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Potential of Soil, Sludge and Sediment for Mineral Carbonation Process in Selinsing Gold Mine, Malaysia

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Abstract: Soil, sludge and sediment that are rich in alkaline earth silicates play significant roles as passive agents for removing carbon dioxide through mineral carbonation process. This study was conducted to characterize the mineralogical component and chemical composition of gold mining wastes and to identify the availability of natural silicate minerals as a feedstock for the mineral carbonation process. Particle-size distribution analysis was performed, and pH of the soil, sludge, and sediment were determined, whereas the mineralogical component and chemical composition of the samples were also analyzed. Results demonstrated that the presence of sepiolite and chlorite-serpentine in the stockpile and mine tailings can sequester carbon dioxide into magnesium carbonates, while the presence of stilpnomelane in the stockpile can be sequestered into iron carbonate. The presence of large amounts of small-size particles (silt fraction) in sludge (78.23%) at the mine tailings was identified to have higher surface area to absorb carbon dioxide. pH conditions of sludge (pH 7.9) and sediment (pH 8.3) from the mine tailings were favorable to enhance carbonate precipitation. Therefore, gold mine wastes have shown the potential for passive sequestration of carbon dioxide, thus, providing more insights into the enhancement of mineral carbonation process and the potential of natural silicate minerals.

Keywords: soil mineralogy; waste materials; divalent cation; mineral carbonation process; feedstock; gold mine

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1. Introduction

Carbon dioxide (CO_2) emission is expected to increase due to economic growth and global energy demand throughout the year. About 21% of global anthropogenic CO_2 emissions are derived from the mining industry [1], making it a significant sector for mitigation approaches in reducing CO_2 release to the atmosphere. The process of extracting metal ore for instance, from an open pit mine will typically emit high amounts of CO_2 into the atmosphere. According to the International Energy Agency (IEA), a decline of about 13% of the cumulative CO_2 emission per year is necessary to curtail global climate change by 2050. Hence, an effective solution is required in order to achieve the CO_2 emission reduction target. Various methods have been developed to reduce CO_2 emission. Among them, carbon capture and storage (CCS) by mineral carbonation process is the most reliable technique in storing CO_2 permanently in stable carbonates form [2–13]. Mining wastes have been seen as potential materials for storing CO_2 by means of the mineral carbonation. Thus, there is a need to discover potential feedstock from various types of mining wastes to enhance the mineral carbonation process in mitigating CO_2 emissions permanently in the form of mineral carbonates.

1.1. Potential of Mining Waste for Carbon Sequestration

Consumption of mining waste such as from within the mine tailings and waste dump might have potential as feedstocks for CCS in trapping CO_2 into solid carbonate and locking it out of the atmosphere. In general, the greatest carbon capture potential from mining wastes is between 1190 and 1204 million tonnes of CO_2 [6]. Feedstocks from industrial mining waste consist of: (1) alkaline earth metal-bearing silicates [14–16]; (2) hydroxide minerals [15,17,18]; and (3) silicate waste rocks that are rich in divalent cations, such as magnesium (Mg), calcium (Ca) and iron (Fe) [2,14–16,18]. For instance, mixture of mining waste and rock that is rich in calcium silicate can facilitate the production of pedogenic carbonate minerals from the formation of artificial soil [19]. Mafic and ultramafic rocks have commonly been explored in previous research due to the presence of high amount of Mg-Ca-Fe. Examples of minerals that have been investigated include olivine [$(Mg,Fe)_2SiO_4$], serpentine [$Mg_3(OH)_4(Si_3O_5)$], and wollastine ($CaSiO_3$) [20–22]. Utilization of mining wastes that are rich in Mg-Ca-Fe for passive carbon sequestration could provide a feasible preference for the reduction of CO_2 emission.

Mine tailings contain potential sources for CO_2 sequestration through the presence of organic carbon from sediments, organic material deterioration and bedrock carbon [23]. The process of mine-tailing conversion to carbonate minerals is known as carbon fixation that involves precipitation of Mg/Ca carbonates and silica, dissolution of Mg/Ca silicates, and dissolution of CO_2 into rainwater [5–7,23,24]. For instance, Mg in the form of magnesium silicates from mine tailings have the potential to bind CO_2 molecules through a mineral carbonation process that encourages reduction of CO_2 concentration in the atmosphere [25–27]. Therefore, excessive amounts of Mg-Ca-Fe and the presence of silicate minerals in mining waste may provide substantial feedstock for CO_2 sequestration by the mineral carbonation process.

Reduction in CO_2 emission requires mechanisms such as rocks and soils that have the ability to remove CO_2 from the atmosphere in the long run [4,20–23]. It is known that rocks and minerals play a significant role as passive agents for reducing CO_2 emission through carbon sequestration [5,23,24]. Several minerals such as wollastonite ($CaSiO_3$), portlandite [$Ca(OH)_2$], and gypsum ($CaSO_4 \cdot 2H_2O$) are present in some urban soils that make them possible for use in carbon capture [3–8]. Role of soils in storing organic carbon into carbonate form, become a potential for carbon sequestration. Soils can sequester CO_2 in three conditions: (1) soils signifying a stable carbon sink because it acts as a host to precipitates of pedogenic carbonate minerals; (2) soils are the site of rock weathering that encourage dissolution of inorganic carbon in the soil mixture [3–6]; and (3) urban soils contain carbonate minerals for carbonation of artificial silicates that are derived from photosynthesis by plants growing on the soil [19]. In this study, the potential of soil, sludge, and sediment from mining areas were evaluated as potential feedstocks to store carbon via the mineral carbonation process.

1.2. The Mineral Carbonation Process

Mineral carbonation is an approach to estimate carbon capture potential of mining waste in carbonates form to sequester CO_2 for the long term. The process of mineral carbonation appears as the result of silicate mineral weathering [4] and also mimic natural chemical conversions of CO_2 from non-carbonate minerals to form stable carbonate minerals [28]. For instance, weathering of silicate minerals occurs in the presence of CO_2 and potential divalent cations such as magnesium (Mg^{2+}), calcium (Ca^{2+}), and iron (Fe^{2+}) to form carbonates as shown in Equation (1) [2,4,9,29,30]:

$$(Mg,Ca,Fe)SiO_3(s) + CO_2(g) \rightarrow (Mg,Ca,Fe)CO_3(s) + SiO_2(s) + H_2O(l) \quad (1)$$

From the mineral carbonation reaction, CO_2 is being eliminated from the atmosphere and stored as solid carbonate mineral [4]. Therefore, the process of mineral carbonation is essentially relevant to be applied in reducing anthropogenic CO_2 emissions.

Nevertheless, the main challenge is to obtain the potential divalent cation due to fast rock/mineral weathering under tropical climate, like in Malaysia. Silicate minerals containing Mg-Ca-Fe are usually favorable because they are abundantly present in several types of alkaline mining wastes [2,29]. Previous research indicated that mineral carbonation mainly occurs in natural silicate minerals, such as olivine, serpentine, limestone, and wollastonite due to high amount of Mg and Ca content [20–22,31]. Besides, natural silicate rocks such as basalt and granite, play a major part to sequester CO_2 into carbonate form. This can be proven when the reaction occurs between CO_2 and silicate rocks containing Ca and Mg to form carbonate, such as limestone.

CO_2 mineralization using natural minerals and industrial waste can enhance carbonation reaction due to the presence of reactive components such as Ca and Mg oxides that can react with CO_2 to form carbonates [16,32]. Natural minerals (e.g., olivine, serpentine, and wollastine) and industrial waste (e.g., steelmaking slag, bottom and fly ashes, and municipal solid waste incinerator) are suitable feedstocks to accelerate mineral carbonation [14,16,32]. Thus, utilization of natural minerals and industrial residue through the mineral carbonation process can help mitigate CO_2 emission.

In order to estimate the potential of carbon sequestration from mining waste, several factors need to be considered, such as mineralogy and chemical composition of the rock or soil. It is important to identify the amount of magnesium oxide (MgO), calcium oxide (CaO), and iron oxide (FeO) available in the sample to reveal how much magnesite ($MgCO_3$), calcite ($CaCO_3$), and magnetite ($FeCO_3$) can be formed through the mineral carbonation process [33]. The presence of silicate minerals that are rich in Mg, Ca, and Fe are typically selected for mineral carbonation based on the mineralogical composition of the mining wastes.

Mineral carbonation is a permanent and safe method to store CO_2 in stable form and provides several advantages that can be utilized on a large scale [2,14,17,34]. Apparently, natural silicate minerals are found worldwide in various types of industrial wastes that can be used for mineral carbonation to store CO_2 [2,15,33]. Therefore, mineral carbonation is among the most promising alternative methods in reducing CO_2 emission to the atmosphere.

1.3. Factors Affecting The Mineral Carbonation Process

In the mineral carbonation process, the formation of carbonates is highly favored in specific considerations. It is important to identify the factors that can enhance mineral carbonation to sequester more CO_2. Several parameters such as temperature [35], pH [3,20,21,36] and particle-size fraction [23,36] can affect carbonation rate to sequester more CO_2 in mine waste. Temperature is one of the considerations influencing carbonation rate and enhances silicate mineral dissolution rate. Rising carbonate temperature can increase the potential of CO_2 uptake because high temperature will encourage evaporation for reaction of mineral carbonates [34,36]. pH is also important for the carbonation reaction of mining residue. Higher pH values are highly preferred to promote dissolution of carbonates [35]. As pH increases, the proportion of carbonate in solution also increases due to carbonate precipitation that is generally favored in alkaline form [35]. Carbonation potential also depends on particle-size fraction. Finer grain size particles promote higher carbonation compared to coarser particles, which is due to higher surface area that could enhance the development of carbonate mineral precipitates [23,36]. Thus, temperature, pH and particle-size of mining wastes are the key parameters that need to be considered to speed up the mineral carbonation process.

Numerous studies have discovered the potential of mining and mineral processing waste for the mineral carbonation reaction, such as chrysotile mine tailings [21,23,26,27,34,36,37], nickel mine tailings [20,21,30,37], red mud (bauxite) [38–41], and asbestos [21,35,42]. However, less research has explored the potential of gold mining wastes that contain a variety of host rock and minerals that can enhance carbon sequestration process. The current research focuses on the role of soil, sludge, and sediment from gold mining wastes as raw materials for the mineral carbonation process. Thus, the objectives of this study were to: (1) characterize the mineral phases and chemical composition of the mining wastes; and (2) identify the availability of natural silicate minerals to be used as feedstock for the mineral carbonation process.

2. Materials and Methods

2.1. Site Description

Selinsing Gold Mine, which is located at Felda Sungai Koyan, Pahang (coordinate: N 4°15′0″, E 101°47′10″) was chosen to represent a gold mine area in Peninsular Malaysia [43,44]. It is a 135 m deep open pit mine, occupying about 776 ha. The mine is situated in the Central Gold Belt of the Malay Peninsula. This gold belt varied in length and in width from 10 km to 20 km, extending along the entire backbone of the peninsula and into Thailand, Cambodia and Laos to the north [45]. The gold belt is characterized by a sequence of felsic tuff and argillite mixed with carbonaceous shale, limestone and quartzite [46]. Other rock types include phyllite, siltstone, cataclasite, mylonite and argillite [44], sandstone, shale, tuffaceous siltstone and tuffaceous conglomerate [45] and serpentinized mafic to ultramafic rocks. The major minerals found in the gold mine were pyrite (FeS_2), quartz (SiO_2) and dolomite [$CaMg(CO_3)_2$] [44]. Individual quartz veins within the gold belts varied from a few cm to 30 m in width and contain other ore minerals, such as free gold, arsenopyrite (FeAsS), chalcopyrite ($CuFeS_2$), sphalerite [(Zn,Fe)S], pyrrhotite (Fe_7S_8), galena (PbS), stibnite, and graphite [44].

2.2. Field Sampling

Field sampling was performed, in which four types of gold mine wastes were collected from six sampling points within Selinsing Gold Mine including the mine tailings, waste dump, stockpiles [super lower grade (SLG), lower grade (LG), high grade (HG)], and borrow pit (Figure 1). The sampling strategy used was based on previous geological information and lithology of mine rock at the various sampling points as shown in Table 1. The information was obtained from Selinsing Gold Mine technical report [46]. The stratification was based on different types of mining waste. Samples were collected at equal surface area, which was about 15 cm depth as a representative sample from each mining waste to reduce grouping and segregation error [47,48]. A total weight of 13.57–17.37 kg of soil samples and 2.20–4.60 kg of sludge and sediment samples were collected at each sampling point and the composite samples were obtained. Descriptions of sampling points at Selinsing Gold Mine are shown in Table 1.

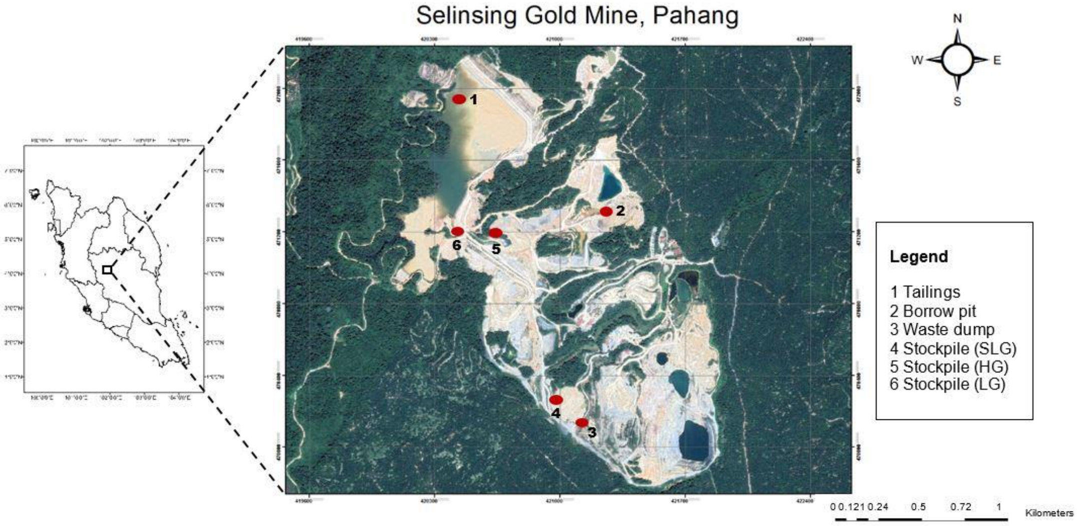


Figure 1. Locations of six sampling points at Selinsing Gold Mine, Pahang, Malaysia using geographic information system (GIS).

Table 1. Descriptions of the sampling points at Selinsing Gold Mine.

Sampling Point	Type of Sample	Total Sample Weight	Characteristic
Tailings	Sludge sediment	2.20 kg 4.60 kg	Waste from treatment plant.
Stockpile High Grade (HG) Lower Grade (LG) Super Lower Grade (SLG)	Soil	13.57 kg	Phyllite, conglomerate. Phyllite, shale. Tuffaceous, shale.
Waste Dump	Soil	17.37 kg	Sedimentary rock, arginite, volcanic.
Borrow Pit	Soil	15.37 kg	Highly silicate clay, argillite, kaolinite, serinite, medium to fine size, highly oxidize.

2.3. Particle-Size Distribution Analysis

The pH of the soil, sludge, and sediment samples were determined using a pH meter at the ratio of 1:2.5 (soil to water), where the amount of raw sample of soil, sludge, and sediment used were 30 g and 75 mL of distilled water. Soil testing was conducted based on the British Standard (BS) 1377, Part 3: 1990, Section 9 [49].

Prior to the analysis, all soil samples were air-dried at room temperature for 24 h, while the sediment and sludge sample were oven-dried at 70 °C for 24 h. Then, the samples were ground into powder form using agate mortar and pestle and later were sieved to 2 mm size fraction using particle sieves to eliminate excess coarse particles and to homogenize the mixture for mineralogical analysis.

Size distribution of the individual particles such as sand, silt and clay fraction in the soil, sludge, and sediment samples were determined by mechanical analysis using pipette method which is regarded as the standard method for particle-size analysis, and is more precise compared to sieve analysis [50,51]. In this method, the suspension of samples were pipetted together with aliquot at 10 cm depth. The clay fraction was kept for X-ray diffraction (XRD) analysis. Calculations of clay weight (Equation (2)), total weight of sand particles (Equation (3)) and silt (Equation (4)) are shown below [50]:

$$\%\ \text{clay} = [(A \times 1000 \div \text{volume of pipet}) - \text{C}] \times (100 \div \text{weight of soil}) \tag{2}$$

$$\%\ \text{sand} = B \times (100 \div \text{weight of soil}) \tag{3}$$

$$\%\ \text{silt} = 100 - \%\ \text{sand} - \%\ \text{clay} \tag{4}$$

A is the weight of clay particles, B is the total weight of all sand fractions, and C is the mean weight of the three replicates. The texture of the soil, sludge and sediment was determined based on the % of the total weight of clay, sand and silt particles, using the software that has been developed and published by Teh and Rashid (2003) for computing the texture chart of the United States Department of Agriculture (USDA). The software was downloaded at http://www.agri.upm.edu.my/~chris/tal [52].

2.4. Mineralogical Analysis

The clay fraction of the soil, sludge and sediment was X-rayed as oriented sample without treatment [50]. The clay was analyzed using X-ray diffractometer (XRD) instrument (model Bruker-AXS D8 Advanced, USA) at 1°/min rate (0.025° step size) and counting for 0.2 s per step over the 5–50° scattering angle range. The range of detection limit was between 1 and 2%. Integrated intensity of peak areas were determined from the single peak function using Diffrac AT EVA software (v.9.0) and its application is important for clay minerals compared to peak height because it depends on crystallinity and deficiencies [53]. The d-spacing was identified using OriginPro 8 software (Originlab Corporation, Northampton, UK).

2.5. Morphological and Chemical Analysis

Surface morphological structure of the minerals of interest in all samples of the mining wastes was observed under scanning electron microscopy (SEM) at the magnification of 2, 5, 10 and 50 µm. The SEM instrument (model Phillips XL30, Amsterdam, the Netherlands). was equipped with energy dispersive X-ray (EDX) to quantify (in %) the elemental composition of the soil, sludge, and sediment used in the study. The values obtained for Mg, Ca, Si, Fe, Al and K were then converted in terms of its oxide equivalence, with detection limits of 0.1%.

3. Results

3.1. pH and Particle-Size Distribution of the Soil, Sludge and Sediment

In general, the pH of the soil at the stockpile, waste dump, and borrow pit in the Selinsing Gold Mine were very acidic, with values ranging from 2.2 to 3.3 (averaging 2.8) (Figure 2). On the other hand, the pH of the sludge and sediment were alkaline in nature, with the values of 7.9 and 8.3, respectively. It is believed that pH plays a significant role in the carbonation reaction of the mining wastes. Therefore, several pH values can be used as a manipulated variable in the mineral carbonation process to measure significant pH values that can accelerate the carbonation reaction.

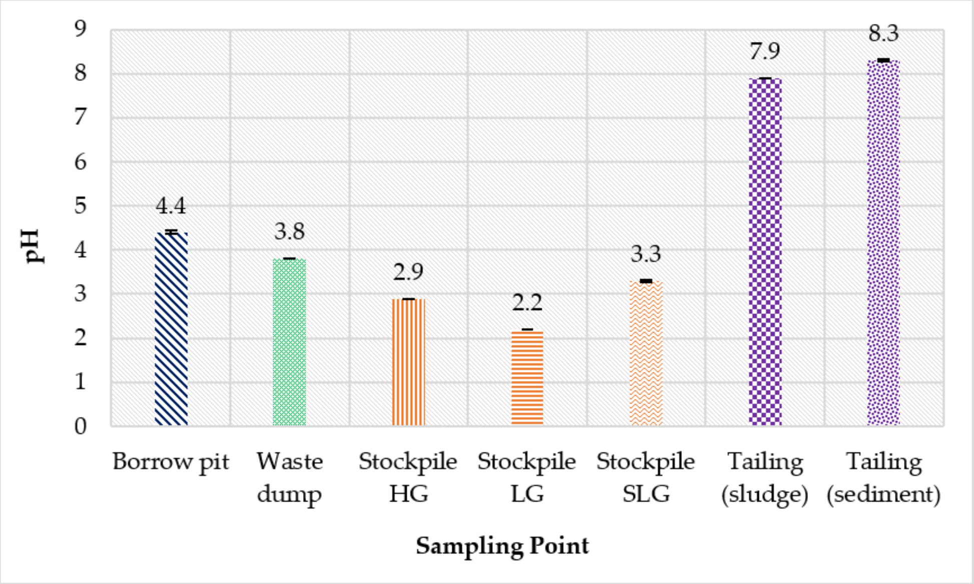


Figure 2. pH of soil, sludge and sediment samples at different types of gold mining waste. Black bars indicate standard deviation from three replicates.

Results of the particle-size analysis showed that the textures of the gold mining wastes were mostly silt loam, while the borrow pit and stockpile LG were silt and sandy loam, respectively (Table 2). In silt loam texture, the highest silt content was found in sludge (78.23%) of mine tailings, followed by waste dump (68.06%), stockpile HG (63.98%), stockpile SLG (60.65%), and sediment (53.21%) of mine tailings. It is noted that both clay (<2 µm) and silt (2–50 µm) were present in fine-size particles which is favorable for the mineral carbonation process. It is believed that particle-size fraction can affect the mineral carbonation process of the gold mining wastes. Therefore, different particle-size fractions of gold mining waste such as using finer and coarser-size particle are required to compare the rate of carbonation reaction.

Table 2. Particle size distribution of the soil, sludge and sediment at Selinsing Gold Mine waste.

	Particle Size Distribution (%)			Soil Texture Class
Sampling Point	**Clay**	**Silt**	**Sand**	**(USDA)**
	(<2 µm)	**(2–50 µm)**	**(>50 µm)**	
Borrow pit	7.81	91.05	1.19	Silt
Waste dump	11.16	68.06	20.66	Silt loam
Stockpile HG	10.80	63.98	25.17	Silt loam
Stockpile LG	19.17	18.08	62.77	Sandy loam
Stockpile SLG	7.25	60.65	32.01	Silt loam
Tailings (sludge)	17.43	78.23 *	4.28	Silt loam
Tailings (sediment)	6.96	53.21	39.74	Silt loam

* Highest percentage among silt loam.

3.2. Mineralogy and Chemical Composition of Soil, Sludge and Sediment

The mineralogical composition of the clay fraction of the soil, sludge and sediment from the different types of mining wastes at Selinsing Gold Mine is summarized in Table 3. The crystalline phases were mainly comprised magnesium silicates (sepiolite, chlorite-serpentine) and iron silicates minerals (illite, stilpnomelane). The major minerals present in the soil at the waste dump, stockpile and borrow pit were illite $(K,H_3O)(Al,Mg,Fe)_2(Si,Al)_4O_{10}[(OH)_2,(H_2O)]$ and kaolinite $[Al_2Si_2O_5(OH)_4]$, with minor amount of quartz (SiO_2) (Figure 3a). The presence of kaolinite in the soil at the stockpile (Figure 4a–c), waste dump (Figure 4d) and borrow pit (Figure 4e) was evidenced by the hexagonal shape crystals observed in the SEM micrographs, respectively. This is consistent with the previous results that kaolinite was present in waste dump of mining areas [54]. Our study also found some aerinite $[(Fe^{+2},Fe^{+3},Al)_3Mg_3(Ca,Na)_4(Si_{13.5}Al_{4.5}O_{42})(OH)_6)\cdot 12H_2O]$ in the waste dump. Additionally, both stilpnomelane $[Fe_2(Si_3O_9)]$ and sepiolite $[Mg_4Si_6O_{15}(OH)_2\cdot 6H_2O]$ were present in the stockpile under investigation (Table 3).

The most dominant mineral in the clay fraction of the sludge and sediment was illite, followed by chlorite-serpentine $[(Mg,Al)_6(Si,Al)_4O_{10}(OH)_8]$ and quartz (SiO_2) (Figure 3b). The presence of chlorite-serpentine in the tailings was depicted by their tabular shape as seen in the SEM micrographs (Figure 4f—in sludge; Figure 4g—in sediment).

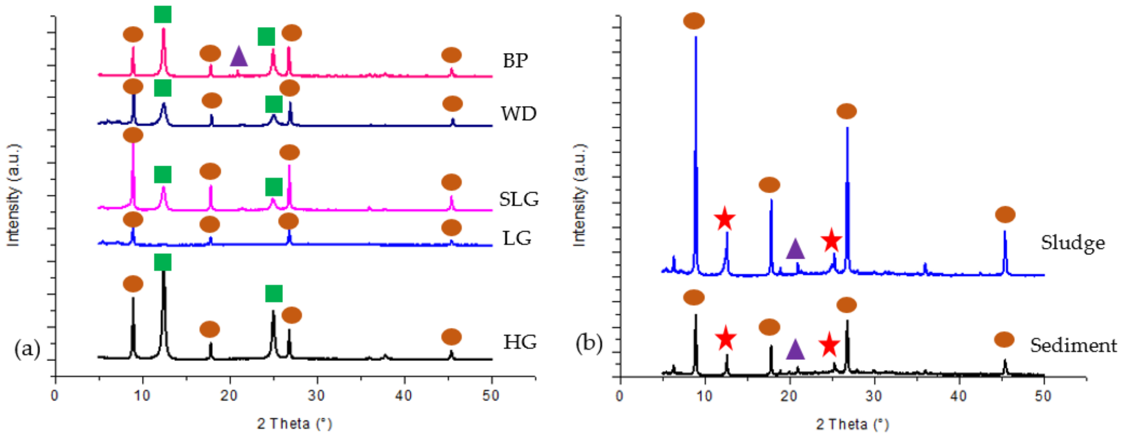
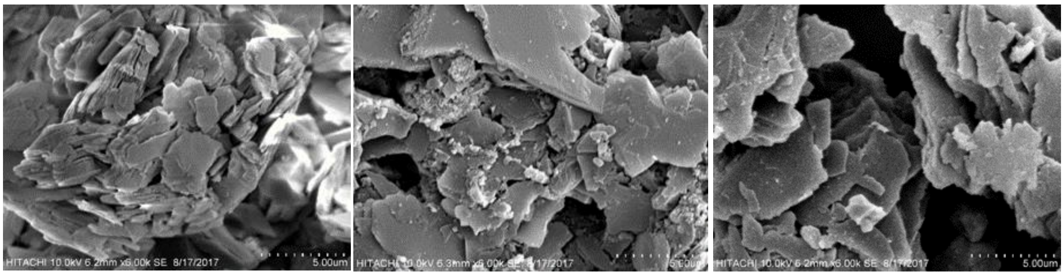


Figure 3. X-ray diffractograms of the soil clay fraction from the: (**a**) stockpile (HG, LG, SLG), waste dump (WD), borrow pit (BP); and (**b**) sludge and sediment from mine tailings. The added symbols signify the peaks for all types of mining wastes: illite (orange circle), kaolinite (green square), chlorite-serpentine (red star) and quartz (purple triangle).

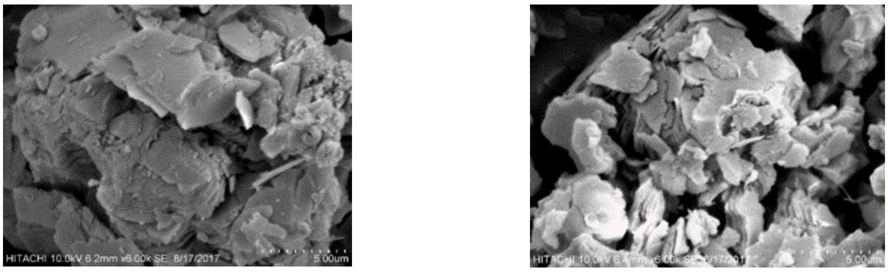
Table 3. Summary of mineral composition of clay fraction in soil, sludge and sediment samples of the waste dump, stockpile, mine tailings, and borrow pit. Symbol of (√) is referring to the presence of minerals in sampling points.

Minerals	Sampling Point			
	Waste Dump	**Stockpile**	**Tailings**	**Borrow Pit**
1. Quartz SiO_2	√	√	√	√
2. Kaolinite $Al_2Si_2O_5(OH)_4$	√	√	√	√
3. Chlorite-serpentine [a] $(Mg,Al)_6(Si,Al)_4O_{10}(OH)_8$			√	
4. Illite [b] $(K,H_3O)(Al,Mg,Fe)_2(Si,Al)_4O_{10}[(OH)_2,(H_2O)]$	√	√	√	√
5. Aerinite [b] $[(Fe^{+2},Fe^{+3},Al)_3Mg_3(Ca,Na)_4(Si_{13.5}Al_{4.5}O_{42})(OH)_6]\cdot 12H_2O$	√			
6. Stilpnomelane [b] $Fe_2Si_3O_9$		√		
7. Sepiolite [a] $Mg_4Si_6O_{15}(OH)_2\cdot 6H_2O$		√		

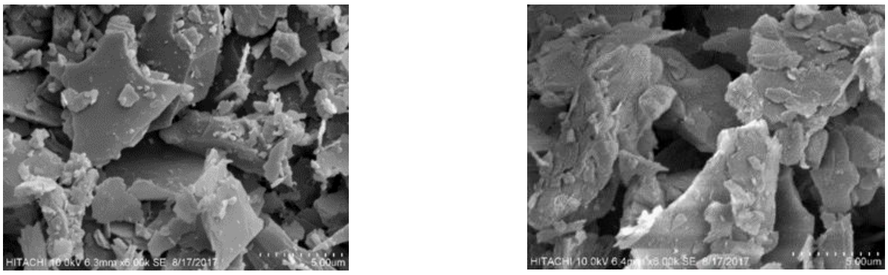
[a] Mg-silicates mineral; [b] Fe-silicates mineral.



(**a**) stockpile (HG) (**b**) stockpile (LG) (**c**) stockpile (SLG)



(**d**) waste dump (**e**) borrow pit



(**f**) sludge (**g**) sediment

Figure 4. Scanning electron micrographs of soil samples at stockpile (**a**) HG, (**b**) LG, (**c**) SLG, (**d**) waste dump, (**e**) borrow pit; and (**f**) sludge and (**g**) sediment from mine tailings at magnification of 5 μm.

The chemical composition of the gold mine wastes determined by SEM-EDX and presented in the form of MgO, CaO, SiO_2, Fe_2O_3, Al_2O_3 and K_2O are given in Table 4. Quartz was one of the dominant minerals in the soil, sludge, and sediment at the gold mine. The highest percentage of SiO_2 was found in borrow pit (75.26%), followed by stockpile SLG (67.90%), stockpile HG (65.61%), sediment (64.70%) and sludge (60.39%) at mine tailings, waste dump (59.53%), and stockpile LG (48.46%). It was also found that soil, sludge, and sediment of mining waste contain major minerals such as illite and kaolinite (Table 3). This was explained by the highest percentage of Al_2O_3 which is coming from kaolinite found in soil at the waste dump (29.93%), followed by stockpile HG (25.84%), stockpile SLG (22.82%), stockpile LG (20.18%), and mine tailings of sludge (18.22%) and sediment (14.60%). Whereas, the percentage of K_2O is coming from illite that was highly present in soil at stockpile LG (7.24%), followed by sludge (7.06%) and sediment (6.63%) at mine tailings, waste dump (6.36%), stockpile SLG (5.72%), borrow pit (4.11%), and stockpile HG (3%).

Findings from chemical analysis also indicated that 2.72% of MgO in soil of stockpile LG and 1.74% in sludge of tailings is the potential divalent cation for the mineral carbonation process to occur (Table 4). This can be supported by the carbonation reaction of MgO in the presence of CO_2 as the following reaction Equation (5) [2,55]:

$$MgO + CO_2 \rightarrow MgCO_3 \text{ (Delta Hr = 118 kJ/mol)} \quad (5)$$

The total 4.46% of MgO was explained by the presence of sepiolite and chlorite-serpentine in soil of stockpile and tailings respectively based on the XRD results (Table 3). Although the percentage was very small, both chlorite-serpentine and sepiolite are the potential silicate minerals for the mineral carbonation process. Ferric oxide (Fe_2O_3) is the potential silicate mineral that is favorable for the mineral carbonation process [29]. The highest percentage of Fe_2O_3 was found in stockpile LG (11.79%), followed by waste dump (3.6%), sludge (3.2%) of mine tailings, stockpile SLG (3.15%), and sediment (3.04%) of mine tailings (Table 4). The total 24.78% of Fe_2O_3 was explained by the presence of stilpnomelane in stockpile, illite in all types of mining wastes, and aerinite at waste dump based on the XRD results (Table 3). Thus, stockpile and tailings of Selinsing Gold Mine have great potential for carbon sequestration due to the presence of silicate minerals (chlorite-serpentine, sepiolite) for the mineral carbonation process to store CO_2 in stable carbonate.

Table 4. Chemical composition of soil, sludge and sediment of the mining waste at Selinsing Gold Mine.

Major Oxide (%)	Sampling Point						
	Stockpile HG	**Stockpile LG**	**Stockpile SLG**	**Waste Dump**	**Borrow Pit**	**Tailings (Sludge)**	**Tailings (Sediment)**
MgO *	-	2.72 *	-	-	-	1.74 *	-
SiO_2	65.61	48.46	67.90	59.53	75.26	60.39	64.70
Fe_2O_3 *	-	11.79 *	3.15 *	3.60 *	-	3.20 *	3.04 *
Al_2O_3	25.84	20.18	22.82	29.93	19.25	18.22	14.60
K_2O	3.00	7.24	5.72	6.36	4.11	7.06	6.63
SO_3	2.53	8.83	-	-	-	1.37	-

* Divalent cation that can influence the mineral carbonation process.

4. Discussion

4.1. Properties of Soil, Sludge, and Sediment for Mineral Carbonation

Alkaline condition can promote the dissolution of minerals into carbonate form in accelerating mineral carbonation [27,35]. During the mineral carbonation process, weak bases such as ammonium hydroxide (NH_4OH) can be added to increase the pH to produce more precipitation [28]. As pH increases, the proportion of carbonate in the solution increases which is due to carbonate precipitation that is generally favored under alkaline condition [3,28,35]. From the carbonation reaction, three solid products such as silica-rich solid, Mg and Ca carbonate were produced at high pH [28]. Therefore, dissolution of minerals in sludge and sediment at high pH conditions is preferable to achieve a higher CO_2 conversion into carbonate form during the carbonation reaction. Other research indicated that carbonation precipitation was enhanced at the pH of 8–10 [21,35], with the optimum pH of 10 required for aqueous mineral carbonation [28,31]. Thus, more stable carbonate can be formed to sequester more CO_2 in the long run. Furthermore, different types of host rocks can affect the rate of carbonate dissolution at various pH values. In contrast, the dissolution reactions in magnesium silicate minerals (e.g., basalt and serpentine) are highly favorable under acidic condition [34,35]. Since the host rocks in Selinsing Gold Mine originated from sedimentary as well as mafic to ultramafic rocks, mineral dissolution rates are facilitated further under alkaline condition. Therefore, we believe that the sludge and sediment at the gold mine tailings, which were alkaline in nature, have a great potential for the mineral carbonation process.

Silt fraction was present in small-size particles that have large surface area to enhance the development of carbonate precipitates from the mineral carbonation process [23,36]. Small size particles are very reactive with CO_2 that can promote mineral carbonation [29]. A previous study had proven that a decrease in particle size to less than 30 μm would enlarge surface area, leading to increased carbonation conversion [33]. Although silt loam contained a low amount of clay content, it still had the potential for mineral carbonation because of the presence of the high amounts of small-size silt particles. Silty soils with small amounts of clay are able to absorb carbon due to the presence of large a surface area [56]. Thus, particle-size distribution in the materials is an important factor in the carbonation process. This points to the fact that sludge at the gold mine tailings has a lot of small-size particles and is a suitable candidate for mineral carbon sequestration.

Results for mineralogical composition indicated that both illite and kaolinite minerals were found widespread in the soils of Selinsing Gold Mine. These minerals are produced from the chemical weathering of alumino-silicates existing under tropical environment [50]. The various rock types containing some alumino-silicates found in the study area were phyllite, conglomerate, shale, tuffs, and carbonate [44,45]. Therefore, a high percentage of Al_2O_3 and SiO_2 had proven the presence of clay minerals such as illite and kaolinite in the gold mining wastes.

4.2. Availability of Silicate Mineral for Mineral Carbonation

Sepiolite present in the clay fraction of the soil in the stockpile (Table 3, Figure 3a), chlorite-serpentine in the sludge and sediment of mine tailings (Table 3, Figure 3b, respectively), and stilpnomelane in stockpile (Table 3) have the potential of being involved in the mineral carbonation process [5,6]. Mineral carbonation is an exothermic reaction, where the products of silicate mineral weathering occur when CO_2 reacts with divalent cations (Mg-Ca-Fe) to form geologically stable metal carbonates [23,36]. This study showed that both chlorite-serpentine (Equation (6)) and sepiolite (Equation (7)) were the silicate minerals containing divalent Mg that reacted with CO_2 to form carbonate [5,6], whereas stilpnomelane (Equation (8)) was the potential silicate mineral that contains divalent cation of Fe to react with CO_2 to form carbonate as shown in the following reactions [29]:

Chlorite-serpentine

$$(Mg,Al)_6(Si,Al)_4O_{10}(OH)_8 + 6CO_2 \rightarrow 6(Mg,Al)CO_3 + 4SiO_2 + 4H_2O \quad (6)$$

Sepiolite

$$Mg_4Si_6O_{15}(OH)_2{\cdot}6H_2O + 6CO_2 \rightarrow 4MgCO_3 + 6H_4SiO_4 + 7H_2O \quad (7)$$

Stilpnomelane

$$Fe_2Si_3O_9 + 2CO_2 \rightarrow 2FeCO_3 + 3SiO_2 \quad (8)$$

The presence of Mg-silicate minerals in the stockpile and mine tailings suggest that CO_2 can be sequestered into magnesium carbonate ($MgCO_3$). In support of this finding, it was believed that all the MgO within the waste rock or soil was capable of sequestering CO_2 to form magnesite [31]. In the context of carbon sequestration, Mg and Ca-rich minerals are required to sequester more CO_2 [21,23]. Therefore, this study has proven the availability of chlorite-serpentine and sepiolite in gold mining waste that can be utilized as a feedstock for the mineral carbonation process.

Iron (Fe) is one of the potential elements that can form stable carbonate through the mineral carbonation process. The presence of Fe-silicate minerals in the stockpile of the gold mine indicated that CO_2 can be sequestered into iron carbonate ($FeCO_3$) as shown in Equation (8) [29]. Therefore, the presence of Mg-silicate minerals (e.g., chlorite-serpentine and sepiolite) and Fe-silicate mineral (e.g., stilpnomelane) in the gold mining wastes can be utilized as feedstocks to increase the mineral carbonation process.

5. Conclusions

This study has explored the potential for the mineral carbonation processes in soil, sludge, and sediment from gold mining waste to sequester more CO_2 for permanent CO_2 sequestration. Selinsing Gold mining wastes in Pahang have a great potential for passive carbon sequestration due to the alkaline nature of sludge and sediment at the gold mine tailings, and large amounts of small-size particles (silt fraction) in the soil, sludge, and sediment. Sludge at the gold mine tailings having a lot of small-size particles is more suitable for mineral carbon sequestration. Furthermore, the process of carbonation is enhanced by magnesium silicate minerals (chlorite-serpentine and sepiolite) and iron silicate mineral (stilpnomelane) present in the materials, which can be used as a carbonation feedstock. Both MgO and Fe_2O_3 are the potential divalent cations that can encourage the formation of magnesium carbonate and iron carbonate, respectively, through the mineral carbonation process. Overall, soil, sludge, and sediment at the gold mining wastes can be used as a feedstock in storing CO_2 permanently by the mineral carbonation process. Although mining wastes have low valuable minerals for industrial purposes, large volume of waste produced from mining operations can be utilized to reduce CO_2 emission through mineral carbonation. The potential of physical and chemical properties of gold mining wastes for the mineral carbonation process specifies long term CO_2 storage, which can be reflected as an environmentally and economically sustainable approach in the future. Further evaluation using laboratory scale experiment on potential of soil, sludge, and sediment of gold mine especially in tailings and stockpile are required to speed up the carbonation reaction to sequester more CO_2.

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