

Adsorption of Nickel and Zinc by Residual Soils

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Abstract: Soil has long been utilized as low cost liner material to prevent contamination from leachate to groundwater media. To find a suitable soil material for this purpose is a great challenge. This study describes the potential use of residual soil to functions as engineered clay liner for waste disposal landfill in Malaysia. Three types of residual soils were investigated namely marine clays (SBMC1, SBMC2), Residual Granites (BGR, KGR) and residual meta-sediments (BBMS1, BBMS2 and PMS). Physical and chemical tests were applied for both granitic soils to determine the physical and chemical properties of soil materials. Physical and chemical tests involved grain size distribution, Atterberg limits, compaction, pH, organic content, specific gravity, Cation Exchange Capacity (CEC) and Specific Surface Area (SSA) as well as Batch Equilibrium Test for adsorption of heavy metals. The best potential soil materials for clay liner is the materials that have high pH value, high organic matter, high liquid and plastics limits, high CEC and SSA values. The best material also highly dominated with clay (in this case PKMC, SBMC1 and SBMC 2). Result show the range of pH values are from 6.95-8.36, range of organic content are from 4.35-6.41%, the specific conductivity values range from 2.13-2.34 and for liquid limit and plastic limit range are from 56.40-84 and 26.86-59.35% respectively; which is high to very high plasticity. Residual soils as low-cost adsorbent materials were also used for removal of Nickel (Ni) and Zinc (Zn) from aqueous solutions. Batch test was used and the effect of heavy metal concentration was studied. Results were analyzed using adsorption isotherm models (i.e., Linear, Langmuir and Freundlich). Based on the correlation coefficient (r^2 values), most of residual soils fitted nicely to Linear, Langmuir and Freundlich models. For Ni, most soils fitted to Langmuir models except for meta-sediment while for Zn fitted to Linear model. Marine clay has the highest adsorption coefficient ranged between $K_L = 0.2380-0.9655 \text{ L kg}^{-1}$ followed by granite and meta-sediment $K_L = 0.0031-0.0168 \text{ L kg}^{-1}$ and $K_L = 0.0016-0.0075 \text{ L kg}^{-1}$ respectively. While for Zn, marine clay also has the best adsorption coefficient ranged between $K_d = 0.0453-0.1249 \text{ L kg}^{-1}$, followed by granite and meta-sediment ranged between $K_d = 0.0027-0.0028 \text{ L kg}^{-1}$ and $K_d = 0.0012-0.0016 \text{ L kg}^{-1}$. The selectivity sequence K_L for Ni is $\text{SBMC2} > \text{SBMC1} > \text{PKMC} > \text{BGR} > \text{PMS} > \text{KGR} > \text{BBMS2} > \text{BBMS1}$ while for Zn, the selectivity sequence of K_d is $\text{SBMC2} > \text{SBMC1} > \text{PKMC} > \text{BBMS1} > \text{BBMS2} > \text{PMS} > \text{BGR} > \text{KGR}$. The study concludes that marine clay is the best material for landfill clay liner due to suitable physical-chemical characteristics and also appeared to be the best natural adsorbent of Ni and Zn of metal concentration in solution.

Keywords: Residual Soils, Heavy Metals, Physical-Chemicals Properties, Adsorption Isotherm

Introduction

Solid wastes are part of environmental pollution's contributor. Landfill; which is an engineered waste

disposal site with specific pollution control is designed to minimize pollution. However, Malaysia's landfills are still using the oldest and very common way of disposing which is open dumping due to the increase of pollutions.

Its occur when solid phase (waste), liquid phase (leachate) and gas phase form due to biological, physical, chemical and other influent factors of waste (Sabahi *et al.*, 2009). Leachate can percolate through the base of landfill, enter the groundwater systems and change the quality of groundwater (Taha *et al.*, 2011). Engineered clay liners are utilized to Malaysia sanitary landfill by using the compacted clay to prevent the seepage and remove heavy metals commonly found in landfill leachate. Many metals ions are essentials as trace elements but in higher concentration it's become toxic. Heavy metals are difficult to remove from environment unlike other pollutant that chemically or biologically degraded (Komy *et al.*, 2013). Therefore, conventional clay liners are design with the focus on minimizes the permeation of leachate through liner. Residual soils are chosen to be clay liner as abundance and availability of this residual soil will make them economically feasible (Rosli *et al.*, 2008). Soil structures and the various size fractions have their own ability to absorb and transfer to the environment (Huang *et al.*, 2014). The capacity of natural soil to adsorb heavy metals has been study and published by many researchers (Antoniadis *et al.*, 2007; Zuhairi *et al.*, 2008). Mobility of heavy metals in soils can be describe by distribution coefficient (Kd) where Kd is one of the input parameters in contaminant transport modeling to model the contaminant flow in groundwater (Zuhairi and Samsudin, 2007). Kd parameter shows the absorption or attenuation of contaminant. The objectives of this study are to investigate the attenuation capability of natural geologic material based on chemical-physical and adsorption properties and proposed the best materials that function as engineered clay liner for landfill application.

Material and Methods

Material

In the present study, eight samples were collected from Selangor area in Malaysia and they consist of three types of residual soils which were; granitic residual soils from Broga (BGR) and Kajang (KGR), meta-sediment residual soils from Puchong (PMS) and two samples from Bukit Badong (BBMS 1 and BBMS 2) and marine clay from Port Klang (PKMC) and two samples from Sungai Besar (SBMC 1 and SBMC 2). All samples were air-dried and sieved into 0.125 mm and 63 μm for soil tested.

Physical-Chemical Characteristics

Physical-chemical tests were conducted according to British Standard Method, BS1377 (1990) and Geotechnical Research Centre, Montreal, Canada (1985). Physical properties test consist of particle size distribution, Atterberg Limits, specific gravity and compaction while chemical properties test consist of soil pH, organic matter, Specific Surface Area (SSA) and Cation Exchange Capacity (CEC).

Sorption Test

Samples were tested using Batch equilibrium test where this test provides a quick method of estimating the contaminant retention capacity of any liner material (Zuhairi and Samsudin, 2007). This test used to study the behavior of natural soil (adsorbent) through a different initial concentration factor.

Preparation

After collecting soil samples, samples were air-dried and passed through 63 μm sieve. To prepare the Ni and Zn metal, stock solutions were made using their nitrates and were further diluted with 10 different concentration 20, 40, 60, 80, 100, 150, 200, 250, 300 and 400 mg L^{-1} under room temperature and controlled with pH 5-6 by adding HNO_3 and NaOH . Nitrate salts were chosen because nitrate has poor ability to complex with metallic cations (Zuhairi *et al.*, 2008). To perform this test, 4 g of soil sample (adsorbent) with 40 mL of metal aqueous solutions (1:10 ratio soil/solution) were added in centrifuge tubes. Samples were shaken 100 RPM under room temperature ($27\pm 1^\circ\text{C}$) for 24 h for attained the equilibrium. After shaking, samples were centrifuged at 1500 RPM for 10 minutes and were filtered using 45 μm nitrocellulose membranes. The pH of this solution also measured using pH meter (Hanna Instrument 2211). Solutions were analyzed using Couple Plasma Mass Spectroscopy (Perkin Elmer Model OPTIMA 3000 with auto sampler).

Data Evaluation

Adsorption isotherms are mathematical models where they describe the distribution of adsorbate among liquid and solid phases, based on assumptions that are related to the heterogeneity, homogeneity of solid surface, the type of coverage and the possibility of interaction between the adsorbate species (Babazadeh *et al.*, 2011). The equilibrium adsorption isotherm is importance in the design of adsorption system (Parida *et al.*, 2012).

Adsorption Isotherm

Adsorption data were analyzed using Linear, Langmuir and Freundlich isotherm models. The concentration of heavy metals adsorbed by soils, q_e can be calculated by using following formula Equation 1:

$$q_e = \frac{(C_o - C_f)V}{W} \quad (1)$$

Where:

C_o and C_f = Initial concentration and equilibrium concentration of metal solution respectively (mg/L)
 V = Volume of solution added (mL)
 W = Mass of air-dried soil (g)

Langmuir isotherm model is valid for mono-layer adsorption onto a surface with a finite number of identical sites and also considered several assumptions like the localized adsorption, similar energies on the surface of active sites where there is no interaction between adsorbed molecules and the surface of heterogeneous catalytic reactions (Parida *et al.*, 2012; Tang *et al.*, 2012). Langmuir models represented in following equations (Galedar and Younesi, 2013) Equation 2:

$$q_e = \frac{K_L A_m C_e}{1 + K_L C_e} \quad (2)$$

The linear equation for Langmuir model given as below Equation 3:

$$C_e / q_e = 1 / K_L A_m + C_e / A_m \quad (3)$$

Where:

KL = Langmuir binding constant (L/mg)

Am = Saturated adsorption amount of metal ions (mg/g)

Ce = Equilibrium concentration of metal ions (mg/L)

Freundlich isotherm model is an empirical equation assuming that the adsorption process takes place on heterogeneous surface (Ruffino and Zanetti, 2009; Li *et al.*, 2010; Sivarama Krishna *et al.*, 2012; Tang *et al.*, 2012). It also used to describe the adsorption of organic and inorganic components in solution (Parida *et al.*, 2012) and represent an ideal situation, which does not include the possible saturation of the sorption sites (Sastre *et al.*, 2007). The non-linear Equation 4 and linear Equation 5 for Freundlich models shows as below:

$$q_e = K_f C_e^{1/n} \quad (4)$$

$$\text{Log} q_e = \text{log} K_f + (1/n) \text{log} C_e \quad (5)$$

Where:

KF = Freundlich sorption coefficient

1/n = Freundlich sorption exponent

Ce = The metal concentration in final contact concentration (mg/L)

Results

Soil Characterization

The characteristics of each soil are tabulated in Table 1 and 2. Based on particle size distribution granitic residual soils have high percentage of sand ranged from 46-63%, followed by meta-sediment residual soil which have ranged between 5-46% of sand while marine clay has the least of sand percentage ranged between 1-46%.

Meta-sediment has the highest percentage of clay and plasticity index compared to marine clay ranged from 12-39 and 32.448-42.90% respectively. Specific gravity also will confirm the types of grain sizes. Specific gravity result shows granitic soils give high values ranged from 2.45-2.66 followed by meta-sediment (2.32-2.67) and marine clay (2.13-2.34). Table 2 also shows pH and organic matter for each soil. The selective sequence from highest pH is PKMC > SBMC2 > SBMC1 > BGR > KGR > BBMS2 > BBMS1 > PM S where marine clay has highest pH values ranging from 6.95-8.36, followed by granitic soils (pH 5.32-5.85) and meta-sediment soils (pH 4.21-5.46). Marine clay has the highest organic matter from 4.35-6.41%, specific surface area (49.18-77.26%) and cation exchange capacity (75.4-115.06 meq/100 g).

Heavy metals adsorption capacities can be expressed in relationship between distribution coefficients, metal removed from solution per gram of the soil samples (mg/g) and the equilibrium of metals concentration in solutions (mg/L). These sorption isotherms provide the sorption behavior of soil suspension when they are at equilibrium with metal solutions (Wang and Nan, 2009). Table 3 and 4 show the summary of three main isotherm parameters that have been applied for Ni and Zn adsorption.

Table 1. Physical characteristics of adsorbent/residual soils

| Physical characteristics | BGR | KGR | PMS | BBMS 1 | BBMS 2 | PKMC | SBMC1 | SBMC2 |
|--------------------------------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|
| Particle size distribution: | | | | | | | | |
| Sand (%) | 54-63 | 46-54 | 40-46 | 10-22 | 5-13 | 19-46 | 1-8 | 7-16 |
| Silt (%) | 32-42 | 41-50 | 21-32 | 62-72 | 75-79 | 46-67 | 73-87 | 66-76 |
| Clay (%) | 1-6 | 2-6 | 25-39 | 14-22 | 12-16 | 8-20 | 12-19 | 15-22 |
| Atterberg Limit: | | | | | | | | |
| Plastic Limit (%) | 38.01-38.61 | 35.03-36.67 | 35.31-37.12 | 52.98-59.68 | 46.10-52.41 | 26.86-31.00 | 49.18-59.35 | 46.09-49.16 |
| Liquid Limit (%) | 48.50-50.00 | 50.60-51.60 | 65.00-69.60 | 91.60-94.20 | 84.50-89.00 | 56.40-59.40 | 79.50-84.00 | 73.50-79.00 |
| Plasticity Index (%) | 9.90-11.99 | 14.63-16.27 | 29.69-32.48 | 34.12-40.82 | 33.98-42.90 | 28.00-30.11 | 20.15-34.22 | 24.34-30.91 |
| Specific Gravity | 2.50-2.59 | 2.45-2.66 | 2.63-2.67 | 2.32-2.50 | 2.37-2.48 | 2.13-2.34 | 2.14-2.29 | 2.23-2.34 |
| Max Dry Density (g/cm ³) | 1.75 | 1.71 | 1.42 | 1.37 | 1.50 | 1.63 | 1.37 | 1.39 |

Table 2. Chemical Characteristics of adsorbent/residual soils

| Chemical characteristics | BGR | KGR | PMS | BBMS 1 | BBMS 2 | PKMC | SBMC1 | SBMC2 |
|--------------------------|-------------|-------------|-------------|-------------|-------------|---------------|-------------|-------------|
| pH | 5.35-5.85 | 5.32-5.54 | 4.21-4.38 | 4.80-5.46 | 4.54-4.92 | 7.94-8.36 | 6.95-7.42 | 7.50-7.67 |
| Organic Matter (%) | 0.22-0.34 | 0.39-0.50 | 0.45-0.54 | 1.42-2.07 | 0.84-1.23 | 4.35-5.14 | 5.31-6.06 | 5.08-6.41 |
| SSA (m ² /g) | 17.96-21.93 | 25.76-26.83 | 32.53-36.81 | 30.74-39.35 | 35.59-40.13 | 49.18-49.99 | 60.28-62.38 | 67.09-77.26 |
| CEC (meq/100g) | 0.79- 1.35 | 1.31- 1.35 | 0.55- 0.96 | 1.64- 1.93 | 2.81- 3.07 | 113.87-115.06 | 91.25-92.32 | 75.40-77.90 |

SSA: Specific surface area; CEC: Cation exchange capacity

Table 3. Adsorption isotherm's result on Nickel (Ni)

| Metals | Soils | Linear equation | | Langmuir equation | | | Freundlich equation | | |
|--------|-------|-----------------|--------|-------------------|--------|--------|---------------------|--------|--------|
| | | K_d | R^2 | K_L | A_m | R^2 | K_F | n | R^2 |
| Ni | BGR | 0.0049 | 0.4324 | 0.0168 | 0.3146 | 0.8847 | 0.7791 | 4.2827 | 0.7087 |
| | KGR | 0.0021 | 0.3452 | 0.0031 | 0.4304 | 0.9803 | 0.6957 | 4.7170 | 0.9684 |
| | PMS | 0.0047 | 0.9397 | 0.0075 | 0.3077 | 0.8265 | 0.6295 | 3.7397 | 0.7916 |
| | BBMS1 | 0.0017 | 0.7252 | 0.0012 | 0.1875 | 0.6705 | 0.6449 | 5.2301 | 0.5536 |
| | BBMS2 | 0.0015 | 0.6808 | 0.0016 | 0.5991 | 0.4667 | 0.4069 | 3.1706 | 0.8217 |
| | PKMC | 0.0269 | 0.5323 | 0.2380 | 1.4562 | 0.9967 | 0.4733 | 0.7508 | 0.9727 |
| | SBMC1 | 0.0667 | 0.5750 | 0.4471 | 1.8861 | 0.9962 | 0.5555 | 0.6063 | 0.9922 |
| | SBMC2 | 0.1436 | 0.5230 | 0.9655 | 1.9406 | 0.9283 | 1.3161 | 0.5424 | 0.9579 |

Table 4. Adsorption isotherm's result on Zinc (Zn)

| Metals | Soils | Linear equation | | Langmuir equation | | | Freundlich equation | | |
|--------|-------|-----------------|--------|-------------------|--------|--------|---------------------|--------|--------|
| | | K_d | R^2 | K_L | A_m | R^2 | K_F | n | R^2 |
| Zn | BGR | 0.0016 | 0.8270 | 0.0059 | 0.6507 | 0.7056 | 27.4789 | 1.9912 | 0.7899 |
| | KGR | 0.0012 | 0.3160 | 0.0025 | 0.2248 | 0.8087 | 9.5521 | 1.1818 | 0.9251 |
| | PMS | 0.0027 | 0.8539 | 0.0056 | 0.5715 | 0.9077 | 28.3270 | 2.0153 | 0.9038 |
| | BBMS1 | 0.0028 | 0.8310 | 0.0031 | 1.1892 | 0.2387 | 158.7084 | 4.0633 | 0.4189 |
| | BBMS2 | 0.0028 | 0.7598 | 0.0027 | 0.4398 | 0.6498 | 17.6116 | 1.4486 | 0.9054 |
| | PKMC | 0.0453 | 0.8295 | 0.2116 | 2.8265 | 0.1450 | 5.0957 | 2.2153 | 0.3884 |
| | SBMC1 | 0.0711 | 0.8317 | 0.6128 | 4.4964 | 0.5107 | 4.7984 | 2.8345 | 0.3967 |
| | SBMC2 | 0.1249 | 0.8133 | 0.6899 | 1.8646 | 0.8030 | 2.3529 | 1.8748 | 0.6699 |

Higher R^2 indicates good agreements with that isotherm. For Ni, the experimental results it fitted to Langmuir and Freundlich isotherms model ($R^2 > 0.7$) except for meta-sediment while for Zn, most soils fitted to Linear equation except for granite residual soil. Correlation between Langmuir and Freundlich models gives better explanations on the influence of soil characteristics (Sastre *et al.*, 2007). This is why authors attached the pH value of solutions after adsorption test. The pH of initial concentration is controlled at pH 5-6.

Discussion

Based on physical-chemical characteristics of residual soils, grain size plays important role in adsorption capacity. Granitic residual soil have highest percentage of sand where coarse-grained soils exhibit lower tendency for adsorption of heavy metal compared to fine-grained soil (Liew and Zuhairi, 2003). Meta-sediment also has the highest percentage of clay and plasticity index which indicates higher adsorption capability compared to marine clay (Zulfahmi *et al.*, 2011). However, marine clay has the smallest value of specific gravity (1.37-1.39%) where this would confirm that marine clay have higher fine particle (Chalermyanont *et al.*, 2009). Fine-grained soils (i.e., PKMC, SBMC1 and SBMC2) have a large surface activity and large surface area such as clay minerals, humid acids and others which enhance adsorption capacity (Liew and Zuhairi, 2003). The surface area of clay will increase the contact area and facilitate

adsorption of positively charged ions. So, clay easily accommodate heavy metals on its active sites through ion exchange and complexation (Lukman *et al.*, 2013). High SSA also allows strong physical and chemical interactions with fluids and dissolved species, which subjected to electrostatic attraction, sorption or specific cation exchange reactions (Oztoprak and Pisirici, 2011). Higher CEC and exchangeable cations (except Ca) among the mineral samples are due to salinity characteristics (Sastre *et al.*, 2007) and higher heavy metal sorption due to negative charge of clay fraction (Chalermyanont *et al.*, 2009). Marine clay has highest pH values ranging from 6.95-8.36, in alkaline region due to calcareous content for this soil. According to (Sastre *et al.*, 2007; Lukman *et al.*, 2013), alkaline pH promotes heavy metal precipitation and adsorption onto clay surfaces. Organic matter in soils also plays an importance role in the adsorption of heavy metal ions even in soils and for this study, marine clay has the highest range of organic matter.

From Table 3 and Langmuir isotherms model, the selectivity sequence for KL on Ni is SBMC2 > SBMC1 > PKMC > BGR > PMS > KGR > BBMS2 > BBMS1 while for Zn, the selectivity sequence of K_d are SBMC2 > SBMC1 > PKMC > BBMS1 > BBMS2 > PMS > BGR > KGR. Marine clay shows high value of K_d due to high clay fraction. Small particle size, with high pH (e.g., pH for PKMC = 7.94-8.36) and high organic matter (e.g., PKMC = 4.35-5.14%) can enhance heavy metals retention. Granite and meta-sediment residual soils have lowest K_d values and the lowest sorption capacity

because they have high percentage of sand fraction. This behavior also related to low percentage of surface area and low in organic matter. According to Zuhairi *et al.* (2008), the relationship between soil properties and adsorption parameters exist in a group of soils with different characteristics.

Figure 1 and 2 shows the pH value of the final solution. Marine clay samples seem to have higher pH value (alkaline) compared to the initial solution. At high pH

values (above point of zero change), surface of marine clay has higher negative charge which results in higher attraction of cations (Ni^+ and Zn^{2+}). For granitic and meta-sediment residual soils, they became more acidic compared to initial solution. At low pH (below the point of zero change), the exchange sites on granite and meta-sediment particle become positive, the Ni^+ and Zn^{2+} cations compete with H^+ ions in the solution for active sites and consequently lower adsorption (Hefne *et al.*, 2010).

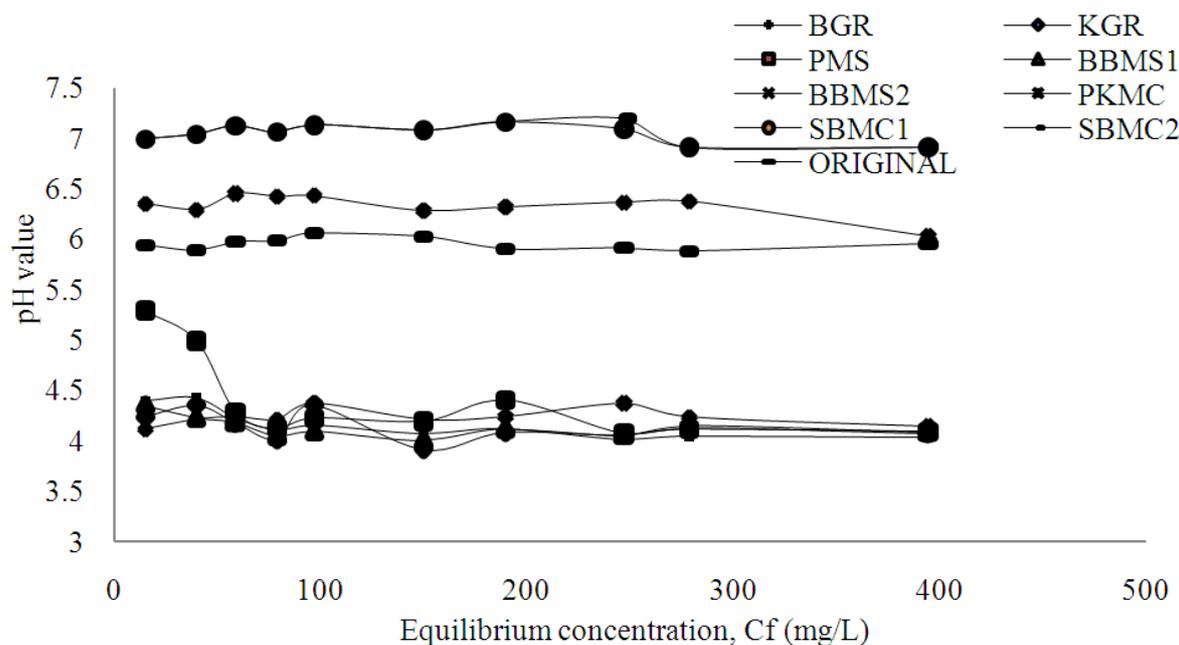


Fig. 1. pH value of final concentration of Ni

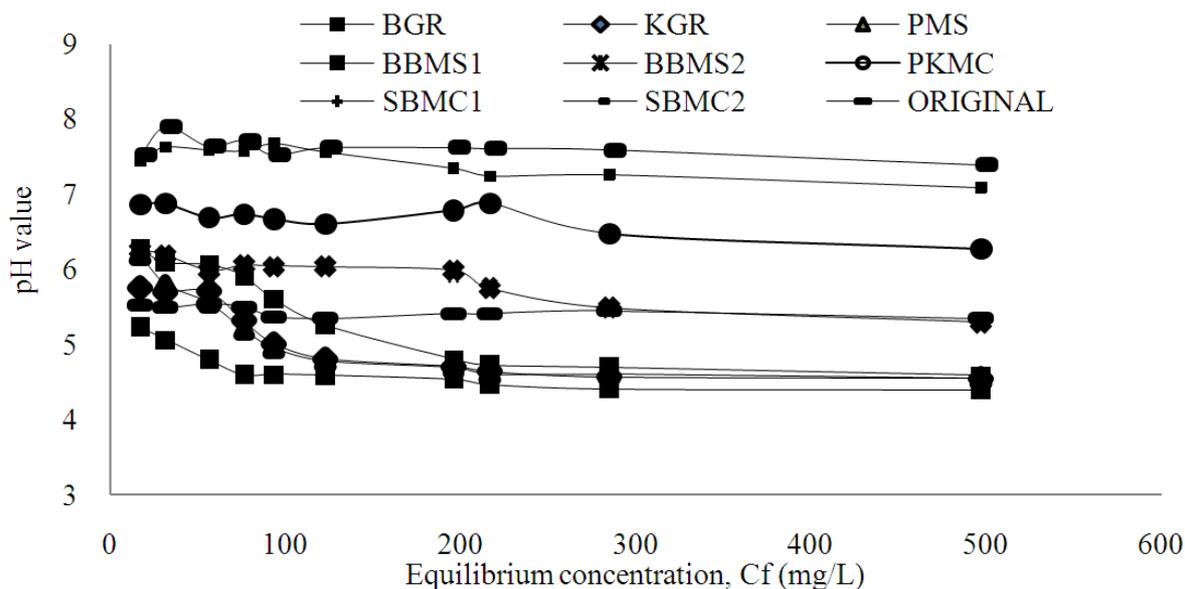


Fig. 2. pH value of final concentration of Zn

Conclusion

Adsorption of Ni and Zn from aqueous state was studied under different concentrations. The Linear, Langmuir and Freundlich isotherm models explaining modest adsorbate-adsorbent interactions. The adsorbent, marine clay appeared to be the best natural adsorbent of Ni and Zn of metal concentration in solution. The rank for residual soil to be the best clay liner are marine clay > granite > meta-sediment. Marine clay has suitable physical and chemical characteristics as we discussed above, due to higher adsorption capacity. Batch adsorption experiment was applied to study the adsorption characteristics of Ni and Zn onto residual soils. For Ni, equilibrium data were well fitted to the Langmuir isotherm and Freundlich isotherm while for Zn, equilibrium data were fitted to Linear equation. Marine clay performance was found to be better than the all other residual soils to adsorption of Ni and Zn. Marine clay having the highest specific surface area (49.18-77.26%), organic matter (4.35-6.41%), cation exchange capacity (75.4-115.06 meq/100 g). The study concludes that marine clay is the best material for landfill clay liner.

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Author's Contributions

Nur 'Aishah Zarime: Conducting the experimental work and drafting this paper,

W.Y. Wan Zuhairi: Is the main supervisor of the study and revise the article and

Sivarama Krishna: Who also revise this article.

Ethics

This article is original and contains unpublished materials. The corresponding author confirms that all of the other authors have read and approved the manuscript and no ethical issues involved.

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