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Mineralogical and Petrographic characterization of the Paleozoic shale in the Kroh Formation, North Perak, Peninsular Malaysia

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Abstract

The increasing global demand for clean energy has made it imperative to explore and exploit unconventional oil and gas resources. Twenty-five percent of the sedimentary rocks in Peninsular Malaysia are Paleozoic shales. Nevertheless, no work has been carried out on shales in the Kroh Formation as regards their mineralogical composition and the effect of minerals in the hydraulic fracturing. Representative samples of black shales from the Kroh Formation were analyzed using X-ray diffraction (XRD) and Field Emission Scanning Electron Microscope (FE-SEM) with energy dispersive X-ray (EDX). The mineralogical composition of the shales has an important effect on the competency of rocks, for drilling and for hydrocarbon production. The FESEM results showed that the shale samples mainly comprised kaolinite which appears as booklets and quartz as round grain with small oval depressions of vary grain sizes. The elemental compositions of the shale samples determined by EDX confirm the minerals identified by FESEM. XRD and FESEM results both identified the minerals as illite, chlorite, pyrite, and a minor amount of dolomite, feldspar, and calcite. Knowing the clay minerals composition of the shale can be useful in planning for drilling and hydraulic fracturing.

Keywords: *Minerology; Petrographic; Paleozoic shale; hydraulic fracturing; morphology.*

1. Introduction

Shale is a fine grain sedimentary rock, containing different types of clay minerals such as illite and kaolinite and non- clay minerals such as quartz and pyrite [1]. Shales are structurally fissile, which give them the tendency to split along relatively smooth surfaces parallel to the bedding plane. Black shale is a dark-colored mud rock containing organic matter, silt- and clay-sized mineral grains that accumulate together [2]. A shale gas reservoir comprises not of a single lithology but a collection of fine-grained rocks capable of storing significant amounts of gas. Most of the black shales are marine and may have a real extent of thousands of square kilometers. Unconventional hydrocarbon resource has become an interesting field of study and especially for it as source rocks which can also turn to be excellent reservoirs for generation and accumulation [3].

Exploration and production of gas from fine-grained rocks such as shale, contemporaneous with technology advancement facilitated exploration of black shale and made them cost benefit. During the last few years, a major concern has been given to unconventional oil and gas shales. Black shales have attracted interest from researchers primarily because of their economic importance in terms of hydrocarbon development potential [4]. Mineral composition and texture can be critical properties that influence the potential of shales, particularly for drilling and production [5]. The mineral contents of the rocks in an unconventional reservoir are important when the operators want to perform hydraulic fracture operations successfully. The presence of expandable clays like smectite causes problems during drilling due to swelling of the clays. Fractures are more common and created more easily in carbonate-rich and silica-rich than in clay-rich shales. Mineralogy alone maybe the first deciding factor when assessing the economic exploitation potential of a shale reservoir [6].

Generally, rocks with low clay and high quartz content have low Poisson's Ratio and high Young's Modulus, thus making them more brittle and more prone to natural fractures and are good for fracking [7]. Many Paleozoic black shales are rich in quartz and can be regarded as siliceous shales or marlstones. Shale can vary considerably in kerogen type, thermal maturity, and mineralogy [8-9]. In spite of the fact that Paleozoic black shales are old and their organic matter and degree of maturation might have been changed by diagenesis/metamorphism, these shales attract attention as potential unconventional oil and gas resources [10-12].

Twenty five percent of sedimentary rocks from Peninsular Malaysia are Paleozoic shales [13]. These black shale formations are exposed in areas such as Langkawi, Kedah, Perlis, North Perak, and many other areas in Malaysia. These areas have been mapped by the Minerals and Geoscience Department, and the lithology and stratigraphy have been studied extensively [14-19]. However, the mineralogy and petrography of these shales have not been studied in detail. Therefore, this study is designed to characterize the mineral composition and petrographic texture of the shale in order to get an idea about the hydraulic fracturing potential based on the minerals that present in the shale.

2. Geological setting

Paleozoic rocks are mainly marine and account for about 25% by area of Peninsular Malaysia. The Paleozoic formations of Peninsular Malaysia are distributed in three northwesterly to northerly trending zones parallel to the general elongation trend of the Peninsula [13]. They are the Western Belt, Central Belt and Eastern Belts (Figure 1). Each of these Belts is characterized by distinctive tectonic, stratigraphy structure and sedimentary history.

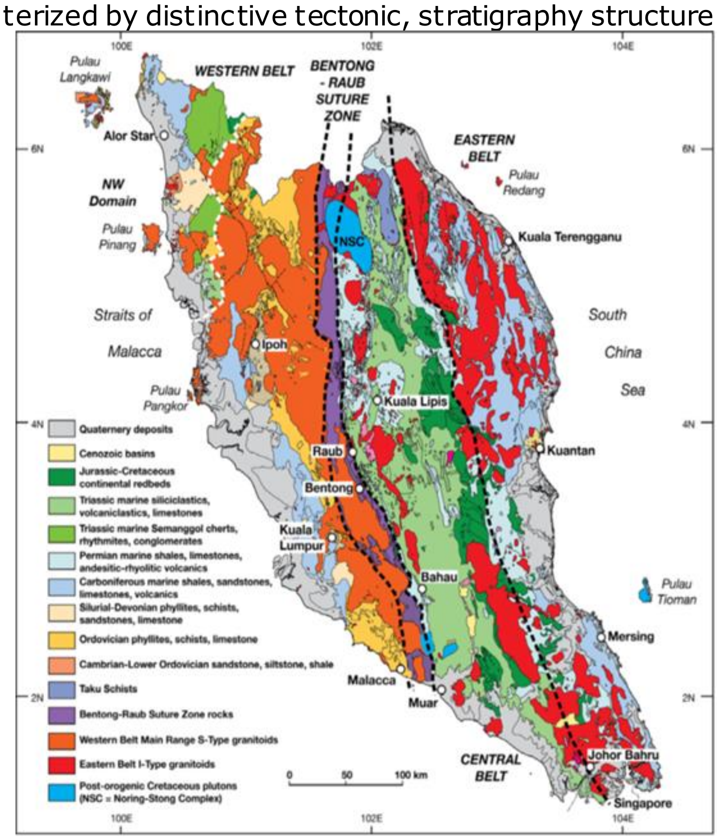

Fig 1. Simplified geological map of the Malay Peninsula showing the study area (*after Tate et al.* [30])

The Western Belt forms a portion of the Sibumasu Terrane, derived from the NW Australian Gondwana margin within the late Cambrian-Early Permian. The Central and Eastern belts represent the Sukhothai Arc constructed within the Late Carboniferous-Early Permian on the margin of the Indochina Block (derived from the Gondwana margin in the Early Devonian) [18].

Paleozoic rocks of the Western Belt are distributed at foothills along both flanks of the Main Range granite batholith stretching from the Malaysian–Thai border southwards to Malacca. The Northwestern Zone of the Western Belt covers Langkawi, Kedah, and Perlis and these are mainly shallow-marine shelf sediments [17]. Lower Paleozoic rocks are confined to the Western Belt, while the Upper Paleozoic rocks occur in the Central and Eastern Belts.

The Kroh Formation is part of the Baling Group. It is characterized by the Early – Middle Silurian and Lower Devonian graptolites, and the Early – Middle Devonian Tentaculites. The Mahang Formation in Kedah is equivalent to the Kroh Formation in Upper Perak. The Kroh Formation is overlain by the Upper Paleozoic rocks. The Upper Paleozoic rocks were deemed as an extension of the Kati Formation [20]. They are extensively exposed in the Pengkalan Hulu, Kelian Intan and Kerunai areas in northern Perak. The rock succession extends eastwards into the Bersia area as mentioned by [21] (Figure 2). Generally, the Kroh Formation is composed of black shale, sub-mature arenite, calcareous shale, and limestone.

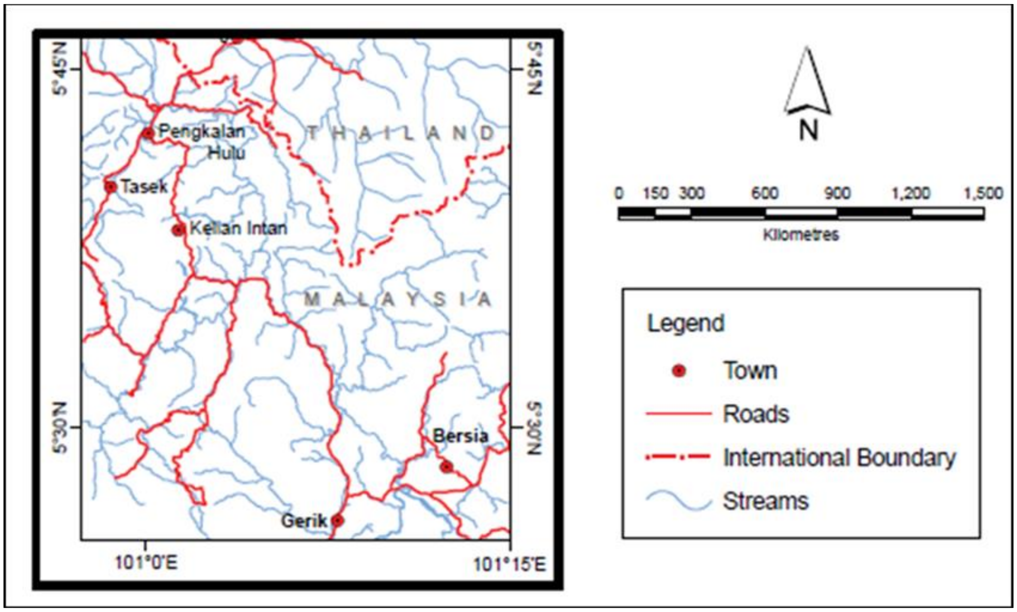

Fig 2. Location map of the study area (*after* [20])

Table 1. Schematic stratigraphic column of the Betong-Pengkalan Hulu Transect area, Malaysia showing the Kroh Formation [19]

PERIOD/EPOCH		FORMATION/UNIT	STRATIGRAPHIC COLUMN	DESCRIPTION
QUATERNERY	HOLOCENE	Alluvium (Qa)		Surficial deposits - unconsolidated gravel, sand, silt and clay of fluviatile and colluvial origin.
	PLEISTOCENE	Nenering beds (Qnn)		Semi-consolidated deposits consist of gravelly, sandy and silty beds, chanelling is common.
TERTIARY				Unconformity
CRETACEOUS		Berapit formation (Kbr)		Conglomerate, grey to reddish, very poorly sorted, subangular to rounded clast of up to 40cm in diameter.
JURASSIC				Unconformity
TRIASSIC				
PERMIAN		Gerik formation (Pgk)		Mainly tuffs of rhyolitic and rhyodacitic composition, occasionally metamorphosed; limestone and calcareous shale lenses occur sporadically; interbeds of tuff, limestone and calcareous shale also occur.
CARBONIFEROUS		Kubang Pasu (Ckp)		Well-bedded areno-argillaceous rocks with subordinate ribbon chert. The rocks had been invariably metamorphosed especially the one closer to the granite body. Ribbon chert
DEVONIAN		Kroh formation (SDkr)		d Argillaceous facies: Mainly shale and phyllite; commonly carbonaceous, with subordinate chert. c Calc-silicate facies: Grey calc-silicate hornfels. b Calcareous facies: Grey to dark grey impure limestone. a Arenaceous facies: Mainly metasandstone, occur sporadically within the argillaceous facies. (It is difficult to establish the order of the rock succession owing to folding, faulting, and the lenticular shape of the rock units as well as a repetitious character of its components)
SILURIAN				

The rocks are divisible into four facies: the argillaceous facies, calc-silicate facies, calcareous facies, and minor arenaceous facies. The widely spread argillaceous facies with a considerable amount of carbonaceous content suggests that the deposition of this rock unit occurred in a euxinic marine environment. Fine-grained materials indicate long distance transportation and the deposition took place in a quiet and undisturbed environment. The absence of benthos fossils and bioturbation indicates that the deposition occurred in a deep marine environment. The calcareous facies and arenaceous facies with subordinate conglomerate might be deposited in the continental shelf, the relatively shallower part of the depositional basin [19].

3. Material and methods

Twelve representative samples were taken from six locations in the Gerik area, Upper Perak (Figure 2). They are carbonaceous shale and have been dated and assigned an Upper Ordovician -Lower Devonian age [22]. All the shale samples were subjected to X-ray diffraction (XRD) and Field emission scanning electron microscope (FESEM) studies. XRD analysis is a useful method to identify minerals that are present in the shales. Shale samples were ground using a milling machine into powder, and the powder was analyzed using a Bruker D8 Discover X-ray Diffractometer according to the Hardy and Tucker method [23]. Field emission scanning electron microscopy (FE-SEM) is a known tool for investigating and imaging the microstructure of rocks [24-27]. The method depends on the interaction of the electrons emitted by the FESEM with the atoms that make up the sample producing signals that contain information about the sample surface topography, composition, and properties such as electrical conductivity. Sample preparation includes polishing the sample into thin blocks and then coating with gold. Energy-dispersive spectrometry(EDX) technique was used to determine the elemental composition of the sample.

4. Results and discussion

4.1. X- Ray diffraction (XRD)

XRD was used to identify the clay minerals and other minerals present in the shale samples. The results of all shale samples from the Kroh Formation have a mineralogical composition which typically consists of kaolinite, illite, and non- clay minerals such as quartz. A large quantity of illite usually indicates older rocks. All of these minerals were identified by the fact that each mineral has unique fingerprints.

Kaolinite is a clay mineral, with the chemical composition ($Al_2Si_2O_5(OH)_4$). It is a layered silicate mineral, with one tetrahedral sheet of silica (SiO_4) linked through oxygen atoms [28], which has diffraction at 7.3Aº basal spacing [23]. Figure 3 shows the XRD result for one sample. Kaolinite is present in variable quantities in all shale samples. The XRD peak suggests that the kaolinite is a clear crystal.

Quartz is very common in shales and is present in all the samples as shown in Figure 3. The quartz has diffractions at 4.2 Aº and 3.3 Aº. The intensities of XRD peaks suggest that quartz is dominant in all the samples. The quartz produced the highest peaks as compared to the other minerals. The height of the peaks produced by a certain mineral does not indicate its quantity. It just shows that the mineral has a good crystalline form [29].

The presence of quartz will be good for hydraulic fracturing because fractures form more readily and more widespread in carbonate-rich and silica-rich shales. Other minerals identified from the XRD analysis include calcite, alkaline feldspar, siderite, dolomite, smectite, muscovite, goethite and pyrite (Figure 3). Table 2 shows the mineralogical composition of 12 rock samples in the Kroh Formation which were analyzed by XRD.

The qualitative analyses of mineral employing a search/match program become increasingly challenging when the mineral is a mixture of several phases, rendering the diffraction pattern complex.

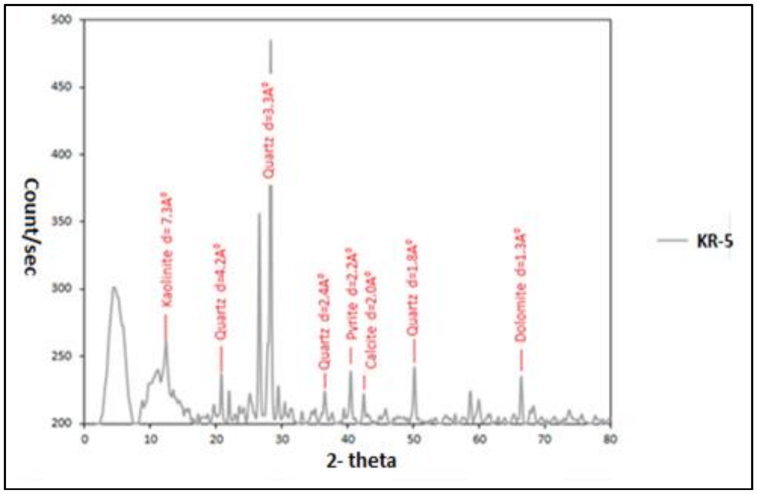

Fig 3. The XRD peak of shale from the Kroh Formation in the KR-3 samples

Table 2. Mineralogical composition of 12 rock samples from the Kroh Formation determined by XRD

Sample ID	Quartz	Kaolinite	Illite	Feldspar	Pyrite	Dolomite	Smectite
KR-1	✓	✓	✓		✓	✓	✓
KR-2	✓	✓					
KR-3	✓	✓		✓	✓		
KR-4	✓	✓			✓	✓	
KR-5	✓	✓	✓	✓			✓
KR-6	✓	✓					
KR-7	✓	✓	✓	✓			✓
KR-8	✓	✓		✓			✓
KR-9	✓	✓				✓	
KR-10	✓	✓					
KR-11	✓	✓		✓			
KR-12	✓	✓					✓

4.2. Field Emission Scanning Electron Microscope with energy dispersive X-ray (SEM-EDX)

Field Emission Scanning electron microscopes (FE-SEM) with energy dispersive X-ray (EDX) were employed to measure the surface topography, microstructure, and chemical composition of rock samples. The FESEM-EDX analysis was carried out on the twelve shale samples from the Kroh Formation. Figure 4 shows the images of shale samples at different magnification factors. The crystal structure can be clearly observed with increasing magnification.

Kaolinite appeared as pseudo hexagonal plates or books in the SEM result (Figure 5 A). The presence of kaolinite was also confirmed by EDX techniques. Results showed that the shale samples are dominated by silica, oxygen, and aluminum ($Al_2Si_2O_5(OH)_4$) and the weight concentration of Si and Al confirmed their identification as kaolinite (Figure 5B). In the SEM, the quartz appeared as rounded grain with small oval depressions and different grain sizes (Figure 6.A). The presence of quartz is also confirmed by EDX techniques. Results showed that the elemental composition of the shale samples is dominated by silica and oxygen (Si-O), and they have high peaks in the spectrum as shown in Figure 6.B. Addition, the weight concentration of these elements is high compared to the other elements present in the samples.

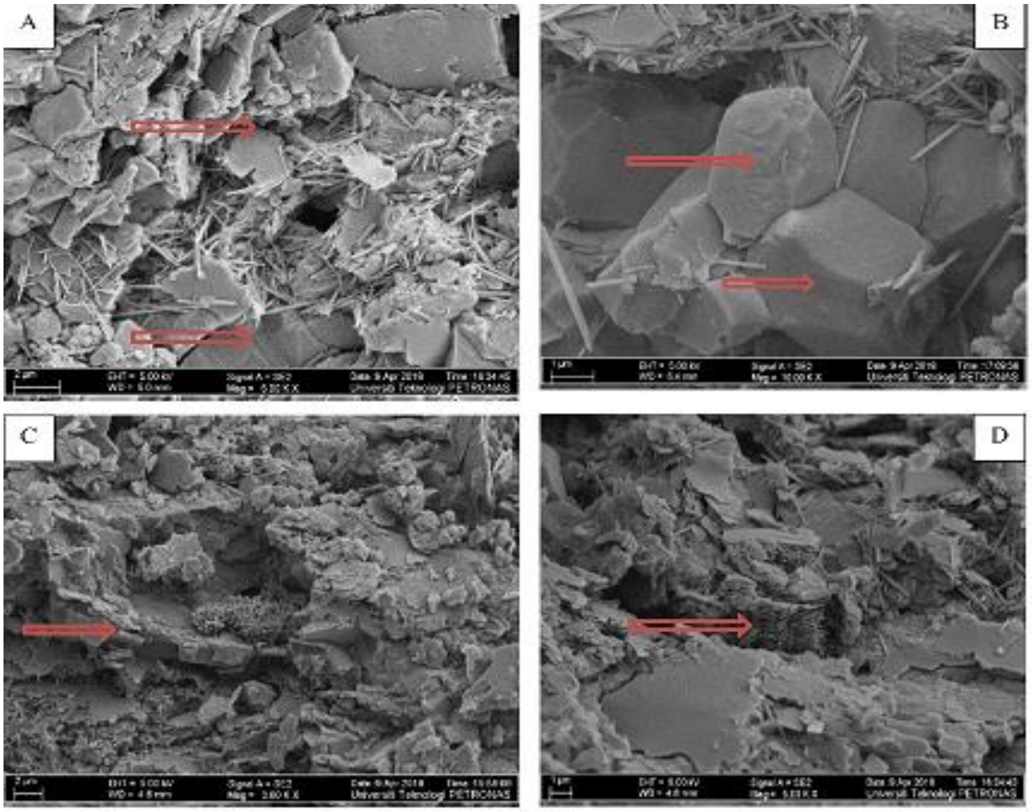

Fig 4. Images of shale samples from the Kroh Formation. (A) Quartz appeared as rounded grain with small oval depressions. (B) Quartz appeared as large grains. (C) Quartz appears as different grain size. (D) Kaolinite appeared as pseudo hexagonal plates or books

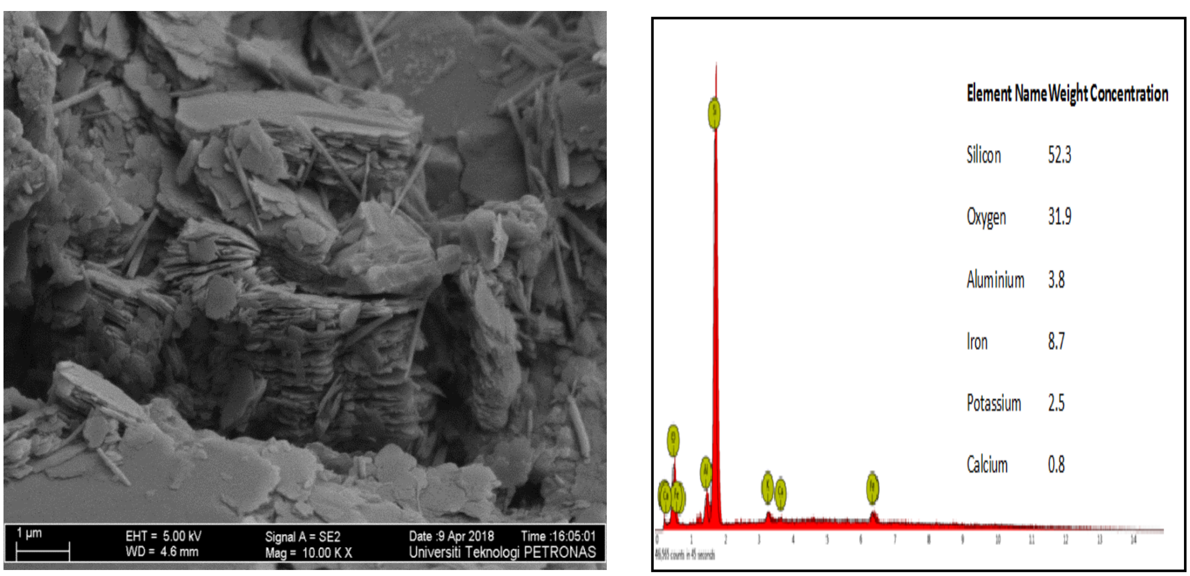

Fig 5. (A). Kaolinite appeared as pseudo hexagonal plates or books in sample KR-3. (B). EDX Spectrum Analysis of KR-3 for kaolinite show the elemental composition of the shale sample

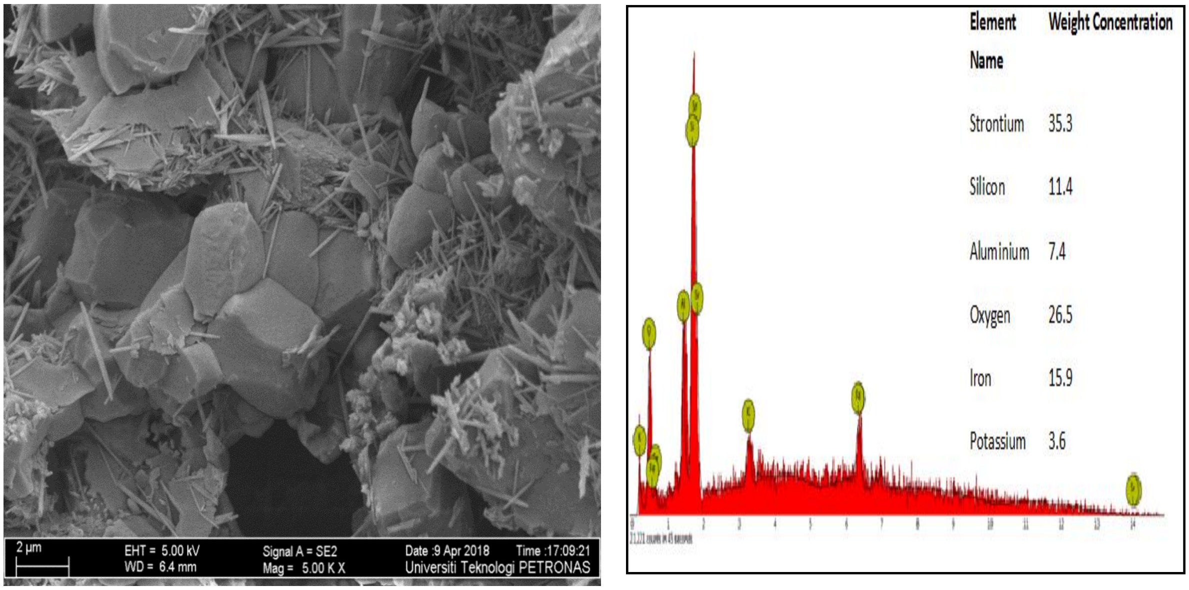

Fig 6. (A). SEM image of sample KR-5 shale sample shows the quartz appeared as large grains. (B). EDX Spectrum Analysis of sample KR-5 for quartz

5. Conclusion

An integrated mineralogical and petrographic investigation of the shale samples from the Kroh Formation in the Upper Perak was performed to determine the minerals and the morphology of the minerals present in the shale. Knowing the composition in the shales of the shale can be useful in planning for drilling and hydraulic fracturing.

FESEM were used to identify minerals to determine the chemical composition of the minerals present in the shale. Shale is the most abundant sedimentary rock and is a combination of a wide variety of minerals that clay minerals found in the samples tested are illite, kaolinite and the non- clay minerals are quartz and pyrite. The peaks of the quartz are highly diffracted because quartz has good crystalline form and shows larger peaks. On the other hand, clays do not diffract as well because they do not have good crystalline form and consequently, their peaks are lower than that of quartz.

The peak height of the quartz does not reflect the quantity. It just means that the quartz has a good crystalline form. It is difficult to get the quantitative mineral composition of the clays. The reason is that they do not have good crystalline forms and do not diffract well.

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