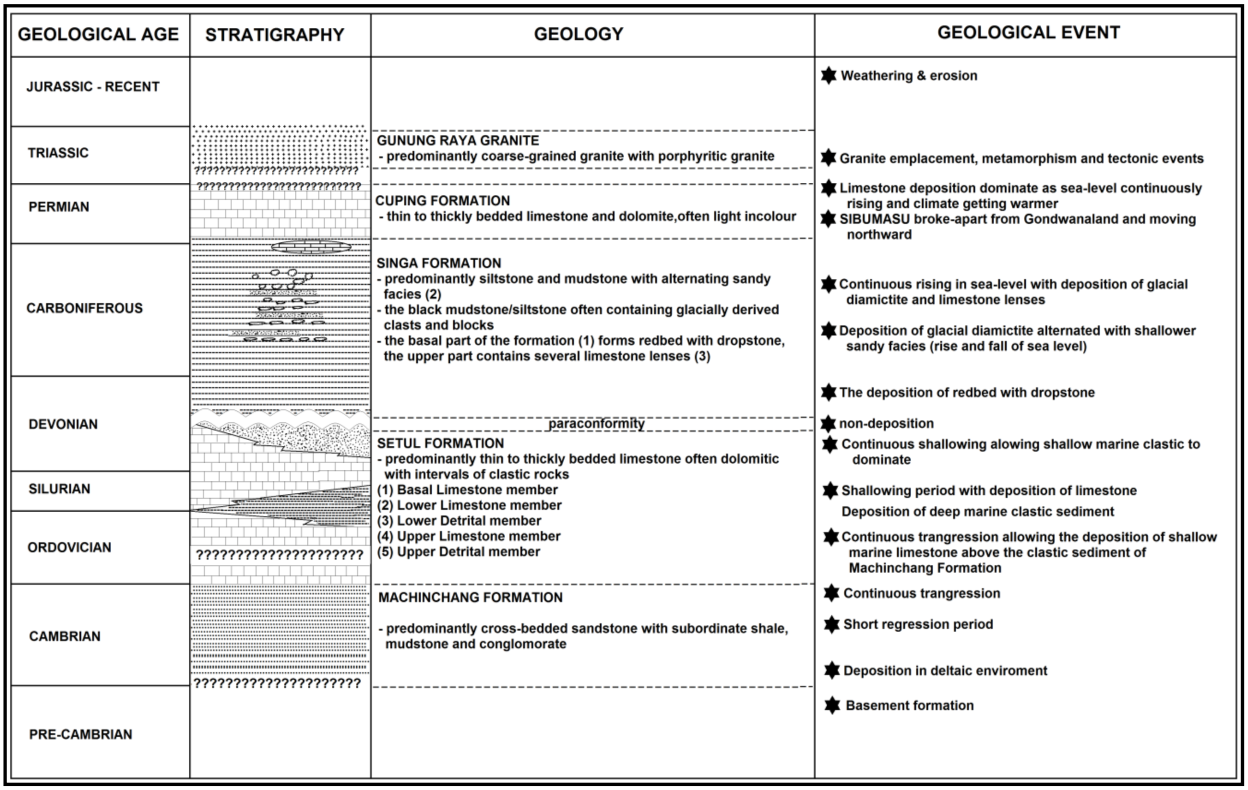


Fig. 2.3: Summary of important Palaeozoic geological history of Langkawi Islands (after Shafeea Leman et al., 2007).

CHAPTER 3

PETROLOGY OF THE LANGKAWI GRANITES

3.1 Introduction

This chapter will describe in detail petrographic analysis of the Langkawi granites collected from two units: Gunung Raya and Kuah. The sample has been classified using QAP diagram and their modal composition indicates that they are monzo – syenogranite (Streckeiensen, 1976; Fig, 3.1).

30 granite samples were collected from Gunung Raya and Kuah units for petrology and geochemical study. Each samples weight about 1-2 kg. Ten thin sections were prepared for all the rock type to decipher the texture, mineralogy, alteration and deformation. Point counting data was done by using a Swift Model E point counter fitted with an automate stage. Normally, the total counts were around 1500-2000 on each specimen so that the whole surface of the thin section was covered. Based on texture and petrography, the Langkawi granites can be divided into two main types; the Gunung Raya granite that shows coarse grain texture and strongly porphyritic texture with K- Feldspar phenocrysts reaching 6 cm in length and the Kuah granite composed predominantly of coarse-grained quartz and contains veins of tourmaline, muscovite and finer-grained quartz. The description of grain size interval of Williams et al., 1974 (mentioned below) shall be followed:

Table. 3.1: Grain size interval (from Williams, 1974)

< 1 mm	Fine-grained
1-5 mm	Medium-grained
5 mm-3 cm	Coarse-grained
> 3 cm	Very coarse-grained

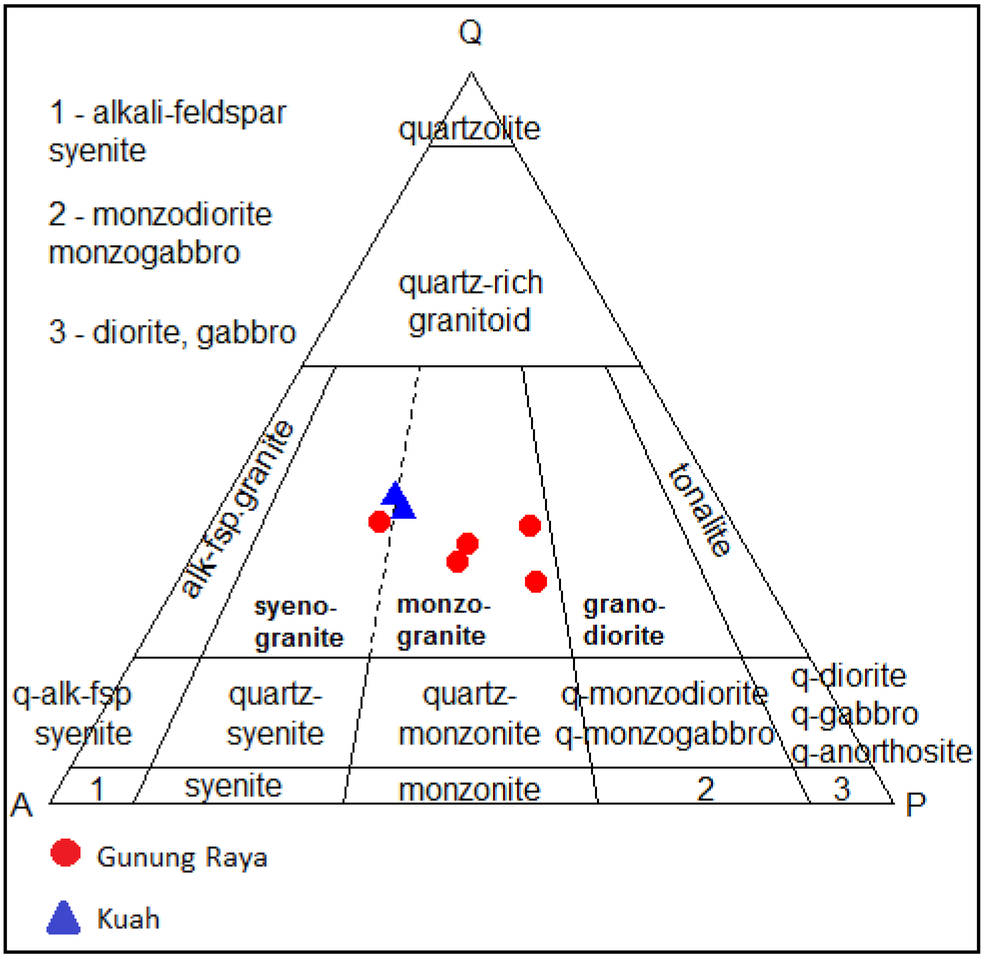


Fig. 3.1: Triangular QAP diagram of modal analysis of Langkawi Granites after Streckeiensen (1976) classification.

3.2 Macroscopic Description

3.2.1 Gunung Raya

The Gunung Raya granites are characterized by strongly porphyritic texture with K- Feldspar phenocrysts reaching up 6 cm in length (Figs. 3.2 and 3.3). It shows the mineral alignment due to the magma movement. In hand specimen, quartz is randomly dispersed. The general colour of the Gunung Raya granite is grey, with patches of biotite mineral. The grain size varies from very coarse- to medium-grained (Fig. 4.3A).

3.3 Microscopic Description

3.3.1 Gunung Raya Granite

The major constituents of the Gunung Raya granites are consist of quartz (32 wt %), K-feldspar (19 wt %), Plagioclase (35 wt %), Biotite (9 wt %), Muscovite (5 wt %). The accessories minerals are zircon, apatite, magnetite and tourmaline. The phenocryst assemblage consists of K-feldpsar and the ratio to matrix is approximately 50:50. Feldspar phenocrysts may reach up to 20 mm in length, with the average groundmass mineral size being 2-3 mm. Most K-feldspar phenocrysts show tartan and simple twinning. Occasionally the interstitial microcline tends to wrap around plagioclase crystals.

Petrographically, majority of the K- feldspar is altered to sericite and secondary muscovite (Fig. 3.4B). Some K-feldspar have the typical appearance in their surface and occur as anhedral grains. Some of the K-feldspars are microcline with cross-hatched texture and associated with K-feldspars size range from 0.5 - 2mm across. Occasionally, microcline occurs as an inclusion in plagioclase. Most of the perthitic microclines are commonly found together with quartz inclusion showing a patchy character, their size ranged between 1 and 12 mm (Fig. 3.4C). In some sections, perthites are found in the altered plagioclase as inclusion and most of them are patchy perthite.

Plagioclases range from 0.5 to 5 mm in size and may occur as discrete phenocrysts. It usually shows albite, carsbad-albite and pericline twinning. Sericitization of plagioclase is particularly prominently developed in the Gunung Raya granite. The plagioclase is partially altered to extensively sericitized, especially in the core of the crystal. (Fig. 3.4D).

Quartz in the Gunung Raya granite is mostly anhedral and sometimes occurs as sub grains with the size ranging 1 – 7 mm. There are two types of quartz: (i) large size shape (5mm) and (ii) aggregate shape (0.5-2mm). K-feldspar, plagioclase, muscovite and zircon occur as inclusions in quartz. Most of the quartz crystals show wavy extinction.

Biotites occur as both euhedral and subhedral grains. The size ranged from 1 to 3 mm. They are brown in colour and exhibit a pale brown to dark brown pleochroism. In some case, biotite altered to secondary muscovite. The alteration is very prominent around the edges of the biotite (Fig. 3.4E). The biotite has been chloritized along the cleavage plane (Fig. 3.4F).

Accessory minerals found in the Gunung Raya granite are zircon, apatite, magnetite and tourmaline. Zircon and apatite occur as the inclusion of biotite and associate with the pleochroic haloes. Magnetite formed as small individual grain and sparsely distributed in the rock. Tourmaline usually occurs as randomly distributed grains scattered throughout the thin section. Tourmaline also occurs as small anhedral crystal and rarely shows subhedral grains. They show brownish green to blue interference colour. The crystal shows pleochroism from light blue to brownish green. The tourmaline also shows very prominent cleavages and has high relief.

The alteration minerals that occur in the Gunung Raya Granite are chlorite, sericite and secondary muscovite. Secondary muscovite and chlorite formed as secondary minerals following the alteration of feldspar and biotite. They show a high interference colour and are usually anhedral. Commonly, sericite formed by replacing the K- feldspar and plagioclase.

3.4 Macroscopic Description

3.4.1 Kuah Granite

The Kuah granite is medium- to fine-grained granite and associated with tourmaline apolopegmatite. It is composed predominantly of coarse-grained quartz and contains veins of tourmaline, muscovite and of finer-grained quartz. The tourmaline is visible in hand specimens and in thin sections, and is identified as black green grains with fine brown rims. The coastal granite exposures are slightly weathered and the tourmaline segregation growing up of black colour from the smooth whitish granite surface. Tourmalinisation is commonly manifested as segregated round clots of 5 cm in diameter which occurs as prominent resistant projections of black colour against the light coloured granite surface in the background (Fig. 3.6). The phenocrysts of quartz and feldspar also occurred in the Kuah granite. The maximum size of quartz is 1.5cm across and it is gray in colour (Fig. 3.7A).

3.5 Microscopic Description

3.5.1 Kuah Granite

Microscopically, the granite is holocrystalline with grain size ranging from fine-to medium-grained. They are non-porphyritic, allotromorphic to hypidiomorphic rocks. The major constituents are alkali feldspar (45 wt %), quartz (35 wt %), tourmaline (15 wt %) and plagioclase (5 wt %). The accessories minerals are muscovite and biotite. The secondary minerals present are muscovite and chlorite. The muscovite also occurs in the veins and commonly borders the tourmaline.

The alkali feldspar occurs as anhedral grains, both of microcline and perthitic type are present. The alkali feldspars are often sericitised (Fig. 3.7D). Inclusions in quartz are common and often sericitised.

Quartz occur as anhedral grains and in the porphyritic type they occur both as phenocryst and also in the matrix (size ranging between 0.5cm – 1.5cm) (Fig. 3.7E). Fine grain quartz also associates with tourmaline. The quartz has highly undulose extinction, and some crystals being spherulitic.

Tourmaline is found throughout the Kuah granite, and commonly segregated into round clots of size (>2mm). These segregations may show radial arrangement of the tourmaline, or may be random (Fig. 3.7B). Unlike the tourmaline observed in the Gunung Raya granite, the tourmaline in the Kuah granite shows euhedral shape and vein into the feldspar or quartz. The crystal has pleochroism from light grayish to bluish in colour. Tourmaline also shows very prominent cleavages.

Plagioclase shows albite twin, occurs as a minor mineral and absent from a few samples from Kuah granite. Plagioclase usually occurs as sub to euhedral crystal, often have polysynthetic or lamella twinning. In the other rock, the plagioclase crystals are enclosed completely by K-feldspar.

Euhedral zircon and anhedral apatite occur among them (Fig. 3.7C). Magnetite is also present and garnet is only seen in one sample from the Kuah granite (Fig. 3.7F).

3.6 Tourmalinization and Greisenization

The Langkawi granites include tourmalinization, greisenization, and intrusion of tourmaline-quartz veins, showing the importance of boron-rich metasomatism during the late stages of magmatism. Boron metasomatism leads to pervasive or selective tourmalinisation, usually connected with Sn-W deposits and breccia pipes. In general, Fe-rich tourmalines are associated with Sn-W deposits of greisen affinity. A detailed account of tourmaline, its association with and relationship to hydrothermal mineral systems can be found in Slack (1996).

For this area, the processes of tourmalinization and greisenization inferred from the texture, are not separated by any major time break, Hutchison and Leow (1963) in accordance with Lindgren's (1933) definition, proposed the term tourmaline greisenization to include the complete metasomatic alteration**.** Greisens are also linked to alkali metasomatic processes, usually in highly fractionated granitic cupolas (Harlov & Austrheim, 2013). Greisen is a hydrothermally altered and metasomatised granite commonly associated with Sn-W mineralisation, and more generally with the waning stages of crystallization of S-type granites. The formation of greisen involves the breakdown of feldspar to produce muscovite, and often the precipitation of quartz, together with minerals such as cassiterite, tourmaline, beryl and topaz, in veins or vugs (Harlov & Austrheim, 2013).

Tourmaline veins occur associated with quartz-vein networks, suggesting that both represent passage of hydrothermal fluids along the zones of structural weakness (Fortey, 1986). One clear result of tourmalinization is the destruction of biotite and its replacement by tourmaline. The ferromagnesiam components in the tourmaline are derived from the destroyed biotite (Fook, 1986). In many cases the country rocks surrounding greisenised granite cupolas have disseminated tourmaline, which tends to be particularly abundant in fractured zones. Quartz-tourmaline-dominated assemblages form pervasive replacements as well as cross-cutting veins and veinlets. The replacements can be on a very fine scale, so that the smallest features and textures are perfectly preserved. This type of tourmaline alteration is linked to the emplacement and crystallisation of B-rich granitic magmas, with the possibility that the B-enrichment may have been inherited from a source region containing evaporates or tourmaline-rich protoliths. Pervasive to selectively pervasive tourmalinisation usually occurs associated with Sn-W deposits (Harlov & Austrheim, 2013).

Tourmaline greisenization is best seen on coastal exposures to the east of the jetty, Kuah (Fig. 3.4A) and also occurs in Gunung Raya granite but is less developed on Gunung Raya granite. In the field, tourmalinisation is commonly manifested as segregated round clots which stands up as prominent resistant projections of black colour against the light coloured granite surface in the background. Some mineralization is associated with the Kuah granite. In the skarn zone of the limestone-granite contact to the east of Kuah, copper-bismuth mineralization was found. No tin mineralization is associated with the Langkawi granite, although traces of Sn in stannite and andradite garnet were detected from the Sn mineralization (Jones, 1981).

Fig. 3.2 : Gunung Raya granite exposed at 7 well water fall.
Loc: seven well water Fall / N 06° 33, 972′, E 099° 40, 444′

Fig. 3.3: Photograph shows strongly porphyritic texture with K-feldspar phenocrysts reaching 6 cm in length.
Loc: seven well water fall / N 06° 33, 972′, E 099° 40, 444′

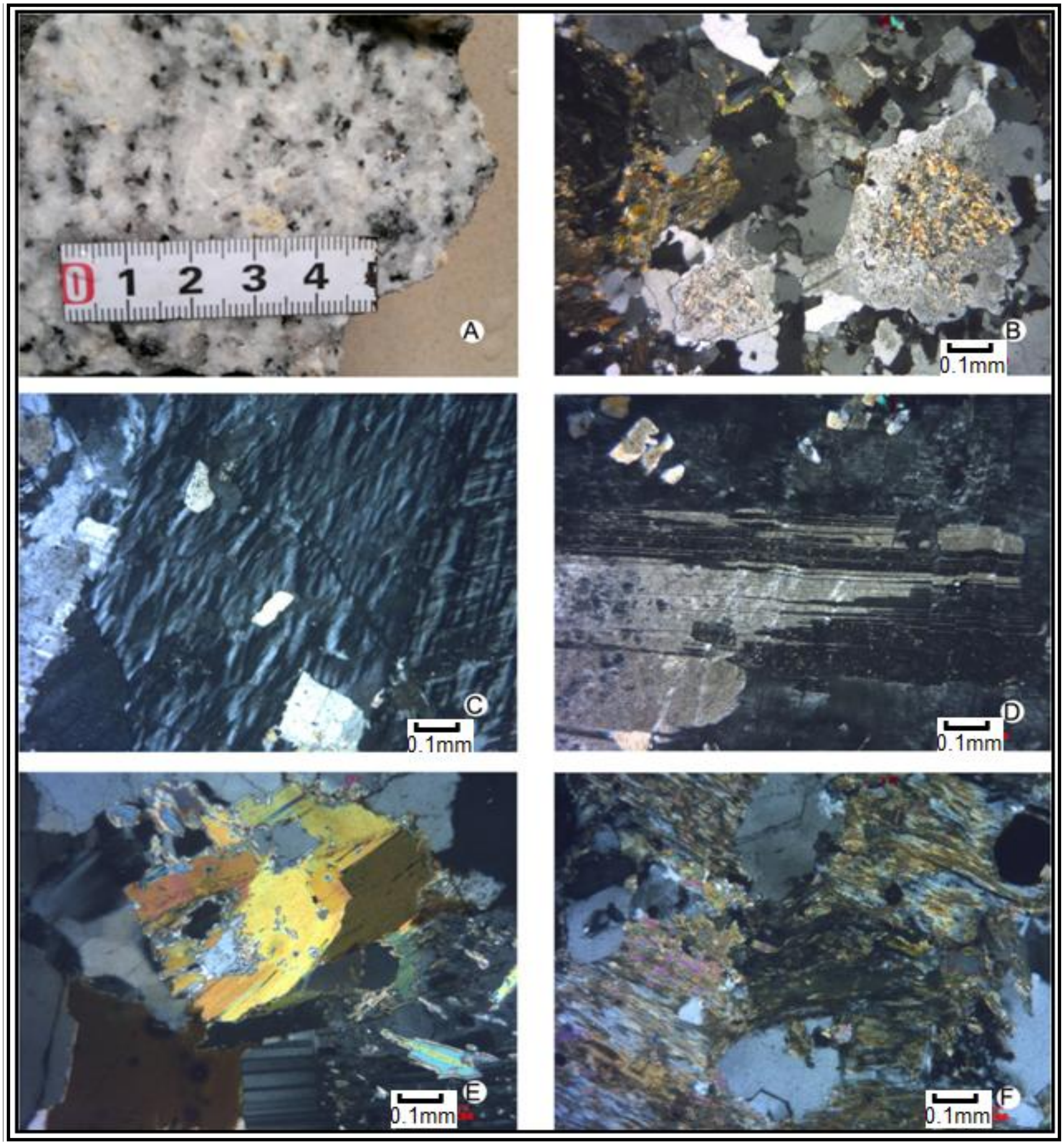


Fig. 3.4 : Photomicrographs of Gunung Raya granite. (under cross nicol). (A) Typical hand specimen of biotite granite from Gunung Raya area, showing strongly porphyritic texture. (B) Alteration of plagioclase to secondary muscovite and sericite. (C) K-feldspar-perthitic microcline. (D) Large subhedral plagioclase showing Carlsbad and Albite twinning. (E) The replacement of biotite with muscovite around the edges of the biotite. (F) Chloritization of biotite in Gunung Raya granite.

Fig. 3.5: Panoramic view of the Kuah granite south east of Langkawi Island.
Loc : East of Kuah Jetty / N 06° 17, 926′, E 099° 51, 129′

Fig. 3.6: Fine-grained tourmaline granite with tourmaline clots.
Loc :East of Kuah Jetty / N 06° 17, 926′, E 099° 51, 129′

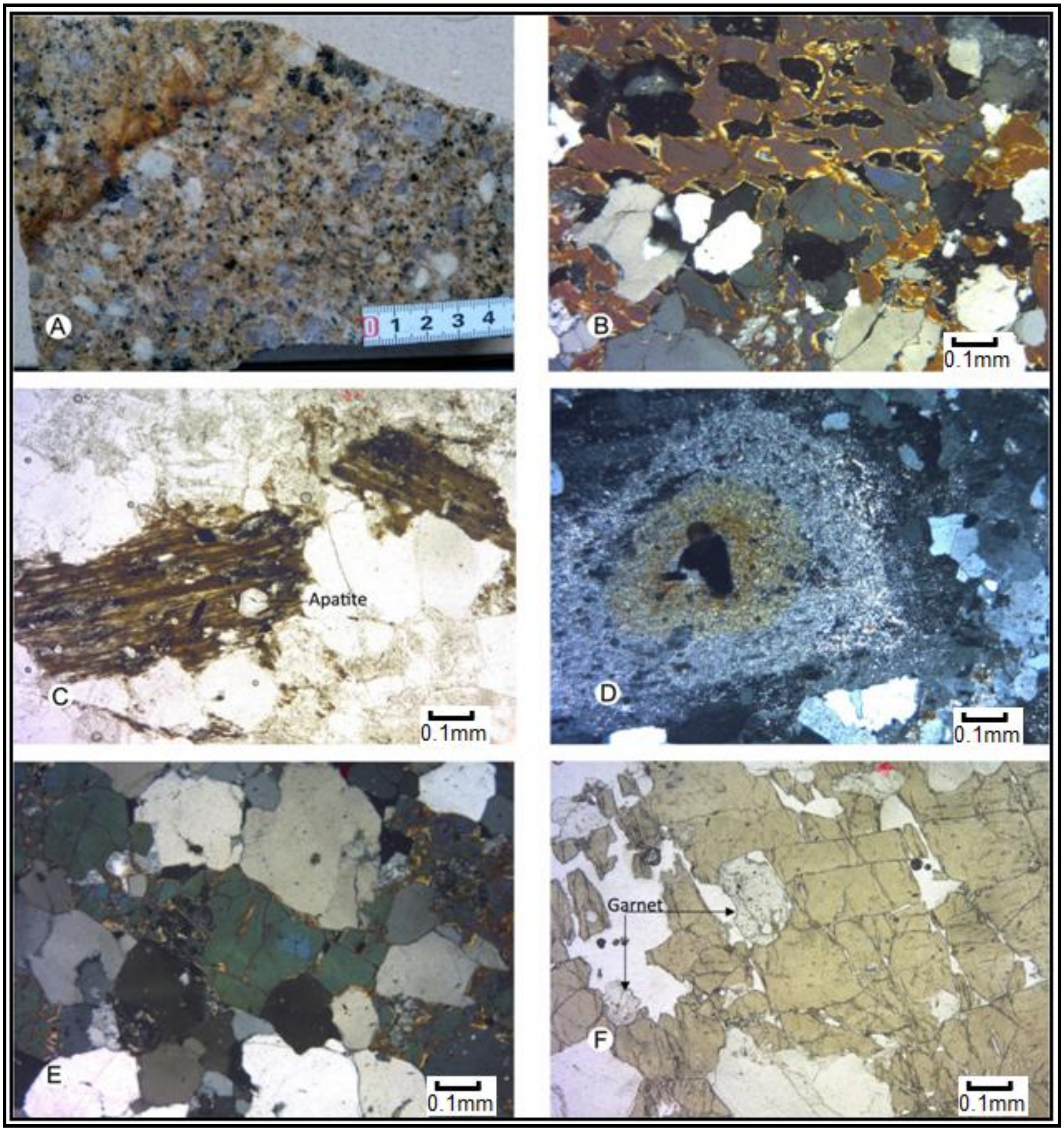


Fig. 3.7: Photomicrographs of Kuah granite. (A) Typical hand specimen of tourmaline granite form Kuah area. (B) Tourmaline form a skeletal network in the Kuah granite. (C) Apatite occurs as an inclusion in chloritized biotite. (D) Sericitization alkali feldspar at its core. (E) Both subhedral tourmaline and anhedral quartz grains occurs in the matrix. (F) Garnet occur as inclusions in tourmaline.

CHAPTER 4

GEOCHEMICAL STUDY OF THE LANGKAWI GRANITES

4.1 Introduction

Geochemical studies of the granites in the study area was carried out to determine any possible differences in the distribution of major and trace elements along with the study of chemical variations in order to achieve a better understanding of their texture, petrogenesis and evolution. A total of 23 samples from Gunung Raya and 7 samples from Kuah granite were analyzed for major, trace and REE elements.

4.2 Analytical techniques

Samples were crushed to a fine powder at the Department of Geology , University of Malaya, Kuala Lumpur Malaysia. The whole-rock compositions were determined at Acme Analytical Laboratories in Vancouver, Canada. Major elements were determined by X-ray fluorescence (XRF) using a Philips PW 1404/10 X-ray spectrometer by fusing with lithium tetraborate and casting into glass discs. The trace element Zr was also measured by X-ray fluorescence using pellets of pressed rock powder. X-ray counts were converted into concentrations by a computer program base on the fundamental parameters method of de Jongh (1973). Precision was of 2-5% for major elements, except Mn and P (5-10%), and 2-5% for Zr.

Inductively coupled plasma mass spectrometry (ICP-MS) has been used successfully for the direct determination of trace elements in granites and thus is required to use sample preparation methods that result in the total dissolution of the sample. The use of hydrofluoric and nitric acids for the digestion of silicate samples is well established, and has been applied to the dissolution of granite, basalts, soil and other rocks. Using this approach for the analysis of granites is problematic since Zr and Hf do not always go into solution. Granite often contains accessory minerals such as zircon, which are very difficult to dissolve. The detection limits are 0.01% for all oxides, 0.1 ppm for Li, Cs, Rb, Cu, Pb, Bi, Ag, Th, U, Zr, Hf, Nb, Ta, Sn, W, Mo and Y, 1 ppm for Be, Ba, Sr, Zn, As, Ce and Cr, and 0.5 ppb for Au. All the major and trace elements listed in the Table. 4.2, 4.3 and 4.4.

4.3 Major Elements Chemistry

Selected major elements using Harker variation diagrams for the Gunung Raya and Kuah granite are shown in Table. 4.2 and Figure. 4.1. The range of SiO_2 of Gunung Raya granite is lower (68.1-75.3 wt %) compared to Kuah granite (75.1 to 77 wt %). This shows that the Kuah granites are more siliceous. Contents of other elements for Gunung Raya granite are typically 2.69 – 4.04% Na_2O, 3.93 – 6.24% K_2O, 0.14 – 0.88% MgO, 1.21 – 5.49% Fe_2O_3, 0.3 – 1.74 % CaO, 0.04 – 0.11% MnO, 0.02 – 0.82% TiO_2 and 0.13 – 0.3 % P_2O_5. In general, the plots shows clear trend of decreasing Al_2O_3, CaO, MgO, Fe_2O_3, P_2O_5, TiO_2 with increasing SiO_2 contents. The Harker diagram shows that K_2O increase while TiO_2, total Fe_2O_3 and Na_2O decrease with increasing SiO_2 content. The Harker diagrams also show that there is a slight difference between the Gunung Raya and Kuah granite in SiO_2 content between 75.3 % and 77 %. The Gunung Raya granite have higher Al_2O_3, CaO, MgO, TiO_2 , P_2O_5 and Fe_2O_3 contents compared to the Kuah granite. The difference between the two granites probably is best illustrated on a P_2O_5 vs SiO_2 diagram (Fig.4.1). Two trends stand out in this diagram and the Gunung Raya samples are separated to those of the Kuah granite. These results show a higher P_2O_5 content of the Gunung Raya granite compare to the Kuah granite at a given SiO_2 concentration. For example, the sample with 75% SiO_2 from Gunung Raya has 0.18% P_2O_5 compared to the Kuah granite which only has 0.09% P_2O_5 with the same SiO_2 content. On the other hand, the Kuah granite have low CaO with the same SiO_2 content compared to the Gunung Raya granite.

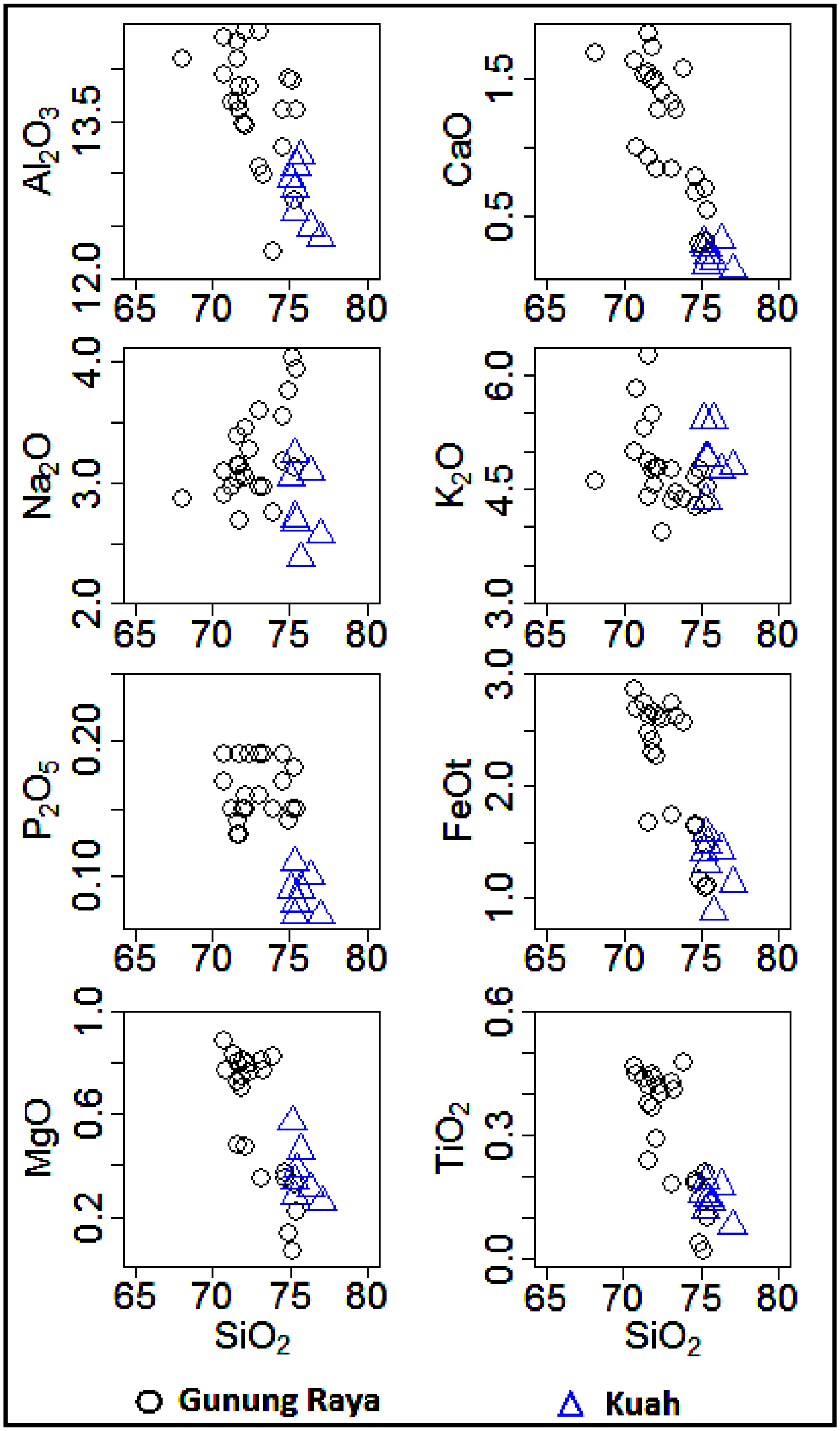


Fig. 4.1: Major elements Harker diagram of the Gunung Raya and Kuah granite from Langkawi Island.

The granite from both units (Gunung Raya and Kuah) generally have high alkali content ($Na_2O + K_2O$) ranging from 7.8 – 9.39 wt % for Gunung Raya granite to 7.38 – 8.46 wt % for Kuah granite. This is clearly shown in the K_2O vs. SiO_2 diagram (Peccerillo and Taylor, 1976) (Fig. 4.2). All the samples from Gunung Raya and Kuah plot between the field of high-K and shoshonite field at high SiO_2. The analyzed samples of both Gunung Raya and Kuah granite show peraluminous character with alumina saturation index (ASI) ranging from 1.02 to 1.17 for the Gunung Raya granite (Fig. 4.3).

In the A-B plot (Debon and Lefort, 1982, Villaseca et al. 1998), A=Al-(K+Na+2Ca) and B= Fe+Mg+Ti, (Fig. 4.4). Majority of Gunung Raya granite samples plot within the weak peraluminous field and a few sample are plot in the moderate and felsic peraluminous field. On the other hand, Kuah granite samples plot exclusively within the felsic peraluminous field.

On Na_2O vs K_2O plot (Fig. 4.5), most rocks from the Gunung Raya and Kuah granite plot in the S type field of Chappell and White (1984). The Na_2O content of the Gunung Raya granite is shown in the d,iagram, fifty percent of the Gunung Raya granite contain more than 3 % Na_2O, whereas the Kuah granite rarely exceeds the value.

All sample plots within the granite field (Fig. 4.6) of the R1 – R2 classification for the volcanic and plutonic rocks after De La Roche et al. (1980) where R1= 4Si – 11(Na+K) – 2 (Fe+Ti) and R2 = 6Ca + 2Mg + Al, expressed in millications. Both granites, especially the Gunung Raya granite grade towards granodiorite, whereas the Kuah granite grade towards alkali granite.

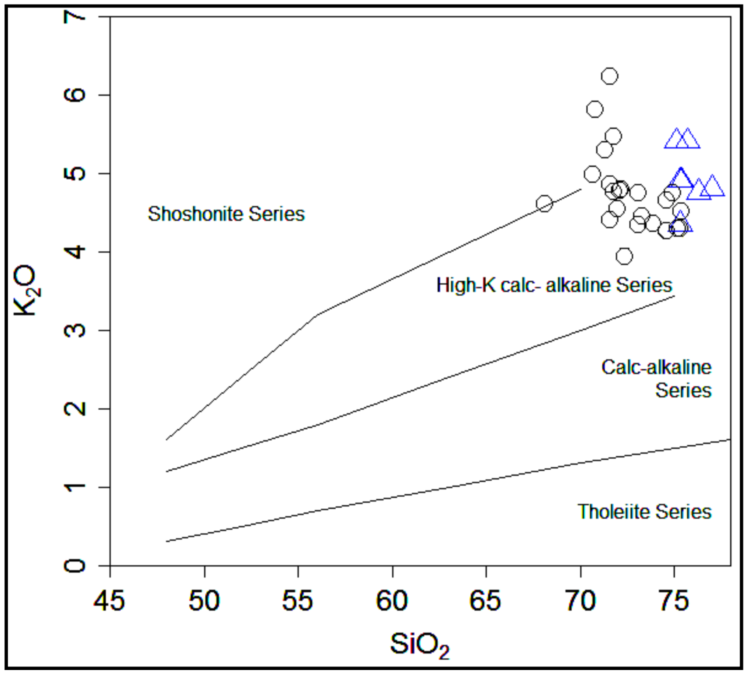


Fig. 4.2 : K_2O vs. SiO_2 diagram of the Gunung Raya and Kuah Granite. Compositional field after Peccerillo and Taylor (1976). (○ - Gunung Raya , △ - Kuah)

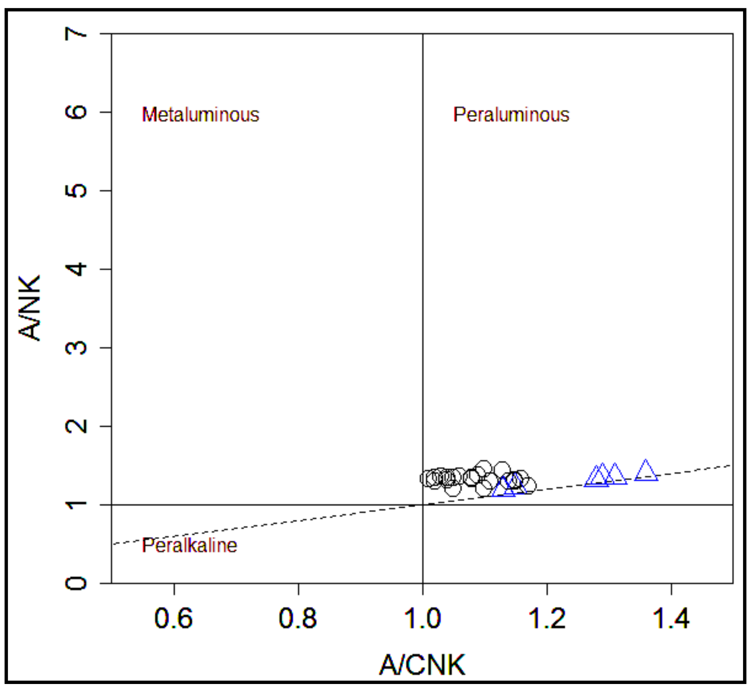


Fig. 4.3: ACNK vs A/NK plot of Gunung Raya and Kuah granite of Langkawi Island. (○ - Gunung Raya , △ - Kuah)

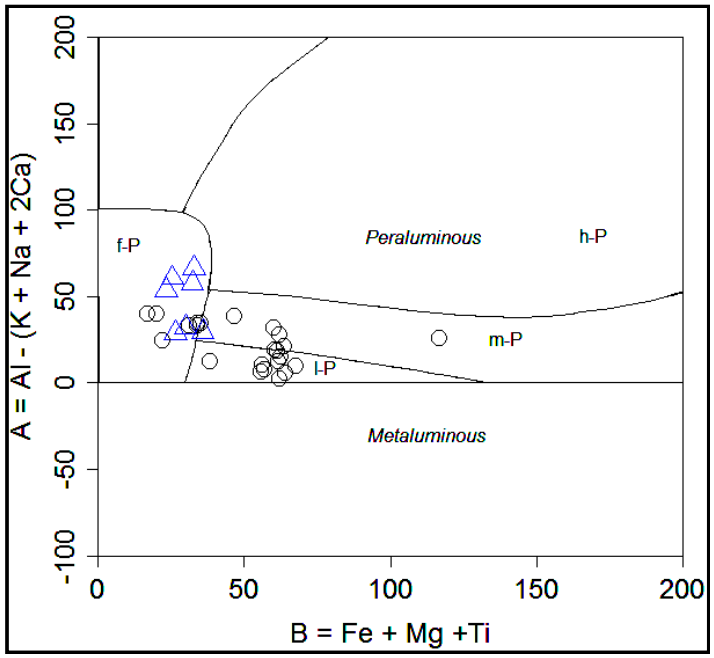


Fig. 4.4 : B-A plot of Gunung Raya and Kuah granite , after Villaseca et al. (1998). Gunung Raya granite plot as weakly peraluminous whereas Kuah granite plot as felsic-peraluminous.

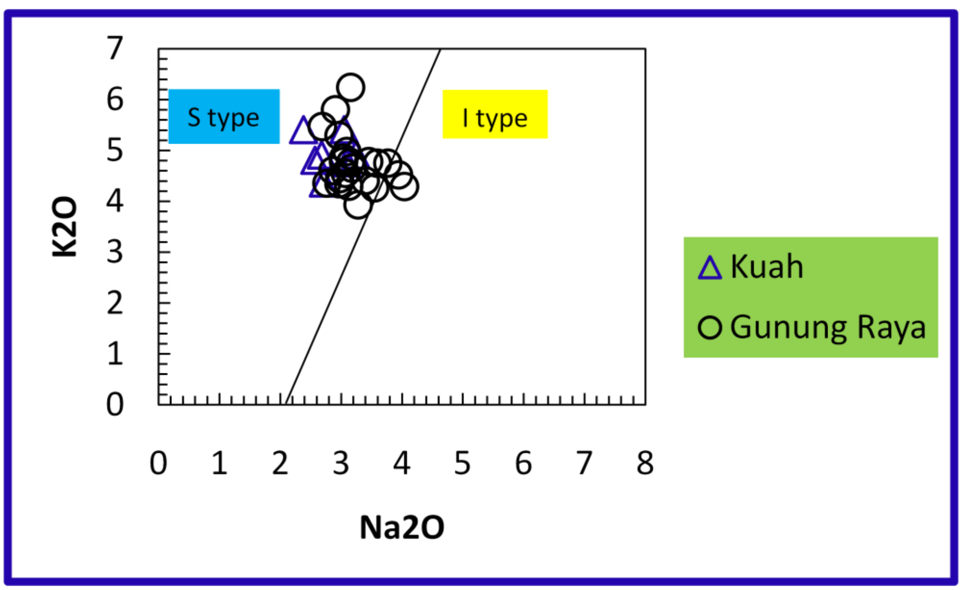


Fig. 4.5: Na_2O vs K_2O plot of the Gunung Raya and Kuah granite. Field of the 'S' and 'I' type granite is after Chappell and White (1983). Note that in general the Gunung Raya granite contain slightly higher Na_2O content compared to the Kuah granite at the same K_2O concentration.

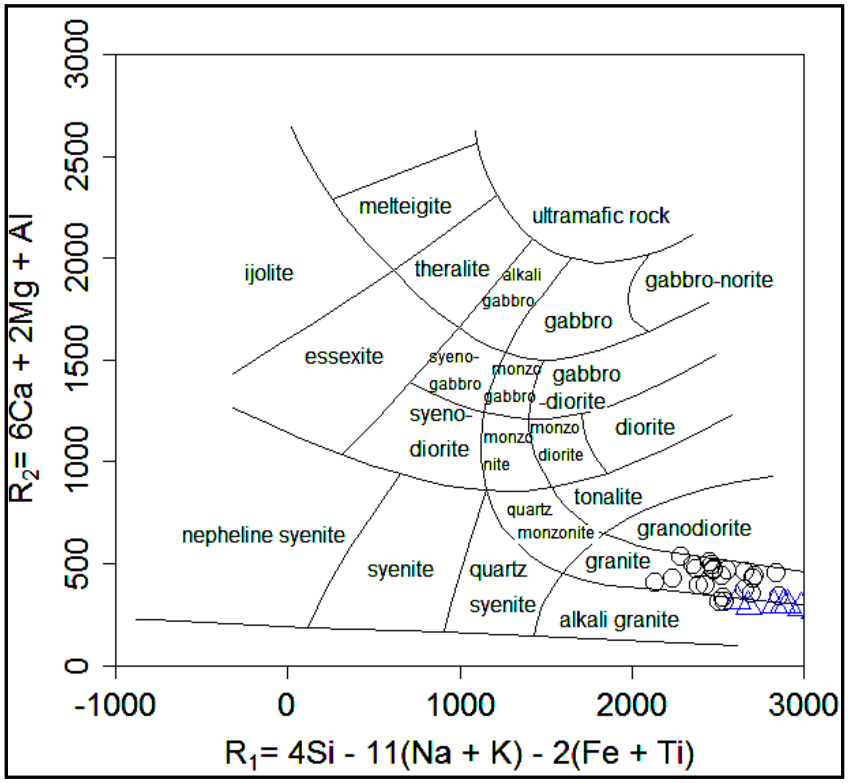


Fig. 4.6 : $R_1 - R_2$ plot of Gunung Raya and Kuah granite, after De La Roche (1980). All samples plot as granite. (○ - Gunung Raya , △ - Kuah)

4.3.1 Norm and aluminium saturation comparison

The Gunung Raya granite shows average Mol Al_2O_3/ ($CaO+Na_2O+K_2O$) or A/CNK ratios of ~ 1.01 - ~ 1.17 respectively. The Kuah granite shows average ratio of ~ 1.13 - ~ 1.31 respectively. The Kuah granite have higher A/CNK ratio which possibly reflects the occurrence of the aluminous minerals muscovite and garnet. According to Shand (1943) all samples from Gunung Raya and Kuah are peraluminous (exceeding 1) and indicate an S-type affinity (exceeding 1.1) following Chappell (1999); Chappell and White (2001).

CIPW norm of the granite is shown in Table 4.2 (Janošek et al., 2006), the Gunung Raya granite show ~28-36% quartz with one sample yielding ~39% (L-2). All samples are granitic in composition. The majority of Gunung Raya granite yield >1% normative corundum, ranging between 1.12 and 2.21%. On the other hand, all the Kuah granites yield >1% normative corundum, ranging between 1.73 and 3.57% respectively. Both Gunung Raya and Kuah granites are having normative corundum > 1%. This could also indicate S-type affinity for these rocks (Chappell and White, 2001).

Table. 4.2: Chemical composition and normative calculations of samples from the Gunung Raya and Kuah granite, Langkawi, Malaysia.

Sample	L1G	L2G	L3G	L4G	L5G	L6 G	L7 G	L8 G	L9 G	L10 G
Type	Granite	Granite	Granite	Granite	Granite	Granite	Granite	Granite	Granite	Granite
Major Elements										
SiO2	74.6	75.3	74.6	72.1	68.1	73.1	73.3	71.6	72.4	70.8
Al2O3	13.26	12.74	13.6	14.35	14.09	13.06	12.98	14.27	13.83	14.3
Fe2O3	1.84	1.64	1.81	2.51	5.49	3.05	2.9	1.86	2.89	2.98
CaO	0.68	0.7	0.79	0.84	1.69	1.33	1.27	0.93	1.4	1
MgO	0.35	0.33	0.37	0.47	1.52	0.81	0.77	0.48	0.76	0.77
Na2O	3.18	3.12	3.55	3.45	2.86	2.96	2.97	3.15	3.28	2.9
K2O	4.65	4.3	4.27	4.78	4.61	4.35	4.45	6.24	3.93	5.81
MnO	0.07	0.06	0.08	0.09	0.11	0.07	0.07	0.05	0.07	0.06
TiO2	0.19	0.21	0.18	0.29	0.82	0.43	0.41	0.24	0.4	0.45
P2O5	0.19	0.18	0.17	0.16	0.3	0.19	0.19	0.14	0.19	0.19
LOI	0.77	0.58	0.82	0.85	0.79	0.74	0.64	0.68	0.92	0.85
SUM	99.78	99.19	100.22	99.88	100.4	100.09	100.01	99.76	100.07	100.15
Trace Elements										
Ba	143	125	118	146	250	224	237	604	201	530
Be	13	21	26	8	5	7	19	7	14	8
Co	2.7	2.4	2.2	2.6	9.1	4.7	4.8	3.6	4.7	4.1
Cs	65.1	57.4	61.5	59	66.1	39.4	46.3	12.9	30.7	35.3
Ga	18.9	17.6	18	18.9	20	16.9	17.3	14.9	18.8	17
Hf	3.2	2.6	3	3.9	7.3	4.6	5.2	3.5	4.5	4
Nb	17.9	16.2	14.9	19.2	21.8	15.1	17.1	9.4	14.7	14.5
Rb	432.4	384.1	406.6	432	454.8	353.5	365.4	347.8	326.8	402.6
Sn	25	23	25	26	12	8	9	6	8	8
Sr	30.7	28.6	31.2	38.2	54.7	63.9	63.6	106.1	68.9	77.7
Ta	4.1	3.8	4.5	3.6	3.3	3.5	3.8	2.4	3.5	2.3
Th	15.4	12.4	12.7	22.5	35.6	22	22.3	14.1	23.1	23.2
U	18.8	8.1	16.1	20.1	12.8	11.2	14.1	9.1	12.6	14.6
V	12	8	9	13	60	27	26	14	27	28
W	8.4	10.5	12.1	6.6	3.3	3.8	4.8	7.8	2.4	1.9
Zr	91.4	82.4	79.2	138.4	263.5	174.5	205.2	103.8	163.4	153.5
Y	20.4	16.6	22.3	32.1	47.6	33.7	32.5	21.7	40.2	39.1
La	19.6	16.4	15.8	27.1	52.6	31.8	32.8	21.6	35.1	34.8
Ce	43.4	35.5	34.9	61.5	111.1	71	70.2	44.4	75.7	75.8
Pr	4.8	3.9	3.82	6.54	12.57	7.87	7.65	4.91	8.49	8.31
Nd	15.5	15.1	14.4	25.4	46.9	27.9	28.7	19.6	32.1	31.5
Sm	3.93	3.36	3.36	5.69	10.4	5.87	5.98	3.65	7.05	6.69
Eu	0.31	0.27	0.37	0.48	0.72	0.7	0.69	0.86	0.79	0.88
Gd	3.64	2.92	3.23	5.08	9.2	6.06	5.91	3.68	6.85	6.18
Tb	0.69	0.54	0.63	0.95	1.57	1.04	1.01	0.62	1.15	1.13
Dy	3.58	2.69	3.68	5.56	8.5	5.7	5.69	3.66	6.92	6.4
Ho	0.81	0.62	0.75	1.23	1.77	1.32	1.27	0.79	1.49	1.46
Er	2.16	1.72	2.54	3.52	5	3.72	3.44	2.27	4.4	4.21
Tm	0.34	0.28	0.37	0.57	0.77	0.59	0.55	0.36	0.66	0.63
Yb	2.33	1.8	2.63	4.35	5.04	3.43	3.61	2.37	4.54	3.94
Lu	0.36	0.31	0.42	0.61	0.72	0.58	0.55	0.38	0.68	0.64
Mo	0.9	1	0.9	0.7	2.4	1.4	1.6	2.1	0.7	1
Zn	35	33	34	45	87	45	45	23	39	43
Normative Mineral										
Quartz	36.86	39.21	35.84	31.69	28.78	35.71	35.66	27.08	34.68	28.94
Corundum	2.21	2.11	2.11	2.36	2.04	1.52	1.42	0.98	2.09	1.88
Orthoclase	27.48	25.41	25.23	28.25	27.24	25.71	26.30	36.88	23.23	34.34
Albite	26.91	26.40	30.04	29.19	24.20	25.05	25.13	26.65	27.75	24.54
Anorthite	2.13	2.30	2.81	3.12	6.42	5.36	5.06	3.70	5.70	3.72
Apatite	0.45	0.43	0.40	0.38	0.71	0.45	0.45	0.33	0.45	0.45

Sample	L11 G	L12 G	L13 K	L14 K	L15 K	L16 K	L17 K	L18 K	L19 K	L20 GQ
Type	Granite	Granite	Granite	Granite	Granite	Granite	Granite	Granite	Granite	Granite
Major Elements										
SiO2	71.6	73.9	76.3	75.4	75.3	75.7	75.3	75.1	77	70.7
Al2O3	13.69	12.26	12.49	13.07	12.63	13.18	12.85	12.95	12.39	13.95
Fe2O3	2.93	2.85	1.59	1.65	1.44	0.98	1.75	1.57	1.26	3.18
CaO	1.55	1.58	0.33	0.22	0.28	0.17	0.15	0.31	0.12	1.64
MgO	0.8	0.82	0.31	0.39	0.28	0.46	0.34	0.57	0.26	0.88
Na2O	3.03	2.76	3.09	2.68	3.24	2.38	2.71	3.05	2.57	3.09
K2O	4.86	4.36	4.76	4.91	4.91	5.41	4.36	5.41	4.81	4.98
MnO	0.06	0.06	0.04	0.05	0.05	0.02	0.03	0.04	0.02	0.06
TiO2	0.42	0.48	0.18	0.15	0.12	0.14	0.19	0.16	0.08	0.47
P2O5	0.14	0.15	0.1	0.08	0.11	0.09	0.07	0.09	0.07	0.17
LOI	0.78	0.79	0.59	1.33	0.7	1.31	1.09	0.91	0.83	0.57
SUM	99.94	100.08	99.85	99.97	99.11	99.84	98.86	100.13	99.43	99.7
Trace Elements										
Ba	412	332	96	95	70	98	57	143	79	405
Be	4	6	14	22	16	17	9	14	23	<1
Co	5.2	4.9	1.5	2	1.5	1.3	2.3	1.2	1.8	5
Cs	27.1	31	30.8	47.4	37.1	22.6	33	35.6	19.8	38.2
Ga	16.7	15.1	15	16.2	15	14.8	15.5	13.2	14.1	16.1
Hf	5.5	4.6	3.9	3.5	3.4	3	3.5	2.6	2.5	5.8
Nb	14.3	14	13.1	14	11.9	12.9	11.4	12.9	7.3	15.2
Rb	297.8	288.3	432.2	450	450.8	475.1	372.1	463	380.2	325.3
Sn	9	7	13	11	10	11	10	11	8	11
Sr	80.8	79.2	23.5	31.9	22.5	17.2	25.7	27.7	11.7	87.4
Ta	2.4	2.3	4	3.4	3.9	3.3	3.6	3.8	3	2.1
Th	21.4	23.5	14.7	14.6	13.7	14.3	15.6	15.1	10.2	24.4
U	4.5	10.5	8.7	10.5	18.8	15.8	7.2	10.6	14.7	8.7
V	32	29	<8	<8	<8	<8	<8	<8	<8	27
W	6.5	3.5	15.2	15.7	12.8	9.6	9	13.7	4.2	27.3
Zr	197.9	164.2	88.5	96.9	89.6	93.1	81.5	67.3	59.8	212.7
Y	32.7	39.2	26.7	24.2	29	47.1	30.1	19.3	40.2	35.7
La	37.7	36.7	15.6	15.1	14	16.4	15.6	13.5	10.4	37.9
Ce	80.3	78.4	34.1	32.5	31.6	36.4	35.6	30	28.1	83.8
Pr	8.85	8.6	3.92	3.93	3.51	4.84	3.87	3.4	3.58	9.17
Nd	32	32.2	16.3	14.9	16.6	17.3	13.9	11.7	16	30.1
Sm	6.47	6.44	3.5	3.67	3.16	4.99	3.8	3.02	4.79	6.77
Eu	0.87	0.9	0.19	0.2	0.2	0.35	0.23	0.18	0.34	0.85
Gd	5.61	6.22	3.28	3.34	3.24	5.64	3.9	2.47	5.74	6.18
Tb	1.01	1.1	0.7	0.68	0.69	1.2	0.81	0.52	1.19	1.1
Dy	5.62	6.09	4.04	4.24	4.6	7.47	5.22	3.3	7.01	6.82
Ho	1.22	1.47	0.93	0.96	0.99	1.69	1.21	0.71	1.69	1.35
Er	3.45	3.77	2.88	2.96	3.2	5.1	3.73	2.04	4.93	3.75
Tm	0.55	0.61	0.48	0.5	0.54	0.83	0.63	0.44	0.81	0.56
Yb	3.35	4.04	3.51	3.34	3.78	5.23	4.39	2.7	5.53	3.8
Lu	0.52	0.58	0.55	0.48	0.57	0.82	0.71	0.42	0.78	0.59
Mo	0.7	1.8	0.6	1.1	1.1	0.5	0.9	0.9	1	1.8
Zn	40	44	14	17	23	2	7	15	5	52
Normative Mineral										
Quartz	31.26	36.97	39.22	40.19	36.96	40.35	42.22	35.39	43.19	29.32
Corundum	0.96	0.49	1.89	3.14	1.74	3.32	3.57	1.73	2.90	0.90
Orthoclase	28.72	25.77	28.13	29.02	29.02	31.97	25.77	31.97	28.43	29.43
Albite	25.64	23.35	26.15	22.68	27.42	20.14	22.93	25.81	21.75	26.15
Anorthite	6.78	6.86	0.98	0.57	0.67	0.26	0.29	0.95	0.14	7.03
Apatite	0.33	0.36	0.24	0.19	0.26	0.21	0.17	0.21	0.17	0.40

Sample	L21 G	L22 G	L23 G	L24 G	L25 G	L26 G	L27 G	L28 G	L29 G	L30 G
Type	Granite	Granite	Granite	Granite	Granite	Granite	Granite	Granite	Granite	Granite
Major Elements										
SiO2	72	71.8	71.3	72.2	73.1	75.4	74.9	75.2	71.6	71.8
Al2O3	13.48	13.84	13.69	13.46	14.35	13.6	13.9	13.89	14.09	13.6
Fe2O3	2.95	2.68	3.04	2.91	1.94	1.24	1.29	1.21	2.75	2.55
CaO	1.51	1.74	1.53	1.28	0.85	0.54	0.3	0.32	1.84	1.49
MgO	0.81	0.7	0.83	0.79	0.35	0.22	0.14	0.07	0.72	0.75
Na2O	3.07	3.15	2.96	3.04	3.59	3.94	3.76	4.04	3.39	2.69
K2O	4.55	4.77	5.3	4.79	4.75	4.52	4.75	4.29	4.41	5.47
MnO	0.06	0.06	0.06	0.06	0.08	0.09	0.04	0.08	0.05	0.07
TiO2	0.44	0.37	0.44	0.42	0.18	0.1	0.04	0.02	0.38	0.45
P2O5	0.15	0.13	0.15	0.15	0.16	0.15	0.14	0.15	0.13	0.19
LOI	0.69	0.59	0.7	0.82	0.66	0.51	0.66	0.54	0.93	1.06
SUM	99.7	99.94	100.06	99.93	100.01	100.36	99.89	99.77	100.33	100.21
Trace Elements										
Ba	325	493	454	313	154	53	6	8	339	601
Be	3	6	4	5	19	19	5	6	8	2
Co	4.1	4.6	4.9	4.4	2.2	1.5	1.3	0.7	4.8	5.2
Cs	31.8	37.1	35.6	25.8	53.1	50.7	44.4	25.2	29.9	38.9
Ga	15.1	14.2	14.8	15.1	15.7	14.6	18.3	18	15.6	13.8
Hf	5.6	4.3	5.4	5.2	2.6	1.3	2.2	2.4	4.1	4.8
Nb	14.2	13.5	13.9	13.5	13.7	7.9	10.9	9.9	11.9	16
Rb	295.2	298.4	319	300.3	361.1	350.3	571.3	506.5	274.2	346.1
Sn	10	10	10	9	17	9	7	7	9	10
Sr	78	94.2	87.1	75	34.1	12	2.7	2.8	86.2	91.2
Ta	2.3	2.6	2.3	2.3	3.5	2.1	2.2	3.6	2.1	1.9
Th	25.4	21	23.2	23.2	13.5	7.9	7.4	6.6	22.7	27.9
U	15	25.8	10.5	11.7	12	22.3	6	5.5	9.8	8.5
V	27	23	27	26	<8	<8	<8	<8	29	30
W	19.3	3.2	6.8	9.4	12.4	8	49.3	32.8	10	6.4
Zr	179.5	150.3	179.9	163.3	85.2	40.2	34.9	34.1	149.7	197.1
Y	36	29.9	35.3	33.1	19.7	15.2	13.2	7.2	28.4	34.2
La	36.2	32.4	36.1	36.3	14.8	7.6	3	4.1	35.3	47.5
Ce	79	68.1	80.1	78.8	34.6	17.1	7.2	8	75.3	100.8
Pr	8.7	7.46	8.48	8.29	3.84	1.87	0.94	0.91	8.23	11.07
Nd	32.1	26.8	31.5	31.1	13.9	7.4	4.5	3.6	30.5	40.2
Sm	6.32	5.73	6.4	6.68	3.37	1.86	1.32	1.06	6	8.44
Eu	0.85	0.89	0.89	0.78	0.37	0.13	<0.02	<0.02	0.88	1.05
Gd	6.12	5.28	5.91	5.63	3.14	2.1	1.41	0.8	5.06	7.43
Tb	1.07	0.93	1.05	1.01	0.6	0.45	0.34	0.19	0.93	1.14
Dy	5.65	5.81	6.1	5.92	3.3	2.63	2.15	1.1	4.93	6.3
Ho	1.28	1.2	1.34	1.31	0.72	0.54	0.52	0.24	1.2	1.28
Er	3.71	3.32	3.95	3.75	2.23	1.6	1.48	0.86	2.97	3.2
Tm	0.6	0.51	0.58	0.55	0.36	0.3	0.26	0.2	0.49	0.5
Yb	3.75	3.05	3.82	3.41	2.47	2.23	2.31	1.97	3.23	3.06
Lu	0.57	0.46	0.58	0.54	0.36	0.34	0.32	0.31	0.49	0.5
Mo	4.4	1.4	2.8	1.8	0.9	0.9	1	0.9	1.4	1.6
Zn	43	42	45	42	33	20	1	<1	44	33
Normative Mineral										
Quartz	32.71	30.82	29.71	32.69	32.15	34.12	34.39	34.92	30.35	31.44
Corundum	1.12	0.64	0.66	1.31	2.14	1.60	2.36	2.38	0.71	1.00
Orthoclase	26.89	28.19	31.32	28.31	28.07	26.71	28.07	25.35	26.06	32.33
Albite	25.98	26.65	25.05	25.72	30.38	33.34	31.82	34.19	28.69	22.76
Anorthite	6.51	7.78	6.61	5.37	3.17	1.70	0.57	0.61	8.28	6.15
Apatite	0.36	0.31	0.36	0.36	0.38	0.36	0.33	0.36	0.31	0.45

Note: CIPW normative calculations according to Janošek et al. (2006).

4.4 Trace Elements Geochemistry

Trace elements using Harker diagrams for Langkawi granite is shown in Figure. 4.7. In general samples from the two plutons clearly fall along two separate and distinct trends. Most trace elements of Gunung Raya is scattered but Sr, Ba, Ce, Zn and Zr produce a straight line. In rocks from the Gunung Raya granite, Sr, Ba, Nb, Co, La, V, Zr, Th, Ce, Nd, Zn, Pb decrease and Rb increase with increasing SiO_2. Trace elements in the Kuah granite show some odd trends. Elements such as Co, Y, Pb increase and Sr, Rb, Nb, Ba, Th, Zr, V, La, Ce, Zn decrease with increasing SiO_2 and Nd produce scattered trend (Fig. 4.8). The trace elements Ba, Sr, Nb, Ce, La, Zr, Zn, Nd, V, Co and Th in both Gunung Raya and Kuah granite plot in two trends respectively. This may indicate that both of Gunung Raya and Kuah granites are from different granitic pulses. This is also confirmed by two separate trends of P_2O_5 vs SiO_2 diagram.

The Kuah granite also shows significantly low Ba, Sr, Nb, Ce, La, Zr, Zn, Nd, V, Co and Th compared to the Gunung Raya granite. One of the distinctive differences between the Gunung Raya and Kuah granite is in the La and Ce contents. These differences are well displayed on La vs Ce diagram (Fig. 4.9). The Zn,Co, and Zr contents from both Gunung Raya and Kuah granites have generally decreased with increasing SiO_2. The decrease of these elements is probably related to fractionation of the opaque phase, biotite and zircon.

Among the trace elements, Rb/Sr ratios are good indicators of magmatic differentiation. Rb/Sr ratio is higher in the Kuah granite average (~20.55) compared to those from the Gunung Raya granite average (~7.48). The high Rb/Sr ratio of the Kuah granite reflects the highly evolved nature of the magma compared to the Gunung Raya granite. The decrease of Ba concomitant with Sr suggests that K-feldspar, biotite and plagioclase are being removed in differentiation sequence for both Gunung Raya and Kuah granite (Fig

.4.13). Other than that, the plot of Sr vs CaO shows the positive slope, which supports that plagioclase is being removed in the differentiation sequences (Fig. 4.15).

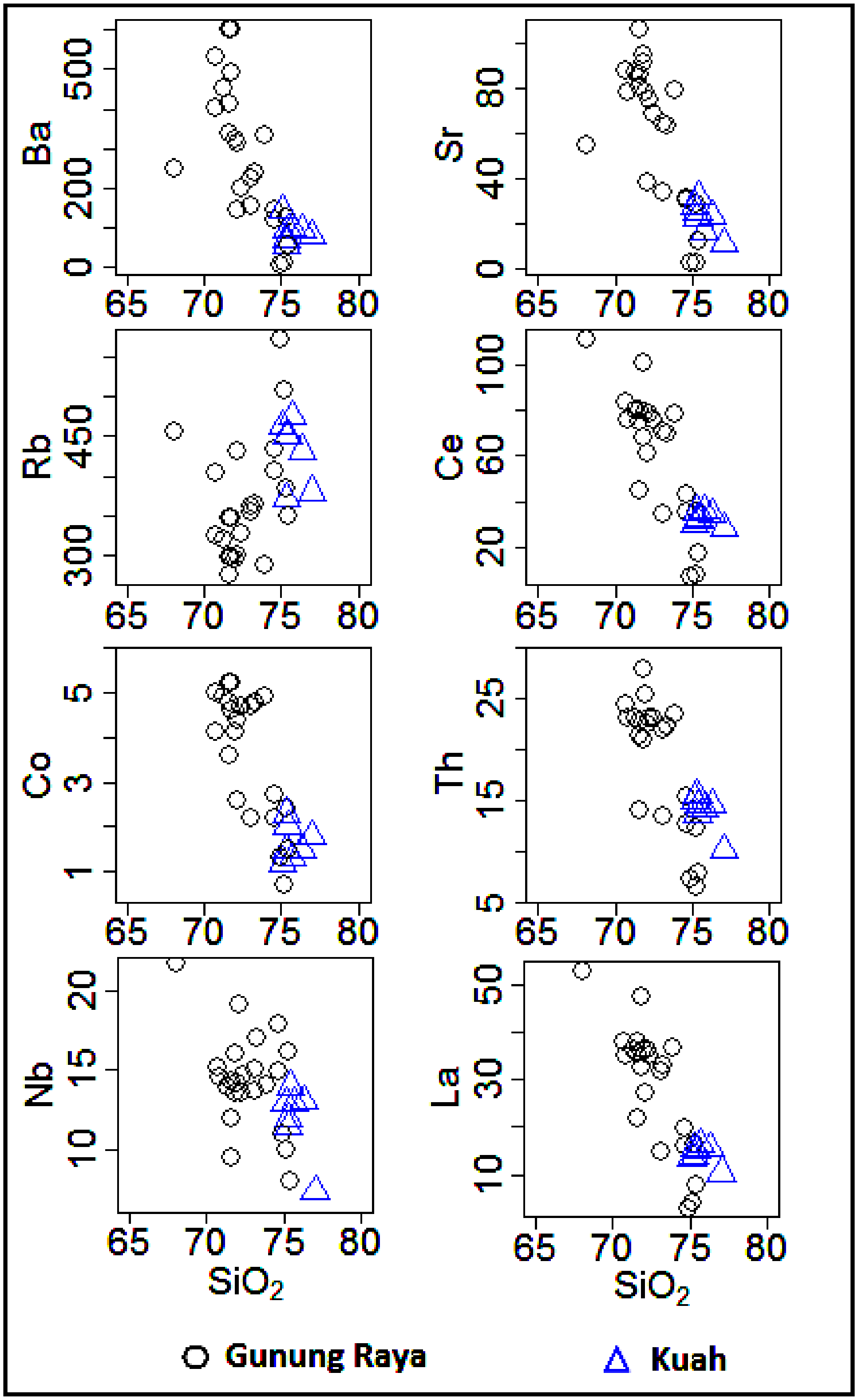


Fig. 4.7 : Trace elements Harker diagram of the Gunung Raya and Kuah granite from the Langkawi Island.

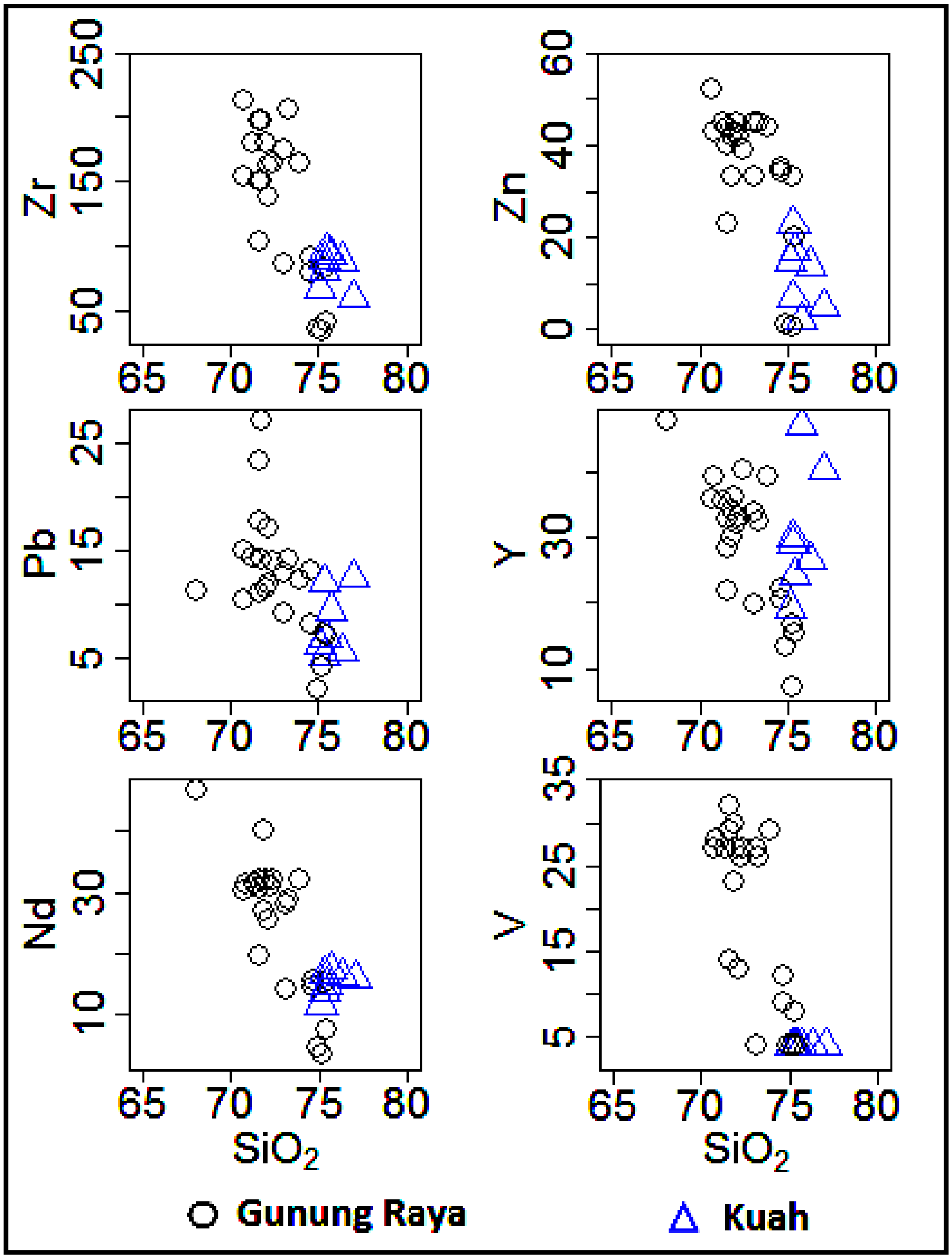


Fig. 4.8 : Trace elements Harker diagram of the Gunung Raya and Kuah granite from the Langkawi Island.

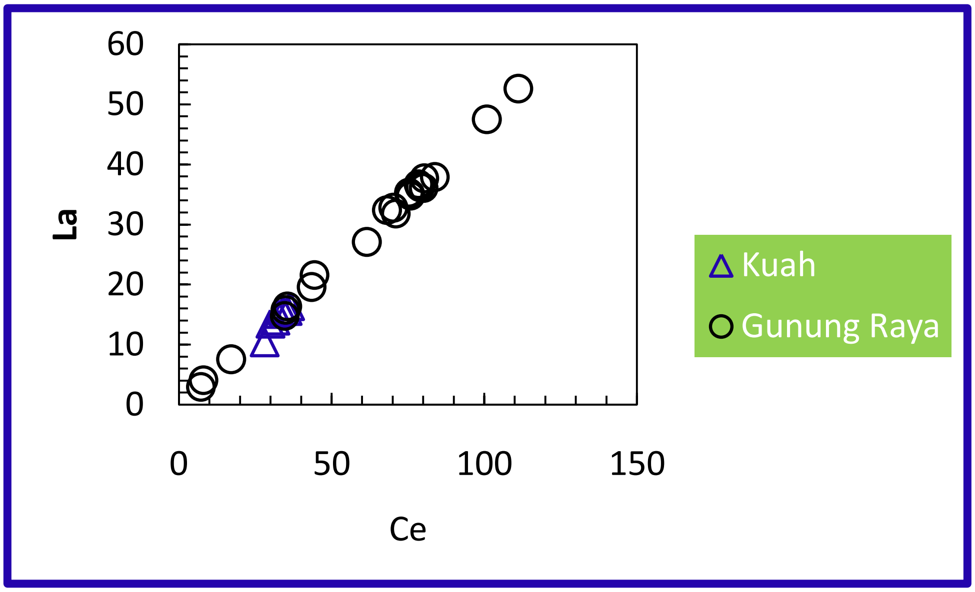


Fig. 4.9 : La vs Ce (in ppm) plot of the Gunung Raya and Kuah granite.

In the multi element variation diagram normalized to the Primitive Mantle (Sun & McDonough, 1989) the Gunung Raya and Kuah granite (Fig. 4.10a&b) show similar trend but Kuah is more regular. However, the Gunung Raya data scatters slightly more than the Kuah data with depletion of Sr, Ti and slightly enrichment of Th, U, K, Pb, P, Nd, Sm and Dy.

Result of the REE analysis and the profiles are plotted in (Fig. 4.11a&b). In general the Langkawi granites have the familiar 'bird-wing' pattern with a negative Eu anomaly in the middle. The Gunung Raya granite differs slightly from the Kuah granite in having higher LREE contents and subsequently a steeper LREE gradient. Nevertheless, the trend is visible in that the Gunung Raya granites have higher total REE (84.06 to 266.86 ppm) compared to the Kuah granite (74.4 to 108.26 ppm).

The $(Eu/Sm)_N$ ratio of the Gunung Raya granite (0.02-0.41) is higher compared to that of Kuah granite (0.14-0.19). The difference is again noticeable in the $(La/Lu)_N$ ratios as the Gunung Raya granites have higher values (0.97-9.87) compared to the Kuah granite

(1.38-3.27). A plot of total REE versus Rb/Sr (Fig. 4.12) also differentiates the Gunung Raya and Kuah granites. The Gunung Raya granite with their lower Rb/Sr ratios are grouped separately from the group of higher Rb/Sr ratio of the Kuah granite.

The chondrite normalized REE spectra of the sample groups, normalized after (Sun & McDonough, 1989). The Gunung Raya and Kuah granites show moderately to strongly fractionated LREE patterns as is indicated, according to Rollinson (1993), by the average $(La/Sm)_N$ = 3.20 and 2.43 respectively. The Gunung Raya and Kuah granites, on the other hand, show moderate to weak HREE fractionation (Rollinson, 1993) expressed by the average $(Gd/Yb)_N$ = 1.20 and 0.77 respectively. All samples show strongly negative Eu anomaly as indicated by the average $(Eu/Eu^*)_N$ ratio (Rollinson, 1993). The value of the negative europium anomaly is for the Gunung Raya and Kuah granites 0.23 – 0.49 and 0.17 – 0.20 respectively.

4.4.1 LIL modeling

The LIL elements Ba,Sr and Rb are of considerable value in determining the type and amount of major phase fractionation in intermediate and acid rocks because they are held predominantly in the major phases. Each element behaves somehow differently; for example, Rb is taken up preferentially by biotite, Ba by biotite and alkali feldspar and Sr by plagioclase and K-feldspar.

Using Rayleigh fractionation equation ($C_1/C_0 = F^{Da-1}$ where C_0 : concentration of element 'a' in the original melt, C_1 : concentration of element 'a' in residual melt, Da : bulk distribution coefficient for element 'a' and F : weight fraction of melt remaining) and the Kd values for rhyolitic liquid (Arth,1976). The net change has been calculated in composition of the liquid after 30% Rayleigh fractionation by removing K-feldspar, hornblende, plagioclase or biotite. The important of K-feldspar, biotite and plagioclase in the differentiation is consistent with large ion lithophile (LIL) modelling.

Inter element vector for LIL modelling is shown in Figure. 4.14. Each of the diagram is the vector diagram representing the net change in composition of the liquid after 30% Rayleigh fractionation by removing K-feldspar, hornblende, plagioclase or biotite. Only one trend has been identified in the early crystallizing phases, the trend is dominated by K-feldspar and plagioclase in both Gunung Raya and Kuah (Fig. 4.14). Thus, the Ba vs Sr log-log plots suggests that crystal fractionation via crystallization of plagioclase + K-feldspar and biotite play an important role in the magmatic evolution of the both Gunung Raya and Kuah granite. The most likely mechanism is crystal fractionation as indicated by LIL modeling.

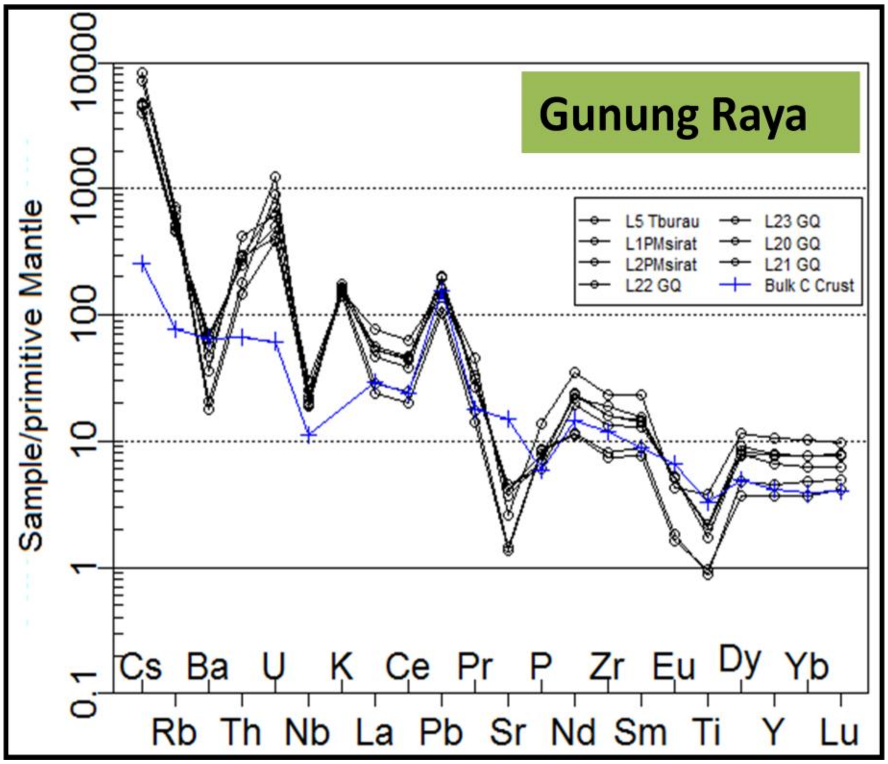


Fig. 4.10a : Multi-element variation plots of Gunung Raya granite, normalization value are from (Sun & Mc Donough, 1989).

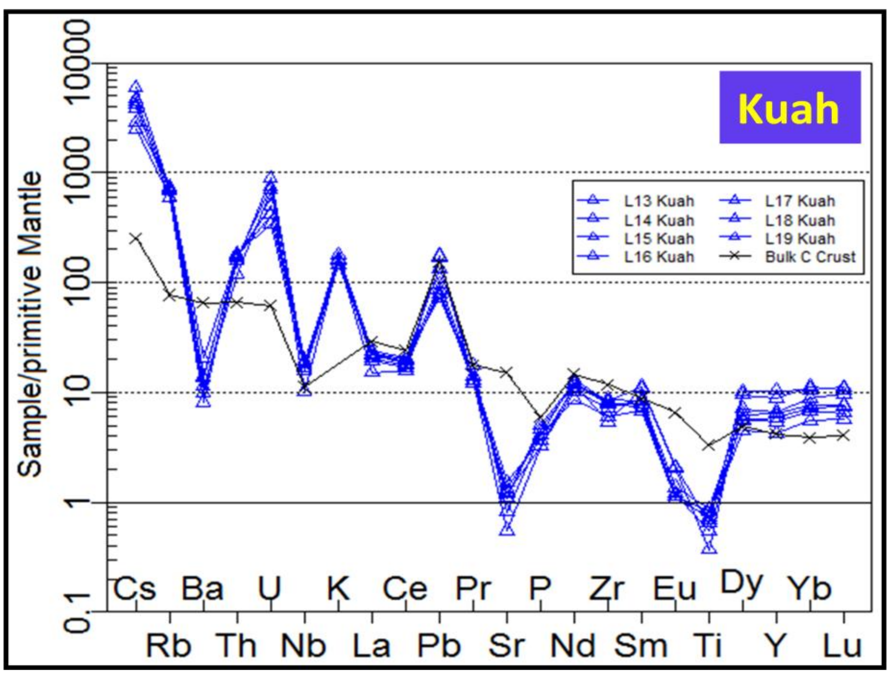


Fig. 4.10b : Multi-element variation plots of Kuah granite , normalization value are from (Sun & Mc Donough, 1989).

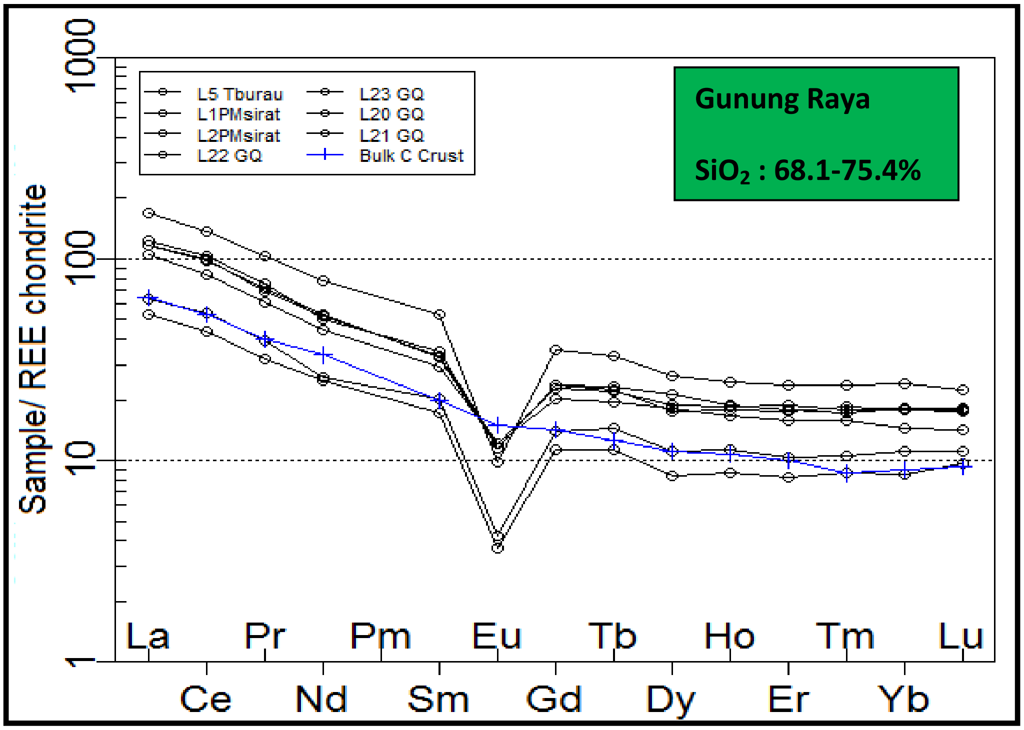


Fig. 4.11a : REE profiles for Gunung Raya granite, normalization value are from Sun and McDonough (1989).

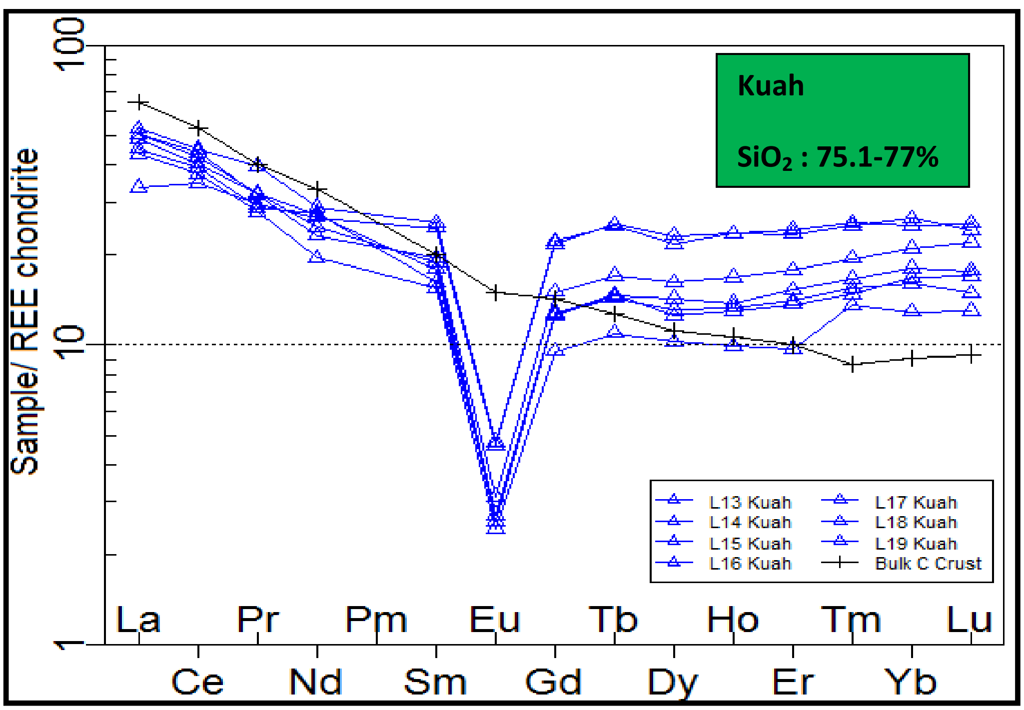


Fig. 4.11b : REE profiles for Kuah granite, normalization value are from Sun and McDonough (1989).

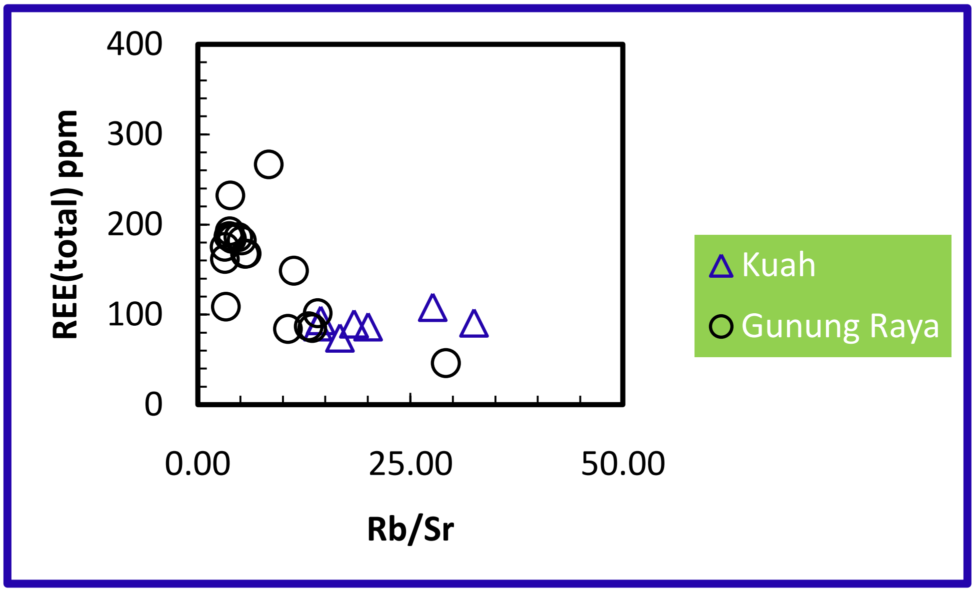


Fig. 4.12 : REE modelling of the Gunung Raya and Kuah magma.

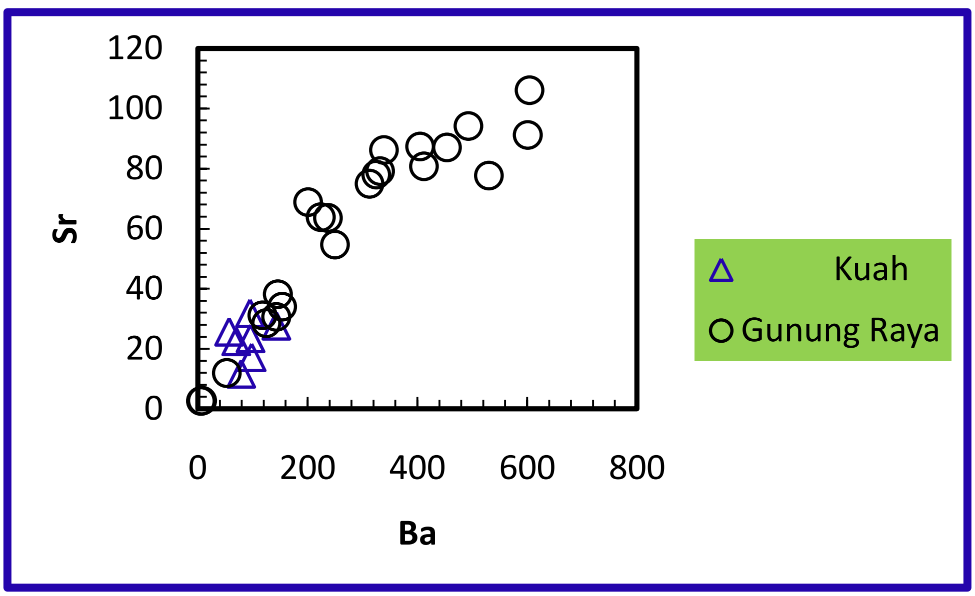


Fig. 4.13 : Sr vs Ba diagram for the Gunung Raya and Kuah granite.

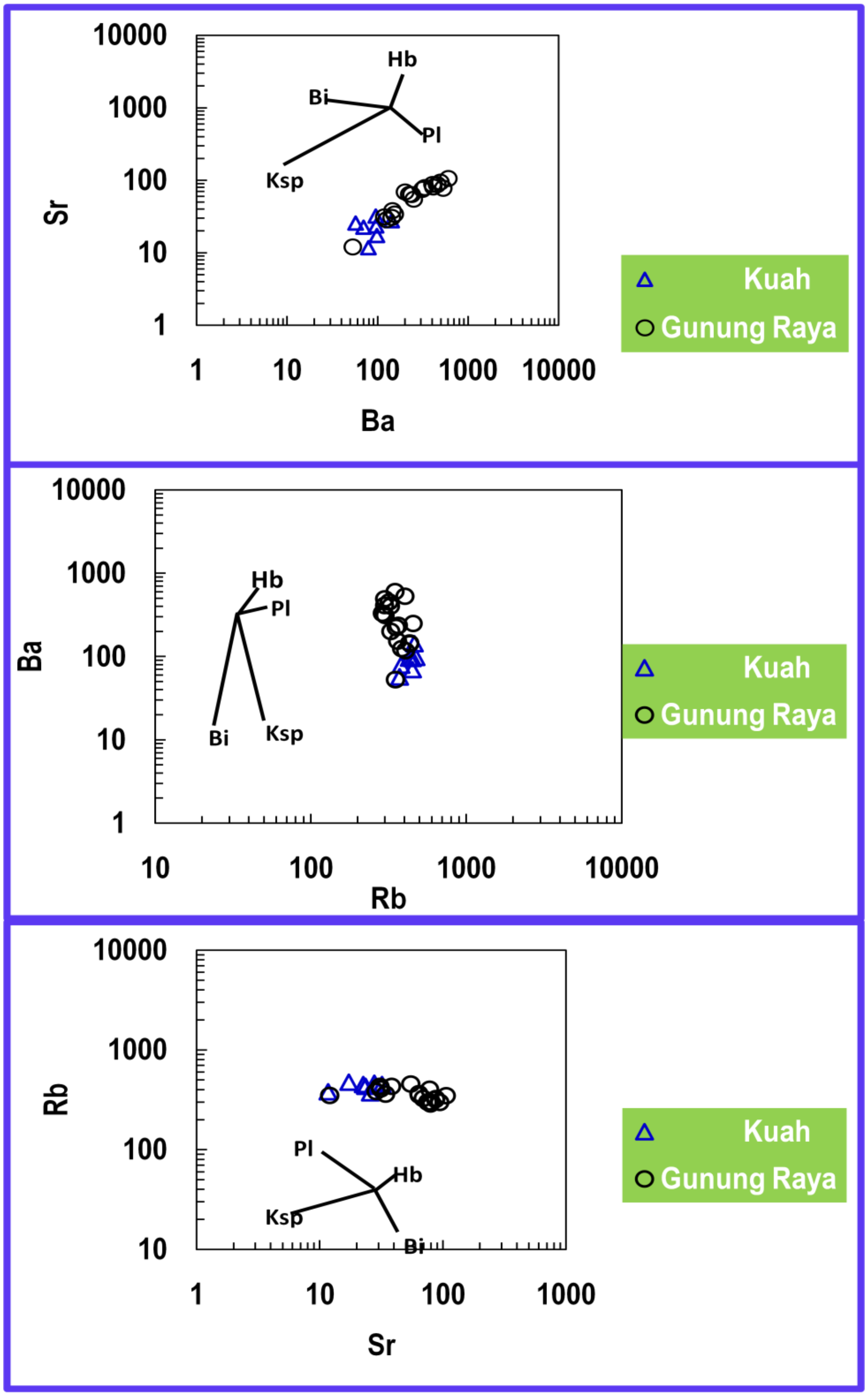


Fig. 4.14 : Large ion Lithophile elements modeling of the Gunung Raya and Kuah granite. Mineral vectors indicate paths of evolved liquid for 30 % of mineral precipitation. Ksp= K-feldspar; Pl=Plagioclase; Bi=Biotite and Hb=Hornblende

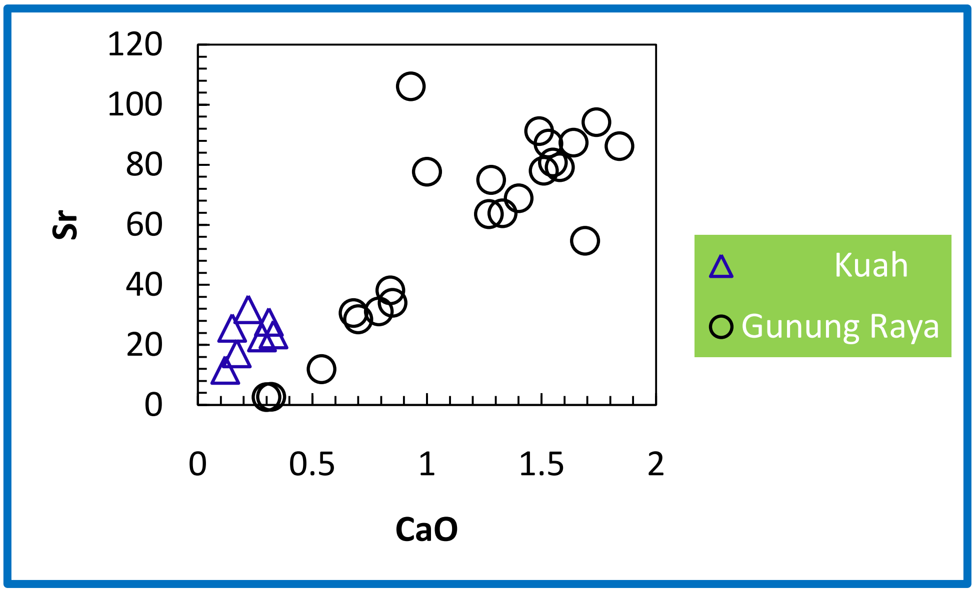


Fig. 4.15 : Sr vs CaO diagram for the Gunung Raya and Kuah granite.

4.5 Zircon Thermometry of Gunung Raya and Kuah granite

Watson and Harrison 1983; Baker et al. 2002; Hanchar and Watson 2003 and Harrison et al. 2007 predicted the saturation of zircon as a function of whole-rock composition and temperature $ln\ D_{zr}^{zircon/melt} = (-3.80 - [0.85\ (M-1)] + 12900/T)$. Saturation temperatures for the rocks in this study were estimated using the experimental calibration of Watson and Harrison (1983) (compositional range: M = (Na + K + 2 Ca)/(Al×Si)=0.9-1.9). All samples from Gunung Raya and Kuah have M<1.9. The results are listed in the Table (4.4) and the plot of M vs Zr(ppm) is shown in (Fig. 4.16a). In the figure , the concentrations of Zr in the Gunung Raya and Kuah granites is compared to the proportions of Zr that can be dissolved in granitoid melts of various compositions at different temperatures (Watson and Harrison, 1983).

According to Watson (1979), about 110 ppm Zr are required for zircon saturation in peraluminous melt (Langkawi granites yield average Zr ppm ~ 127). This implies that high Zr contents lead to overestimate the crystallization temperature of the peraluminous granitic melt. A 100 ppm Zr saturation level implies liquids temperature of ~750°C, a typical estimate of metasedimentary melting temperature producing S-type melts (ie. Wyllie, 1977; Clemens and Wall, 1981). The Gunung Raya granite has higher Zr contents (24-263ppm) and thus higher saturation temperature of 677-830°C with increasing SiO_2. It can clearly see in the plot of SiO_2 vs TZr.sat.C which shows the gradual decrease with evolution of the magma (Fig. 4.16b). The Kuah granites have lower Zr contents (59-96ppm) and yield saturation temperature of (725-766°C).

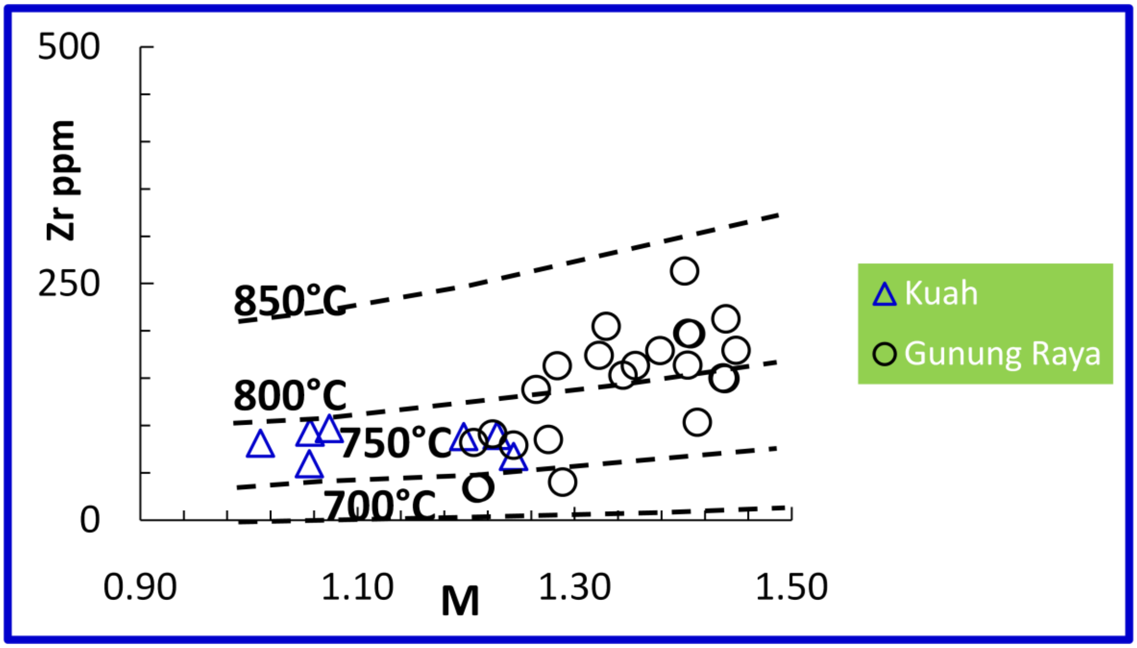


Fig. 4.16a : Zr concentrations in the granites plotted against M, the cation ration (Na+K+2Ca)/(Al×Si). The line marked 700-850°C indicates the proportion of Zr that can be dissolved in granitoid melts of different compositions, expressed by the M parameter (Watson and Harrison, 1983).

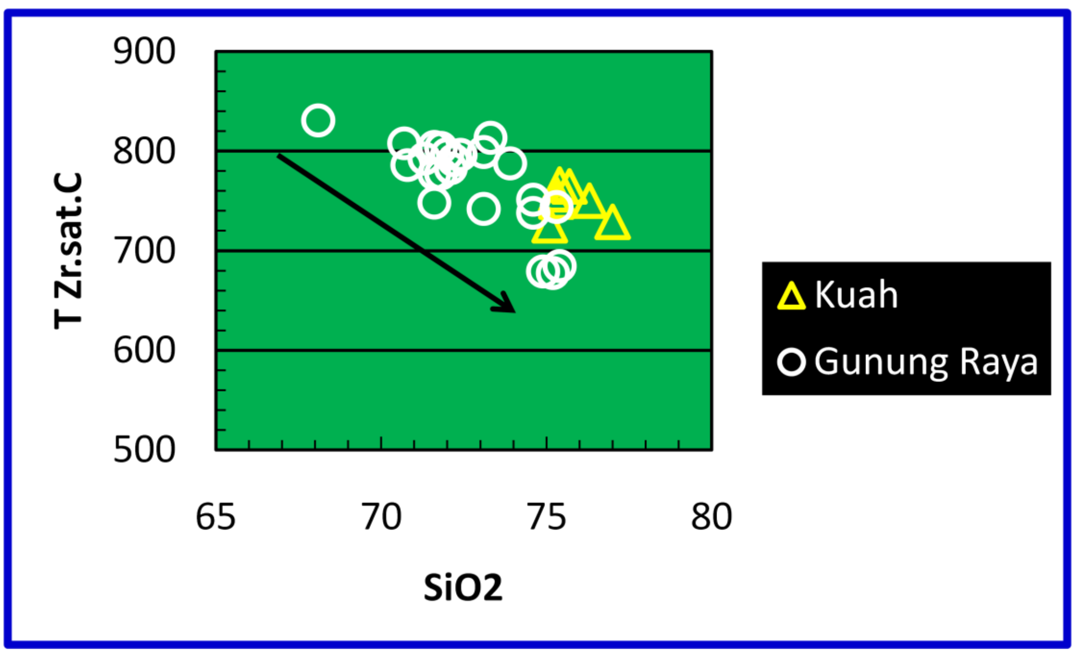


Fig. 4.16b : Zircon saturation temperature vs SiO_2 for the Gunung Raya and Kuah granite.

Table. 4.5: Chemical data used to describe Zr saturation behavior.

Sample	M	Zr(PPM)	TZr Sat.C	Specific temperature of Zircon saturation			
				Zr(Sat)700°C	Zr(Sat)750°C	Zr(Sat)800°C	Zr(Sat)850°C
L1PMsirat	1.22	91.4	751	47.1	90.1	162	276.7
L2PMsirat	1.21	82.4	744	46.4	88.7	159.6	272.6
L3PMsirat	1.24	79.2	738	47.9	91.5	164.7	281.3
L4PMsirat	1.26	138.4	783	48.7	93.2	167.7	286.3
L5 Tburau	1.40	263.5	830	54.7	104.6	188.3	321.5
L6 Tburau	1.32	174.5	799	51.2	97.9	176.1	300.7
L7 Tburau	1.33	205.2	813	51.5	98.4	177.1	302.4
L8 Tburau	1.41	103.8	748	55.3	105.7	190.2	324.8
L9 Tburau	1.28	163.4	796	49.5	94.7	170.4	291
L10Tburau	1.34	153.5	786	52.2	99.7	179.4	306.4
L11 Tewa	1.40	197.9	804	54.9	104.9	188.8	322.3
L12 Tewa	1.40	164.2	787	54.9	104.9	188.8	322.4
L13 Kuah	1.20	88.5	750	46.1	88	158.4	270.5
L14 Kuah	1.07	96.9	766	41.5	79.2	142.6	243.5
L15 Kuah	1.23	89.6	749	47.3	90.4	162.6	277.7
L16 Kuah	1.06	93.1	764	40.8	78.1	140.5	239.8
L17 Kuah	1.01	81.5	756	39.3	75.1	135.1	230.7
L18 Kuah	1.24	67.3	725	47.9	91.5	164.7	281.3
L19 Kuah	1.06	59.8	728	40.8	78	140.3	239.7
L20 GQ	1.44	212.7	808	56.6	108.1	194.5	332.1
L21 GQ	1.38	179.5	797	53.7	102.7	184.7	315.5
L22 GQ	1.44	150.3	777	56.4	107.8	194	331.3
L23 GQ	1.45	179.9	792	57	108.9	196	334.7
L24 GQ	1.36	163.3	790	52.7	100.7	181.2	309.4
L25 PK	1.28	85.2	742	49.2	94.1	169.2	289
L26 PK	1.29	40.2	684	49.8	95.1	171.1	292.2
L27 Tewa	1.21	34.9	679	46.6	89.1	160.4	273.9
L28 Tewa	1.21	34.1	677	46.5	88.9	160	273.3
L29 GQ	1.44	149.7	777	56.5	108	194.3	331.9
L30 GQ	1.41	197.1	803	55	105.1	189.2	323

4.6 Discussion

The range of SiO_2 for the Gunung Raya and Kuah granite are 68.1 to 75.3 and 75.1 to 77 % respectively. This shows that the Kuah granite are more siliceous. Fractional crystallization during the magmatism was recognized in the Harker diagram pattern since chemical composition of trace and rare earth elements of igneous rocks are mainly controlled by mineral/melt partition coefficients. The negative correlations between Al_2O_3, CaO, P_2O_5, MgO, FeO^T, MnO,TiO_2 and SiO_2 (Fig. 4.1) suggest that the granitic rocks are likely the result of fractional crystallization during magmatic evolution. The fractional crystallization is also confirmed by the Sr depletion and Eu negative anomaly which indicate continuous plagioclase fractionation during differentiation. The decrease in P_2O_5, MgO and FeO during magmatic evolution indicates separation of apatite and mafic mineral (such as biotite) during crystallization. Early fractionations of apatite and ilmenite were evidenced by the systematic decreasing trend of P_2O_5 and TiO_2 along with FeO with increasing SiO_2. Langkawi granites are characterized by low total Fe values (average ~2.29) that reflect the typically reduced nature of the source of S-type granitoids. Langkawi granite show fundamental S-type features like peraluminous compositions, high K_2O/Na_2O, low total Fe and restricted composition range dominated by high-SiO_2 granitoids.

The trace elements Ba, Sr, Nb, Ce, La, Zr, Zn, Nd, V, Co and Th in both Gunung Raya and Kuah granite plot in two trends respectively. This may indicate that Gunung Raya and Kuah granites may come from several separate granitic pulses. Zn,Co, and Zr generally decrease with increasing SiO_2. The decrease of these elements is probably related to fractionation of the opaque phase, biotite and zircon. Rb/Sr ratio are higher in the Kuah granite(14.11-32.50) compared to those from the Gunung Raya granite (29.7-43.17). The high Rb/Sr ratio of the Kuah granite reflects the highly evolved nature of the magma compared to the Gunung Raya granite.

Both Gunung Raya and Kuah unit are characterized by the high-K calc-alkaline affinity, Ba, Sr and Nb negative trends and enrichment of Rb, K and La which are compatible to those of typical crustal melt (Chappell and White, 1992). Furthermore, distinct negative anomalies for Nb, Sr, and Ti are showing typical upper crustal compositions (Rollinson, 1993). In addition the negative anomalies in Ba, Nb, Sr, and Ti that become increasingly marked towards the most felsic and fractionated granitic facies: such trends are typical of collisional peraluminous granitoids (Thompson et al., 1984).

Figures 4.11 a&b, show chondrite normalized REE spectra according to Sun and McDonough (1989), Langkawi granites have the familiar 'bird-wing' pattern with a negative Eu anomaly in the middle. For the Gunung Raya and Kuah granites the average normalized (Ce/Yb)N= 4.73 and 2.18 respectively, indicating strong depletion of HREE compared with LREE (Rollinson, 1993). REE patterns of the Gunung Raya and Kuah granites, the gently sloping trend of both rock types can be explained by low REE-fractionation in the magma. The gently sloping trend of Gunung Raya and Kuah granites could possibly be explained by depletions of the LREE by hydrothermal processes generating a non-typical (La/Yb)N-slope (Fig. 4.11 a&b). The slight enrichment of HREE compared to the MREE is possibly due to garnet present in the granite since the lighter REE´s (La-Sm) are incompatible in the garnet structure whereas the heavier (Eu-Lu) are compatible (Blatt et al., 2006). Garnet in the Kuah granite is more abundant than in the Gunung Raya granite which might explain the lower average (Gd/Yb)N-values in the Kuah granite(0.77) compared to the Gunung Raya granite (1.20). A moderately negative Eu anomaly is normally subscribed to fractionation of plagioclase due to the similar ionic radius and charge of Eu2+ and Ca2+ (Rollinson, 1993;

Blatt et al., 2006). In the case of the Gunung Raya and Kuah granites, the negative Eu anomaly cannot as easily be subscribed to plagioclase fractionation if their (La/Yb)N-slope is to be explained by low magmatic fractionation and not by other processes. If magmatic processes did generate the atypical REE-pattern of the Gunung Raya and Kuah granites it is here proposed that when partial melting occurred, plagioclase remained in the source producing a Eu poor melt as described by Rollinson (1993).

4.7 Tectonic Implication

Tectonic evolution of Langkawi can be briefly interpreted as follow. The emplacement of granite during late Triassic times causing distension of the previous structures and the super-imposition of a dome structure in the Langkawi Islands.

Figure 4.17 shows the geotectonic discrimination plots of Pearce et al. (1984), based on some granites from different tectonic setting. The samples included in this study plot on the borders between the syn-collisional granite (syn-COLG), volcanic arc granite (VAG) and within plate granite (WPG) fields. The Gunung Raya and Kuah granites plot dominantly in between VAG-field and COLG-field.

Most of the Langkawi granite samples are plot into the Syn-collision field in the tectonic classification of Pearce et al., (1984) and Harris et al., (1986) which is shown in Figures 4.18 and 4.19. This evidence is in agreement with the formation of the Main Range granite (Ghani, 2009). Most of the Main Range granites dated by U-Pb zircons, which show 209±1 - 215±6 Ma from Penang ,198±2 Ma - 219±9 Ma from Ulu Kali (Liew & Page, 1985) and 210±7 - 215 Ma from Kuala Lumpur (Searle et al., 2012). This dating clearly corresponds to the Late Triassic (Carnian-Norian). This Late Triassic age is taken as the timing of the crustal thickening, partial melting along with the beginning of collision of Sibumasu with Indochina during the Indosinian Orogeny (Oliver et al., 2013). The end of the postcollisional magmatism is constrained to the earliest Jurassic by the age of the youngest S-type granite (198 ± 2 Ma, Liew and McCulloch, 1985) in the Western Belt (Fig. 4.32).

On the other hand K/Ar and Rb/Sr dates of Langkawi grantie range between 217 ± 8 Ma and 209 ± 6 (Bignell & Snelling, 1977). The age 217 Ma peak coincides with the oldest S-type granite date of 219 ± 5/9 Ma (U-Pb zircon age from Liew and McCulloch, 1985) from the Western Belt Main Range. These dates imply that magmatism was generally coeval with the Indosinian Orogeny. Thus, it can be assumed that the Main Range and Langkawi granites were at a relatively high level in the crust by the end of the Triassic-early Jurassic.

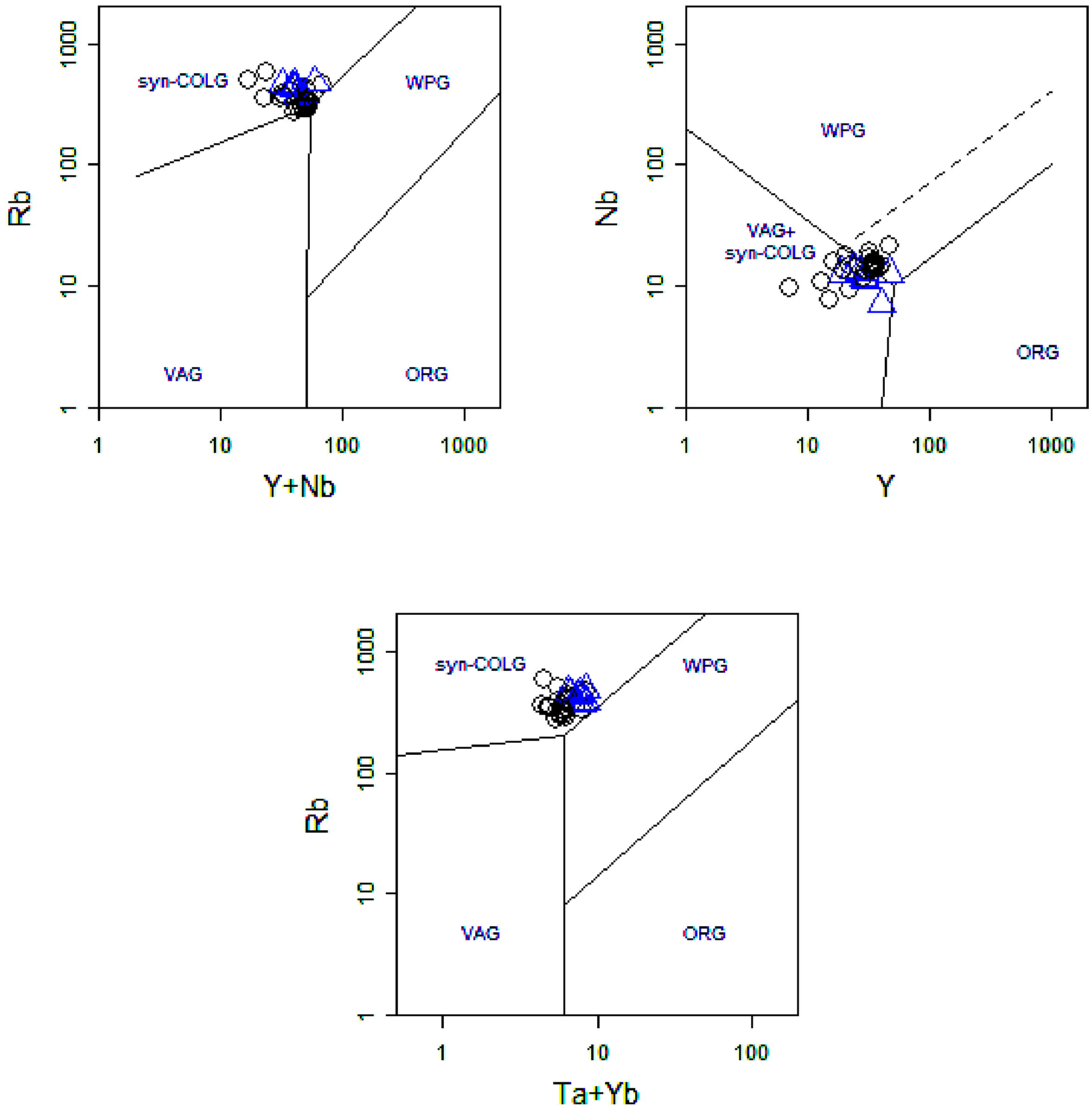


Fig. 4.17 : Geotectonic indication after Pearce et al. (1984). Symbols are, black circles: Gunung Raya granite, blue triangles Kuah granite, syn-COLG: syn-collisional granite, WPG: within plate granite, VAG: volcanic arc granite, ORG: orogenic granite. All samples plot on the border between the syn-COLG and VAG fields. .
(O - Gunung Raya , △ - Kuah)

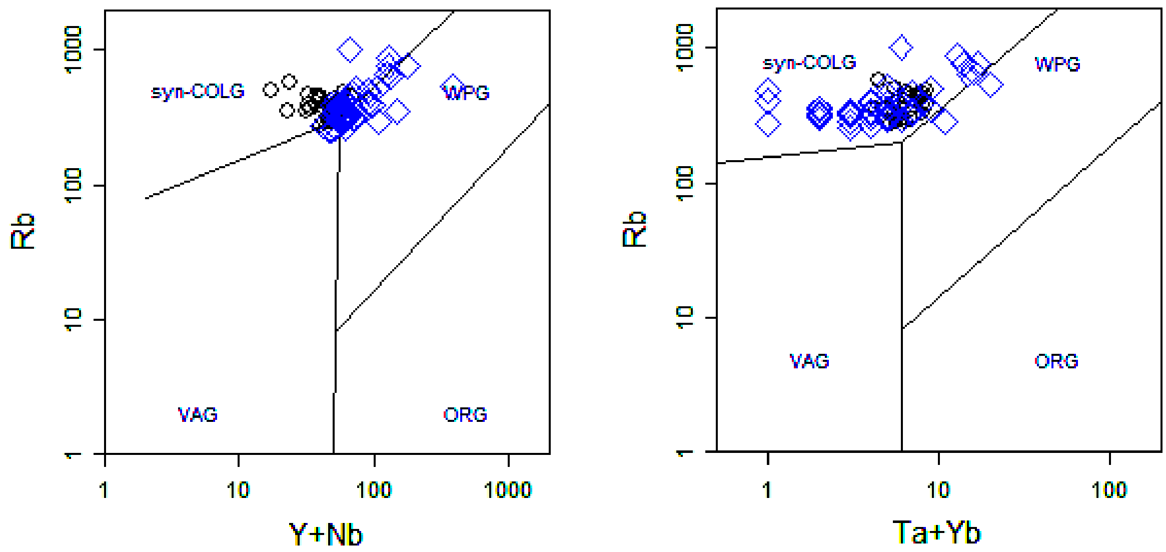


Fig. 4.18 : Geotectonic indication after Pearce et al. (1984). Symbols are, black solid circles: Gunung Raya granite, blue solid triangles Main Range granite.
(O - Langkawi , ◇ - Main Range)

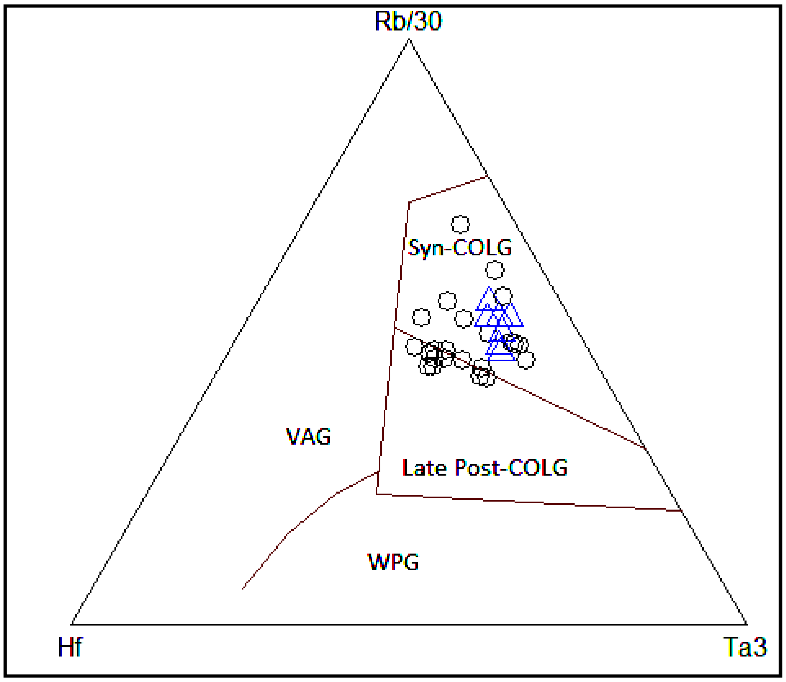


Fig. 4.19 : Geotectonic indication after Harris et al. (1986). Symbols are, black circles: Gunung Raya granite, blue triangles: Kuah granite, syn-COLG: syn-collisional granite, late-post COLG: late to post collisional granite, VAG: volcanic arc granite, WPG: within plate granite. The majority of Gunung Raya and Kuah data plots in the syn-COLG field. Only 6 samples from Gunung Raya granite pot in Late Post – COLG field.

4.8 Geochemical difference between the Langkawi and Main Range granites

4.8.1 Major elements

A data of the Main Range granites has been taken from Liew, 1983; Cobbing et al., 1992 and 30 samples of the Langkawi granites from the present study area. The comparison of SiO_2 content between the Langkawi granite (68.1 -77%) and the Main Range granite (67.28-77.46) clearly indicates that the Langkawi granites has higher evolution of magma than that of Main Range granites (Ghani, 1999). Selected Harker diagram for both major element oxides of the Main Range and the Langkawi granites is given in the Figure. 4.20, which shows Al_2O_3, CaO, P_2O_5, FeO^t, MgO and TiO_2 decrease whereas Na_2O increases with increasing SiO_2.

The ACNK value ($Al_2O_3/CaO+Na_2O+K_2O$, Shand, 1943) for the Main Range granite range from 0.94 (midly metaluminous) to 1.18 (peraluminous) (Fig. 4.21). On the other hand, the Langkawi granite ranges from weak/moderate peraluminous (1.02-1.17) to strongly peraluminous (~1.36). The values increase with SiO_2. Out of 56 samples, 10 samples from Main Range fall below the ACNK=1 line and are metaluminous. The remaining 46 samples are peraluminous. On the other hand, all the samples from the Langkawi granite plot in the peraluminous field. Both the Langkawi and Main Range granite generally have high alkali content (Na_2O + K_2O) ranging between 7.12 – 8.51 wt % for the Langkawi granite and 7.33 – 8.88 wt% for the Main Range granite. This is clearly shown in the K_2O vs. SiO_2 diagram (Peccerillo and Taylor, 1976) (Fig. 4.22), 50% of the granites from Main Range fall into the field of high-K granite with high SiO_2 and other 50% is in the shoshonite field. Only 6 samples from Langkawi granite fall in shoshonite field and remaining samples are fallen in the field of high-K granite with high SiO_2.

Villaseca et al. (1998) established the diversity of crustally derived peraluminous series on a A-B plot of Debon and Lefort (1983) (Fig. 4.23). They have divided the peraluminous granitoids into four types namely high-peraluminous, moderate-peraluminous, weak-peraluminous and felsic-peraluminous. The B-A plot shows that Langkawi granites plot within the weak-peraluminous field and felsic peraluminous field. On the other hand, the Main Range granite lies towards the metaluminous field.

The Na_2O content of Langkawi granite is ranging from 2.68 – 4.04%, and for Main Range granite is (2.24 – 3.97). The Langkawi granite contain slightly higher Na_2O content compared to the Main Range granite at the same K_2O concentration. This is evident from the Na_2O vs K_2O plot (Fig. 4.24), where only 12 out of 30 from Langkawi and 9 out of 56 samples from Main Range plot in the I –type field. All samples plot within the granite field (Fig. 4.25) of the R1-R2 classification diagram for volcanic and plutonic rocks after De La Roche et al. (1980), where R1= 4Si - 11(Na+K) – 2(Fe+Ti) and R2= 6Ca + 2Mg + Al, expressed in millications. Only 7 out of 56 samples from Main Range granite plot in the granodiorite field. Majority of the Langkawi granites are graded towards alkali granite.

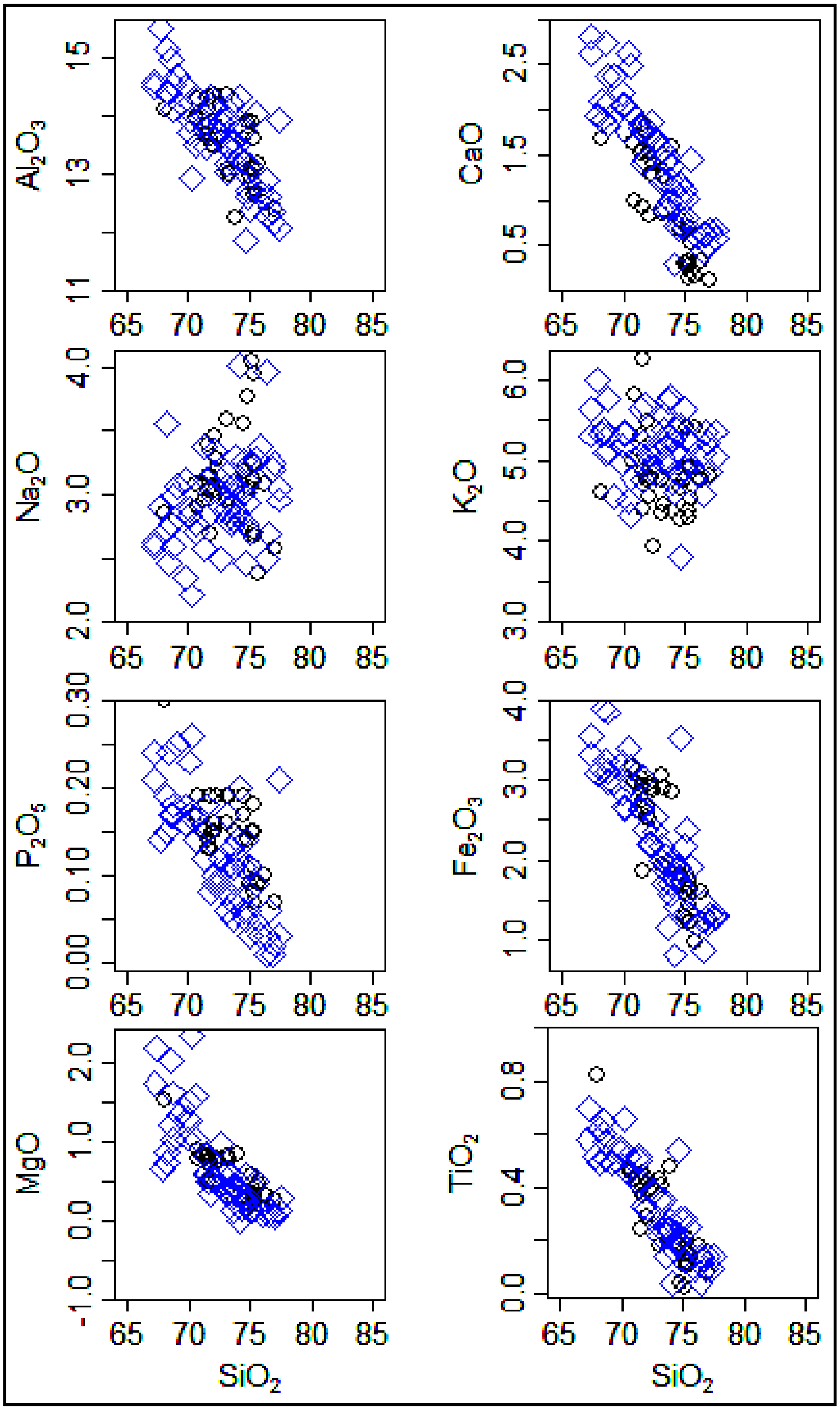


Fig. 4.20 : Major elements Harker diagram of the Langkawi and Main Range granite.
○ - Langkawi , ◇ - Main Range)

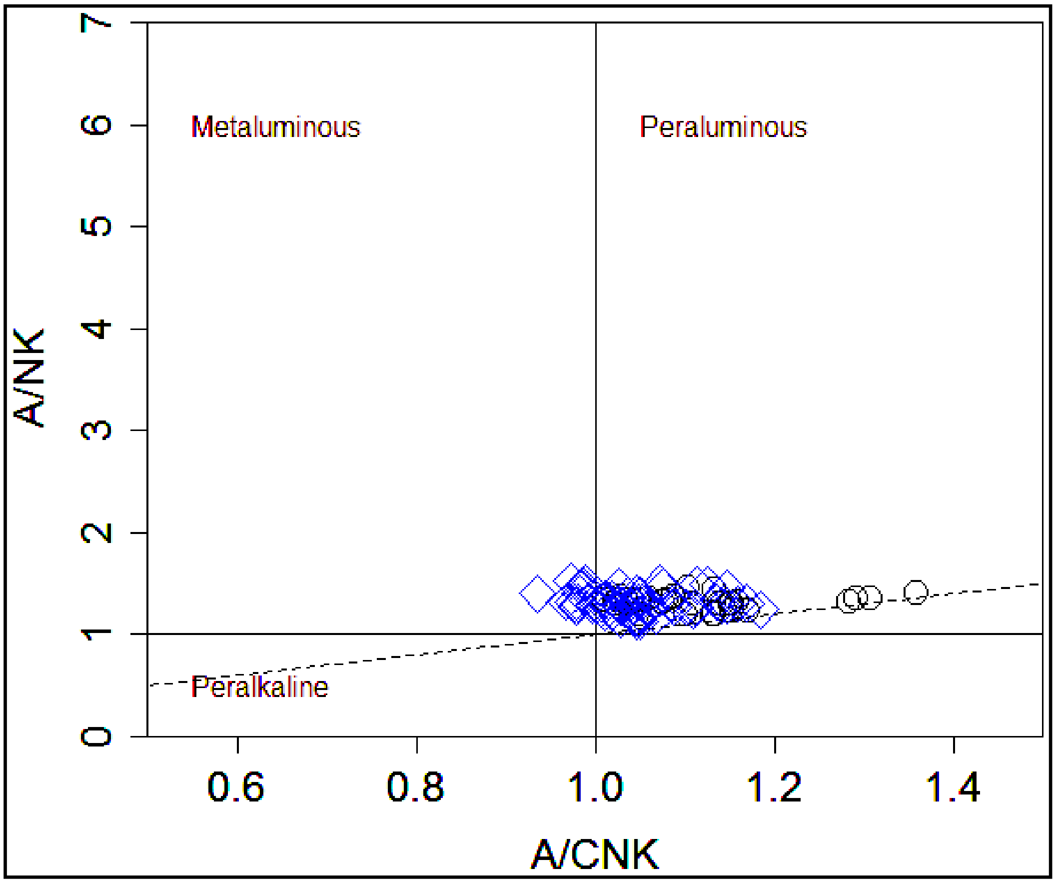


Fig. 4.21 : ACNK vs A/NK plot of Langkawi and Main Range granite.
(O - Langkawi , ◇ - Main Range)

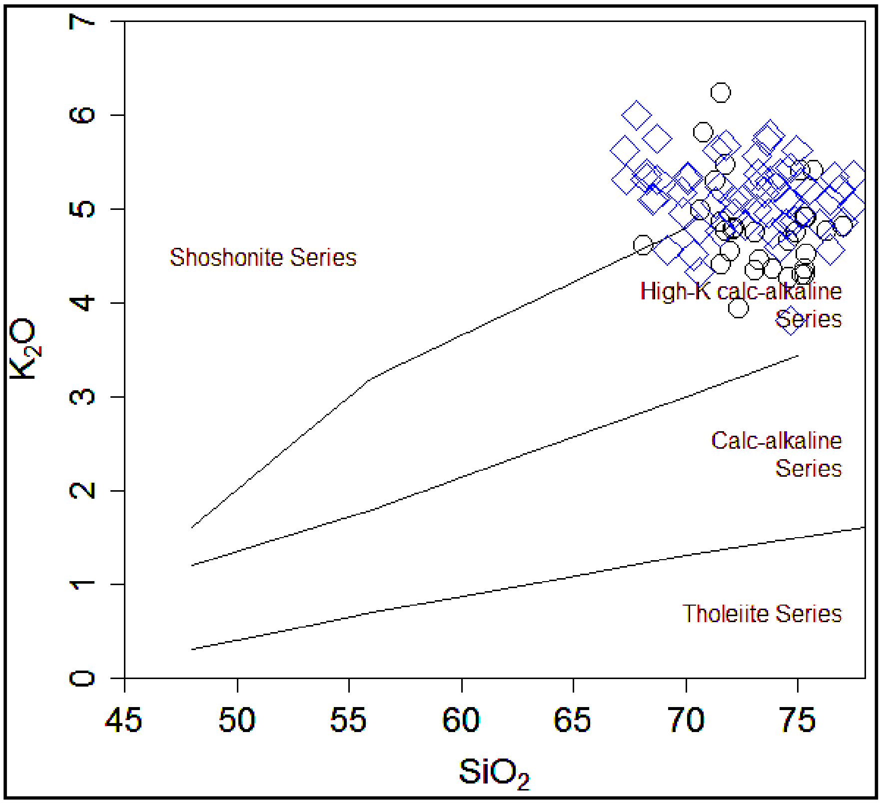


Fig. 4.22 : K_2O vs. SiO_2 diagram of the Langkawi and Main Range Granite. Compositional field after Peccerillo and Taylor (1976).
(○ - Langkawi , ◇- Main Range)

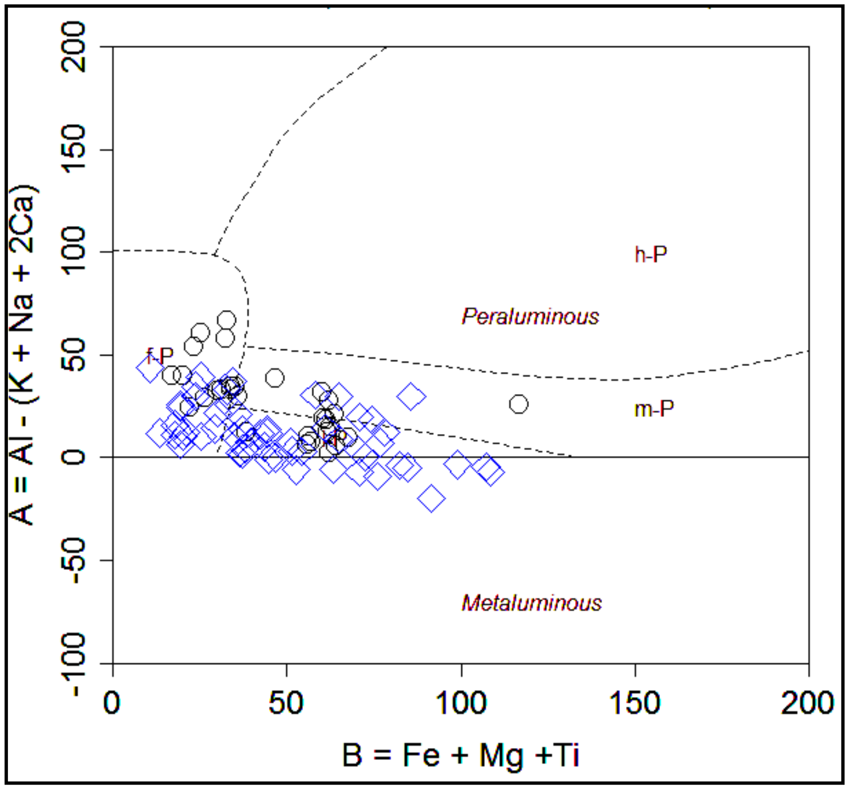


Fig. 4.23 : A - B plot of Langkawi and Main Range granite , after Villaseca et al. (1998).
(O - Langkawi , ◇ - Main Range)

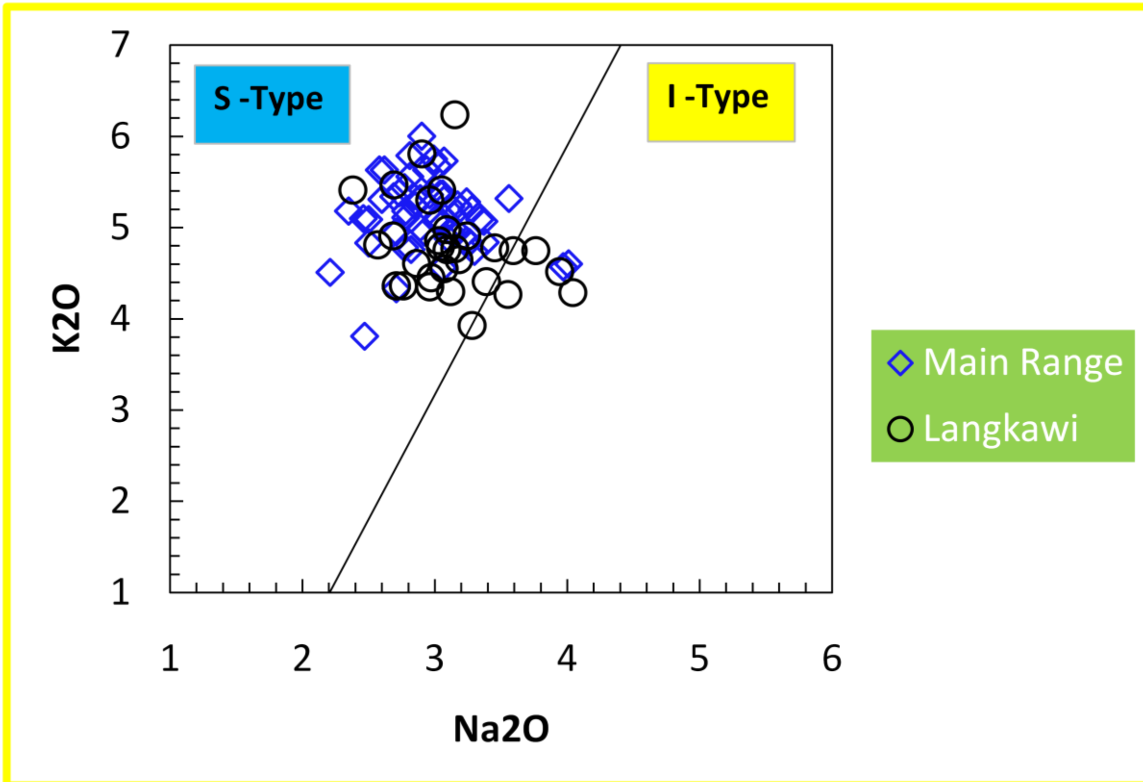


Fig. 4.24 : Na_2O vs K_2O plot of the Langkawi and Main Range granite. Field of the 'S' and 'I' type granite is after Chappell and White (1983). Note that in general the Langkawi granite contain slightly higher Na_2O content compared to the Main Range granite at the same K_2O concentration.

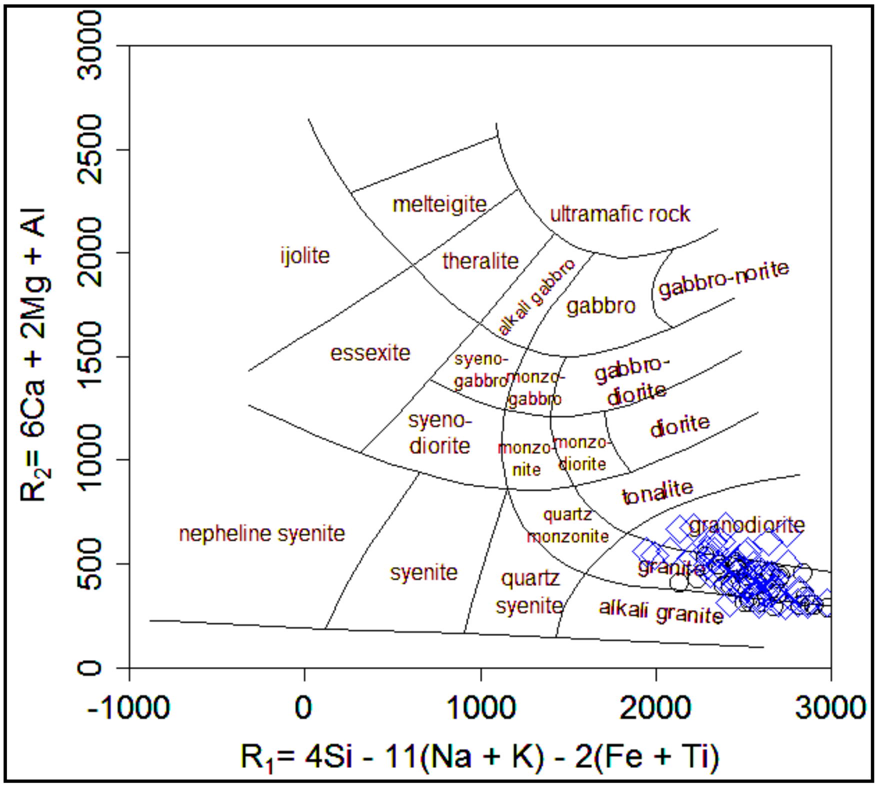


Fig. 4.25 : $R_1 – R_2$ plot of the Langkawi and Main Range granite, after De La Roche (1980). All samples plot as granite.
(O - Langkawi, ◇ - Main Range)

4.8.2 Trace Elements

In general most of the trace elements from Langkawi and Main Range granites shows decreasing trend (Fig. 4.26). Many of trace elements overlap, particularly Rb,Ce, Ba, and La. A clear decreasing trend of both Langkawi and Main Range granite trend is shown by Ba, Sr, Ce, Zr, and Zn with increasing SiO_2. On the other hand, Rb vs SiO_2 plot shows 'J' shape and Nb , La vs SiO_2 display slight scattered trend. Majority of the trace elements; Ba, Sr, Rb, Ce, Zr, Zn, Nb and La of Main Range granite have higher value than those of the Langkawi granite.

Rb/S ratio are higher in the Langkawi granite (average ~10.75%) compared to those of the Main Range granite (average ~ 7.63%). However there are some overlap between 70 to 77% SiO_2. The high Rb/Sr ratio of Langkawi granite reflects the highly evolved nature of the magma compared to the Main Range granite. Positive slope of Sr vs CaO supports that plagioclase is being removed in the differentiation sequence (Ghani, 2003)(Fig. 4.27).

Furthermore, one of the distinctive differences between the Langkawi and Main Range granite is in the La and Ce contents. The Main Range granite samples have high La content at a given Ce and both granites plot a parallel trend to each other (Fig. 4.28). Notably all the granite from Langkawi and Main Range shows the same trend, which is probably related to fractionation of accessory phases ($Kd^{La}_{allanite}$=2594; $Kd^{Ce}_{allanite}$=2278), (Ghani, 2004).

4.8.3 LIL modeling

Large iron lithophile content of the Langkawi granite is more irregular compared to the Main Range granite. This is evidenced from Ba, Sr, and Rb vs SiO_2 plots (Fig. 4.29). Generally, the rocks from the Langkawi granite have low Rb, Sr and Ba (<1000ppm). However in detail Rb content in the Langkawi granite (<500ppm) is lower compared to the Main Range granite (<1000ppm). The importance of K-feldspar, plagioclase and biotite in the differentiation is consistent with large ion lithophile (LIL) modelling. Interelement LIL log variation diagram for pairs Rb-Sr, Ba-Sr and Ba-Rb are shown in Figure. 4.30. It is shown in the vector diagram representing the net change in composition of the liquid after 30% Rayleight fractionation by removing K-feldspar, hornblende, plagioclase or biotite. In all diagrams the trends are consistent with K-feldspar and plagioclase and biotite. Thus the LIL log-log plot suggests that crystal fractionation plays an important role in the magmatic evolution of the both Langkawi and Main Range magma. LIL modeling (Fig. 4.30) of the Langkawi and Main Range show that the magmatic evolution is controlled by K-feldspar, plagioclase and biotite.

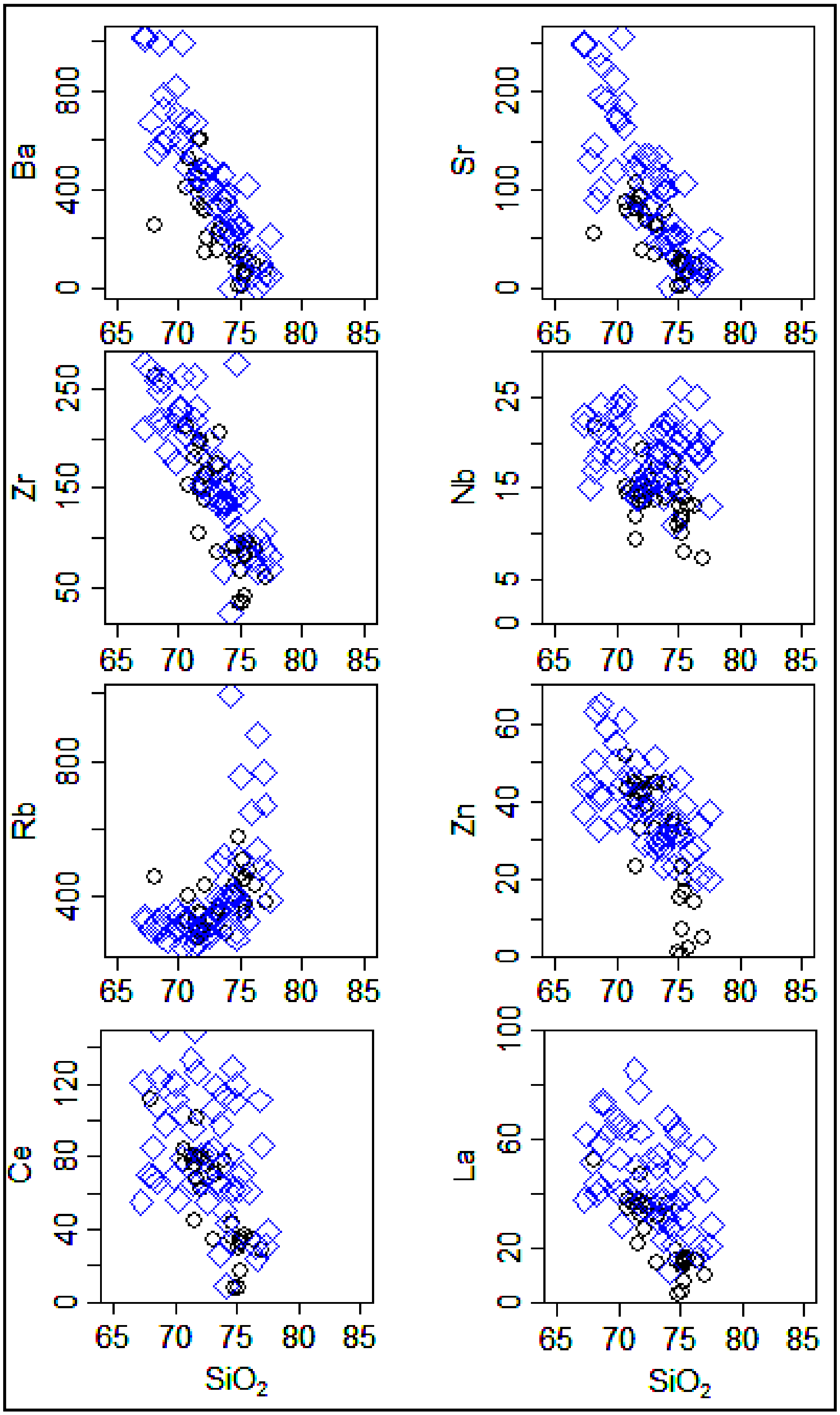


Fig. 4.26 : Trace elements Harker diagram of the Langkawi and Main Range granite.
(○ - Langkawi, ◇ - Main Range)

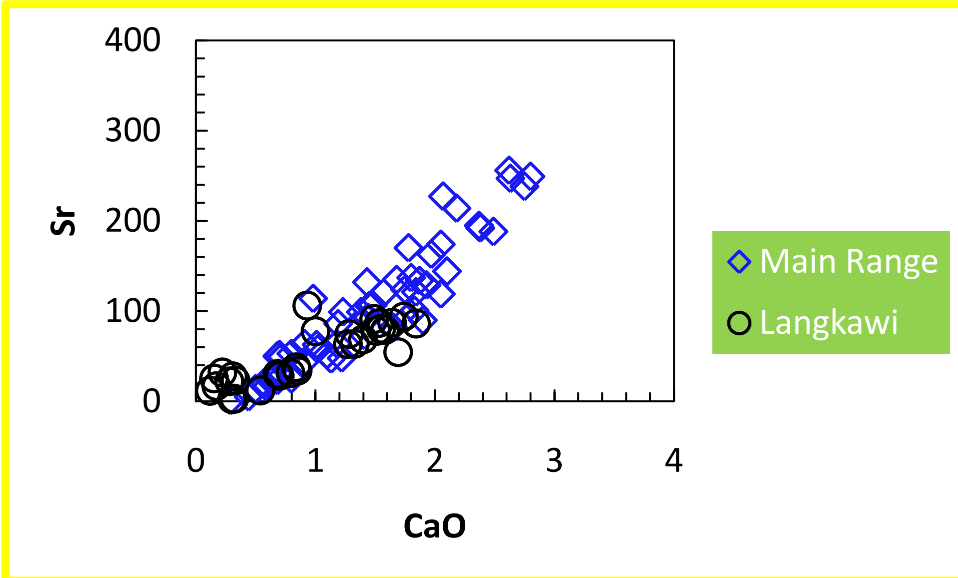


Fig. 4.27 : Sr vs CaO diagram for the Langkawi and Main Range granite.

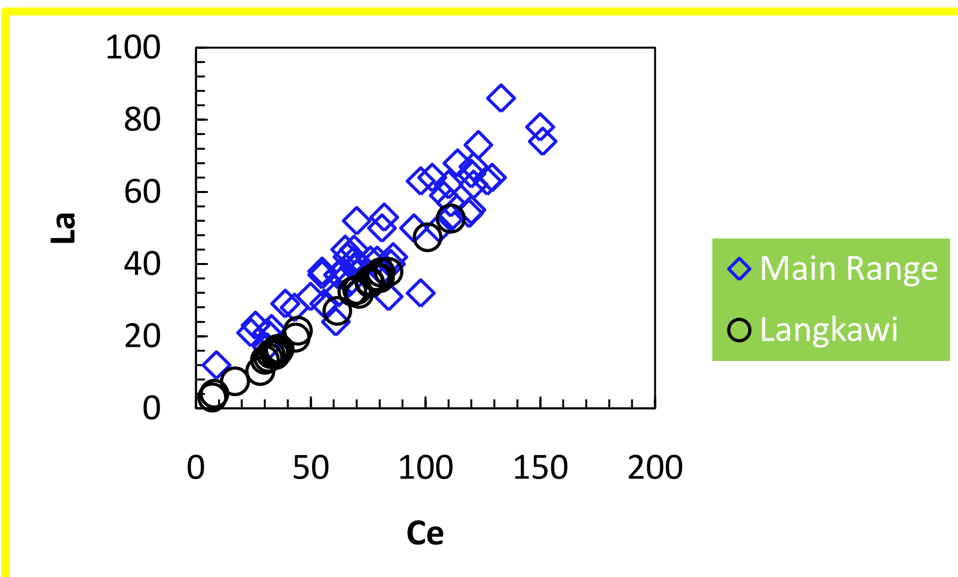
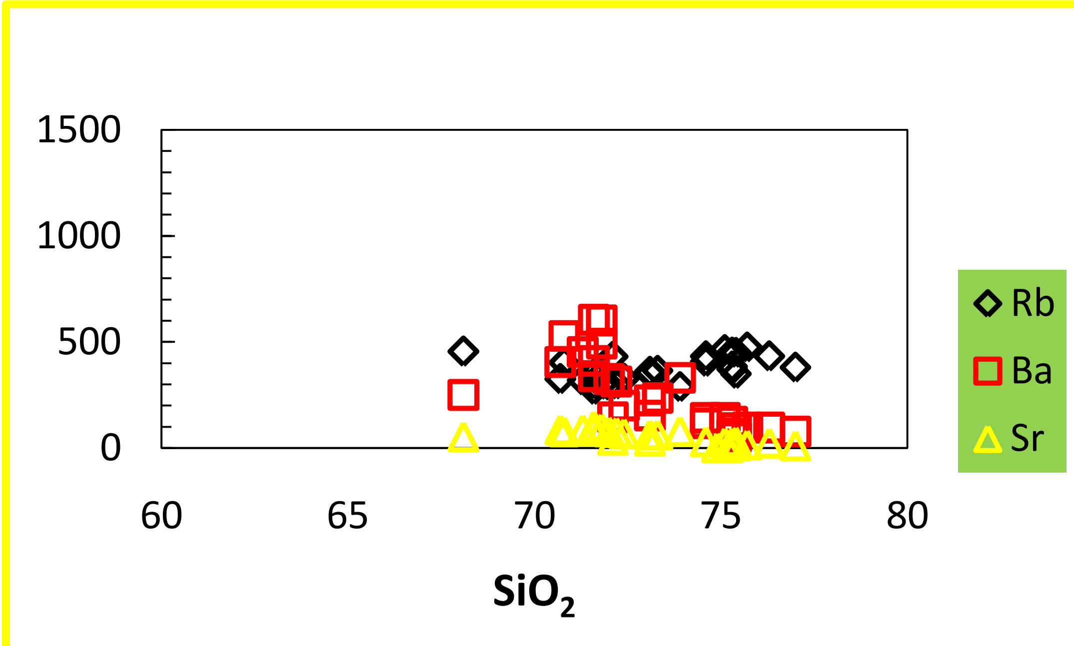


Fig. 4.28 : La vs Ce (in ppm) plot of the Langkawi and Main Range granite.



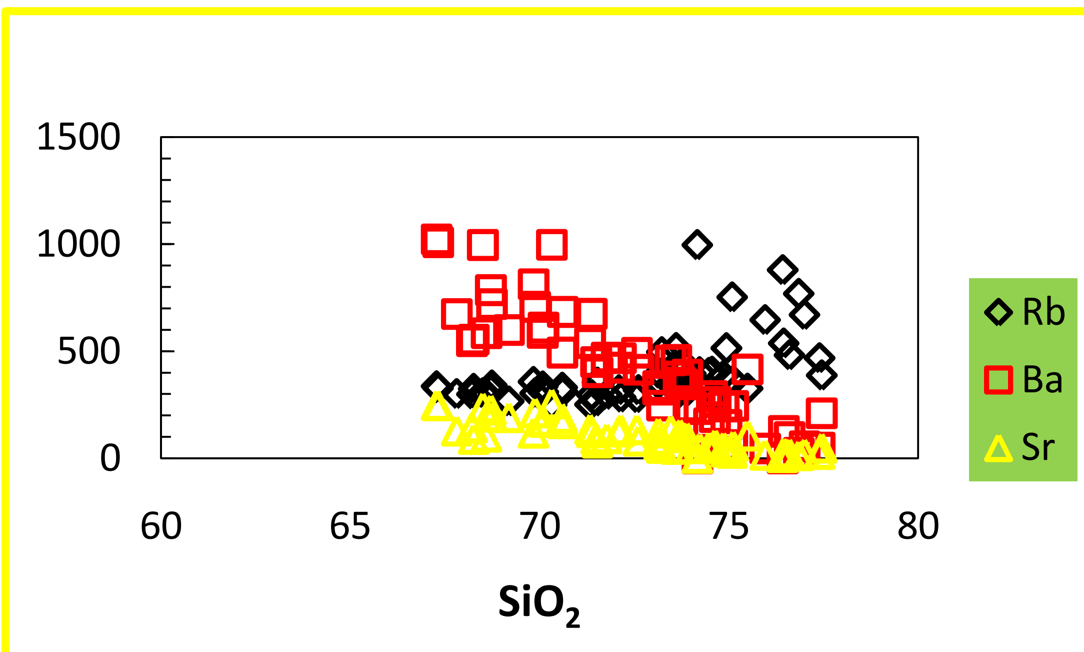


Fig. 4.29 : Large ion Lithophile elements (Rb, Sr, Ba) plot for the Langkawi and Main Range granite.

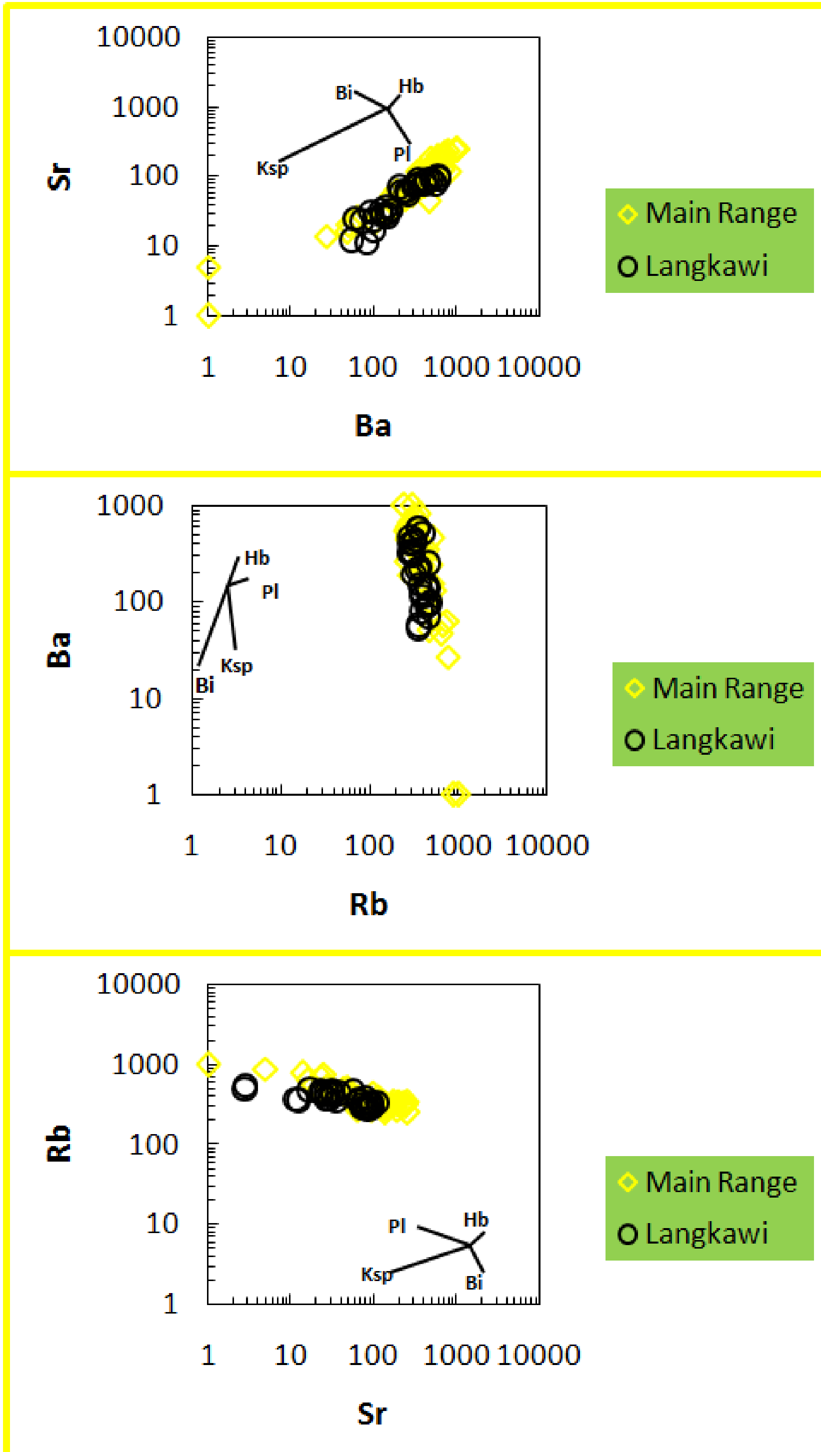


Fig. 4.30 : Large ion Lithophile elements modeling of the Langkawi and Main Range granite. Mineral vectors indicate paths of evolved liquid for 30 % of mineral precipitating. Ksp= K-feldspar; Pl=Plagioclase; Bi=Biotite and Hb=Hornblende

4.9 Zircon Saturation

The saturation temperature of the Langkawi and Main Range granites were calculated base on zircon saturation thermometry (Watson and Harrison, 1983)(Fig. 4.31). The plot shows that there is different zircon saturation. The Langkawi granite with lower average Zr contents (~127.3 ppm) yield saturation temperature of (677-830°C), which show the gradual decrease with evolution of the magma. The Main Range granite have higher average Zr contents (~ 164.9ppm) and yield saturation temperature of (654-830°C).

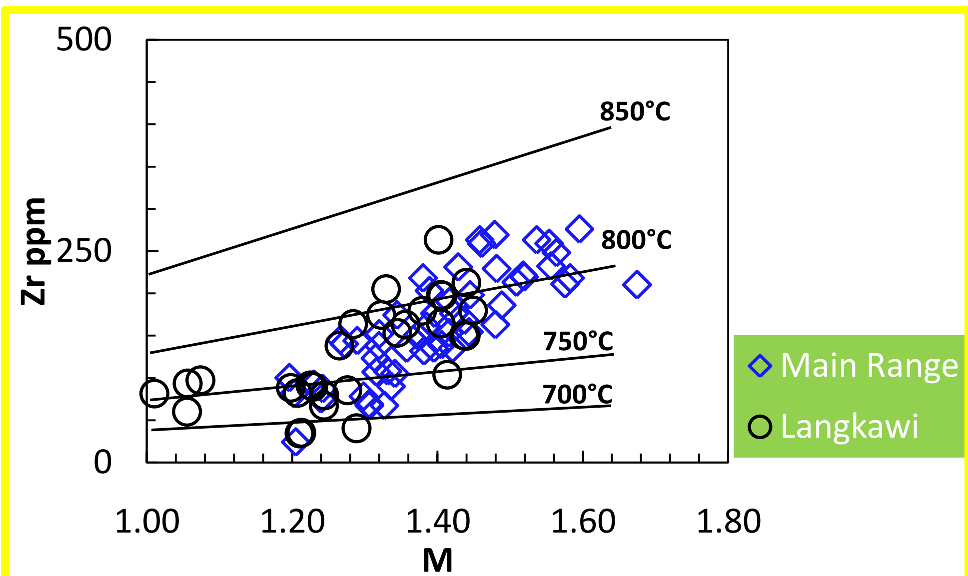


Fig. 4.31: Zr concentrations in the granites plotted against M, the cation ration (Na+K+2Ca)/(Al×Si). The line marked 700-850°C indicate the proportion of Zr that can be dissolved in granitoid melts of different compositions, expressed by the M parameter (Watson and Harrison, 1983).

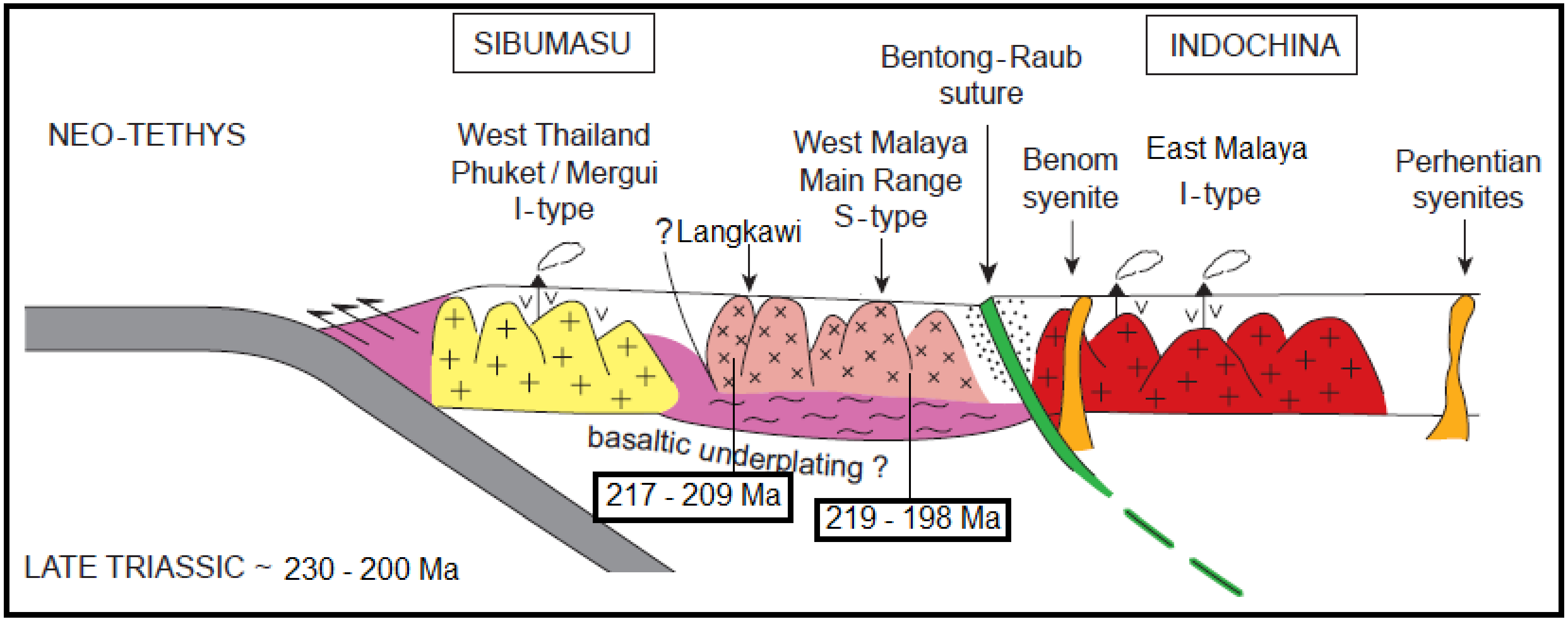


Fig. 4.32 : Plate tectonic evolution of Northern Malaysian Peninsula from Late Triassic 230-200 Ma (after M. Searle et al., 2012). Age dates from, (Bignell & Snelling, 1977; Liew and McCulloch, 1985; Searle et al., 2012; Oliver et al., 2013), Illustrates the late Triassic closure of Palaeo-Tethys along the Bentong–Raub Suture, and crustal thickening of the West Malaya province associated with intrusion of the Triassic S-type granites in the Main Range of Malay

CHAPTER 5

CONCLUSION AND DISCUSSION

5.1 Petrology

By the means of petrology and geochemistry it is possible to elucidate the geological evolution of the Langkawi granite. From the petrological studies of the Gunung Raya and Kuah granites, several conclusions can be made as follow:

- Occurrence of euhedral plagioclase and biotite in the both Gunung Raya Granite and Kuah Granite indicate that they are formed at the earliest phases to be crystallized.
- The microcline has inclusions of quartz and plagioclase which suggest the late growth of microcline and also shows that the microcline has continuing growth in the Gunung Raya granite.
- Secondary muscovite, chlorite and sericite are formed due to hydrothermal activity in the both Gunung Raya and Kuah granite.
- The mineral assemblage quartz+secondary muscovite in the both Gunung Raya and Kuah granites is instead suggested being a product from the breakdown of potassium feldspar and primary precipitation from peraluminous fluids.
- Exsolution of sodic and potash feldspars has resulted in the production of perthite in which the sodic plagioclase occurs as lenses or bands across the microcline. For such crystallization to have taken place there must have been initially a silicate melt which suggest a magmatic origin for Langkawi granite.

- Garnet present in only one specimen from Kuah granite. It occurs with tourmaline.
- The presence of garnet and muscovite in Kuah granite is also indicative of S-type affinity of rocks (Chappell & White, 2001).
- In the Kuah granite, tourmalinization is the destruction of biotite and its replacement by tourmaline. The ferromagnesium components in the tourmaline are derived from the destroyed biotite.

5.2 Geochemistry

Higher SiO_2 content of the Kuah granite have more evolved nature of the magma compared to the Gunung Raya granite, as confirmed by Rb/Sr ratio. Similarly, the Main Range granite also shows the highly evolved rocks with SiO_2 content. Both the Gunung Raya and Kuah granites were occurred due to fractional crystallization during the magmatic evolution. Other than that the Sr depletion and Eu negative anomaly indicate the continuous plagioclase fractionation during the differentiation. In addition to that the Sr vs CaO plot shows positive slope, which is supporting that, the plagioclase is being removed in the differentiation sequence. The negative correlation of Al_2O_3, CaO, Ba and Sr with SiO_2 in the both Langkawi granites suggests that the plagioclase would be the main phase of crystal fractionation. The high content of Al_2O_3 and CaO in the Gunung Raya granite may be related to the high modal content of calcic plagioclase. On the basis of large iron lithophile diagram, both the Gunung Raya and Kuah units show the same minerals vectors trend. This indicates that the K-feldspar, biotite and plagioclase are being removed in differentiation sequence. In comparison, magmatic evolution of the Main Range granite was also controlled by K-feldspar, plagioclase and biotite, those are the important mineral assemblage in the magmatic evolution.

Most of the major oxides and trace elements of the Langkawi granites show a linear, smooth and coherent variation trends, which suggest that the main petrogenetic process is likely related to partial melting or crystal fractionation. Furthermore, both Gunung Raya and Kuah units display high peraluminousity (ASI>1.1%) coupled with high normative corundum (>1%). This could also indicate the S -type affinity of both granites (Chapple & White, 1984). Most granites from Gunung Raya and Kuah fall into the field of high-K granite with high SiO_2 but some samples from the Gunung Raya unit have been in the shoshonite field, which indicate that the Langkawi granites have higher K_2O content compared to the original 'S' type granite (Chappell & White, 1999).

Two different trends of P_2O_5 vs SiO_2 diagram indicate that the Gunung Raya and Kuah magma were formed from different magmatic pulses. In addition to that the trace elements Ba, Sr, Nb, Ce, La, Zr, Zn, Nd, V, Co and Th in both the Gunung Raya and Kuah granites plot in two trends respectively. This may also assume that these granites consist of several separate granitic pulses.

In the zircon thermometry of Langkawi granite implies that the Gunung Raya granite has yielded zircon saturation temperature of average ~765 °C and the Kuah granite, ~748°C, which is comparable to the typical estimate of metasedimentary melting temperature of S –type melts (Wyllie, 1977; Clemens & Wall, 1981).

On the other hand, both the Gunung Raya and Kuah units are characterized by the high-K calc-alkaline affinity, Ba,Sr and Nb negative trends and enrichment of Rb, K and La which are compatible to those of typical crustal melt (Chappell & White, 1992). This is supported by the negative trend of Nb, Sr and Ti are typical crustal compositions (Rollinson, 1993).

In the tectonic discrimination diagram of Rb vs Y+Nb, Nb vs Y and Rb vs Ta+Yb, both the Gunung Raya and Kuah samples from Langkawi Island plot in syn-COLG (Syn-Collisional Granite, Pearce et al., 1984). K/Ar and Rb/Sr dates 217 ± 8 Ma and 209 ± 6 (Bignell & Snelling, 1977), suggesting that the Langkawi granite can be related to the late Triassic continent collision event.

5.3 Comparison with Main Range granite

SiO_2 content (68.1 -77%) of Langkawi granite is comparable to the Main Range granite (67.28-77.46) and it is indicative of the highly evolved nature of the Langkawi granite (Ghani, 1999). This evidence is also supported by high Rb/Sr ratio of the Langkawi granite. The Langkawi granite contain slightly higher Na_2O content compared to the Main Range granite at the same K_2O concentration. This is evidenced from the Na_2O vs K_2O plot (Fig.4.24), where only 12 out of 30 from Langkawi and 9 out of 56 samples from Main Range plot in the I –type field. The Main Range granite consists of mixed “I” and “S” type features and thus the Main Range granites cannot be designated as exclusively “S” type (Ghani, 2000).

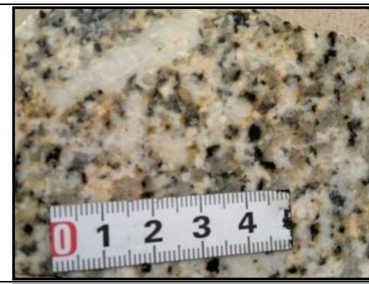
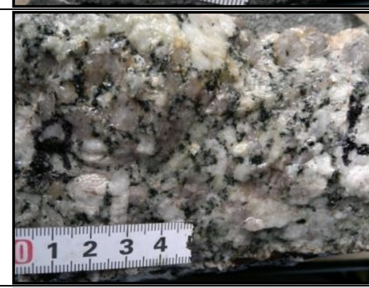
According to the LIL modelling, crystal fractionation plays an important role in the magmatic evolution of both the Langkawi and Main Range magma. K-feldspar, biotite and plagioclase are being removed in differentiation sequence for the both Langkawi and Main Range granite during magmatic evolution.

5.4 Significance of differences between the Langkawi and Main Range granites

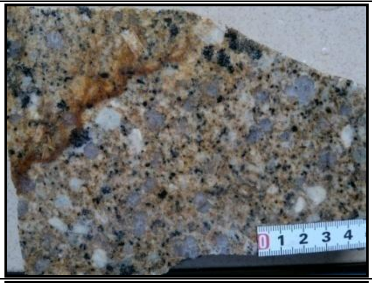
The Langkawi granite is characterized by high SiO_2 content (68.1 – 77 wt %) according to Chappel & White (1984) is characteristic of rocks which derived from SiO_2-rich sources. Among the both Gunung Raya and Kuah granites, the Kuah granite (75.1 – 77 wt % SiO_2) have more silica and evolved magma than that of the Gunung Raya granite (68.1 – 75.3 wt % SiO_2). The Kuah granites are more differentiated, indicated by the higher Rb/Sr. In general both Langkawi granites have higher Rb/Sr ratio (average ~10.75 %) compared to that of the Main Range granite (average ~7.63%). This reflects that the Langkawi granite magma is highly evolved compared to the Main Range granite magma. Harris et al. (1993) found that high Rb/Sr ratios (4-10) is suggestive of granite formation at vapour-absent condition (dehydration melting), whereas Rb/Sr ratios <3.5 suggest vapour-present melting. Accepting the validity of their modeling, it would appear that the most of the Langkawi granite (Rb/Sr ratio average ~10.75 %) were formed by vapour-absent melting with fractionation of plagioclase.

In the Peninsular Malaysia, strongly differentiated granites, characterized by their Rb/Sr ratios are mineralized (Yeap, 1974). In Langkawi the strongly differentiated Kuah granite is also mineralized. Although tin is not found, copper-bismuth mineralization is recorded from the skarn-granite contact near Kuah. Geochemical studies suggest that the Gunung Raya and Kuah magmas derived from the different magmatic pulses but they were controlled by the same mineral assemblages. On the other hand, a comparison of LIL modeling between the Langkawi and Main Range magma are controlled by same mineral assemblages during crystallization, suggesting that both Langkawi and Main Range magmas may also derived from the same magma sources. The Langkawi granites have relatively low sodium, Na_2O less than 3.2 % in rocks with approximately 5 wt % K_2O, decreasing to less than 2.2 wt % in rocks with approximately 2 % K_2O and Mol $Al_2O_3/(Na_2O+K_2O+CaO)$ ratio of greater than 1.1 with normative corundum greater than 1 % CIPW corundum. Thus it can be assumed that the Langkawi granite is exclusively S-type granite i.e. derived by partial melting of metasediments. The tectonic discrimination diagram of Pearce et al., (1984) clearly indicates the plot of the Langkawi and Main Range granites falling within the syn-collisional granites. This supports their S-type characteristics and in association with the tin mineralization and in relation to the Indosinian Orogeny.

Appendix (1)

Location	Sample		Description
N 06° 21 ,636' E099° 41 , 780' Pantai Kok (Msirat)	L-1		- very coarse to medium grained muscovite biotite granite - porphyritic texture with K-Feldspar phenocrysts reaching 6cm in length, can be seen as subhedral in shape.
N 06° 33, 972' E 099° 40, 444' Seven well water fall	L- 5		- very coarse to medium grained muscovite biotite granite - clearly seen the comparism to metasediments
N 06° 33, 972' E 099° 40, 444' Seven well water fall	L-6		- very coarse to medium grained muscovite biotite granite - porphyritic texture with K-feldspar phenocrysts
N 06° 25, 126' E 099° 46, 804' Teluk ewa	L- 11		- very coarse to medium grained muscovite biotite granite - porphyritic texture with K-Feldspar phenocrysts reaching 6cm in length, can be seen as subhedral in shape

Appendix (2)

Location	Sample		Description
N 06° 17, 926' E 099° 51, 129' Kuah jetty	L- 13		- medium to fine grained tourmaline Microgranite - The segregations of tourmaline are coarse grained and commonly broken up by the quartz into fragment
N 06° 17, 926' E 099° 51, 129' Kuah Jetty	L- 14		- tourmalinisation is commonly manifested as segregated round clots of one or two inches in diameter
N 06° 24, 298' E 099°48 ,140' Quarry Gunung Raya	L- 21		- very coarse to medium grained muscovite biotite granite - porphyritic texture with K-Feldspar phenocrysts reaching 6cm in length, can be seen as subhedral in shape

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